Supplement for

"Markedly different impacts of primary emissions and secondary aerosols formations on aerosol mixing states revealed by simultaneous measurements of CCNC, V/HTDMA and SP2"

Table S1. The relationship among MAF, NF_H, NF_V and NF_{noBC} at different particle size quantified by the correlation coefficient (r), the normalized mean bias (NMB) and the regression parameters including the slope and the intercept. MAF at SSs of 0.08%, 0.14% and 0.22% were used in comparison for particle size of 200 nm, 150 nm and 100 nm, respectively. 1-NF_{CBC} rather than NF_{CBC} is used, as NF_{CBC} is mainly distributed the range from 0 to 0.2.

Combination	Dp	r	NMB(%)	Slope	Intercept
NF_V-NF_H	50nm	0.438	32.5	0.168	0.772
	100nm	0.484	19.3	0.250	0.670
	150nm	0.482	19.8	0.257	0.647
	200nm	0.657	20.5	0.357	0.567
MAF-NF_H	100nm	0.553	11.4	0.276	0.620
	150nm	0.446	13.3	0.270	0.610
	200nm	0.675	2.25	0.726	0.204
MAF-NF_V	100nm	0.529	-4.9	0.530	0.362
	150nm	0.636	-3.0	0.558	0.334
	200nm	0.629	-9.9	0.914	-0.01
NF_{noBC}-MAF	200nm	0.504	9.25	0.307	0.564
NF_{noBC}-NF_H	200nm	0.564	19.0	0.307	0.591
NF_{noBC}-NF_V	200nm	0.658	-1.9	0.671	0.248
	250nm	0.597	-3.8	0.462	0.405
	300nm	0.706	-2.1	0.525	0.347

	350nm/335nm	0.346	3.21	0.220	0.604

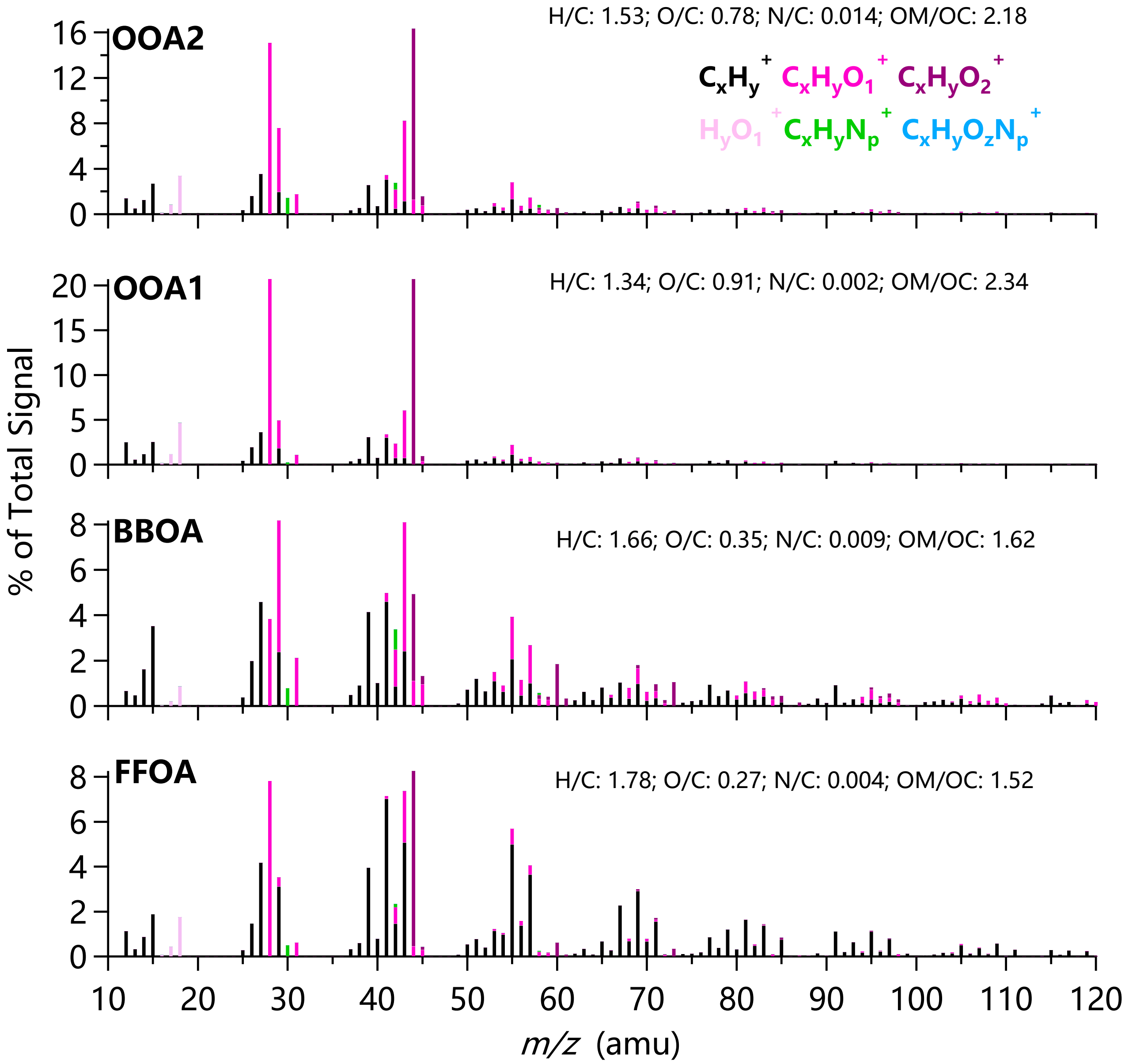

Fig. S1. The mass spectra of OA factors.

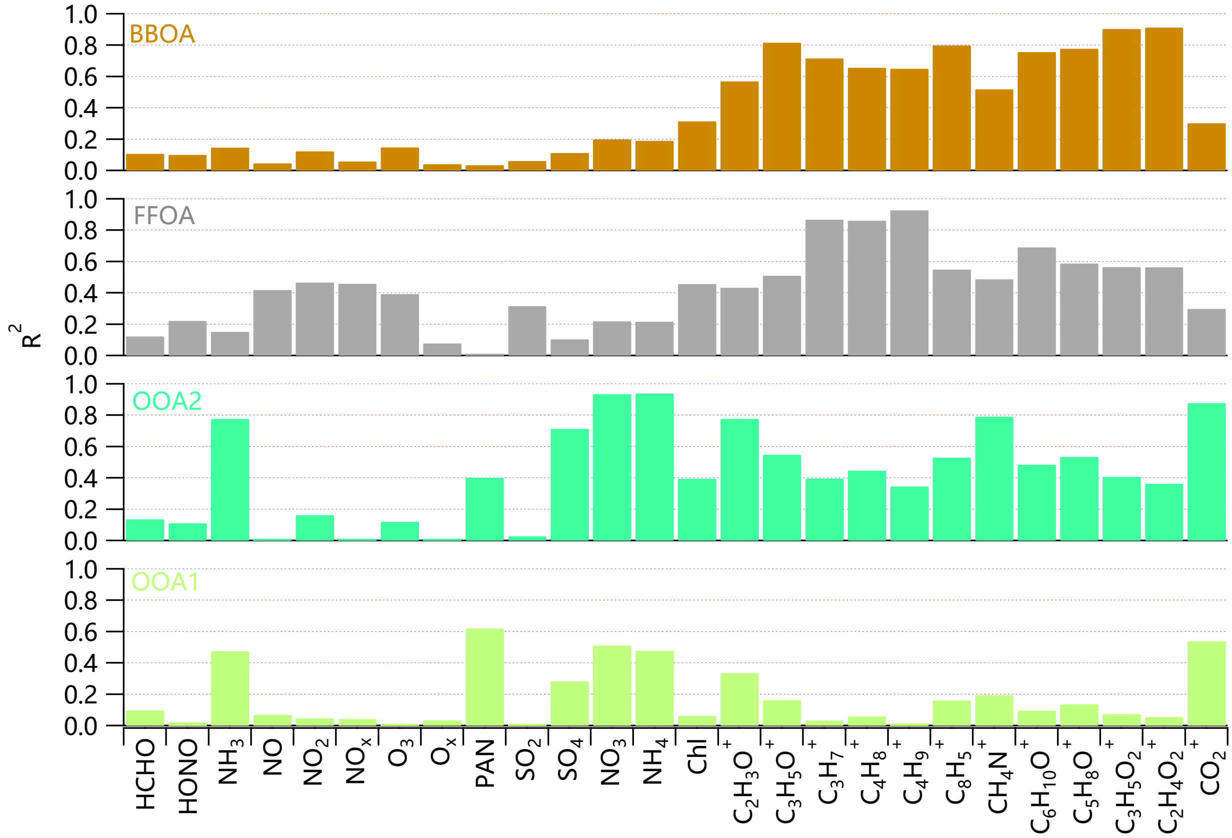

Fig. S2. The correlations between OA factors and external species.

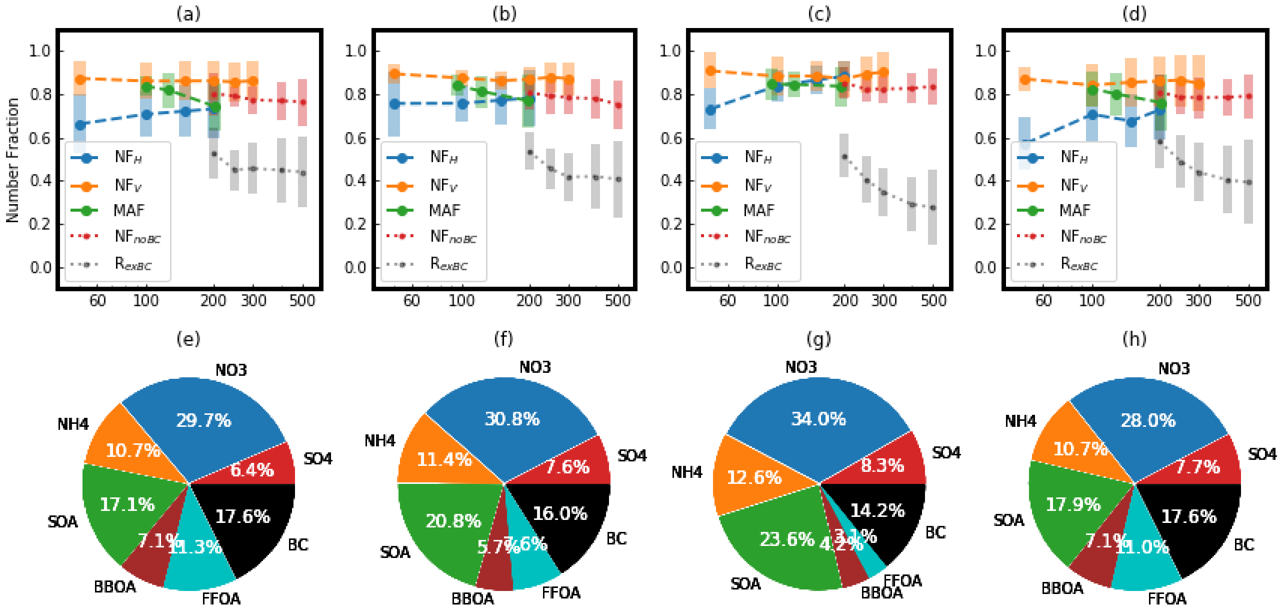

Fig. S3. (i, k, m and o): Average size-dependence of hygroscopic particles (NF$_H$, blue), volatile particles (NF$_V$, orange), CCN (MAF, green), BC free particles(NF$_{noBC}$, red) and ratio of thinly coated BC in total BC particles (R$_{exBC}$, black) during the 0-6, 6-12, 12-18 and 18-24 hours in the heavily polluted period. (j, l, n and p): Corresponding mass fraction of aerosol chemical compositions (identified by colors) during the three periods during the 0-6, 6-12, 12-18 and 18-24 hours in the heavily polluted period.

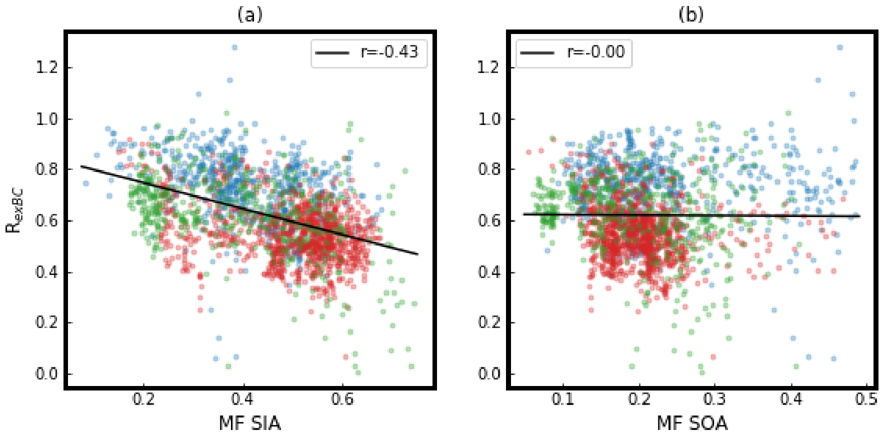

Fig. S4. The correlation between the difference among the four aerosol mixing state parameters and MF of secondary aerosol chemical composition during different periods. Moderately polluted period: Blue; Heavily polluted period: Red; Clean period: Green.

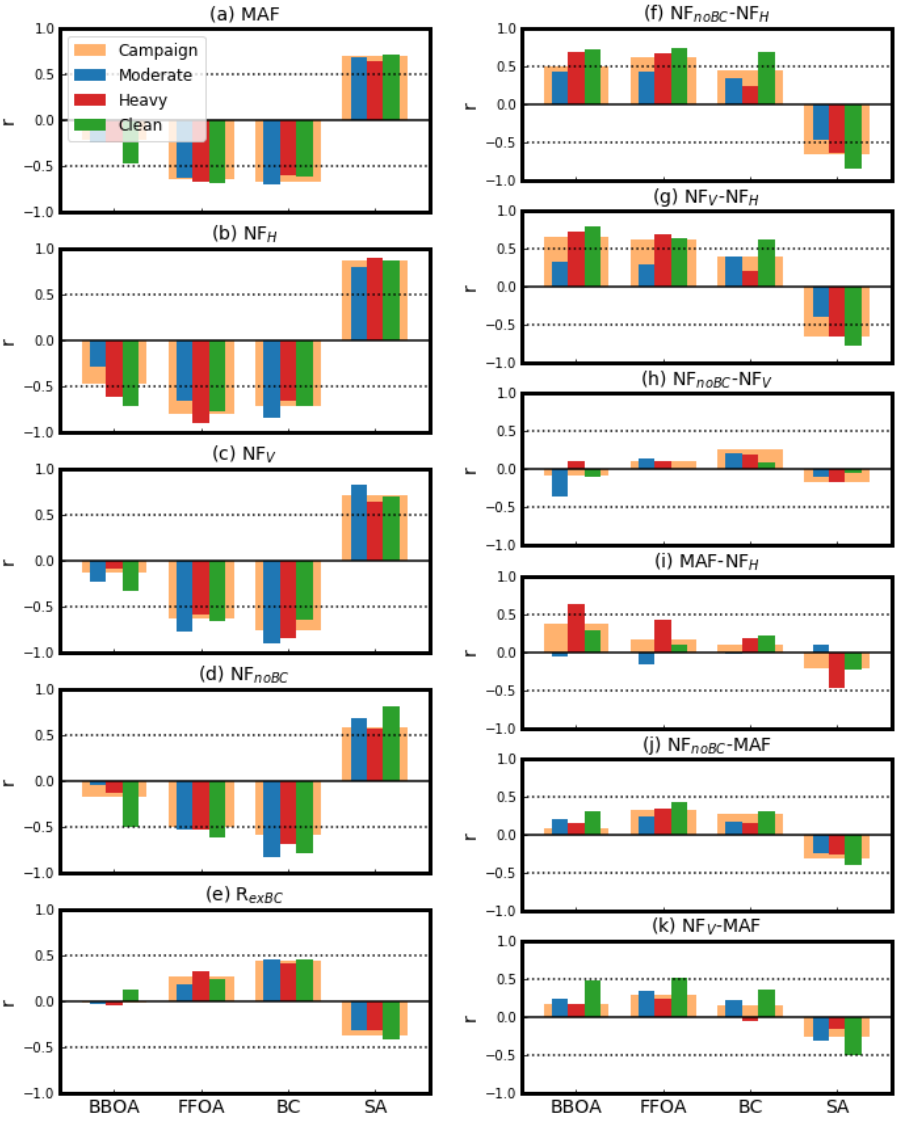

Fig. S5. (a-e) The correlation coefficient (R) between aerosol mixing state parameters and MF of primary aerosol chemical composition during different periods (f-k) The correlation coefficient (R) between the difference among the four aerosol mixing state parameters and MF of primary aerosol chemical composition during different periods. Moderately polluted period: Blue; Heavily polluted period: Red; Clean period: Green; Whole campaign: Orange.

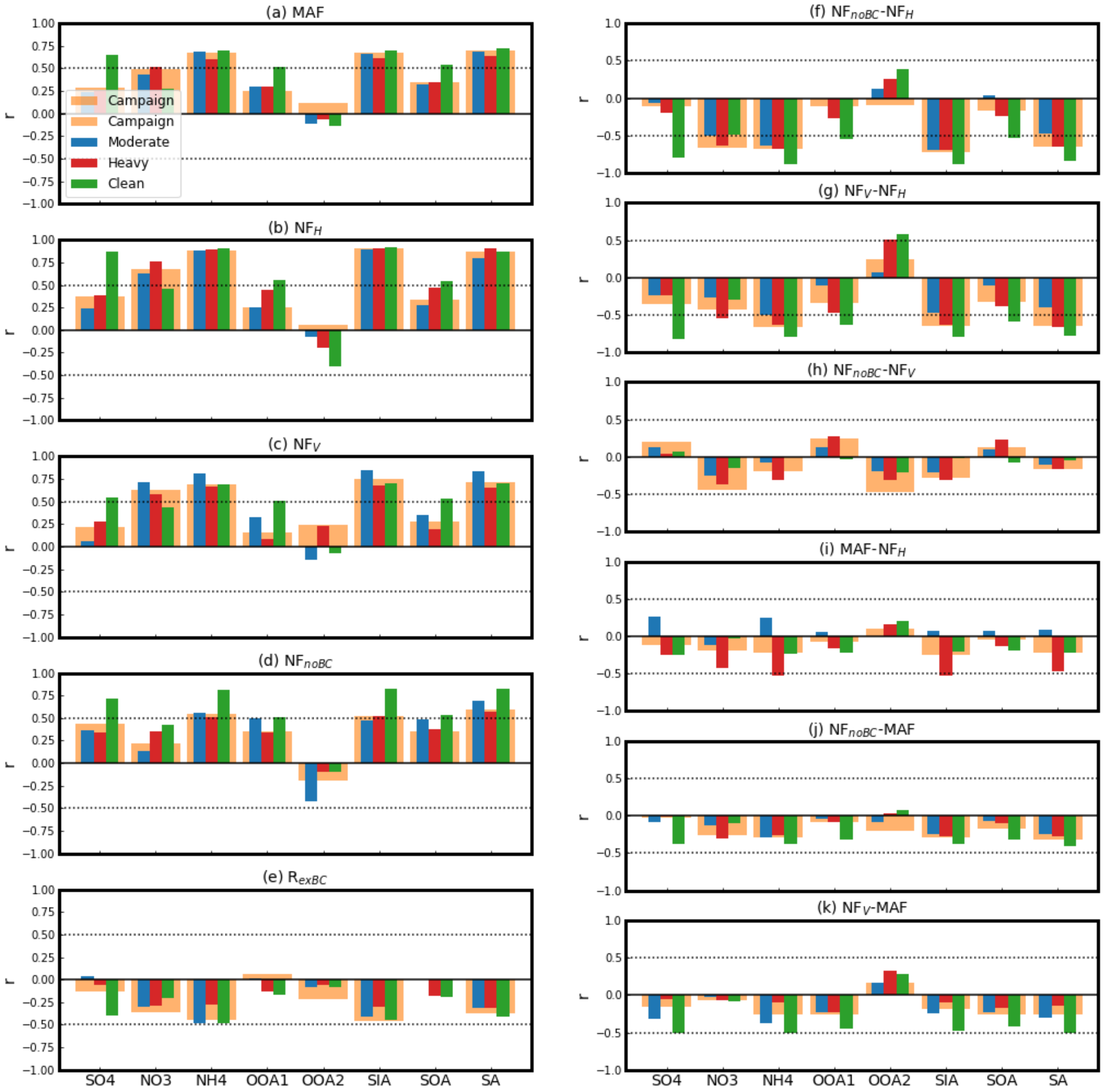

Fig. S6. (a-e) The correlation coefficient (R) between aerosol mixing state parameters during different periods (f-k) The correlation coefficient (R) between the difference among the four aerosol mixing state parameters and MF of secondary aerosol chemical composition during different periods. Moderately polluted period: Blue; Heavily polluted period: Red; Clean period: Green; Whole campaign: Orange.

1. Calibration of SP2

In this study, Aquadag soot particles was used for calibrating the measured incandescence signal of the SP2 as reported by Gysel, et al. [1]. Briefly, the soot mass is determined through the aerosol density as shown in Fig.S3a and aerosol mobility diameter determined by the DMA. The relationships between measured incandescence signal heights and black carbon mass at different diameters were shown in Fig. S3 (b) and (c). The size of rBC refers to the mass equivalent diameter for DMA selected BC-containing particles is converted from the mass of rBC by assuming that the density of rBC is 1.8 g/cm3, which is the median ρ value recommended by Bond, et al. [2].

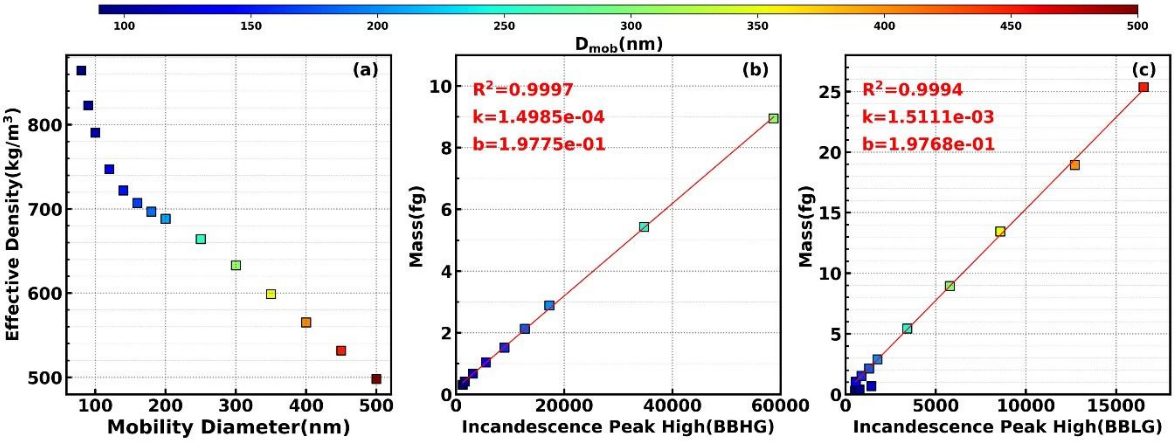

Figure S7. (a) Effective densities of Aquadag soot particles; (b) and (c) Relationships between soot mass and incandescence signals of low gain and high gain channels.

Also, multiple charge corrections were conducted for BC containing aerosols and BC free aerosols using the method reported by Zhao, et al. [3].

(1). Gysel, M.; Laborde, M.; Olfert, J. S.; Subramanian, R.; Gröhn, A. J., Effective density of Aquadag and fullerene soot black carbon reference materials used for SP2 calibration. *Atmos. Meas. Tech.* **2011,** *4*, (12), 2851-2858.
(2). Bond, T. C.; Bergstrom, R. W., Light absorption by carbonaceous particles: An investigative review. *Aerosol Science and Technology* **2006,** *40*, (1), 27-67.
(3). Zhao, G.; Tan, T.; Zhu, Y.; Hu, M.; Zhao, C., Method to quantify black carbon aerosol light absorption enhancement with a mixing state index. *Atmos. Chem. Phys.* **2021,** *21*, (23), 18055-18063.