



Supplement of

Markedly different impacts of primary emissions and secondary aerosol formation on aerosol mixing states revealed by simultaneous measurements of CCNC, H(/V)TDMA, and SP2

Jiangchuan Tao et al.

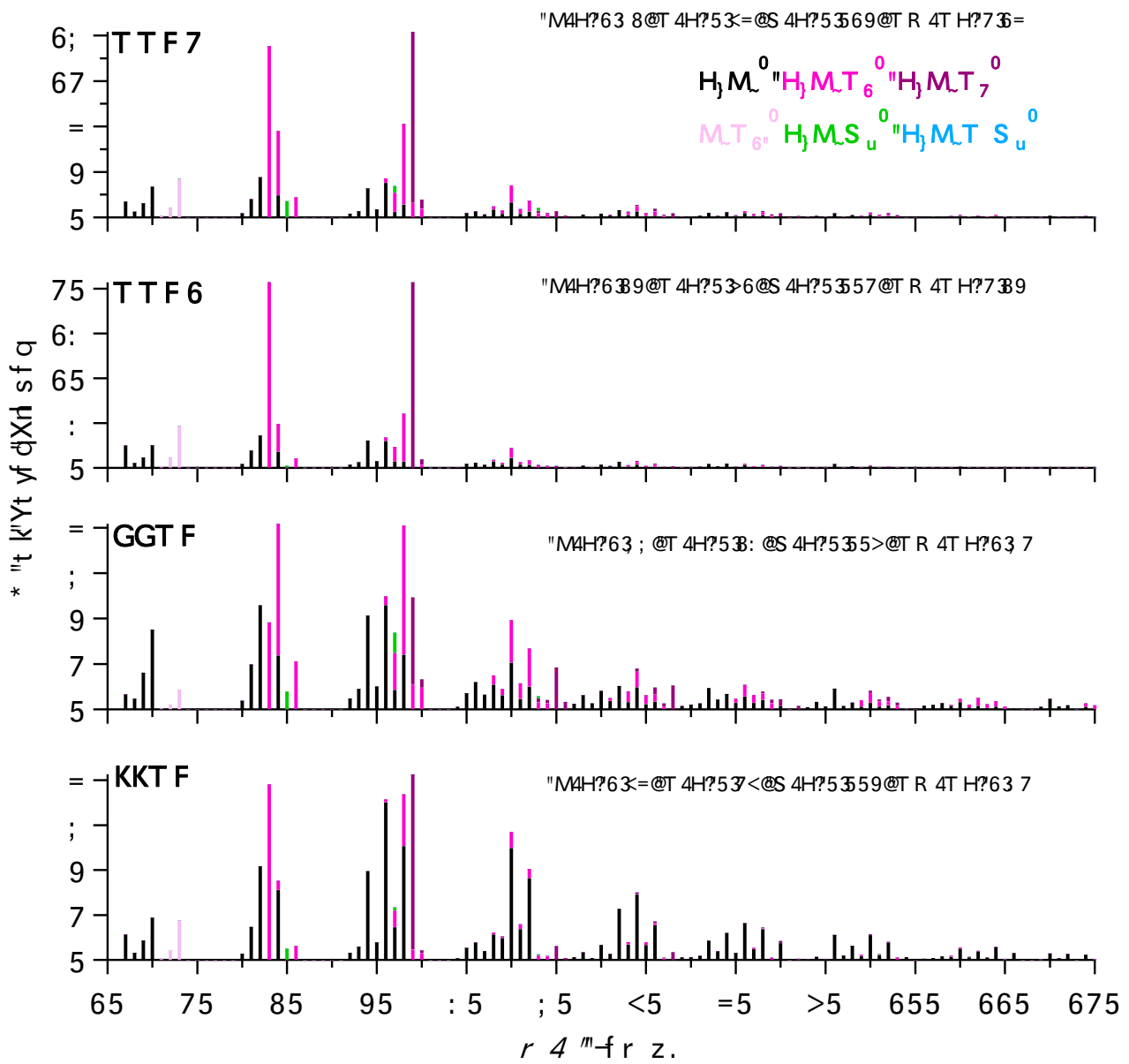
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1 Table S1. The relationship among MAF, NF_H , NF_V and NF_{noBC} at different particle size quantified by
 2 the correlation coefficient (r), the normalized mean bias (NMB) and the regression parameters
 3 including the slope and the intercept. MAF at SSs of 0.08%, 0.14% and 0.22% were used in
 4 comparison for particle size of 200 nm, 150 nm and 100 nm, respectively. $1-NF_{CBC}$ rather than NF_{CBC}
 5 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

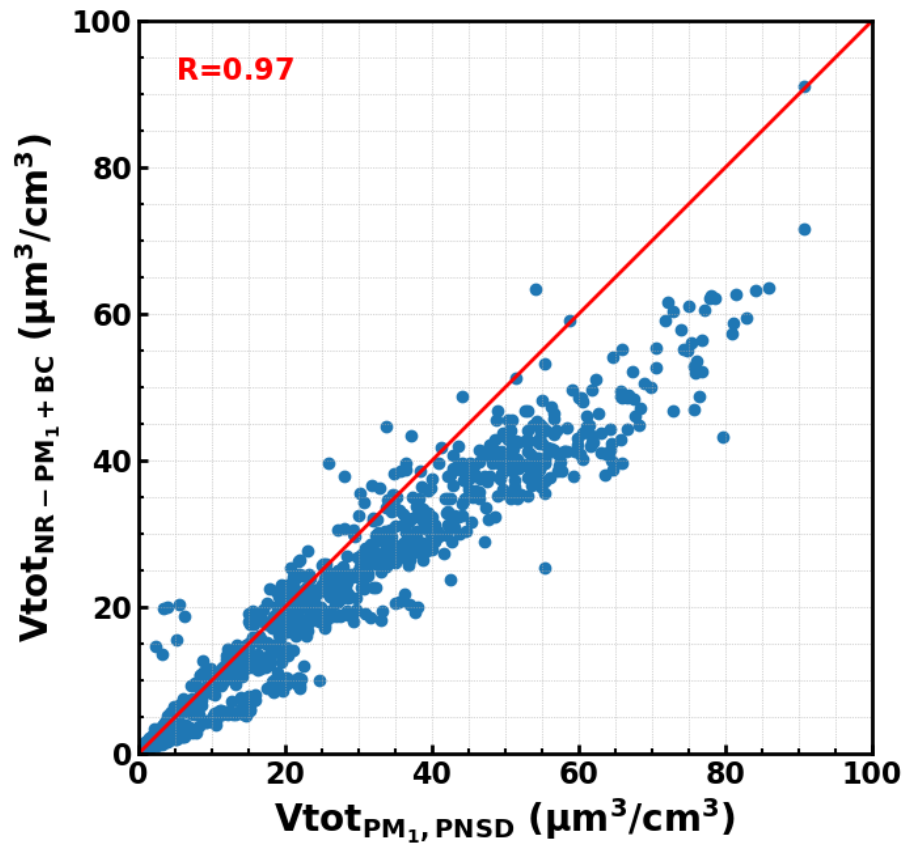
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8 Fig. S1. The mass spectra of OA factors.

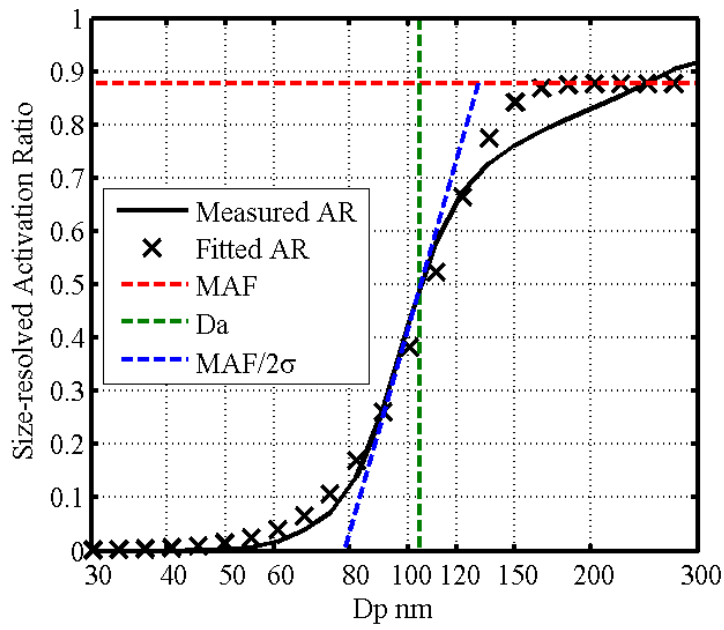
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16 Fig. S3. Comparison between aerosol volume concentration derived from measurements of PNSD and
17 aerosol chemical compositions.

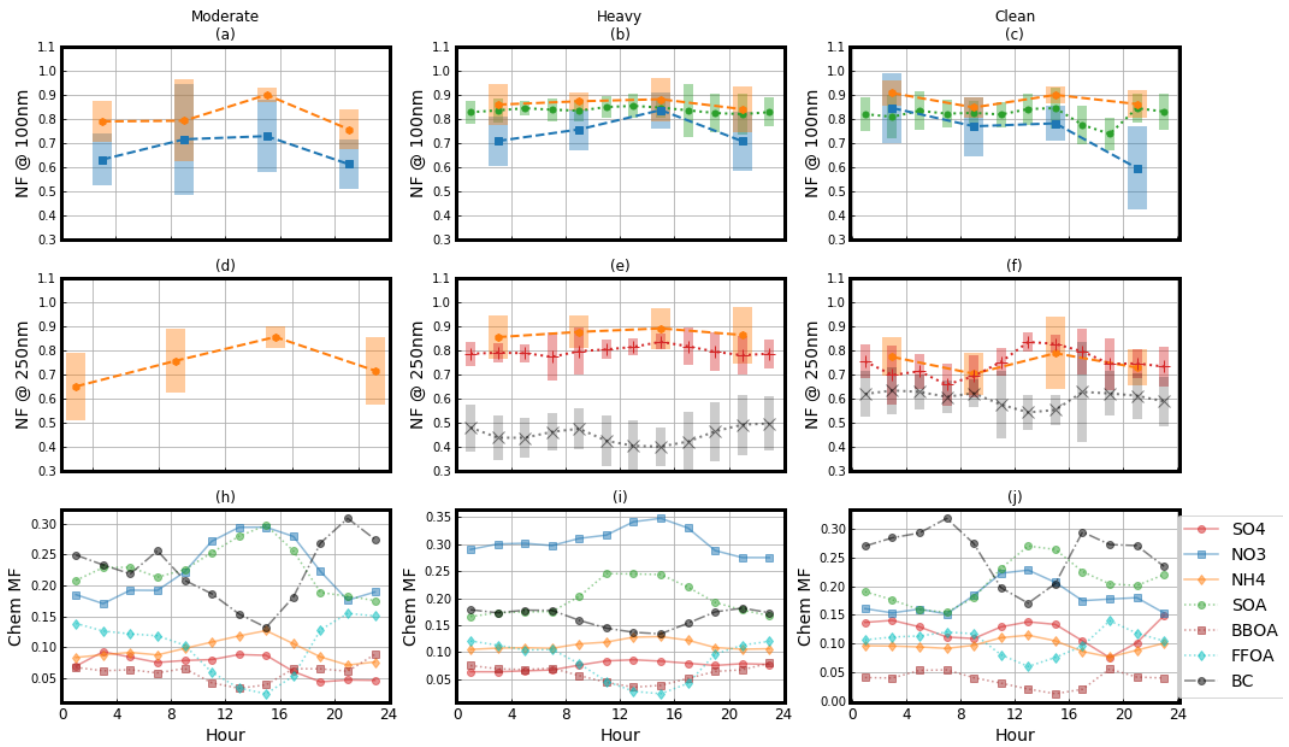
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20 Fig. S4. Schematic of the parameterization scheme of SPAR curves. The black solid curve and the
 21 black crossing are the measured SPAR and fitted SPAR with the parameterization scheme. The red,
 22 green and blue dashed lines indicate the fitting parameters of Maximum Activation Fraction (MAF),
 23 the midpoint activation diameter (Da) and s , respectively.

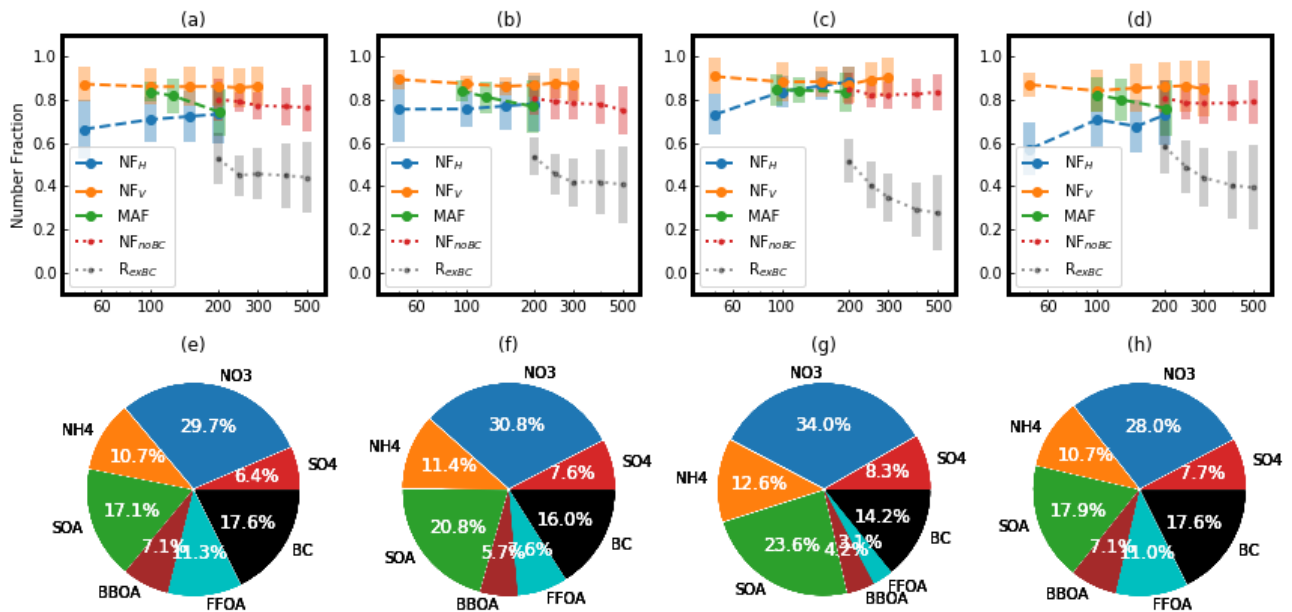
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26 Fig. S5. (a-l) Diurnal variations of aerosol mixing state parameters (identified by color and marker) at
 27 different particle sizes (50, 150, 200 and 300 nm) during the three periods. The shaded areas indicate
 28 the standard deviations. (m-o) Diurnal variations of mass fractions of aerosol chemical compositions
 29 (identified by color and marker) during the three periods.

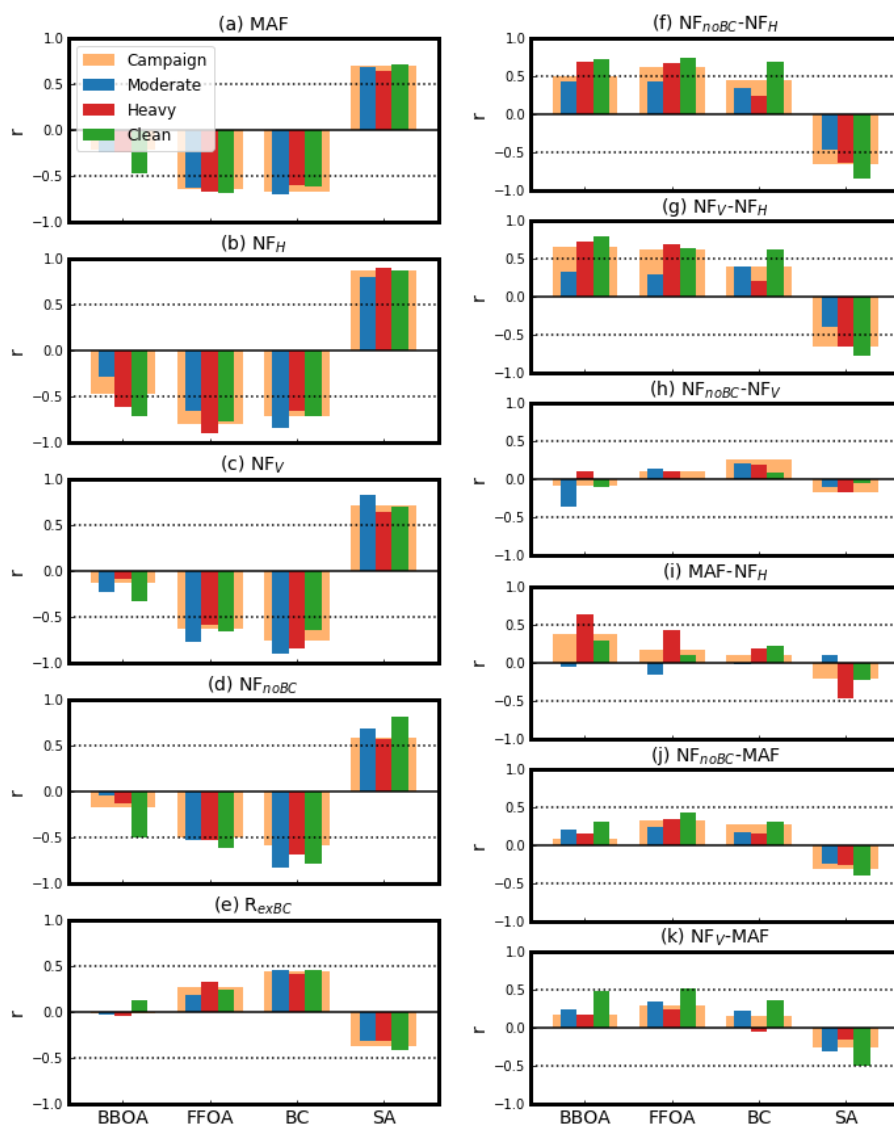
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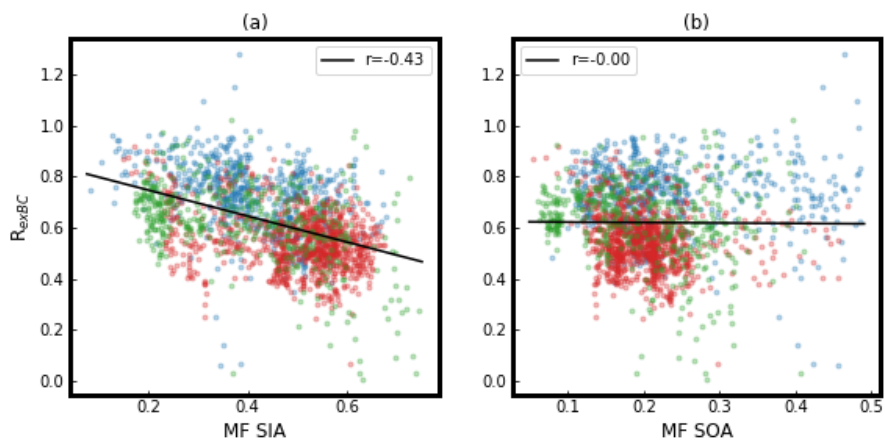
32 Fig. S6. (i, k, m and o): Average size-dependence of hygroscopic particles (NF_H, blue), volatile
 33 particles (NF_V, orange), CCN (MAF, green), BC free particles (NF_{noBC}, red) and ratio of thinly coated
 34 BC in total BC particles (R_{exBC}, black) during the 0-6, 6-12, 12-18 and 18-24 hours in the heavily
 35 polluted period. (j, l, n and p): Corresponding mass fraction of aerosol chemical compositions
 36 (identified by colors) during the three periods during the 0-6, 6-12, 12-18 and 18-24 hours in the
 37 heavily polluted period.

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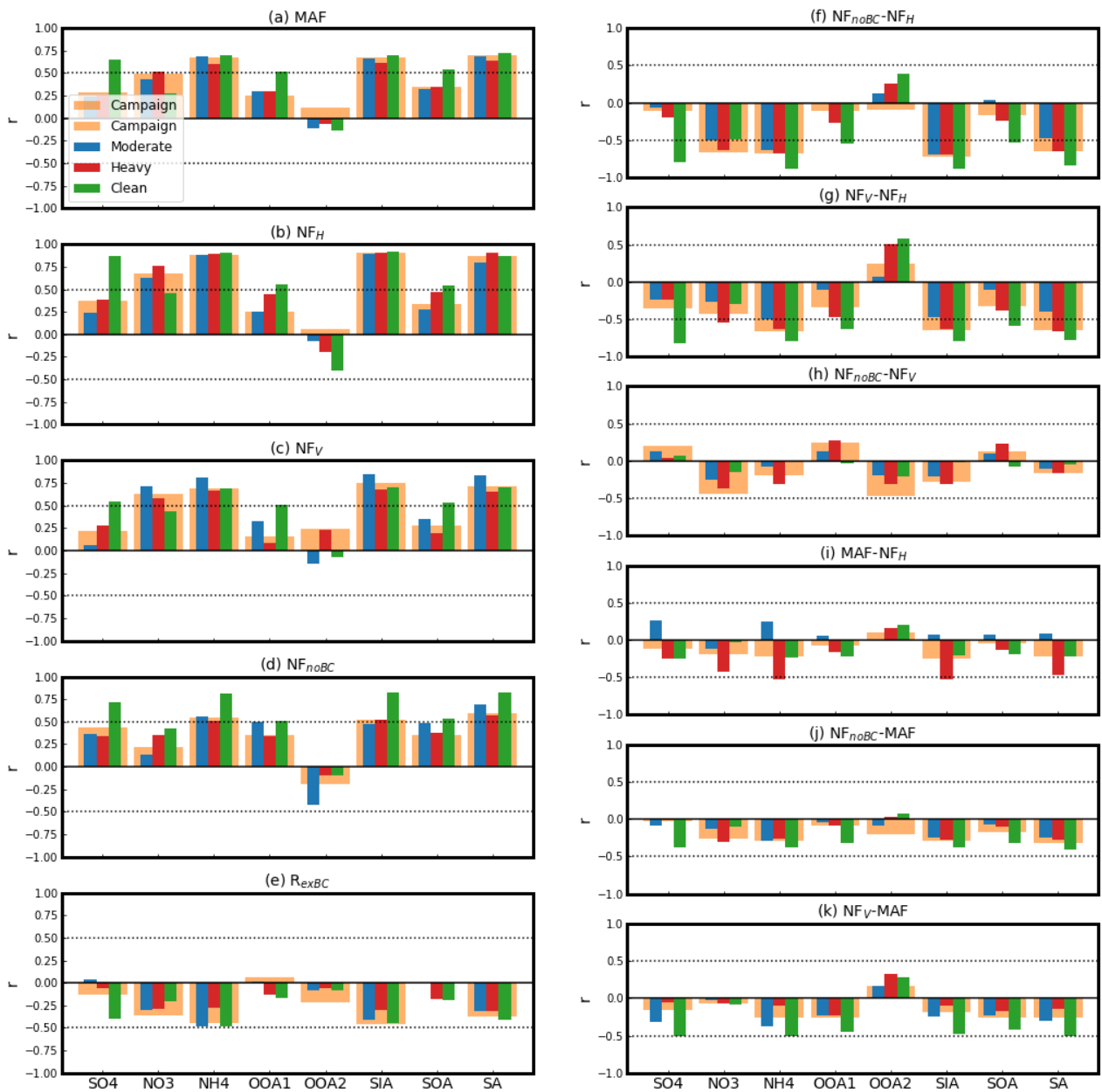
40 Fig. S7. (a-e) The correlation coefficient (R) between aerosol mixing state parameters and MF of
 41 primary aerosol chemical composition during different periods (f-k) The correlation coefficient (R)
 42 between the difference among the four aerosol mixing state parameters and MF of primary aerosol
 43 chemical composition during different periods. Moderately polluted period: Blue; Heavily polluted
 44 period: Red; Clean period: Green; Whole campaign: Orange.



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46 Fig. S8. The correlation between the difference among the four aerosol mixing state parameters and
47 MF of secondary aerosol chemical composition during different periods. Moderately polluted period:
48 Blue; Heavily polluted period: Red; Clean period: Green.

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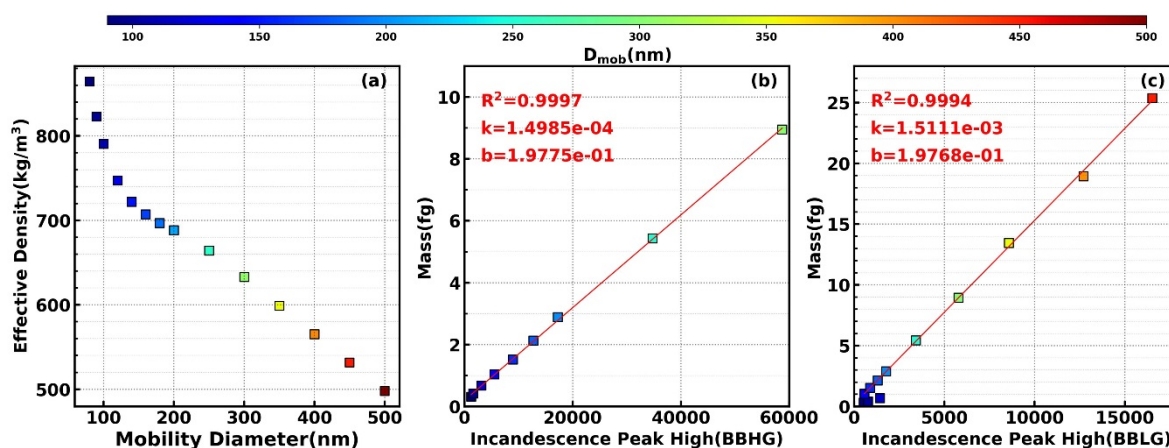
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Fig. S9. (a-e) The correlation coefficient (R) between aerosol mixing state parameters during different periods (f-k) The correlation coefficient (R) between the difference among the four aerosol mixing state parameters and MF of secondary aerosol chemical composition during different periods. Moderately polluted period: Blue; Heavily polluted period: Red; Clean period: Green; Whole campaign: Orange.

57 S1. Calibration of SP2

58 In this study, Aquadag soot particles was used for calibrating the measured incandescence signal
59 of the SP2 as reported by Gysel et al. (2011). Briefly, the soot mass is determined through the aerosol
60 density as shown in Fig. S10 (a) and aerosol mobility diameter determined by the DMA. The
61 relationships between measured incandescence signal heights and black carbon mass at different
62 diameters were shown in Fig. S10 (b) and (c). The size of rBC refers to the mass equivalent diameter
63 for DMA selected BC-containing particles is converted from the mass of rBC by assuming that the
64 density of rBC is 1.8 g/cm³, which is the median ρ value recommended by Bond and Bergstrom (2006).



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66 Figure S10. (a) Effective densities of Aquadag soot particles; (b) and (c) Relationships between soot
67 mass and incandescence signals of low gain and high gain channels.

68 Also, multiple charge corrections were conducted for BC containing aerosols and BC free
69 aerosols using the method reported by Zhao et al. (2021).

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71 Reference:

72 Bond, T. C. and Bergstrom, R. W.: Light Absorption by Carbonaceous Particles: An Investigative
73 Review, *Aerosol Science and Technology*, 40, 27–67, <https://doi.org/10.1080/02786820500421521>,
74 2006.

75 Gysel, M., Laborde, M., Olfert, J. S., Subramanian, R., and Gröhn, A. J.: Effective density of Aquadag
76 and fullerene soot black carbon reference materials used for SP2 calibration, *Atmos. Meas. Tech.*, 4,
77 2851–2858, <https://doi.org/10.5194/amt-4-2851-2011>, 2011.

78 Zhao, G., Tan, T., Zhu, Y., Hu, M., and Zhao, C.: Method to quantify black carbon aerosol light
79 absorption enhancement with a mixing state index, *Atmos. Chem. Phys.*, 21, 18055–18063,
80 <https://doi.org/10.5194/acp-21-18055-2021>, 2021.