

## Supplementary Information for

### The large proportion of black carbon (BC)-containing aerosols in the urban atmosphere

*Lu Chen*<sup>a</sup>, *Fang Zhang*<sup>a, \*</sup>, *Peng Yan*<sup>b</sup>, *Xinming Wang*<sup>c</sup>, *Lu Sun*<sup>d</sup>, *Yanan Li*<sup>b</sup>,  
*Xiaochun Zhang*<sup>b</sup>, *Yele Sun*<sup>e</sup>, and *Zhanqing Li*<sup>f</sup>

<sup>a</sup> College of Global Change and Earth System Science, Beijing Normal University, Beijing 100875, China

<sup>b</sup> Meteorological Observation Center of China Meteorological Administration, Beijing, 100081, China

<sup>c</sup> State Key Laboratory of Organic Geochemistry, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou 510640, China

<sup>d</sup> Department of Atmospheric Sciences, Texas A&M University, College Station, TX, USA

<sup>e</sup> State Key Laboratory of Atmospheric Boundary Layer Physics and Atmospheric Chemistry, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing, 100080, China

<sup>f</sup> Earth System Science Interdisciplinary Center and Department of Atmospheric and Oceanic Science, University of Maryland, College Park, MD, USA

Correspondence to: F. Zhang, [fang.zhang@bnu.edu.cn](mailto:fang.zhang@bnu.edu.cn)

**This file includes**

Supplementary text

Figures, S1 to S6

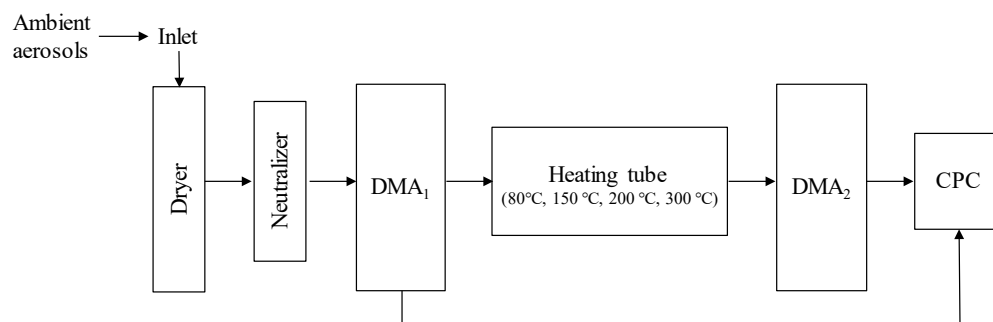
Table S1

References

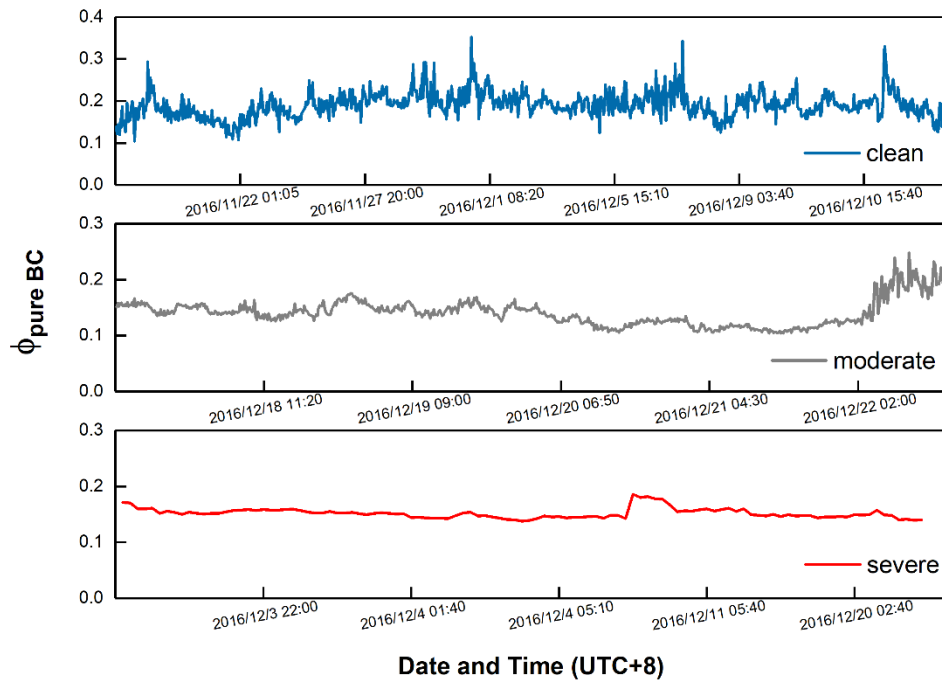
## **1. Supplementary text**

### **1.1 Measurements**

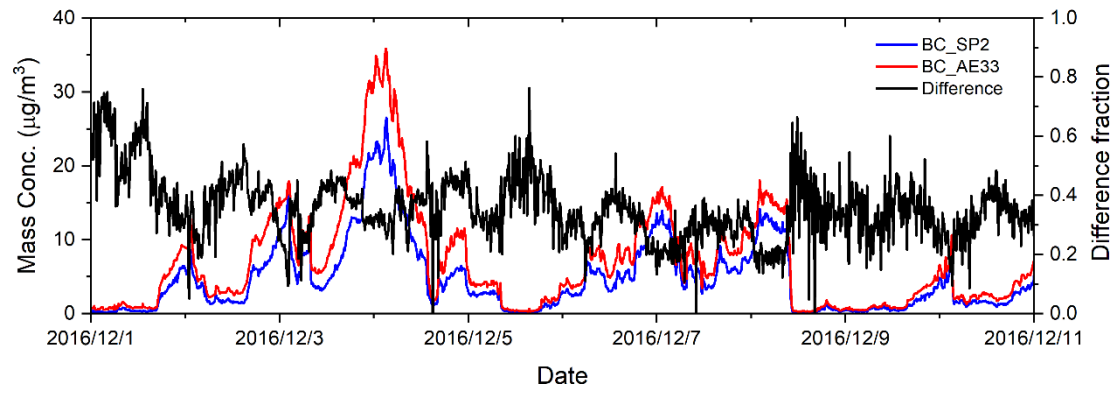
During the field measurements, the ambient aerosols enter the PM<sub>2.5</sub> inlet, then the collected sample aerosols are passed through a drying tube to reduce the humidity to less than 20%. After drying, the dry aerosols enter a neutralizer to balance the charge of the particles (Wiedensohler, 1988), then enter the first DMA and select mono-disperse aerosol of a specific particle size. We set the mobility diameter ( $D_p$ ) from 40 to 300 nm (40, 60, 80, 110, 150, 200, 250, and 300 nm). The selected mono-disperse aerosols go via two path. The first is to directly enter the condensation particle counter (CPC) to obtain the particle number size distribution of the dry aerosols. The second is to enter a heating tube to remove the volatile substances. The heated aerosols pass through the second DMA and CPC so that the number size distribution could be measured (Fig. S1). Here we set 4 heating temperature gradients, including 80 °C, 150 °C, 200 °C, and 300 °C. This study focuses on data obtained only at 300 °C, because the residual material after volatilization at this temperature is considered to be mainly BC (Burtcher et al., 2001; Cheng et al., 2009; Frey et al., 2008; Kondo et al., 2006; Philippin et al., 2004; Smith and O'Dowd, 1996; Wehner et al., 2009). More details about the VTDMA system used in our study were described by Wang et al. (2017).



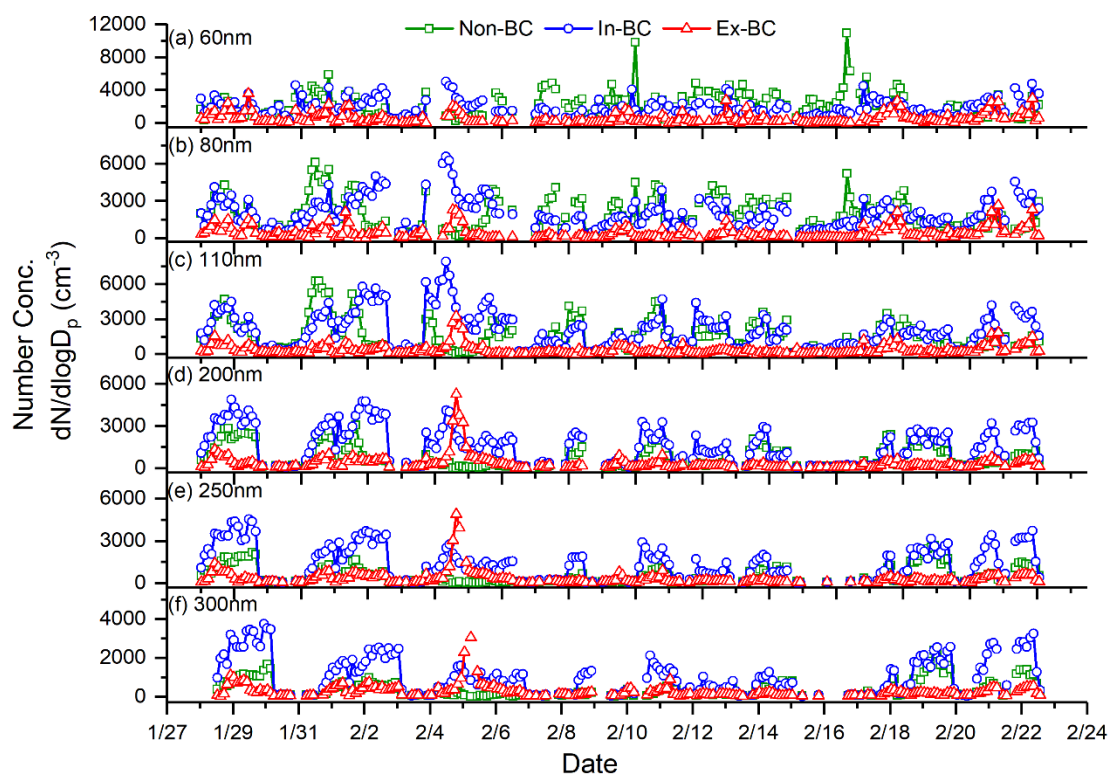
**Figure S1.** A schematic diagram of the volatility tandem differential mobility analyzer (VTDMA).



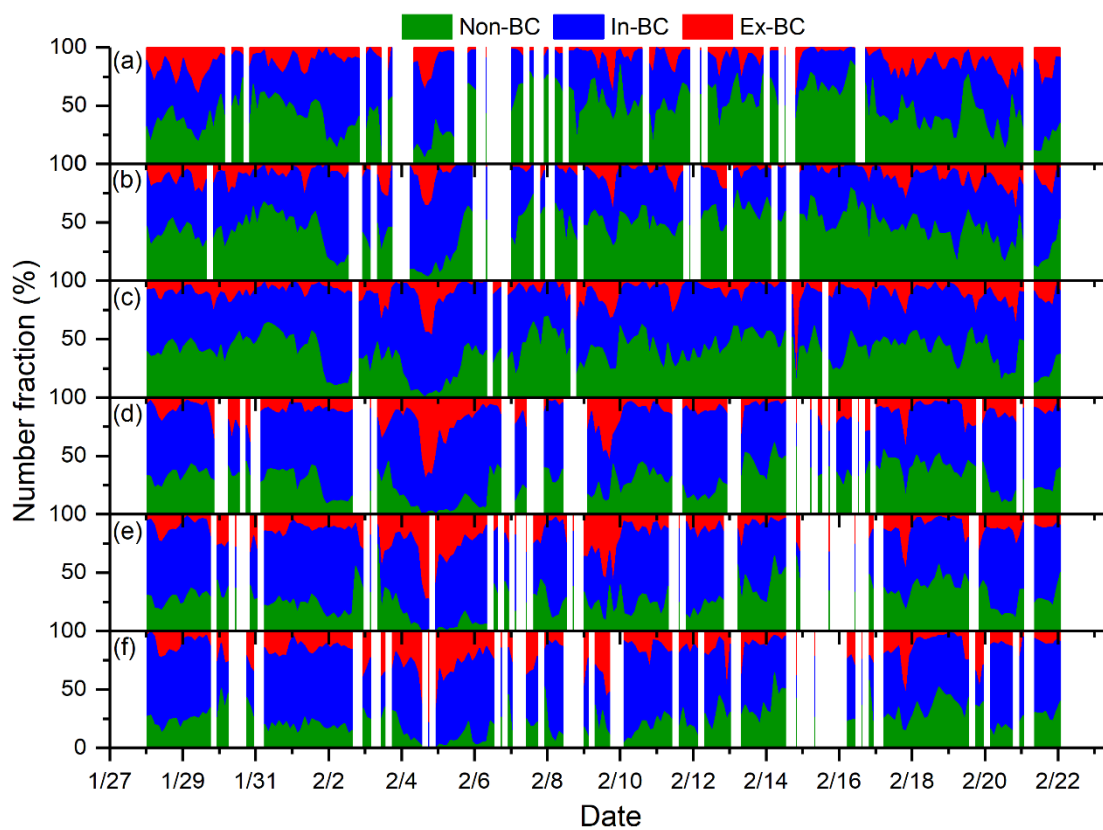
**Figure S2.** The time series of  $\phi_{\text{pure BC}}$  during clean (blue), moderate (grey) and severe (red) periods measured by SP-AMS.



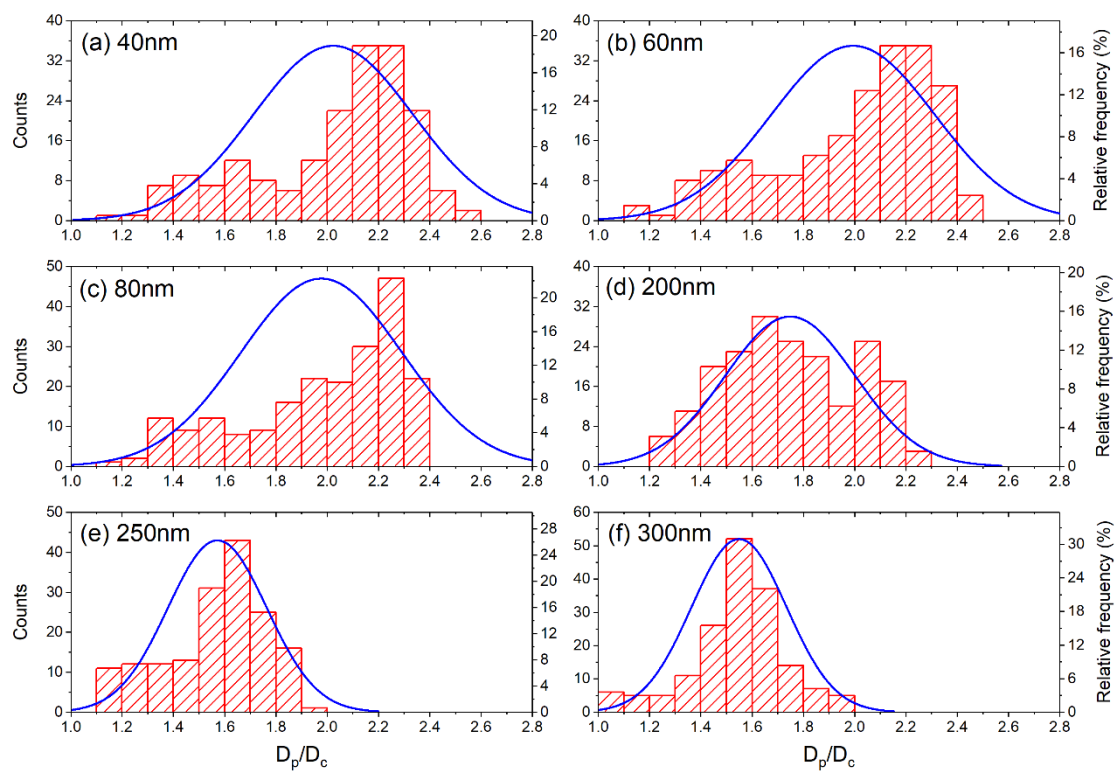
**Figure S3.** Time series of mass concentration of BC measured by SP2 (in blue) and AE33 (in red), and the difference between the two instruments (in black).



**Figure S4.** Temporal variation of number concentration of Non-BC (in green), In-BC (in blue), and Ex-BC (in red) in the range of 60-300 nm particles.



**Figure S5.** Temporal variation of number fraction of Non-BC (in green), In-BC (in blue), and Ex-BC (in red) in the range of 60-300 nm particles.



**Figure S6.** The counts (red) and frequency (blue) distribution of the ratios of  $D_p/D_c$  of BC-containing particles with  $D_p$  of 40-300 nm.



**Table S1.** A summary of  $E_{\text{abs}}$  at different sites derived from previous studies.

Site	Type	Method	$\lambda$ (nm)	$D_p/D_c$	$E_{\text{abs}}$	Reference
Beijing	Urban	MAE ratio	550	$\sim 2.2$	$\sim 2.1$	This study
Beijing	Urban	MAE ratio	365	2.25	3.2-5.3	Cheng et al. (2017)
Beijing	Urban	MAE ratio	880	1.5-2.3	1.5-2.0	Zhang et al. (2018)
Beijing	Urban	TD	630	1.27-1.35	$1.59 \pm 0.26$	Xie et al. (2019)
Guangzhou	Suburban	MAE ratio	550	1.5	$1.5 \pm 0.48$	Wu et al. (2018)
Xianghe	Suburban	MAE ratio	550	2.1-2.7	1.6-1.9	Zhang et al. (2016)
Shouxian	Rural	TD	532	2.4-2.8	$2.3 \pm 0.9$	Xu et al. (2018)
Toronto	Suburban	TD	550	2.1-2.6	1.6-1.9	A. Knox et al. (2009)
California	Urban	TD	532	2	1.06	Cappa et al. (2012)
Boulder	Forest fire	TD	532	1.45	1.4	Lack et al. (2012)
Costa Rica	troposphere	TD	550	$\sim 1.15$	$1.31 \pm 0.1$	Schwarz et al. (2008)

## References

- Burtscher, H., Baltensperger, U., Bukowiecki, N., Cohn, P., Hüglin, C., Mohr, M., Matter, U., Nyeki, S., Schmatloch, V., Streit, N., Weingartner, E., 2001. Separation of volatile and non-volatile aerosol fractions by thermodesorption: instrumental development and applications. *J. Aerosol Sci.* 32, 427-442.
- Cappa, C.D., Onasch, T.B., Massoli, P., Worsnop, D.R., Bates, T.S., Cross, E.S., Davidovits, P., Hakala, J., Hayden, K.L., Jobson, B.T., Kolesar, K.R., Lack, D.A., Lerner, B.M., Li, S.-M., Mellon, D., Nuaaman, I., Olfert, J.S., Petäjä, T., Quinn, P.K., Song, C., Subramanian, R., Williams, E.J., Zaveri, R.A., 2012. Radiative Absorption Enhancements Due to the Mixing State of Atmospheric Black Carbon. *Science* 337, 1078-1081.
- Cheng, Y., He, K.B., Engling, G., Weber, R., Liu, J.M., Du, Z.Y., Dong, S.P., 2017. Brown and black carbon in Beijing aerosol: Implications for the effects of brown coating on light absorption by black carbon. *Sci. Total Environ.* 599-600, 1047-1055.
- Frey, A., Rose, D., Wehner, B., Müller, T., Cheng, Y., Wiedensohler, A., Virkkula, A., 2008. Application of the Volatility-TDMA Technique to Determine the Number Size Distribution and Mass Concentration of Less Volatile Particles. *Aerosol Sci. Technol.* 42, 817-828.
- Knox, A., Evans, G.J., Brook, J.R., Yao, X., Jeong, C.H., Godri, K.J., Sabaliauskas, K., Slowik, J.G., 2009. Mass Absorption Cross-Section of Ambient Black Carbon Aerosol in Relation to Chemical Age. *Aerosol Sci. Technol.* 43, 522-532.
- Kondo, Y., Komazaki, Y., Miyazaki, Y., Moteki, N., Takegawa, N., Kodama, D., Deguchi, S., Nogami, M., Fukuda, M., Miyakawa, T., Morino, Y., Koike, M., Sakurai, H., Ehara, K., 2006. Temporal variations of elemental carbon in Tokyo. *J. Geophys. Res.: Atmospheres* 111, D12.
- Lack, D.A., Langridge, J.M., Bahreini, R., Cappa, C.D., Middlebrook, A.M., Schwarz, J.P., 2012. Brown carbon and internal mixing in biomass burning particles. *Proc. Natl. Acad. Sci.* 109, 14802-14807.
- Philippin, S., Wiedensohler, A., Stratmann, F., 2004. Measurements of non-volatile fractions of pollution aerosols with an eight-tube volatility tandem differential mobility analyzer (VTDMA-8). *J. Aerosol Sci.* 35, 185-203.
- Schwarz, J.P., Spackman, J.R., Fahey, D.W., Gao, R.S., Lohmann, U., Stier, P., Watts, L.A., Thomson, D.S., Lack, D.A., Pfister, L., Mahoney, M.J., Baumgardner, D., Wilson, J.C., Reeves, J.M., 2008. Coatings and their enhancement of black carbon light absorption in the tropical atmosphere. *J. Geophys. Res.: Atmosphere* 113, D03203.
- Smith, M.H., O'Dowd, C.D., 1996. Observations of accumulation mode aerosol composition and soot carbon concentrations by means of a high-temperature volatility technique. *J. Geophys. Res.: Atmosphere* 101, 19583-19591.
- Wang, Y., Zhang, F., Li, Z., Tan, H., Xu, H., Ren, J., Zhao, J., Du, W., Sun, Y., 2017. Enhanced hydrophobicity and volatility of submicron aerosols under severe emission control conditions in Beijing. *Atmos. Chem. Phys.* 17, 5239-5251.
- Wehner, B., Berghof, M., Cheng, Y.F., Achtert, P., Birmili, W., Nowak, A., Wiedensohler, A., Garland, R.M., Pöschl, U., Hu, M., Zhu, T., 2009. Mixing state of nonvolatile aerosol particle fractions and comparison with light absorption in the polluted Beijing region. *J. Geophys. Res.: Atmosphere* 114, D00G17.
- Wiedensohler, A., 1988. An approximation of the bipolar charge distribution for particles in the submicron size range. *J. Aerosol Sci.* 19, 387-389.

Wu, C., Wu, D., Yu, J.Z., 2018. Quantifying black carbon light absorption enhancement with a novel statistical approach. *Atmos. Chem. Phys.* 18, 289-309.

Xie, C., Xu, W., Wang, J., Liu, D., Ge, X., Zhang, Q., Wang, Q., Du, W., Zhao, J., Zhou, W., Li, J., Fu, P., Wang, Z., Worsnop, D., Sun, Y., 2019. Light absorption enhancement of black carbon in urban Beijing in summer. *Atmos. Environ.* 213, 499-504.

Xu, X., Zhao, W., Qian, X., Wang, S., Fang, B., Zhang, Q., Zhang, W., Venables, D.S., Chen, W., Huang, Y., Deng, X., Wu, B., Lin, X., Zhao, S., Tong, Y., 2018. The influence of photochemical aging on light absorption of atmospheric black carbon and aerosol single-scattering albedo. *Atmos. Chem. Phys.* 18, 16829-16844.

Cheng, Y., He, K.B., Engling, G., Weber, R., Liu, J.M., Du, Z.Y., Dong, S.P., 2017. Brown and black carbon in Beijing aerosol: Implications for the effects of brown coating on light absorption by black carbon. *Sci. Total Environ.* 599-600, 1047-1055.

Zhang, Y., Zhang, Q., Cheng, Y., Su, H., Li, H., Li, M., Zhang, X., Ding, A., He, K., 2018. Amplification of light absorption of black carbon associated with air pollution. *Atmos. Chem. Phys.* 18, 9879-9896.

Zhang, Y., Zhang, Q., Cheng, Y., Su, H., Kecorius, S., Wang, Z., Wu, Z., Hu, M., Zhu, T., Wiedensohler, A., He, K., 2016. Measuring the morphology and density of internally mixed black carbon with SP2 and VTDMA: new insight into the absorption enhancement of black carbon in the atmosphere. *Atmos. Meas. Tech.* 9, 1833-1843.