

## Supplemental Information for

# Constraining the particle-scale diversity of black carbon light absorption using a unified framework

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## 10 Data from field and laboratory experiments

*Cappa, et al. 2012.*

The mixing state of internally mixed BC aggregates measured by Cappa *et al.* 2012 (1) was found by first fitting a power-law function to mean  $R_{BC}$  as function of photochemical age ( $-\log([\text{NO}_x]/[\text{NO}_y])$ ), which is shown in figure 1(a). The fitted equation is given by:

$$R_{BC} = (12.215 \pm 0.396) \left( -\log \left( \frac{[\text{NO}_x]}{[\text{NO}_y]} \right) \right)^{0.554 \pm 0.067} \quad (S1)$$

20 Mass absorption cross-section of BC ( $\text{MAC}_{BC}$ ) at wavelength ( $\lambda$ ) of 532 nm is then calculated from figure S-17 of Cappa *et al.* 2012, which shows  $\text{MAC}_{BC}$  enhancement ( $E_{abs}$ ) as a function of photochemical age. Photochemical age was converted to  $R_{BC}$  using equation S1, and  $E_{abs}$  was converted to  $\text{MAC}_{BC}$  using the reported value for  $\text{MAC}_{BC}$  of pure BC ( $7.75 \text{ m}^2/\text{g}$ ). The morphology of particles from this study was inferred using lognormal fits of the size distributions given in figure S-13 of Cappa *et al.* 2012. The single particle BC mass was then calculated assuming BC density of  $1.8 \text{ g}/\text{cm}^3$  (2).

25 *Saliba, et al. 2016.*

Measured  $\text{MAC}_{BC}$  as a function of BC mixing state was taken from Figure 4 of Saliba *et al.* 2016 (3). The mixing state was then converted from organic to black carbon mass ratio (OA:BC) to  $R_{BC}$  using:

$$R_{BC} = \text{OA}:\text{BC} \quad (S2)$$

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The morphology of particles from this study was inferred using lognormal fits of the size distributions given in figure S9 of Saliba *et al.* 2019. The single particle BC mass was then calculated assuming BC density of  $1.8 \text{ g}/\text{cm}^3$  (2).

*Xie, et al. 2019.*

35 Measured  $E_{abs}$  as a function of  $R_{BC}$  was taken from Figure 1a of Xie *et al.* 2019 (4). enhancement was converted to  $\text{MAC}_{BC}$  using a reference  $\text{MAC}_{BC}$  value of  $6.55 \text{ m}^2/\text{g}$  at  $\lambda = 630 \text{ nm}$ , given that the authors state that the thermodenuder-derived  $E_{abs}$  was well correlated with  $E_{abs}$  calculated using reference  $\text{MAC}_{BC}$  (4).

*Cui, et al. 2016.*

40 Measured  $E_{abs}$  as a function of BC mixing state was taken from Figure 5c of Cui *et al.* 2016 (5). The mixing state was converted from organic to elemental carbon ratio (OC/EC) to  $R_{BC}$  using:

$$R_{BC} = \text{OC}/\text{EC} \quad (S3)$$

45 Absorption enhancement was converted to  $\text{MAC}_{BC}$  using  $\text{MAC}_{BC}$  of pure BC =  $4.02 \text{ m}^2/\text{g}$  at  $\lambda = 678 \text{ nm}$ , given in figure 2 of Cui *et al.* 2016 (5).

*Denjean, et al. 2020.*

50 Measured  $E_{\text{abs}}$  as a function of BC mixing state was taken from Figure 2 of Denjean *et al.* 2020 (6). Absorption enhancement was converted to  $\text{MAC}_{\text{BC}}$  using  $\text{MAC}_{\text{BC}}$  of pure BC =  $7.7 \text{ m}^2/\text{g}$  at  $\lambda = 550 \text{ nm}$ . The morphology of particles from this study was inferred using lognormal fits of the size distributions given in figure 1 of Denjean *et al.* 2020. The average mass was then calculated assuming BC density of  $1.8 \text{ g}/\text{cm}^3$  (2).

*Zanatta, et al. 2018.*

55 Average  $\text{MAC}_{\text{BC}}$  and  $R_{\text{BC}}$  were found using reported values for average coating thickness, coating density, and  $\text{MAC}_{\text{BC}}$  in Zanatta *et al.* 2018 (7). The morphology of particles from this study was inferred using reported size distributions given in Table 2 of Zanatta *et al.* 2018. The single particle BC mass was then calculated assuming BC density of  $1.8 \text{ g}/\text{cm}^3$  (2).

*Liu, et al. 2015.*

60 Measured  $\text{MAC}_{\text{BC}}$  as a function of BC mixing state was taken from Figure S2 of Liu *et al.* 2015 (8). We utilize  $\text{MAC}_{\text{BC}}$  derived using standardized major axis at  $\lambda = 781 \text{ nm}$  in order to avoid potential influence of absorbing coatings on  $\text{MAC}_{\text{BC}}$ . The morphology of particles from this study was inferred using the limits of BC diameter given in Figure 1d of Liu *et al.* 2015. The single particle BC mass was then calculated assuming BC density of  $1.8 \text{ g}/\text{cm}^3$  (2).

*Cappa, et al. 2019.*

65 Measured  $\text{MAC}_{\text{BC}}$  as a function of BC mixing state was taken from Figure S4 and S6 of Cappa *et al.* 2019 (9). We analyze measurements of  $\text{MAC}_{\text{BC}}$  at  $\lambda = 532 \text{ nm}$  only.

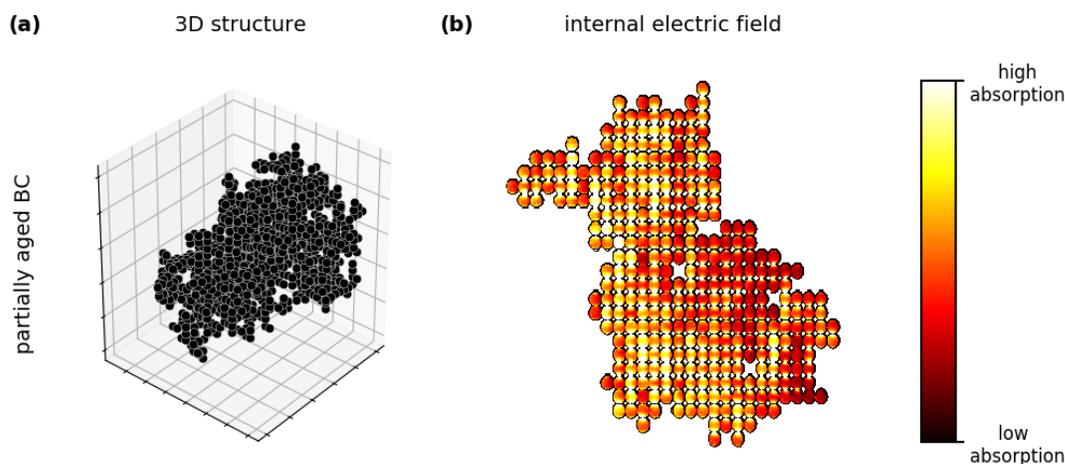
*Shiraiwa, et al. 2010.*

70 Measured  $E_{\text{abs}}$  as a function of BC mixing state was taken from Figure 2 of Shiraiwa *et al.* 2010 (10). The mixing state was converted from the ratio of total particle diameter to black carbon diameter using BC density of  $1.8 \text{ g}/\text{cm}^3$  and coating density of  $1.2 \text{ g}/\text{cm}^3$  (2). The morphology of particles from this study was inferred using reported BC core diameter.

*Zhang, et al. 2018.*

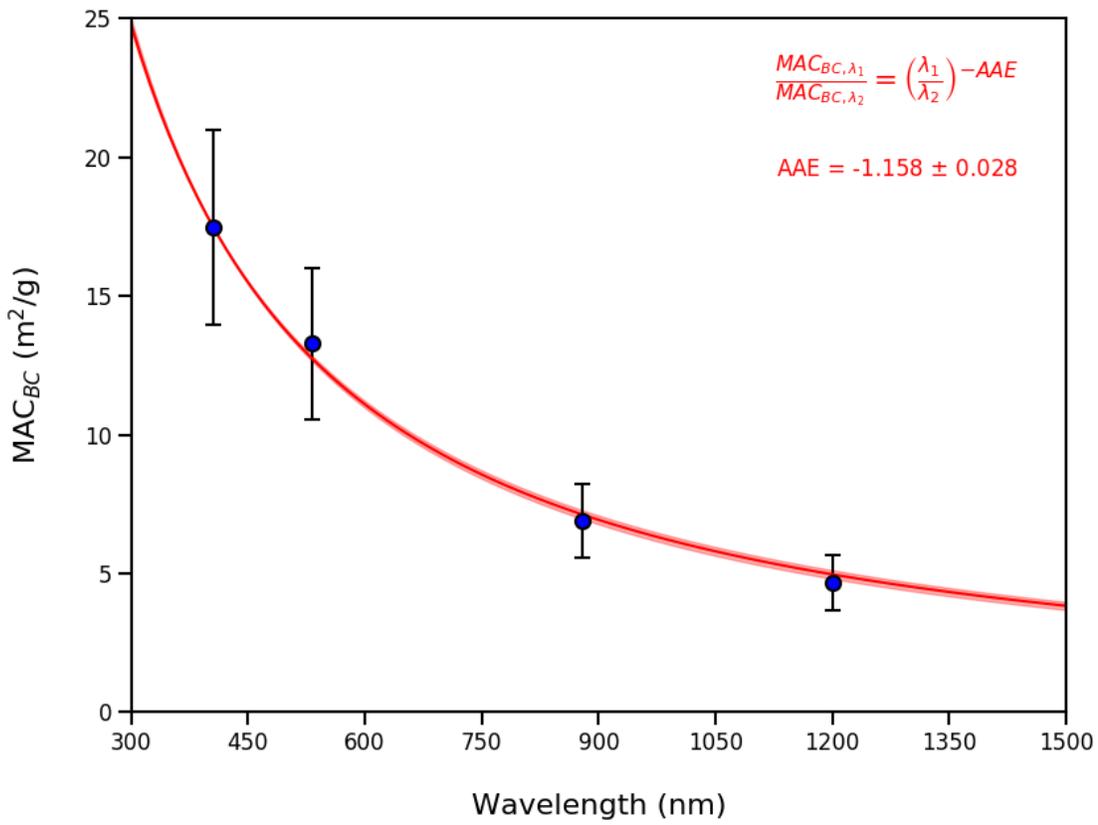
75 Measured  $E_{\text{abs}}$  as a function of BC mixing state was taken from Figure S3 of Zhang *et al.* 2018 (11). The mixing state was converted from organic to elemental carbon mass ratio to  $R_{\text{BC}}$  using equation S3. Absorption enhancement was converted to  $\text{MAC}_{\text{BC}}$  using  $\text{MAC}_{\text{BC}}$  of pure BC =  $4.7 \text{ m}^2/\text{g}$  at  $\lambda = 880 \text{ nm}$ . The morphology of particles from this study was inferred using the reported range of BC core diameters (100-150 nm).

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**Figure S1: (a) 3D structure of a partially collapsed aggregate. (b) Internal light absorption of the partially collapsed aggregate with light incident from the left.**

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**Figure S2: Data used to calculate AAE for fractal BC aggregates with  $\rho_{BC} \leq 1$ , error bars show one standard deviation. Solid line shows fitted equation given in figure, error of AAE is reflective of 95% confidence interval.**