



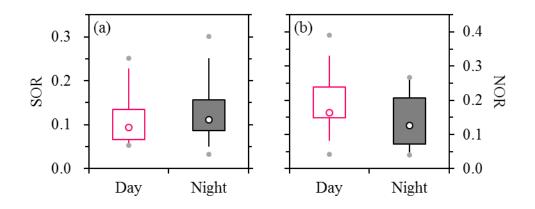
## Supplement of

## Measurement report: Diurnal variations of brown carbon during two distinct seasons in a megacity in northeast China

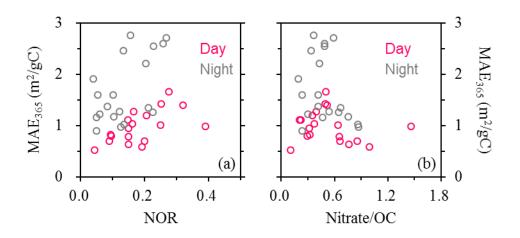
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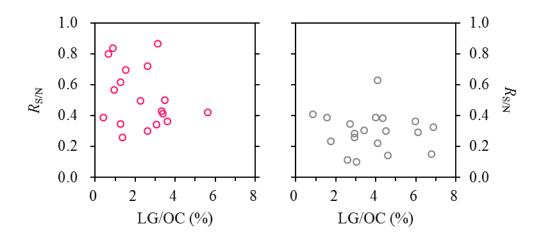
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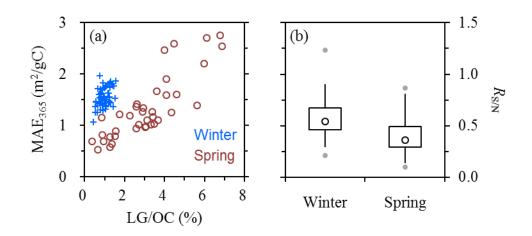
**Figure S1.** Diurnal variations of (a) SOR and (b) NOR in spring. NOR were considerably higher during the day, pointing to enhanced photochemistry. Daytime and nighttime SOR were more comparable, presumably due to the rare occurrence of high RH conditions that are favorable for heterogeneous sulfate formation.



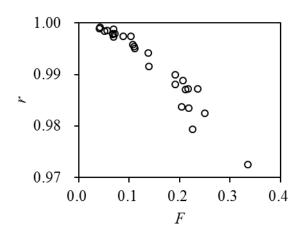
**Figure S2.** Dependences of MAE<sub>365</sub> on (a) NOR and (b) the nitrate to OC ratio in spring. As indicated by the diurnal variations of NOR, nitrate formation was mainly contributed by photochemistry. However, MAE<sub>365</sub> was independent of NOR or nitrate/OC, suggesting that photochemistry should not be an important influencing factor for the springtime MAE<sub>365</sub>.



**Figure S3.** Relationships between  $R_{S/N}$  and LG/OC in spring, with daytime and nighttime results shown in the left and night panels, respectively.



**Figure S4.** Comparisons of (a) the MAE<sub>365</sub> vs. LG/OC relationship and (b)  $R_{S/N}$  between winter and spring.



**Figure S5.** Dependence of *r* on *F* during agricultural fire episodes in spring. *r* was determined by regressing  $\ln(ATN_{\lambda})^*$  on  $\ln(\lambda)$ , while *F* was a measure of the significance of the ~365 nm absorption peak. *r* showed a clear decreasing trend with the increase of *F*. The same trend was observed when plotting *r* against *K*, another indicator for the significance of the ~365 nm absorption peak.

Compared parameters	<i>p</i> value of <i>t</i> -test	Indication	
Winter campaign			
Daytime and nighttime MAE <sub>365</sub>	0.004	More absorbing BrC at night	
Daytime and nighttime LG/OC (LG/EC)	0.001 (0.000)	Increased residential burning emissions at night	
Daytime and nighttime R <sub>S/N</sub>	0.000	Increased vehicle emissions at night	
Daytime and nighttime SOR in the RH range of 70–80%	0.417	Relatively weak influence of photochemistry on sulfate formation	
Daytime and nighttime NOR in the RH range of 70–80%	0.005	Relatively strong influence of photochemistry on nitrate formation	
Daytime and nighttime AAE	0.000	Stronger wavelength dependence of BrC absorption at night	
Daytime and nighttime sulfate/OC	0.011	Decreased SOC/OC ratios at night	
Spring campaign			
Daytime and nighttime MAE <sub>365</sub>	0.000	More absorbing BrC at night	
Daytime and nighttime LG/OC	0.006	Increased agricultural fire emissions at night	
Daytime and nighttime <i>R</i> <sub>S/N</sub>	0.000	Increased vehicle emissions at night	
Daytime and nighttime SOR	0.489	Insignificant diurnal variations of sulfate formation	
Daytime and nighttime NOR	0.083	Insignificant diurnal variations of nitrate formation	
<i>r</i> values for typical samples and open burning episodes [derived from linear regression of $\ln(ATN_{\lambda})^*$ on ln ( $\lambda$ )]	0.000	Agricultural fire-induced non-linearity for BrC's absorption spectra shown on ln-ln scale	
Inter-campaign			
LG/K <sup>+</sup> in winter and spring	0.000	Different biomass burning ways in the two seasons (i.e., residential and open burning, respectively)	
LG/OC in winter and spring	0.000	Stronger impacts of biomass burning in spring	
SOR in winter and spring	0.050		
NOR in winter and spring	0.012	Significant seasonal variations of nitrate formation	
Wintertime MAE <sub>365</sub> and MAE <sub>365</sub>	0.000	Less absorbing BrC in spring with the	

**Table S1.** Summary of *t*-test results for the comparisons involved in the main text. A *p* value of below 0.05 indicates statistically significant difference at the 95% confidence level.

	Daytime	Nighttime	All
Temperature ( $^{\circ}$ C)	$-16.23 \pm 5.13$	$-21.28 \pm 5.54$	$-18.72 \pm 5.87$
Relative humidity (%)	$70.73 \pm 8.10$	$81.10 \pm 5.03$	$75.83 \pm 8.51$
OC (µgC/m <sup>3</sup> )	21.53 ±8.54	$21.27 \pm 11.94$	$21.40 \pm 10.26$
MSOC (µgC/m <sup>3</sup> )	19.74 ±7.73	$19.56 \pm 10.81$	$19.65 \pm 9.29$
EC (μgC/m <sup>3</sup> )	$3.12 \pm 1.69$	$2.62 \pm 1.96$	$2.87 \pm 1.83$
$LG (\mu g/m^3)$	$0.45 \pm 0.24$	$0.57 \pm 0.44$	$0.51 \pm 0.35$
$SO_4^{2-}(\mu g/m^3)$	$9.49 \pm 4.28$	$7.97 \pm 5.03$	8.74 ±4.69
$NO_{3}^{-}(\mu g/m^{3})$	9.53 ±5.84	9.56 ±8.24	$9.55 \pm 7.06$
$Cl^{-}(\mu g/m^{3})$	$2.60 \pm 1.22$	$2.25 \pm 1.19$	2.43 ±1.21
$NH_{4}^{+}$ (µg/m <sup>3</sup> )	8.10 ±3.96	$7.25 \pm 4.80$	$7.68 \pm 4.38$
$K^+$ (µg/m <sup>3</sup> )	$1.01 \pm 0.41$	$1.08\ \pm 0.78$	$1.05 \pm 0.62$
$(b_{\rm abs})_{365}~({\rm Mm}^{-1})$	30.00 ±13.34	$32.48 \pm 20.94$	31.22 ±17.39
MAE <sub>365</sub> (m <sup>2</sup> /gC)	$1.48 \pm 0.18$	$1.61 \pm 0.15$	$1.55 \pm 0.18$
$AAE^{a}$	$6.76 \pm 0.11$	$7.33 \pm 0.14$	$7.04 \pm 0.32$
SOR	$0.14 \pm 0.05$	$0.16 \pm 0.09$	$0.15 \pm 0.07$
NOR	$0.13 \pm 0.05$	$0.11 \pm 0.06$	$0.12 \pm 0.06$
$R_{ m S/N}$	$0.69\ \pm 0.20$	$0.46 \pm 0.13$	$0.58 \pm 0.20$
$LG/K^+$	$0.42 \pm 0.11$	$0.50 \pm 0.10$	$0.46 \pm 0.11$
LG/OC (%) <sup>b</sup>	$0.88 \pm 0.22$	$1.10 \pm 0.26$	$0.99 \pm 0.26$
LG/EC	$0.15 \pm 0.05$	$0.22 \pm 0.06$	$0.18 \pm 0.07$

**Table S2.** Summary of observational results in January, 2021.

Note:

<sup>a</sup> Calculated over 310–460 nm;

<sup>b</sup> On a basis of carbon mass.

	Daytime	Nighttime	All
Temperature ( $^{\circ}$ C)	13.82 ±4.47	7.33 ±4.16	10.58 ±5.38
Relative humidity (%)	$42.65 \pm 17.35$	62.21 ±17.43	$52.43 \pm 19.81$
OC ( $\mu$ gC/m <sup>3</sup> )	$16.64 \pm 14.57$	$22.49 \pm 19.99$	$19.57 \pm 17.51$
MSOC (µgC/m <sup>3</sup> )	14.36 ±13.06	$20.23 \pm 18.27$	$17.30 \pm 15.94$
EC (μgC/m <sup>3</sup> )	$1.35 \pm 0.94$	$1.71 \pm 0.95$	$1.53 \pm 0.95$
LG (µg/m <sup>3</sup> )	$1.20 \pm 1.69$	$2.53 \pm 3.16$	$1.87 \pm 2.59$
$SO_4^{2-}  (\mu g/m^3)$	3.28 ±2.13	$3.38 \pm 1.99$	3.33 ±2.03
$NO_3^-(\mu g/m^3)$	$8.97 \pm 9.07$	$10.79 \pm 9.10$	9.88 ±9.01
$Cl^{-}$ (µg/m <sup>3</sup> )	1.79 ±2.51	$4.84 \pm 5.88$	3.32 ±4.72
$NH_4{}^+~(\mu g/m^3)$	$4.35 \pm 3.88$	$6.33 \pm 5.48$	5.34 ±4.79
$K^+$ (µg/m <sup>3</sup> )	$1.05 \pm 1.21$	$1.31 \pm 1.26$	$1.18 \pm 1.22$
$(b_{\rm abs})_{365} ({\rm Mm}^{-1})$	$17.22 \pm 20.53$	43.66 ±53.16	30.44 ±41.95
MAE <sub>365</sub> (m <sup>2</sup> /gC)	$0.98 \pm 0.31$	$1.69 \pm 0.65$	$1.33 \pm 0.62$
AAE <sup>a</sup>	—	—	—
SOR	$0.11 \pm 0.06$	$0.13 \pm 0.07$	$0.12 \pm 0.06$
NOR	$0.18 \pm 0.09$	$0.14 \pm 0.08$	$0.16 \pm 0.08$
$R_{ m S/N}$	$0.52 \pm 0.19$	$0.30 \pm 0.13$	$0.40 \pm 0.20$
$LG/K^+$	$0.98 \pm 0.45$	$1.58 \pm 0.61$	$1.28 \pm 0.61$
LG/OC (%) <sup>b</sup>	$2.38 \pm 1.36$	$3.85 \pm 1.71$	$3.11 \pm 1.70$
LG/EC	$0.65 \pm 0.46$	$1.14 \pm 1.00$	$0.89 \pm 0.81$

Table S3. Summary of observational results in April, 2021.

Note:

<sup>a</sup> AAE were not provided due to the frequent occurrences of agricultural fires, which could result in distinct peak at ~365 nm for the light absorption spectra of BrC.

<sup>b</sup> On a basis of carbon mass.