



Supplement of

Light absorption properties and potential sources of particulate brown carbon in the Pearl River Delta region of China

Zhujie Li et al.

Correspondence to: Haobo Tan (hbtan@gd121.cn) and Jun Zheng (zheng.jun@nuist.edu.cn)

The copyright of individual parts of the supplement might differ from the CC BY 4.0 License.

Parameters	±σ (%)
$D_{ m p,DMA}$	1.2ª
NCPC	3.3 ^a
$D_{ m p,APS}$	3.0 ^b
$N_{ m APS}$	3.3 ^b
$\widetilde{m}_{non}=1.50$	0.3 ^c
$\widetilde{m}_{r, BC} = 1.80$	3.3 ^c
$\widetilde{m}_{i, \text{ BC}}=0.54$	13.3°
ρ _{BC} =1.5	11 ^d
r _{ext} =0.58	45 ^e
$\Phi_{ m N,CV}$	20.7^{f}

Table S1. Uncertainties of each input parameter, which was given in terms of one relative standard deviation (σ).

^a The measurement uncertainties of the DMA for $D_{p,DMA}$ and the CPC for N_{CPC} , which were recommended by Wiedensohler et al. (2012).

^b The measurement uncertainty of the APS and the uncertainty during the transformation from aerodynamic diameter to Stokes diameter recommended by Peters and Leith (2003).

^c The uncertainties of the refractive indices of non-light absorbing materials and BC core recommended by Cheng et al. (2006).

^d The uncertainty of ρ_{BC} at a 99% confidence level, ranging from 1.00 g cm⁻³ to 2.00 g cm⁻³ (Ma et al., 2012).

^e The uncertainty of r_{ext} is taken from the optical closure study results of Tan et al. (2016).

^f The uncertainty of $\Phi_{N,CV}$ is evaluated from six diameters of the VTDMA measurement conducted in Guangzhou by Cheung et al. (2016).

Table S2. Average relative standard deviation (σ_{Mie}) of the calculated Abs₈₈₀, AAE_{BC,370-520} and AAE_{BC,520-880} with Monte Carlo simulation.

	Abs ₈₈₀	AAE _{BC,370-520}	AAE _{BC,520-880}
σ _{Mie} (%)	15.4	7.9	6.6



Figure S1. Time series of aerosol extinction coefficients, scattering coefficients and SSA at 532 nm.



Figure S2. The time series of size distribution of the derived external BC and internal BC core.



Figure S3. The size distribution of $\Phi_{N,CV}$.



Figure S4. Time series of particle AAE measured by Aethalometer and AAE_{BC} derived by Mie model calculation.



Figure S5. a) The time series of the uncertainties of Abs_{880} , $AAE_{BC,370-520}$, and $AAE_{BC,520-880}$ from the Monte Carlo simulation; b) Time series of the calculated BrC absorption contribution (black dots) with the uncertainty range (the red-colored shadow areas), derived from that lowerand upper-limits of AAE_{BC} obtained from the Monte Carlo simulation. The black dots were the calculated BrC absorption contribution results using the Mie model at five wavelengths, respectively.



Figure S6. Map of 3-day backward trajectory and the fire counts for (a) 5 to 7 December 2014 (b) 12 to 14 December 2014 and (c) 24 to 26 December 2014. Fire count data were from the MODIS Collection 6 Active Fire Product in Fire Information for Resource Management System (FIRMS). Backward trajectories for the past 3-day were calculated using NOAA's Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model.

K⁺ source analysis:

A correction method was applied to acquire K^+ concentrations from biomass burning (K_{BB}^+) followed Jung et al. (2014), and used Na⁺ as a tracer for sea salt and Ca²⁺ for a tracer for crustal materials:

$$K_{BB}^{+} = (K^{+} - [K^{+} / Na^{+}]_{seawater} \times Na^{+} - [K^{+} / Ca^{2+}]_{crustal} \times nssCa^{2+})/(1 - 0.1 \times [K^{+} / Ca^{2+}]_{crustal})$$

where the $[K^+/Ca^{2+}]_{crustal}$ is 0.12; $[K^+/Na^+]_{seawater}=0.036$ is the mass ratio of potassium to sodium in seawater, and nssCa²⁺ represents non-sea-salt Ca²⁺ as presented by Pio et al. (2007). Figure S8 shows that there was a well correlation between K⁺ and K_{BB}⁺, and K_{BB}⁺ was just ~1.5% lower than K⁺. This discrepancy was small, indicating that the effect for sea salt and crustal

materials to K⁺ concentrations was negligible.



Figure S7. The correlation between the K^+ and K_{BB}^+ .



Figure S8. Box-whisker plots of diurnal trends in K^+ concentration (a), NH_4^+ concentration (b), NO_3^- concentration (c), $K^+/PM_{2.5}$ (d), $NH_4^+/PM_{2.5}$ (e) and $NO_3^-/PM_{2.5}$ (f). The red traces represent the variation in average value. The upper and lower boundaries of the box represent the 75th and 25th percentiles, respectively; the whiskers above and below each box represent an error of one standard deviation.

References:

Cheng, Y. F., Eichler, H., Wiedensohler, A., Heintzenberg, J., Zhang, Y. H., Hu, M., Herrmann, H., Zeng, L. M., Liu, S., Gnauk, T., Brüggemann, E., and He, L. Y.: Mixing state of elemental carbon and non-light-absorbing aerosol components derived from in situ particle optical properties at Xinken in Pearl River Delta of China, Journal of Geophysical Research: Atmospheres, 111, doi:10.1029/2005JD006929, 2006.

Cheung, H. H. Y., Tan, H., Xu, H., Li, F., Wu, C., Yu, J. Z., and Chan, C. K.: Measurements of non-volatile aerosols with a VTDMA and their correlations with carbonaceous aerosols in Guangzhou, China, Atmospheric Chemistry and Physics, 16, 8431-8446, 10.5194/acp-16-8431-2016, 2016.

Jung, J., Lee, S., Kim, H., Kim, D., Lee, H., and Oh, S.: Quantitative determination of the biomass-burning contribution to atmospheric carbonaceous aerosols in Daejeon, Korea, during the rice-harvest period, Atmospheric Environment, 89, 642-650, 10.1016/j.atmosenv.2014.03.010, 2014.

Ma, N., Zhao, C. S., Müller, T., Cheng, Y. F., Liu, P. F., Deng, Z. Z., Xu, W. Y., Ran, L., Nekat, B., van Pinxteren, D., Gnauk, T., Müller, K., Herrmann, H., Yan, P., Zhou, X. J., and Wiedensohler, A.: A new method to determine the mixing state of light absorbing carbonaceous using the measured aerosol optical properties and number size distributions, Atmos. Chem. Phys., 12, 2381-2397, 10.5194/acp-12-2381-2012, 2012.

Peters, T. M., and Leith, D.: Concentration measurement and counting efficiency of the aerodynamic particle sizer 3321, J. Aerosol Sci., 34, 627-634, https://doi.org/10.1016/S0021-8502(03)00030-2, 2003.

Pio, C. A., Legrand, M., Oliveira, T., Afonso, J., Santos, C., Caseiro, A., Fialho, P., Barata, F.,
Puxbaum, H., Sanchez-Ochoa, A., Kasper-Giebl, A., Gelencser, A., Preunkert, S., and Schock,
M.: Climatology of aerosol composition (organic versus inorganic) at nonurban sites on a westeast transect across Europe, J. Geophys. Res.-Atmos., 112, 10.1029/2006jd008038, 2007.

Tan, H., Liu, L., Fan, S., Li, F., Yin, Y., Cai, M., and Chan, P. W.: Aerosol optical properties and mixing state of black carbon in the Pearl River Delta, China, Atmospheric Environment, 131, 196-208, <u>http://dx.doi.org/10.1016/j.atmosenv.2016.02.003</u>, 2016.

Wiedensohler, A., Birmili, W., Nowak, A., Sonntag, A., Weinhold, K., Merkel, M., Wehner, B., Tuch, T., Pfeifer, S., Fiebig, M., Fjaraa, A. M., Asmi, E., Sellegri, K., Depuy, R., Venzac, H., Villani, P., Laj, P., Aalto, P., Ogren, J. A., Swietlicki, E., Williams, P., Roldin, P., Quincey, P., Huglin, C., Fierz-Schmidhauser, R., Gysel, M., Weingartner, E., Riccobono, F., Santos, S., Gruning, C., Faloon, K., Beddows, D., Harrison, R. M., Monahan, C., Jennings, S. G., O'Dowd, C. D., Marinoni, A., Horn, H. G., Keck, L., Jiang, J., Scheckman, J., McMurry, P. H., Deng, Z., Zhao, C. S., Moerman, M., Henzing, B., de Leeuw, G., Loschau, G., and Bastian, S.: Mobility particle size spectrometers: harmonization of technical standards and data structure to facilitate high quality long-term observations of atmospheric particle number size distributions, Atmos. Meas. Tech., 5, 657-685, 10.5194/amt-5-657-2012, 2012.