

Optical properties of black and brown carbon and their contribution to aerosol light absorption

Sang-Woo Kim

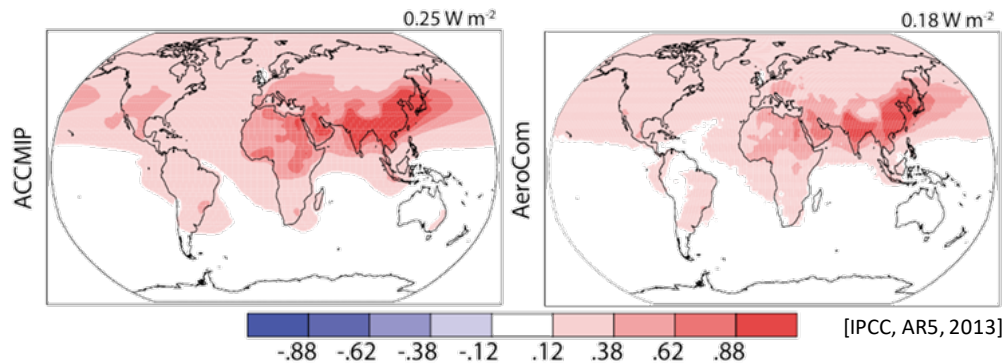
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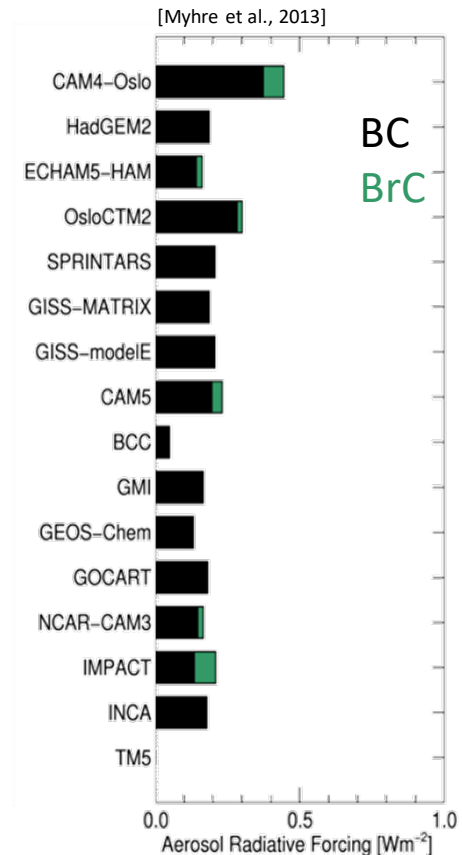
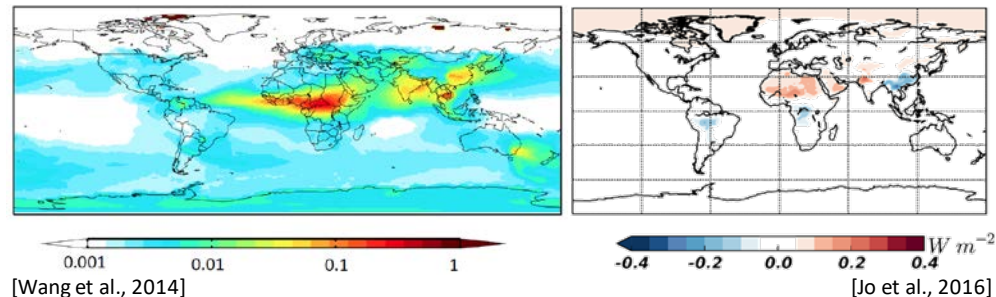
Chaeyoon Cho, Meehye Lee, Saehee Lim, Wenzheng Fang, Örjan Gustafsson,
August Andersson, Rokjin J. Park and Patrick J. Sheridan

Black Carbon (BC) and Brown Carbon (BrC)

Direct radiative forcing by Black Carbon (BC)



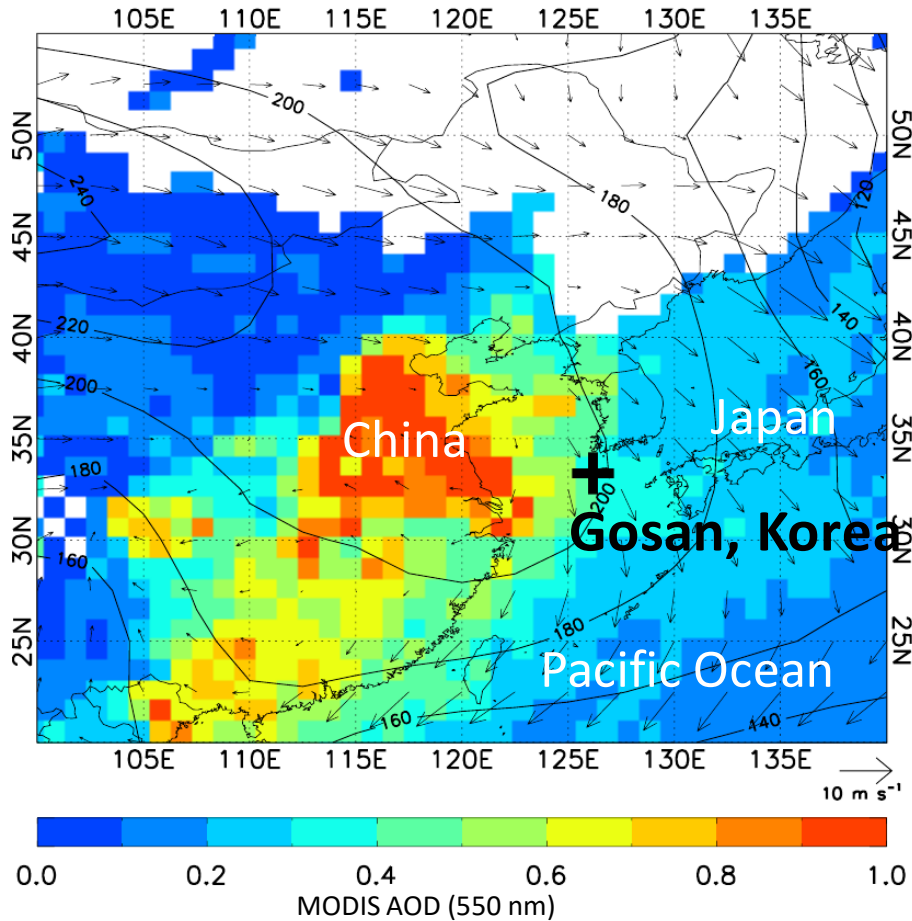
Direct radiative forcing by Brown Carbon (BrC)



$$\text{Absorption Coefficient [m}^{-1}\text{]} = \text{Mass [g m}^{-3}\text{]} \times \text{Mass Absorption Cross-Section [MAC; m}^2 \text{g}^{-1}\text{]}$$

- MAC of $7.5 \pm 1.2 \text{ m}^2 \text{g}^{-1}$ for fresh BC [Bond and Bergstrom, 2006] is adapted to most global aerosol models.
- Some models have also used a broad range of MAC_{BC} ranging from 2.3 to $18 \text{ m}^2 \text{g}^{-1}$ to account for the de-coating or coating-enhancement of ambient BC absorption.

Outline



Surface In-situ measurements

- Characterize the light absorption optical properties of BC and BrC from field measurements
- Estimates of MAC of BC (MAC_{BC}) and BrC (MAC_{BrC})
- Determine the contributions of BC and BrC to total aerosol light absorption coefficient

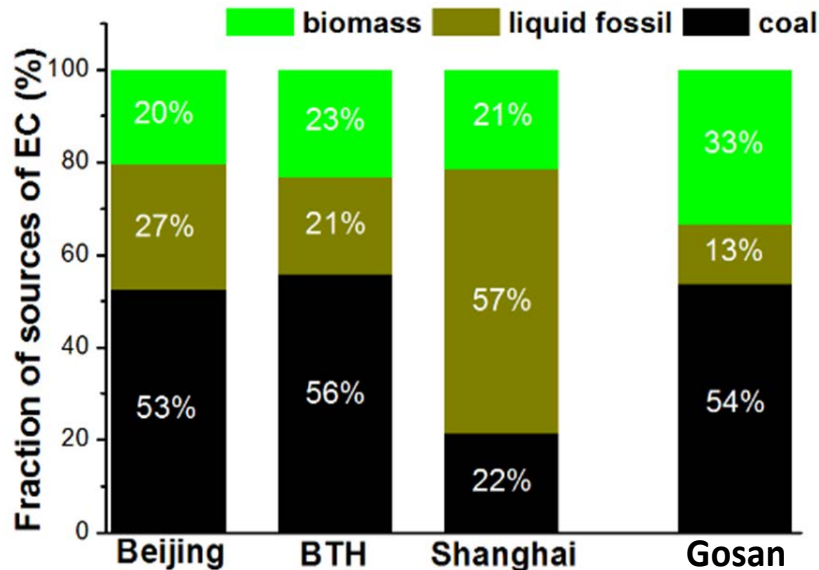
Column AERONET measurements

- Constrain the AOD and AAOD of BC and BrC over East Asia
- Determine the contributions of BC and BrC to AAOD

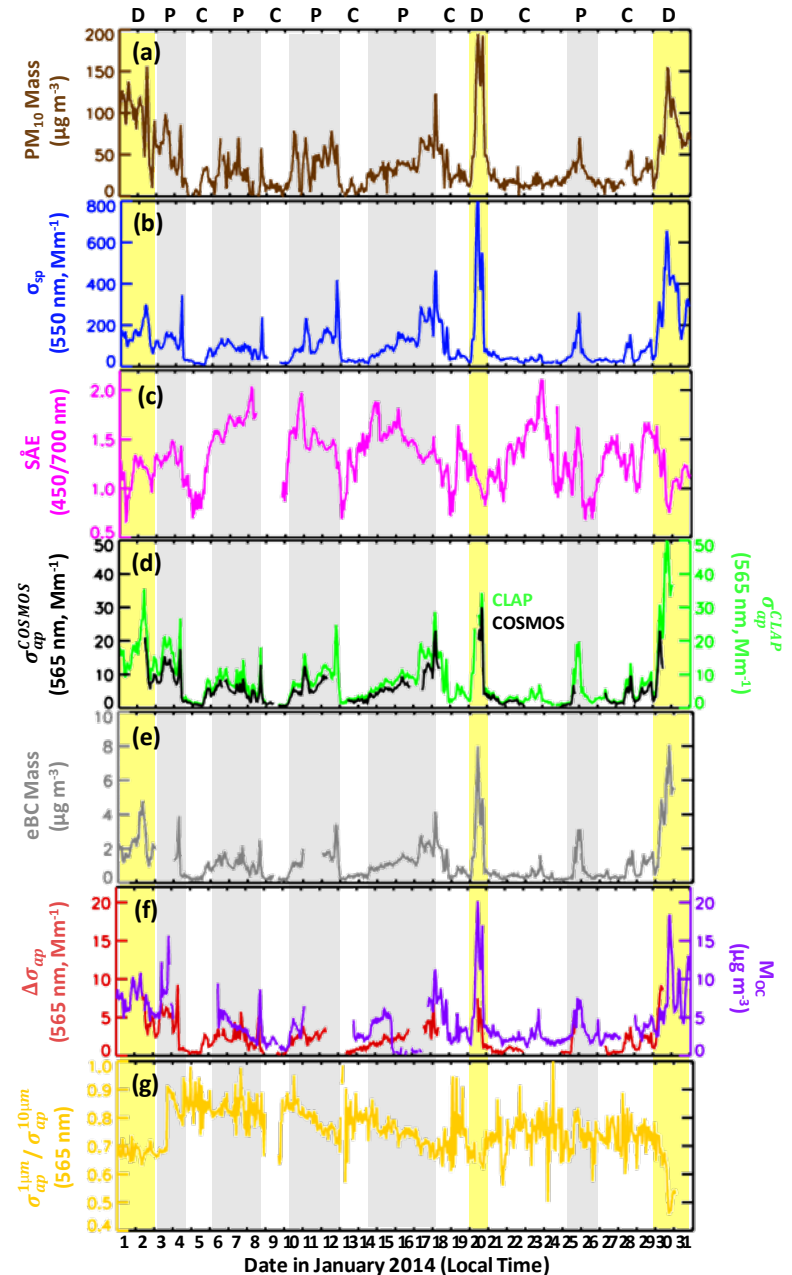
In-Situ Aerosol Light Absorption Measurements during GoPoEx 2014

- Aerosol Absorption Coefficient (*Aethalometer, CLAP, COSMOS*)
- EBC Mass Concentration (*Aethalometer*)
- Aerosol Scattering Coefficient (*Nephelometer*)
- OC, EC mass concentration (*Sunset OC/EC analyzer & PM2.5 sampler*)
- Carbon Isotope Analysis

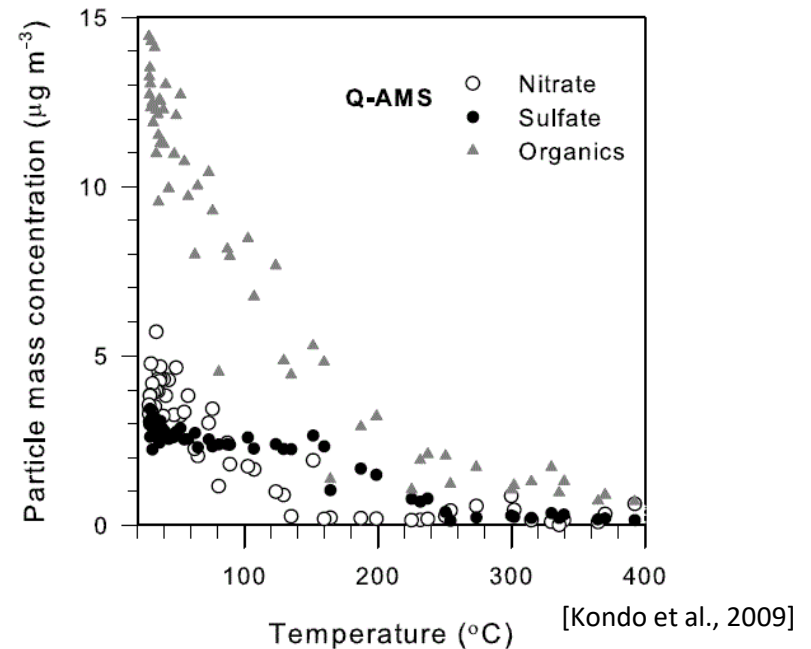
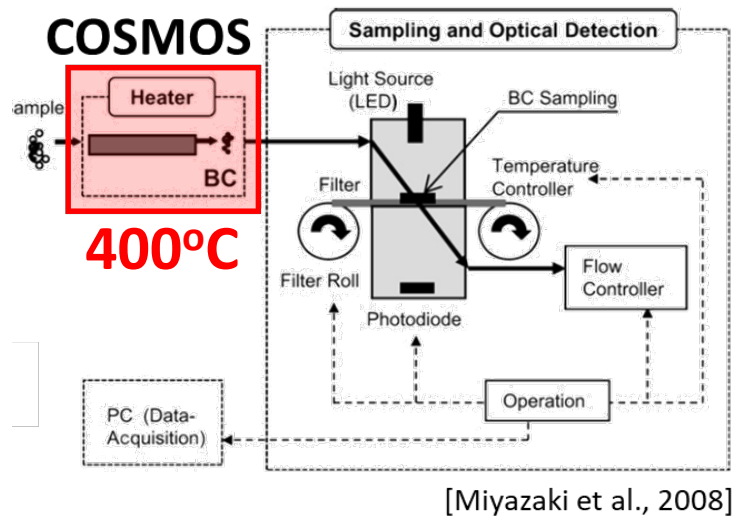
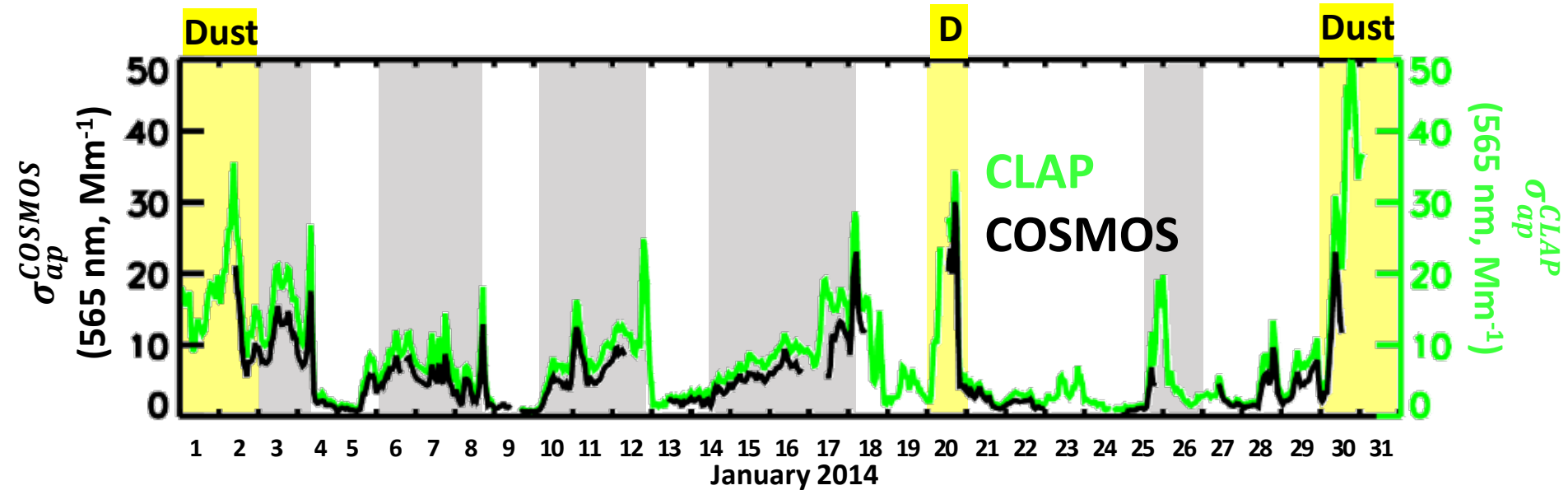
Carbon Isotope Analysis



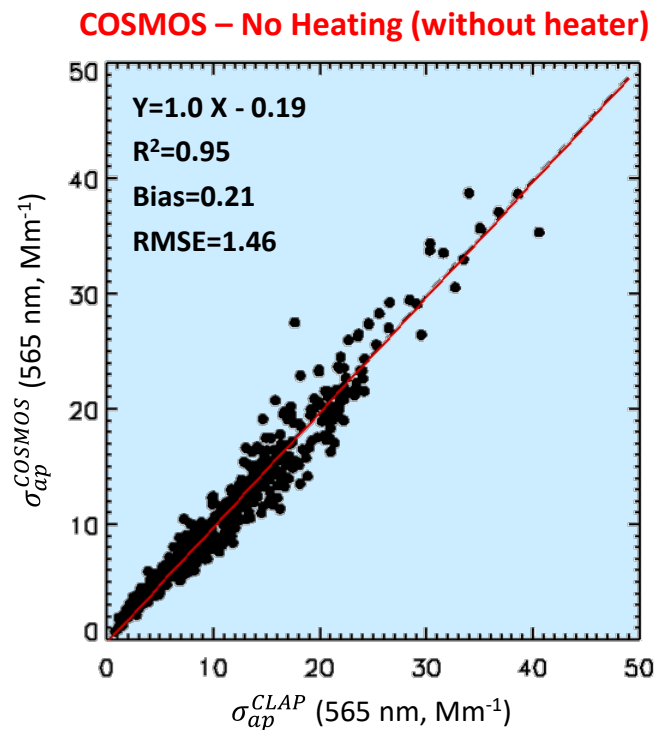
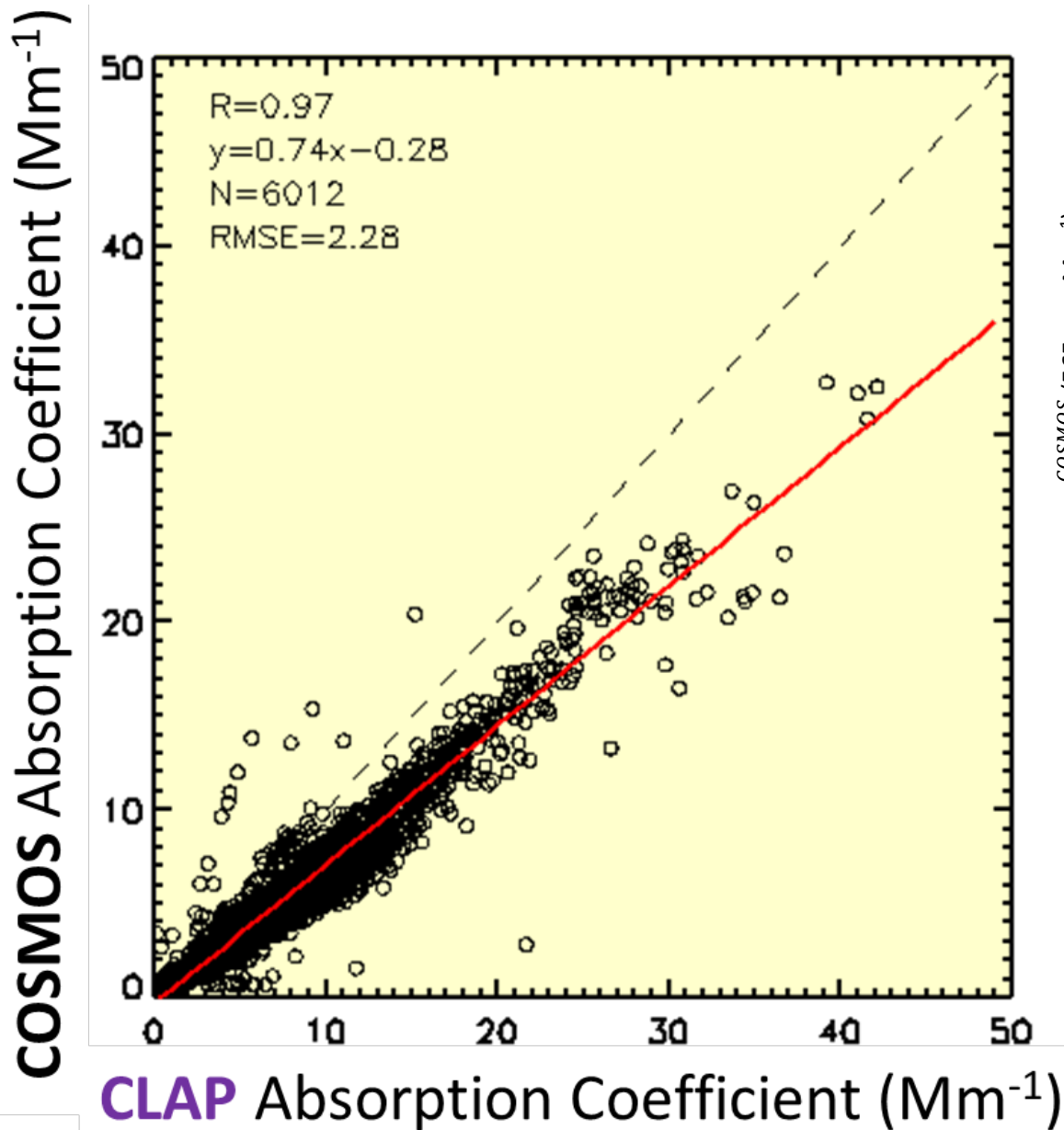
[Fang et al., Scientific Reports, 2017]



Aerosol Light Absorption Properties during GoPoEx 2014



Light Absorption Measurements by CLAP and COSMOS



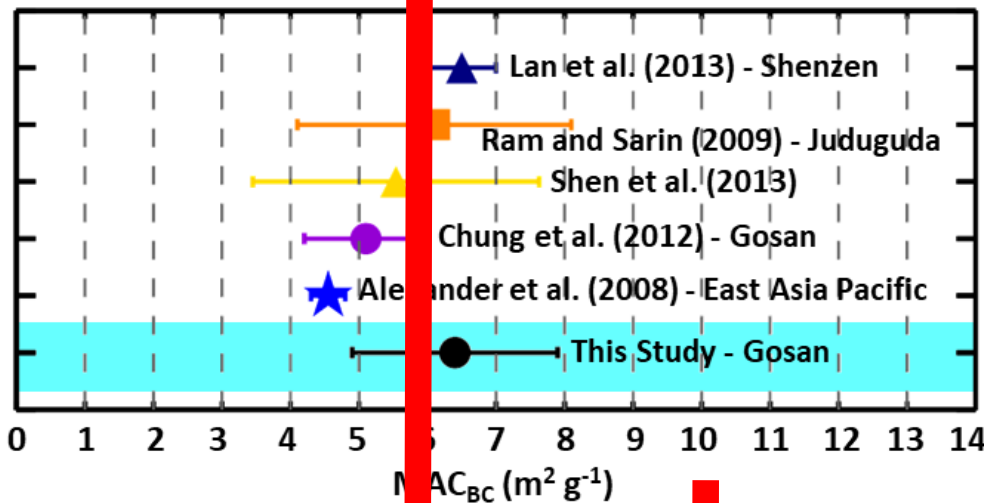
MAC_{BC}

MAC_{BC} (565 nm)

$$= \frac{\sigma_{ap}^{COSMOS}(565 \text{ nm})}{\text{EC mass conc.}}$$

= 6.4 m² g⁻¹

Background



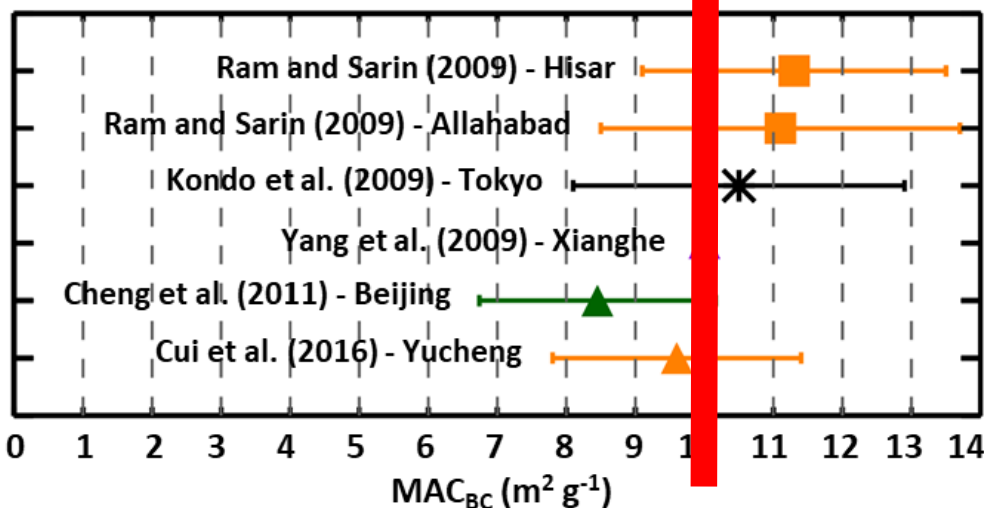
Wavelength (nm)

520
532
550
565
632
650
678

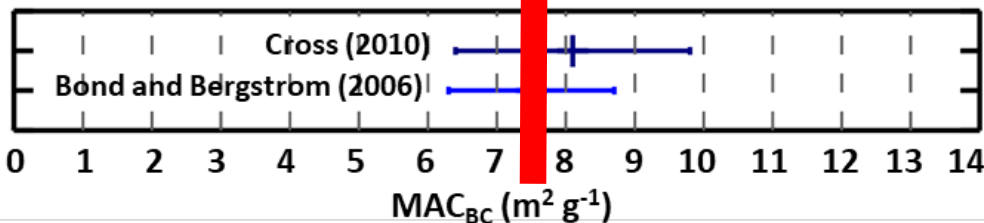
Location

● Korea
▲ China
■ India
✱ Japan
★ East Asia Pacific

Urban



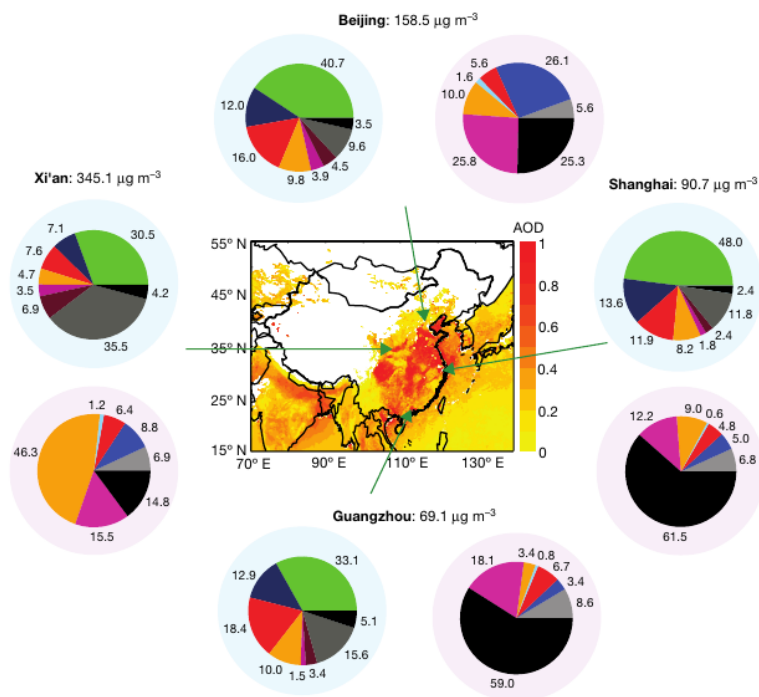
Lab experiment



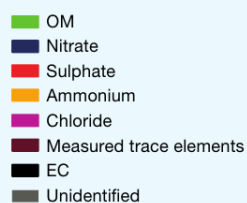
MAC_{BC} observed in Urban Areas

Abundant primary BC + Co-emitted non BC constituents

from motor vehicles, industrial factories, heating (winter)



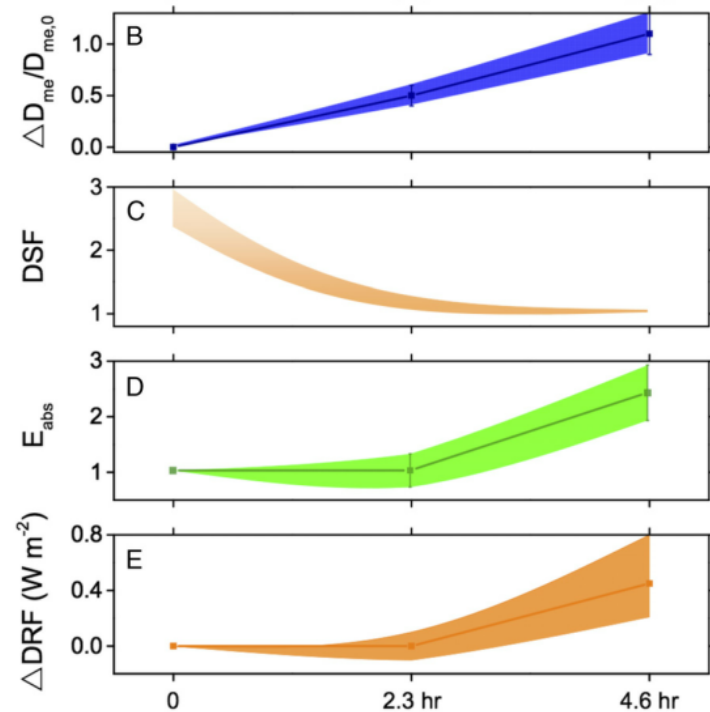
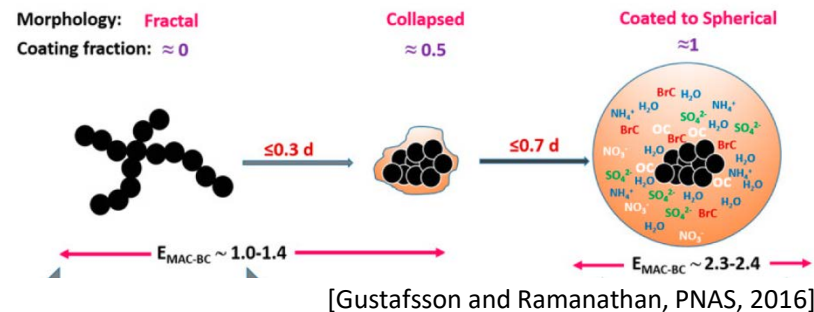
Composition (%)



Sources/factors (%)

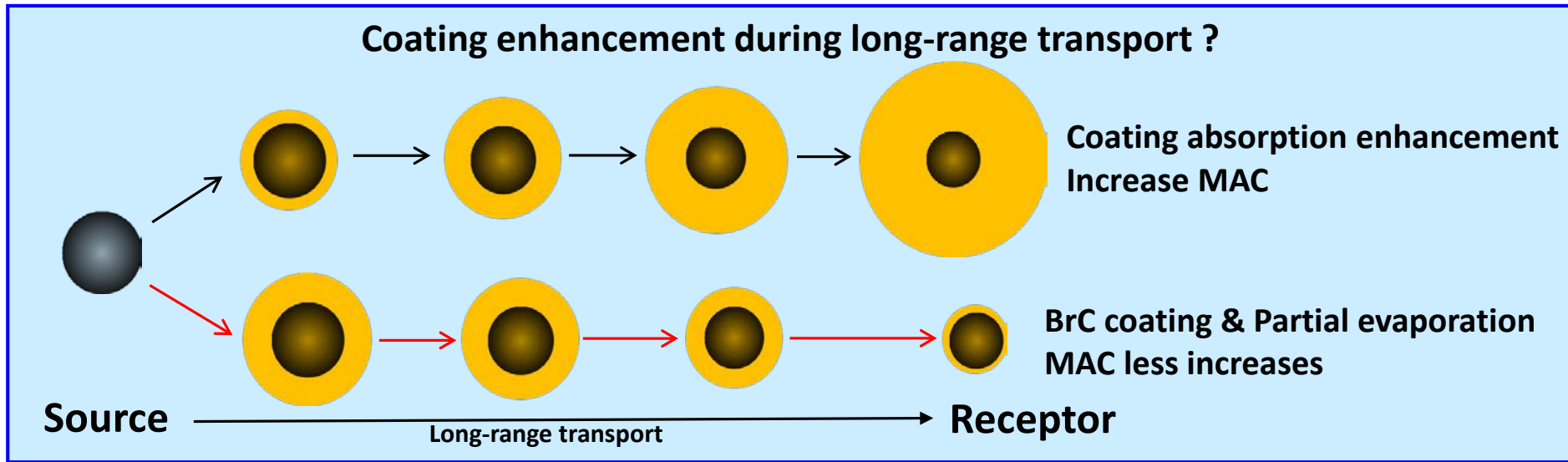


Coating-enhancement of BC absorption

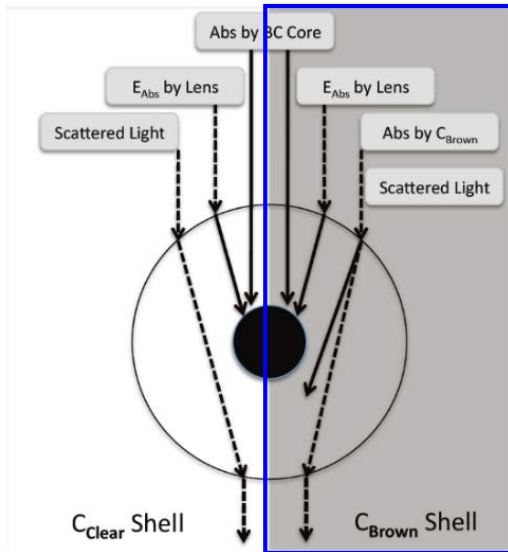


[Peng et al., PNAS, 2016]

MAC_{BC} observed in Background Sites



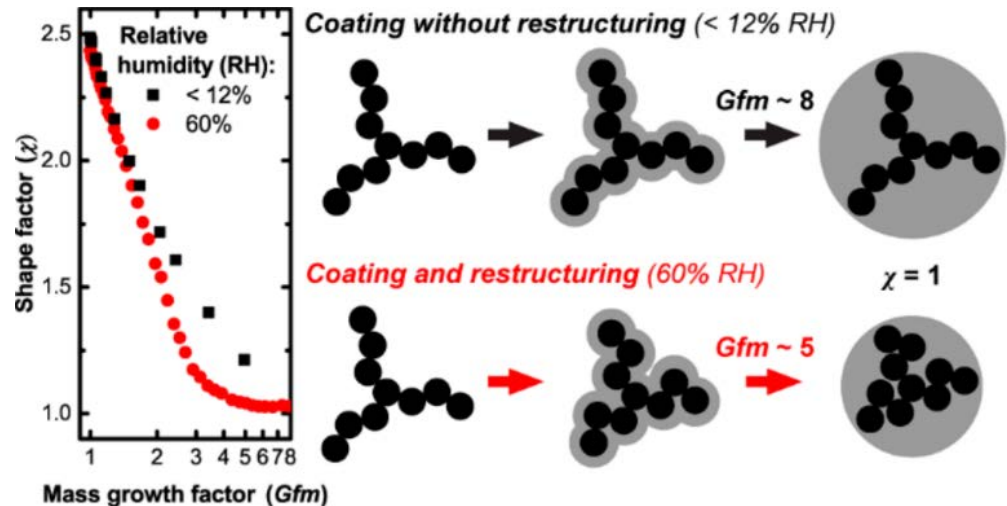
Coating effect of BC absorption by BrC



[Lack et al, ACP, 2010]

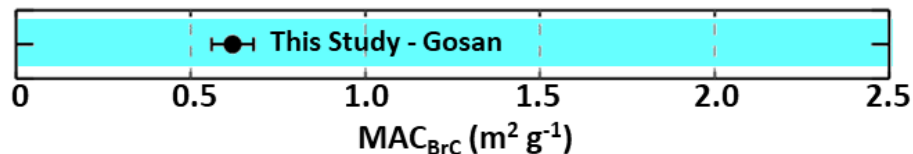
Less coagulation & partial evaporation of the coating materials under dry conditions

[Leung et al., EST, 2017]

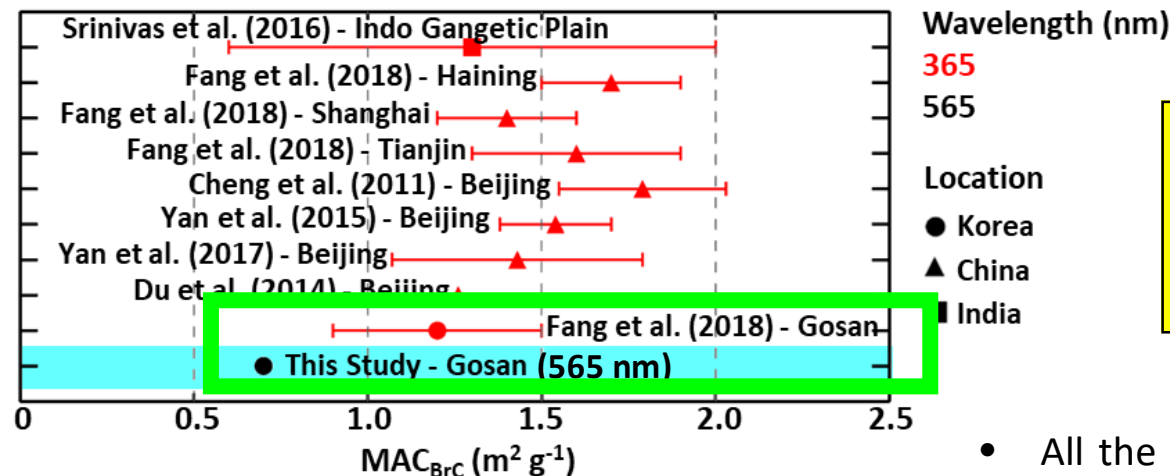


MAC_{BrC}

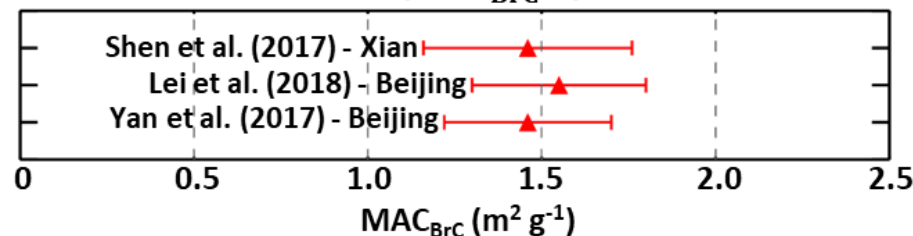
In-situ measured BrC ($MAC_{BrC}^{in-situ}$) at 565 nm



Water-extracted BrC (MAC_{BrC}^{WSOC})



Methanol-extracted BrC (MAC_{BrC}^{MeOH})



$$\begin{aligned}
 &MAC_{BrC}^{insitu} (565 \text{ nm}) \\
 &= \frac{\Delta\sigma_{ap}(565 \text{ nm})}{M_{BrC}} \\
 &= \frac{\Delta\sigma_{ap}(565 \text{ nm})}{M_{eBC} - M_{BC}} \\
 &= \frac{\Delta\sigma_{ap}(565 \text{ nm})}{M_{eBC} - (\sigma_{ap}^{COSMOS} \div MAC_{BC})} \\
 &= 0.62 \text{ m}^2 \text{ g}^{-1}
 \end{aligned}$$

$$\begin{aligned}
 &MAC_{BrC}^{WSOC} (565 \text{ nm}) \\
 &= \frac{\Delta\sigma_{ap}(565 \text{ nm})}{M_{WSOC}} = 0.7 \text{ m}^2 \text{ g}^{-1}
 \end{aligned}$$

- All the results given in here have substantial experimental (e.g., in-situ/ex-situ optical absorption measurement) and analytical (e.g., extraction of BrC) uncertainties, in addition to a lack of understanding of the sources, chemical transformations, and associated optical properties (e.g., photo-bleaching effect; Fang et al., 2017) of BrC.

ATMOSPHERIC SCIENCE

Photochemical degradation affects the light absorption of water-soluble brown carbon in the South Asian outflow

Sanjeev Dasari¹, August Andersson¹, Srinivas Bikkina¹, Henry Holmstrand¹, Krishnakant Budhavant^{1,2,3}, Sreedharan Satheesh³, Eija Asmi^{4,5}, Jutta Kesti⁴, John Backman⁴, Abdus Salam⁶, Deewan Singh Bisht⁷, Suresh Tiwari⁷, Zahid Hameed^{2,8}, Örjan Gustafsson^{1*}

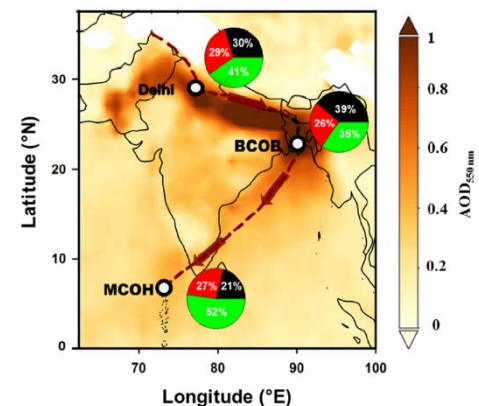


Fig. 1. Meteorology and general aerosol characteristics during the SAPOEX-16.

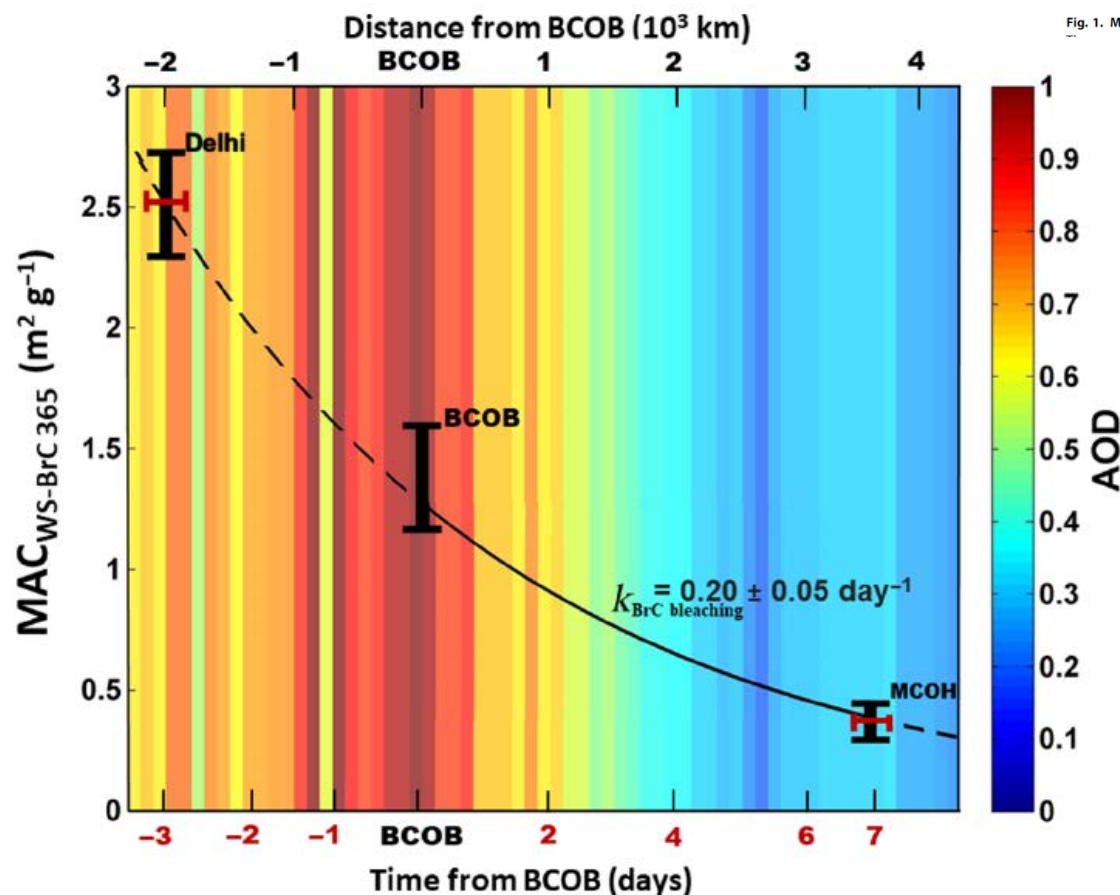
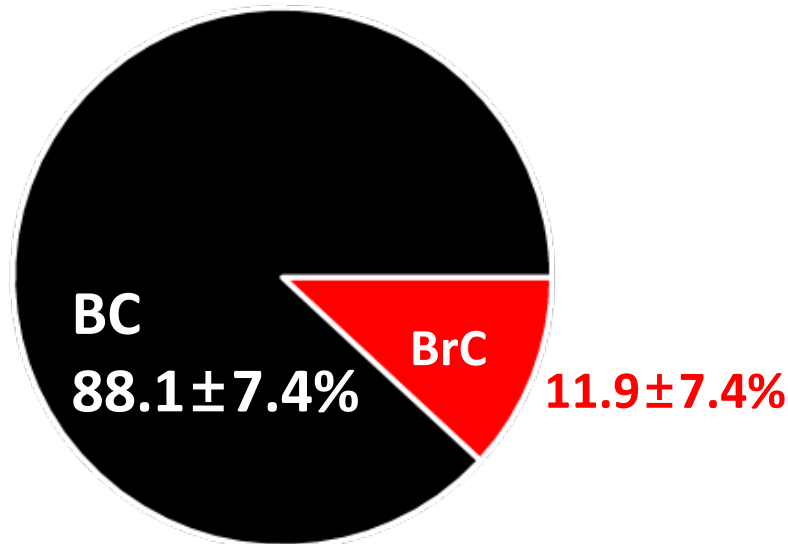


Fig. 5. Bounding the light absorption of WS-BrC in the South Asian outflow.

Contribution of BC and BrC to Light Absorption Coefficient

Total Absorption (565nm)



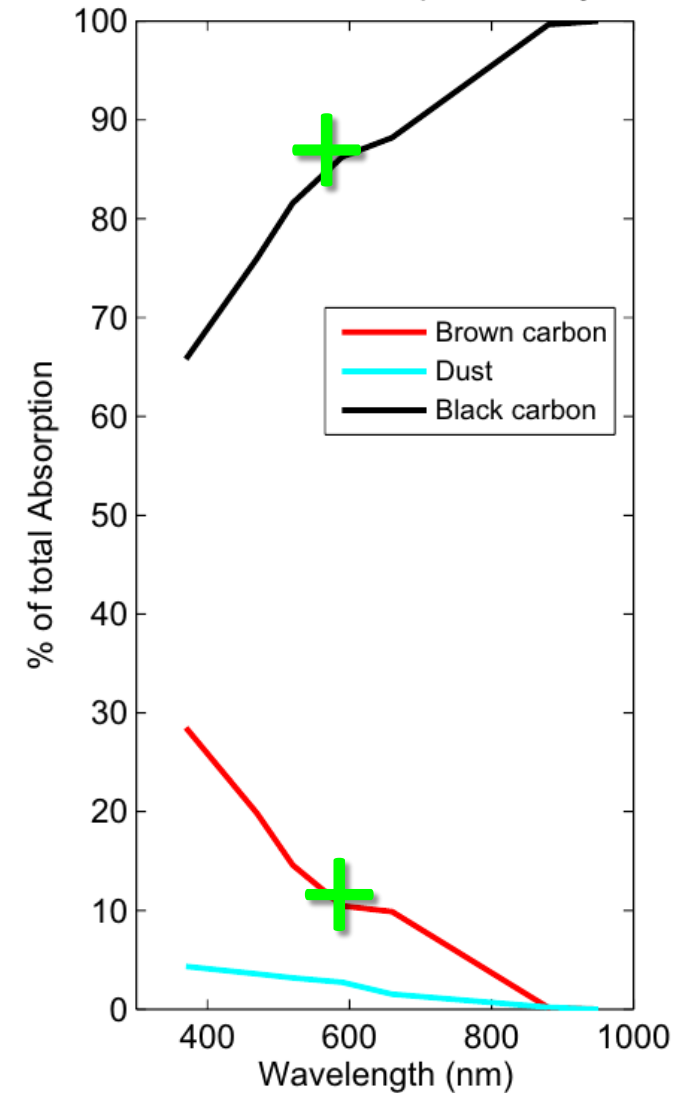
$MAC_{BC} = 6.4 \text{ m}^2 \text{ g}^{-1}$

$MAC_{BrC} = 0.62 \text{ m}^2 \text{ g}^{-1}$

BrC contribution estimates

- Yang et al., 2009: **10 %** at visible wavelength
- Bahadur et al., 2012: **less than 10 %** at 675 nm
- Feng et al., 2013: **18 %** at 500 nm

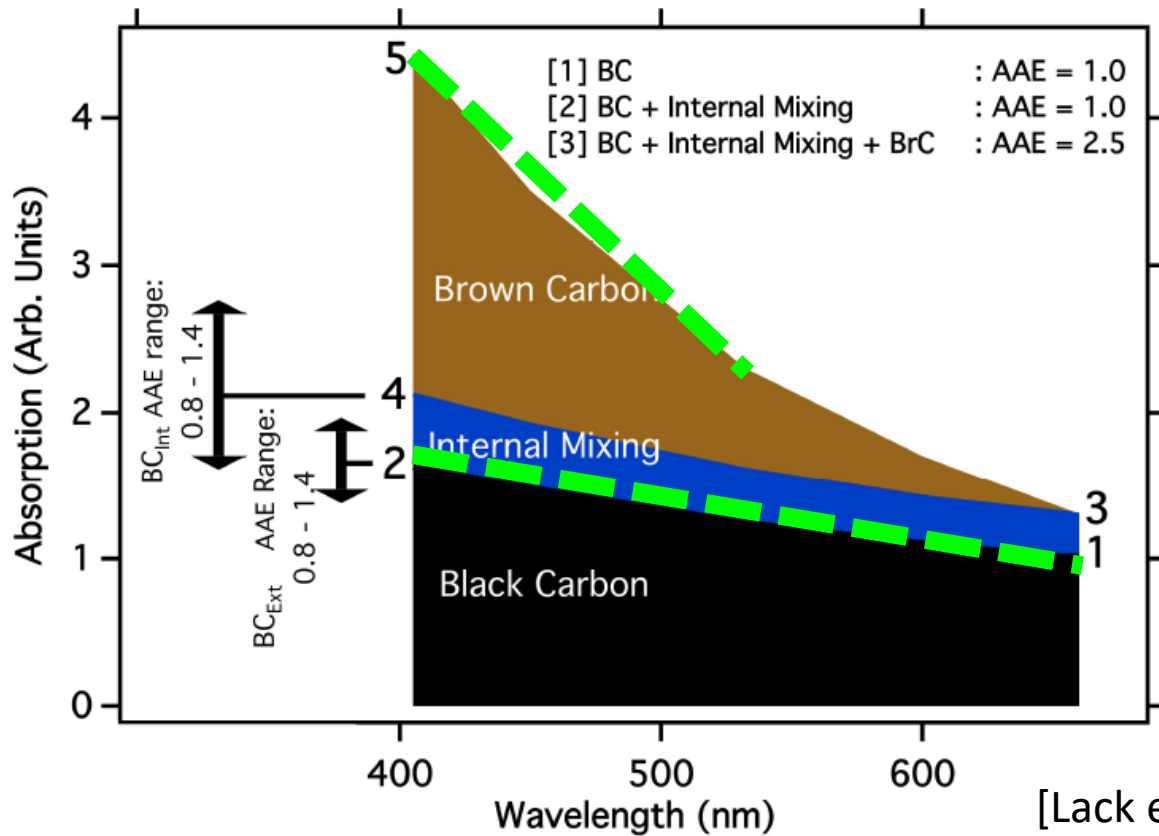
Contribution to Total Absorption – Project Median



[Yang et al., ACP, 2009]

Column Aerosol Light Absorption from AERONET

- Aerosol optical depth (AOD) and absorption AOD (AAOD) at multiple wavelengths
- AOD and AAOD are more closely related to the column abundance of BC and BrC

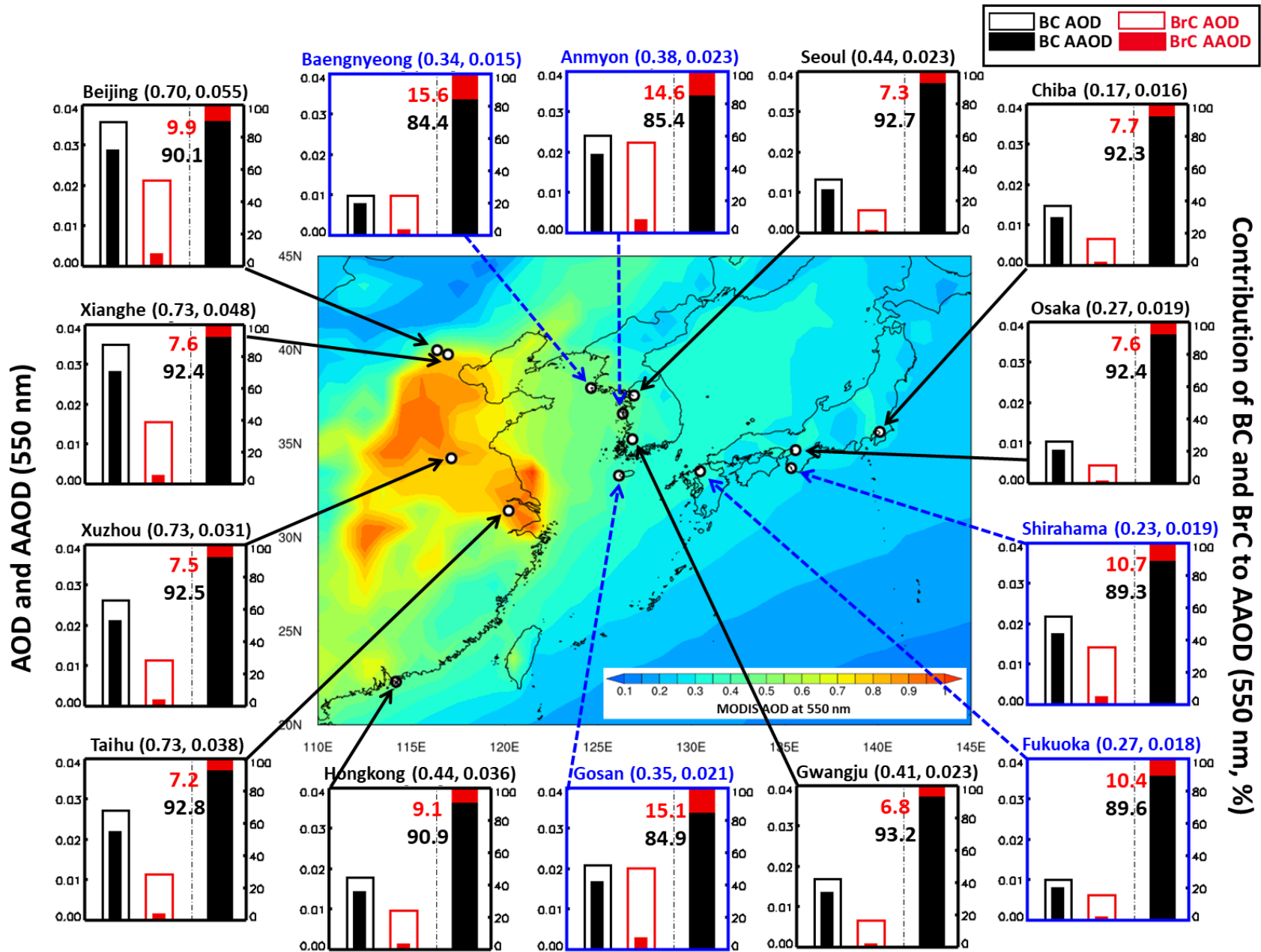


- BrC primarily absorbs solar radiation at near-ultraviolet wavelengths, and has negligible to weak light absorption at longer wavelengths.
- BrC's AAE significantly differs from that of BC.

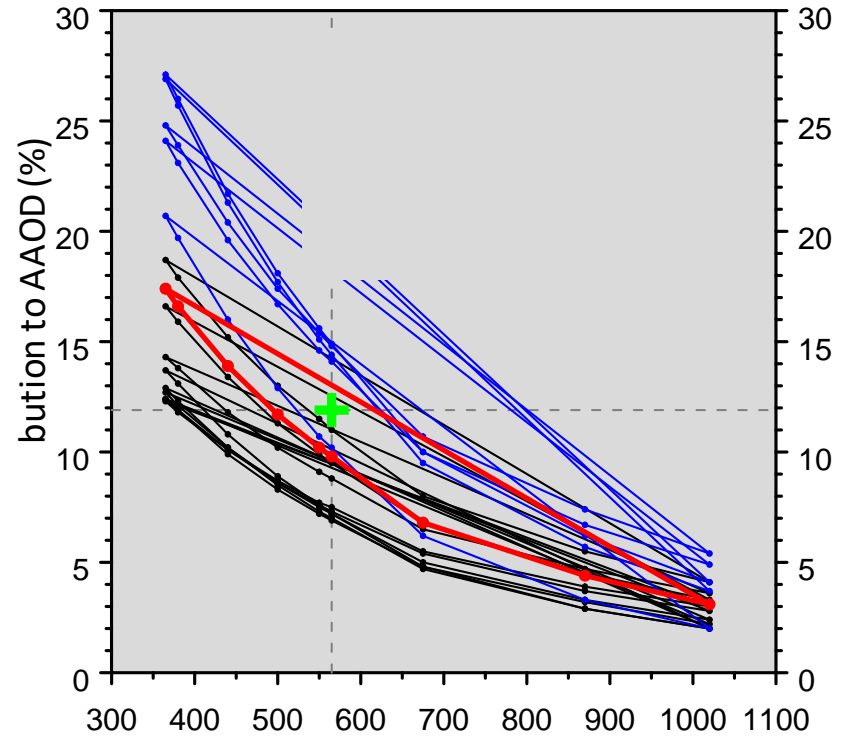
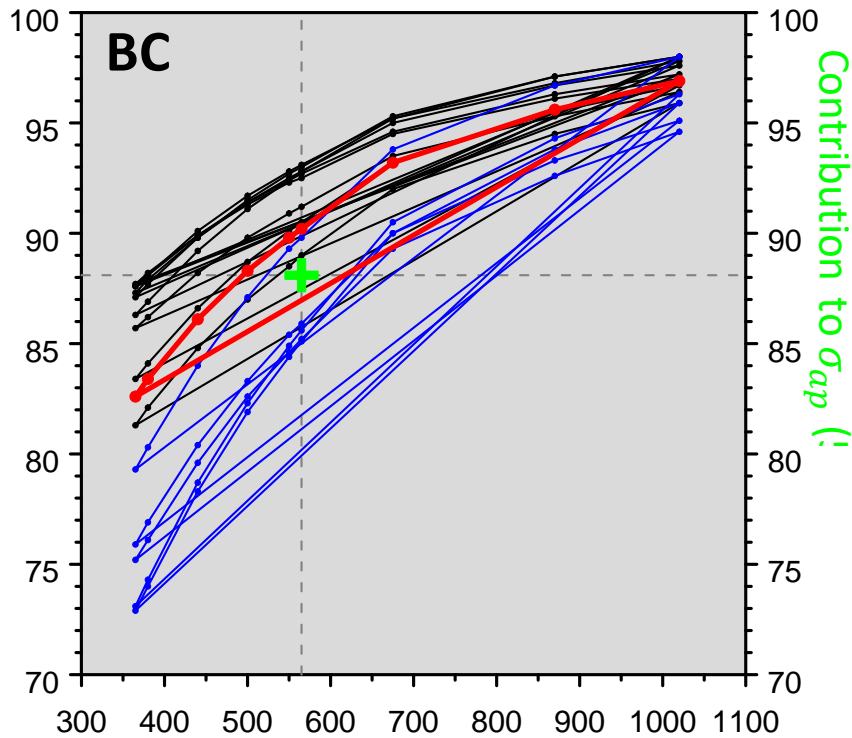
Wavelength Dependency of Absorption for BC and BrC

- 1) **Removed the dust absorption** by the wavelength dependency of absorption for dust particles, because the dust AOD is nearly wavelength independent, and dust AAOD increases steeply at shorter wavelengths compared with carbonaceous aerosols (Russell et al., 2010; Bahadur et al., 2012; Chung et al., 2012b).
- 2) The AAOD, calculated as “ $AAOD=(1-SSA)\times AOD$ ”, was **separated to the individual contributions of BC ($AAOD_{BC}$) and BrC ($AAOD_{BrC}$)** using their spectral dependence on absorption, normally represented by the absorption Ångström exponent (AAE; the so-called “AAE approach;” Bahadur et al., 2012; Chung et al., 2012b).
- 3) The AAOD for BC ($AAOD_{BC}$) and BrC ($AAOD_{BrC}$) were finally converted into **the AOD for BC (AOD_{BC}) and BrC (AOD_{BrC})**, respectively, by using the SSA for BC and BrC suggested by Magi et al. (2009) and Chung et al. (2012).

Contribution of BC and BrC to AOD



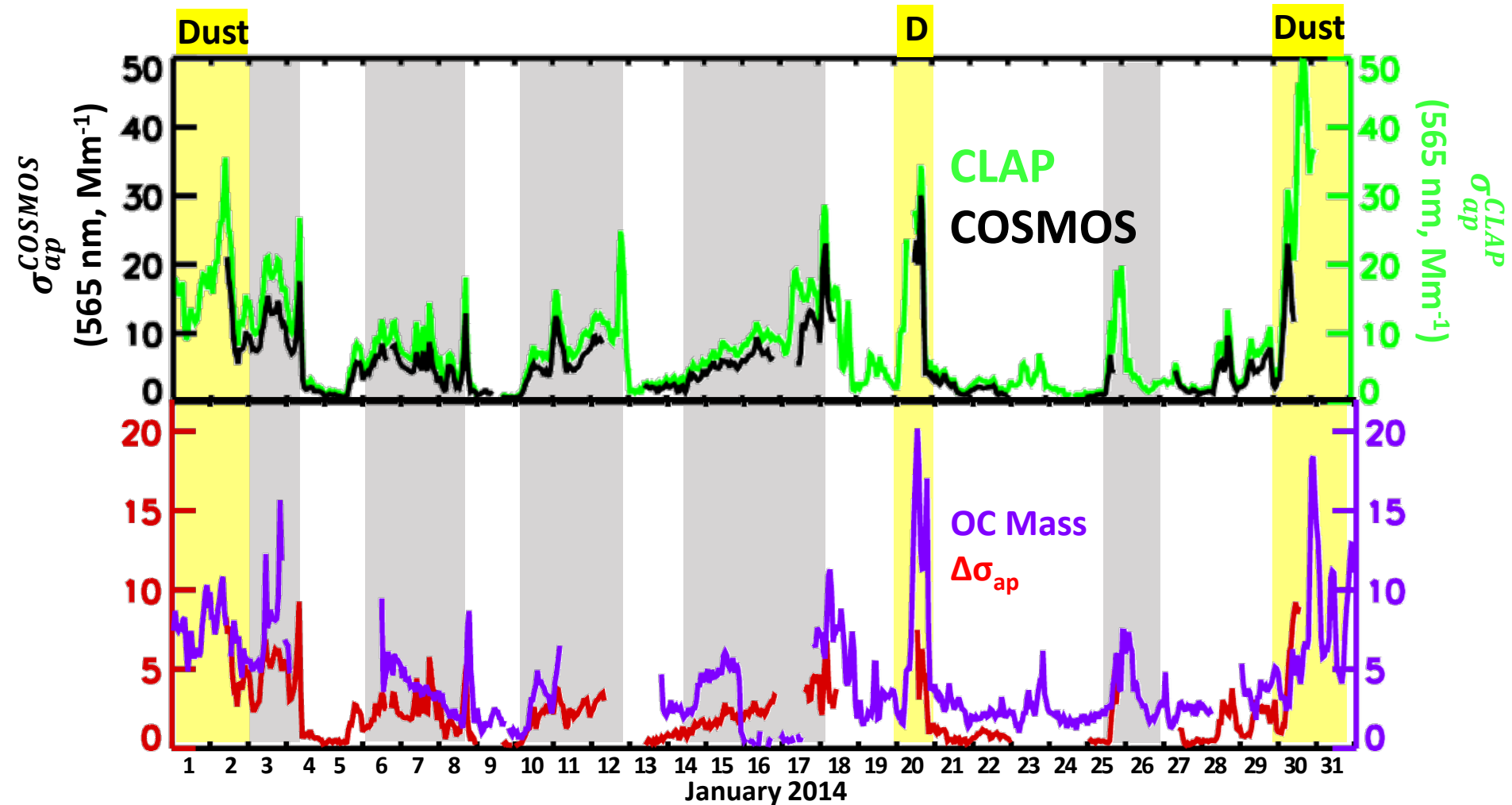
Contribution of BC and BrC to AAOD as a function of Wavelength



Short Summary

- Aerosol absorption coefficient from COSMOS was approximately 15-20% lower due to volatile light-absorbing (“BrC”) aerosols.
- The **MAC** of **BC** and **BrC** was estimated to be **6.4 m² g⁻¹** and **0.62 m² g⁻¹** from in-situ aerosol measurements during GoPoEx 2014, respectively.
- The **contribution of BC to total aerosol light absorption coefficient** at 565 nm was estimated to be **88 %**, while **BrC** accounted for **12 %**.
- The contribution of **BC** and **BrC** to AAOD at 550 nm, constrained by AERONET observations at 14 sites by using different spectral dependences, was estimated to be **85 %** and **15 %**, respectively.
- A high BC contribution to AAOD appeared in urban sites, whereas the contribution of BrC to AAOD was higher in background sites.

Aerosol Light Absorption Properties during GoPoEx 2014

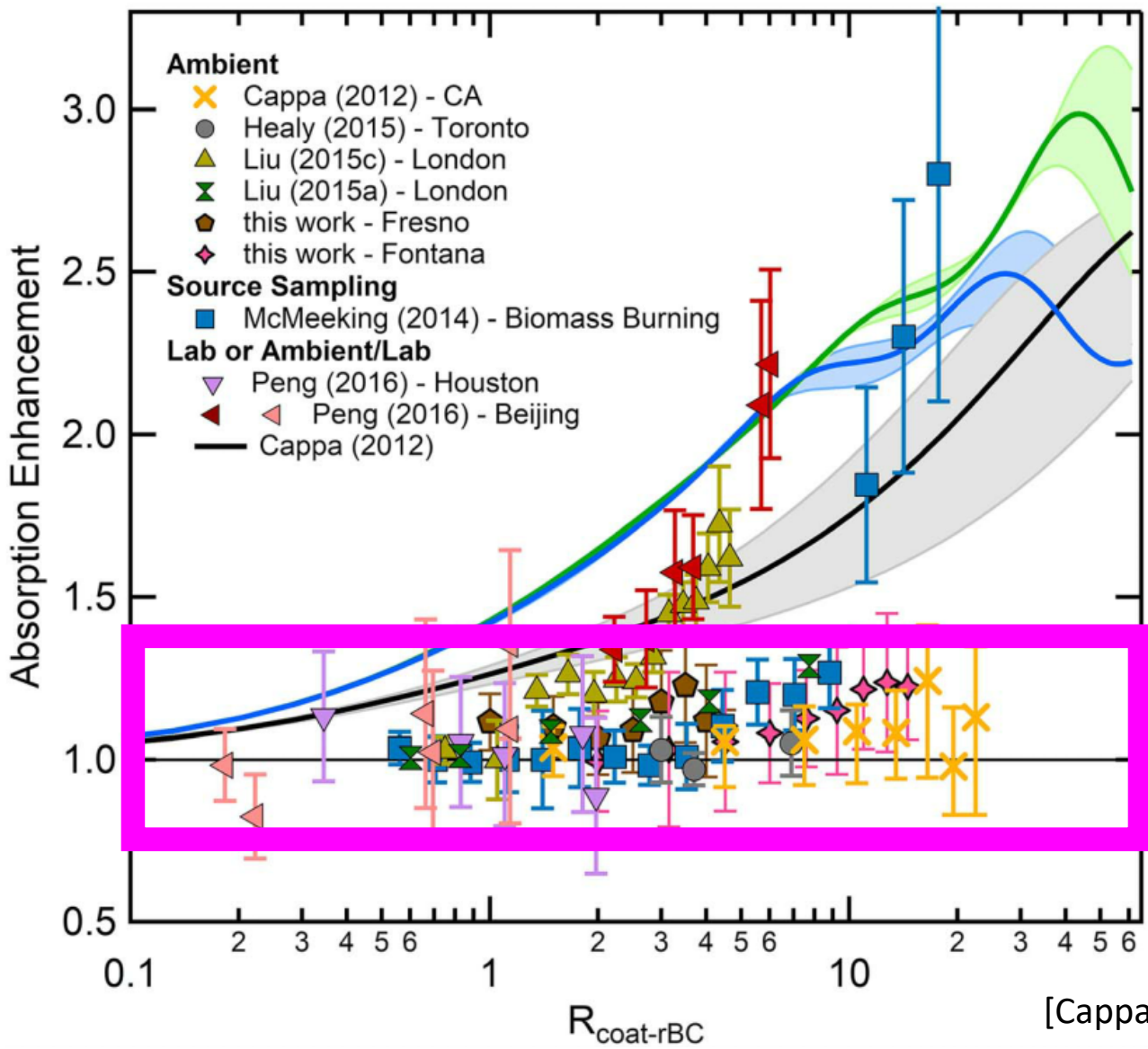


- Absorption coefficient difference ($\Delta\sigma_{ap}$; i.e., BrC absorption) is calculated by:

$$\Delta\sigma_{ap} = \sigma_{ap}(CLAP) - \sigma_{ap}(COSMOS)$$

- Temporal variation of BrC absorption is well match with OC mass concentration.

Discrepancy between modeled and observed absorption enhancement



[Cappa et al., JGR, 2019]

Contribution of BC and BrC to AAOD as a function of Wavelength

