PAUL SCHERRER INSTITUT

LABORATORY OF ATMOSPHERIC CHEMISTRY

Black and brown carbon: small particles with big issues

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20170425

Outline

- General introduction of Black carbon and Brown carbon
- Black carbon research—Introduction and my ongoing projects
- Brown carbon absorption— an report of previous work

Atmospheric aerosols

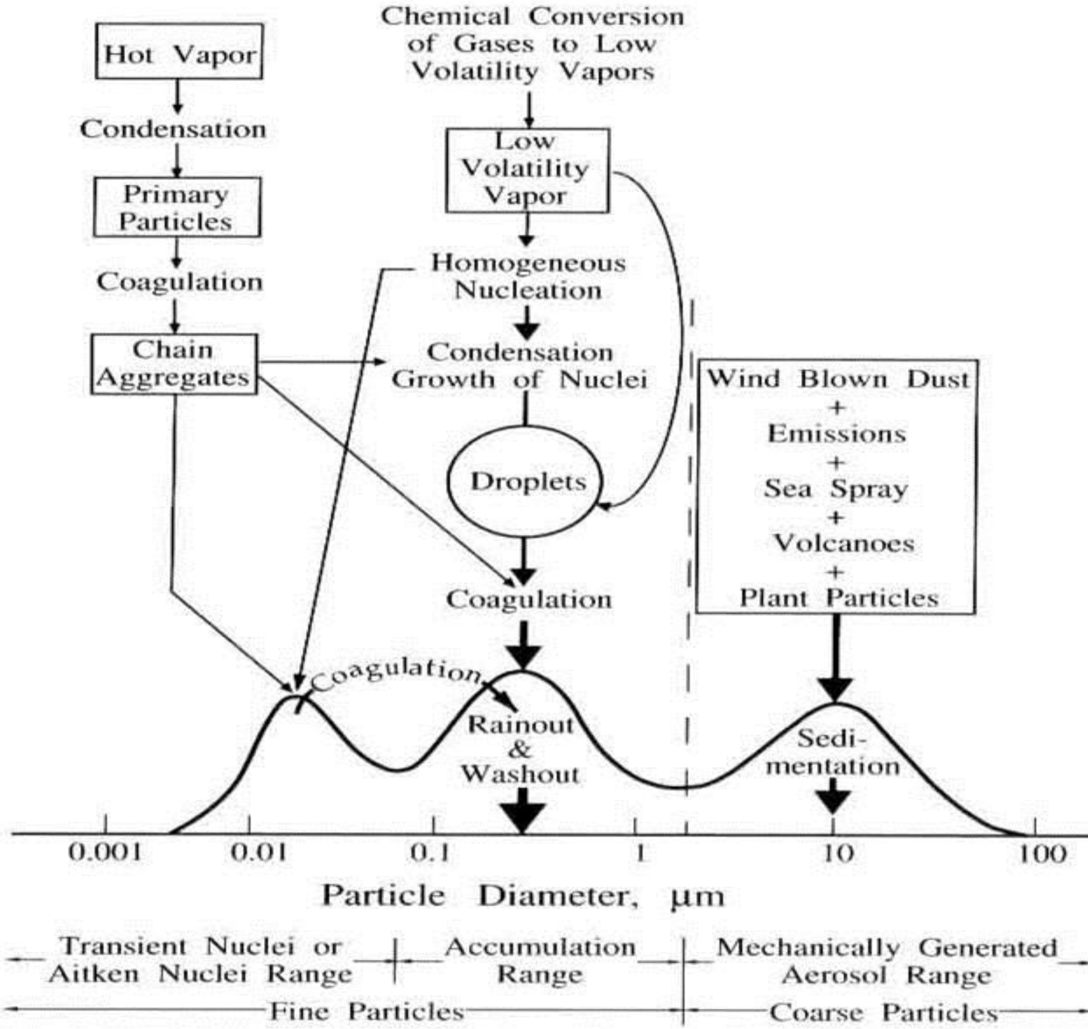
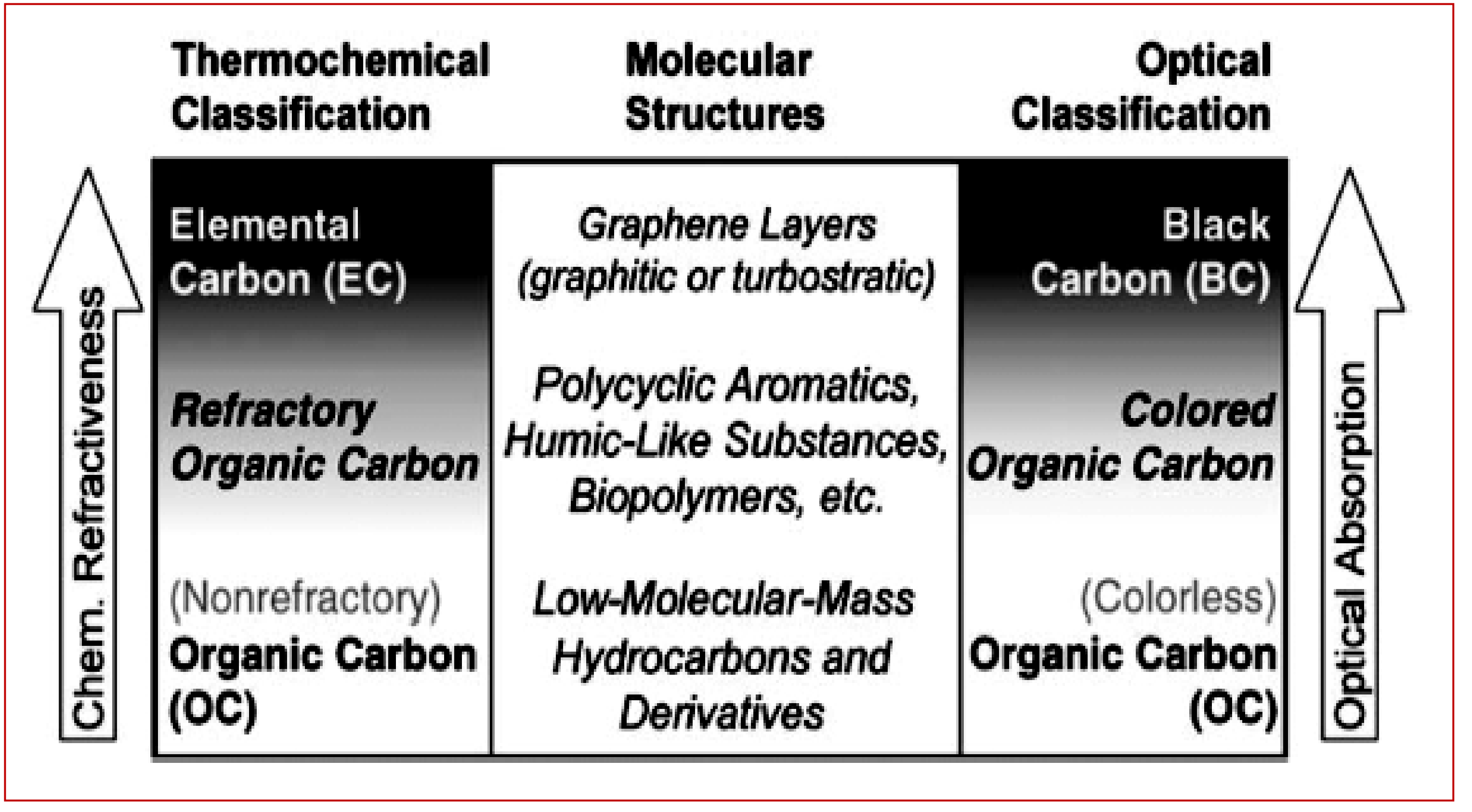


FIGURE 2.16 Idealized schematic of the distribution of particle surface area of an atmospheric aerosol (Whitby and Cantrell, 1976). Principal modes, sources, and particle formation and removal mechanisms are indicated.

Black carbon and brown carbon are mainly in fine particles.

Classification of carbonaceous aerosol

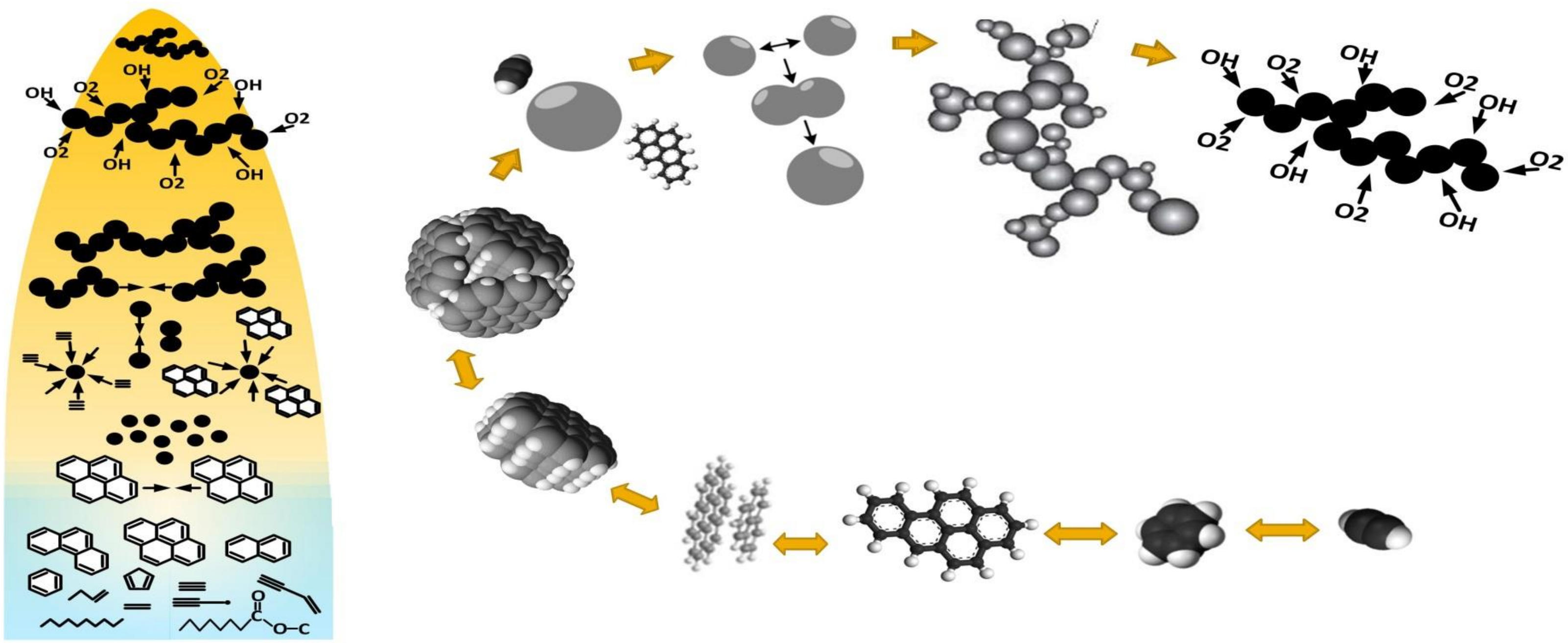


(Ulrich Poschl, AC, 2005)

Black carbon

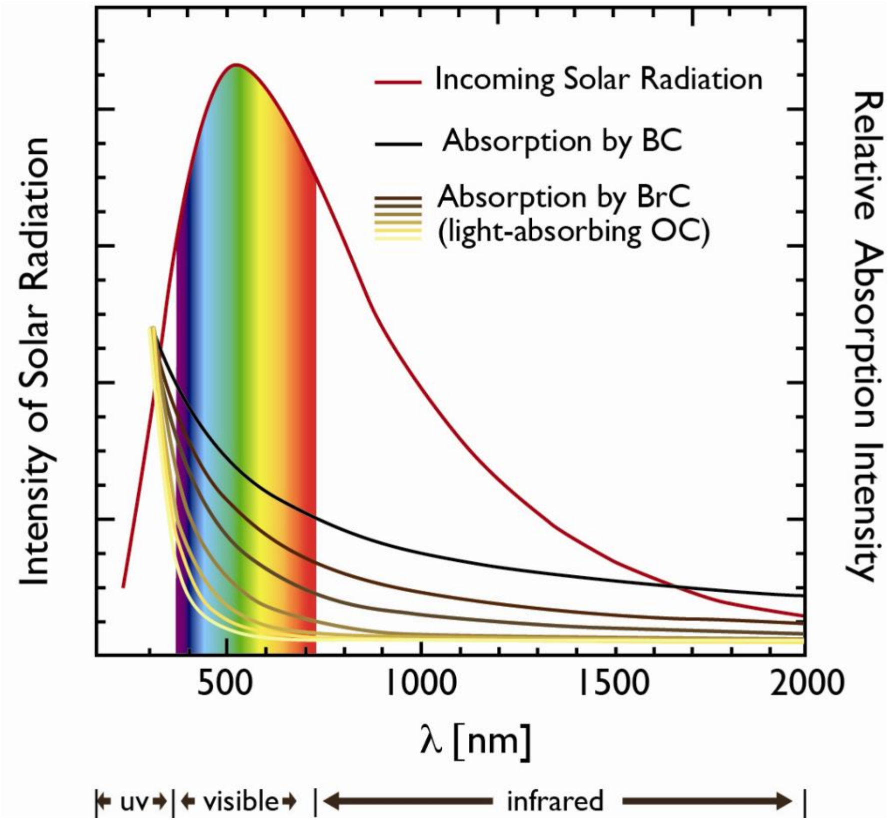
Source and formation of BC

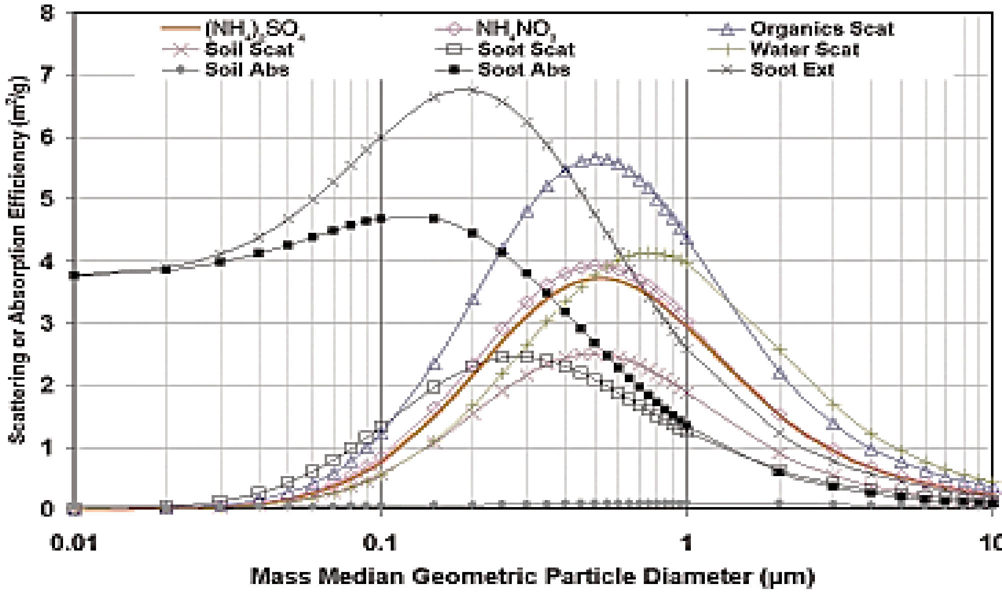
(google images)



(black formation from combustion- google image)

Optical properties

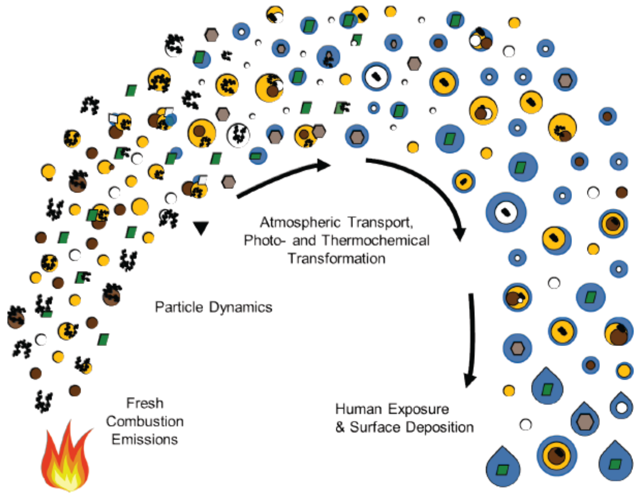


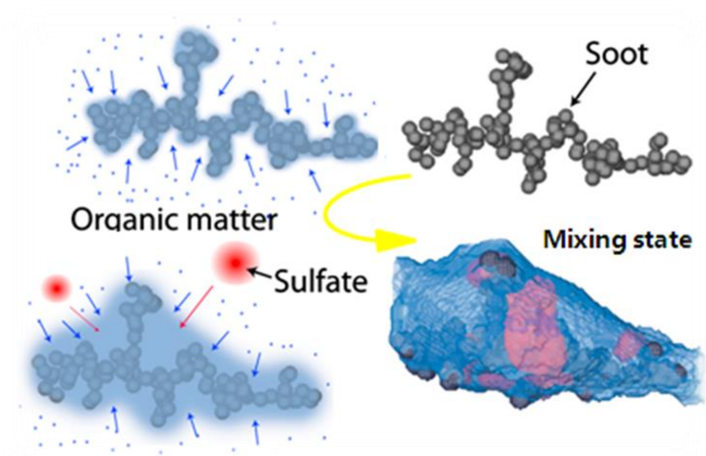


(Watson et al., 2002)

- **BC can absorption light at all wavelengths.**
- **Black carbon is the dominant absorbing component since it has the strongest mass absorption cross-section**

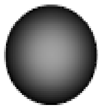
Atmospheric aging- properties change

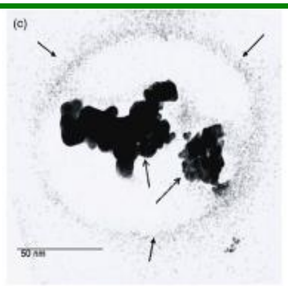




(U.S. EPA, 2012)

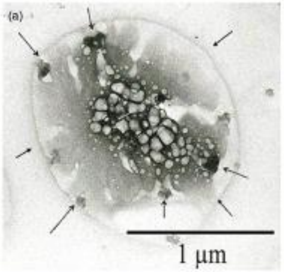
External mixing





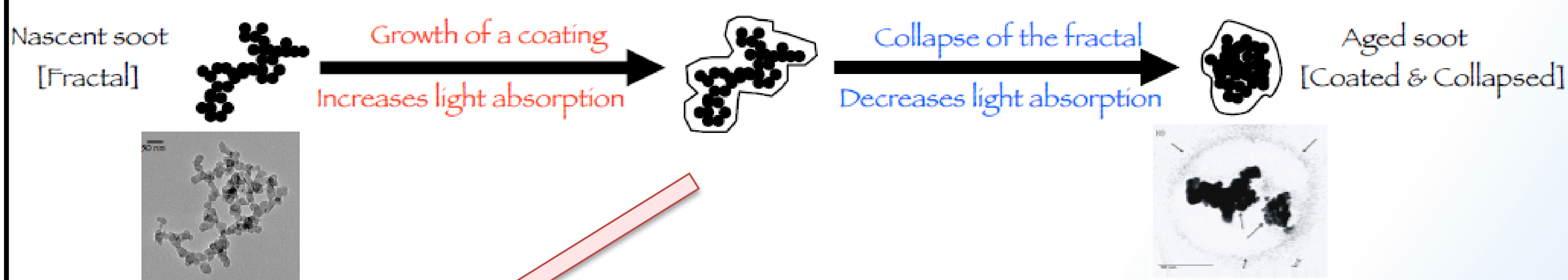
Fresh: hydrophobic

Internal mixing



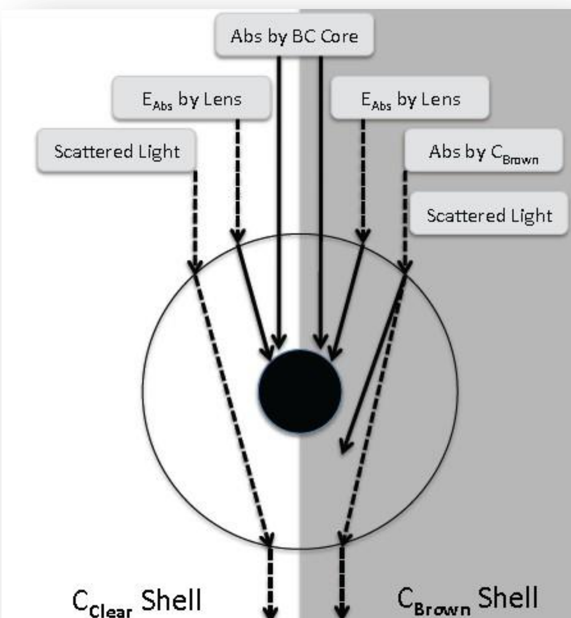
Aged: hydrophilic

Complicated Optical properties through aging: coating and morphology effect



But finally the coating effect will win

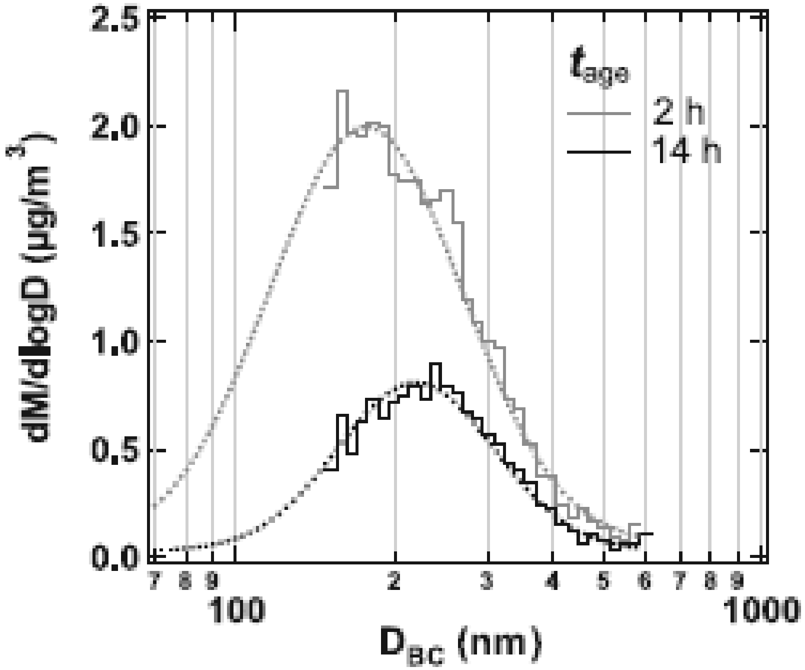
Lensing effect



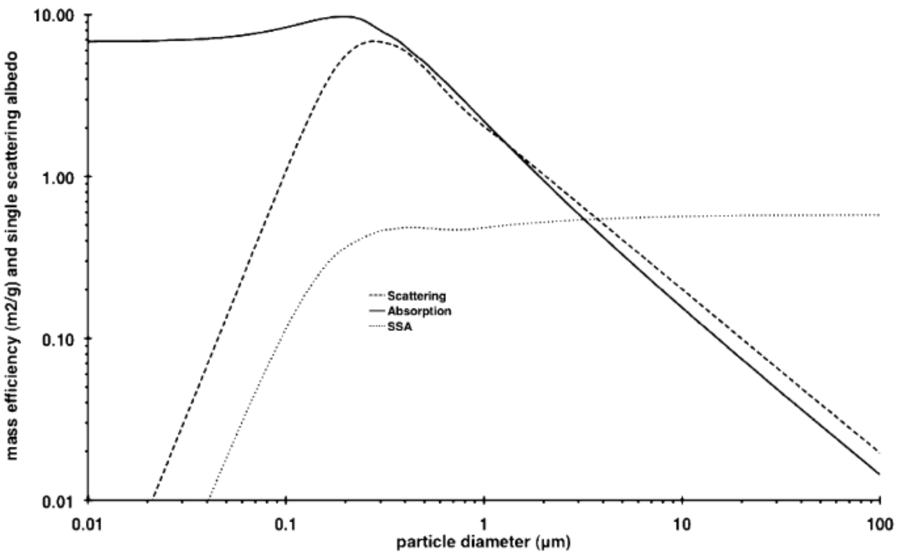
(Sedlacek and Subramanianv's slide)

Uncertainty regarding the contribution of soot to aerosol radiative forcing is large due to complicated optical properties of BC!

Complicated Optical properties through aging: size effect



(Moteki et al., 2007)



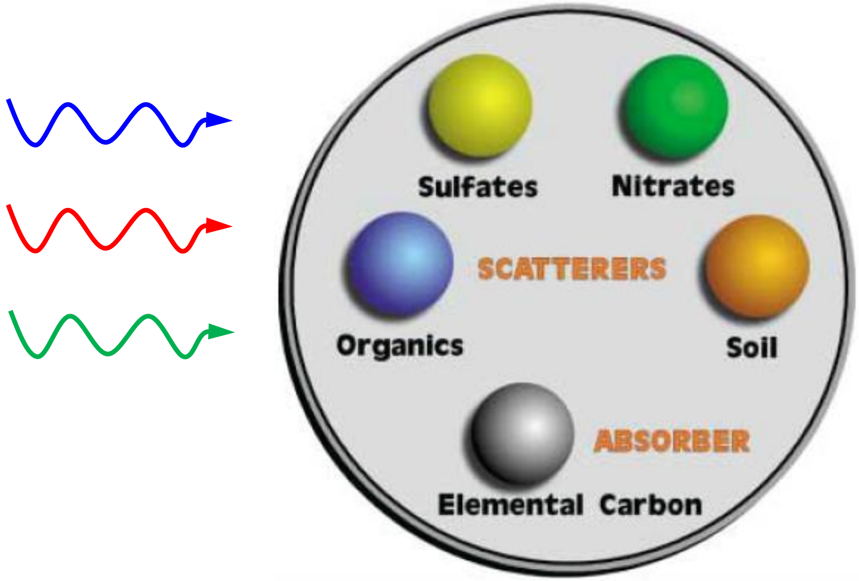
(Seinfeld and Pandis, 2006)

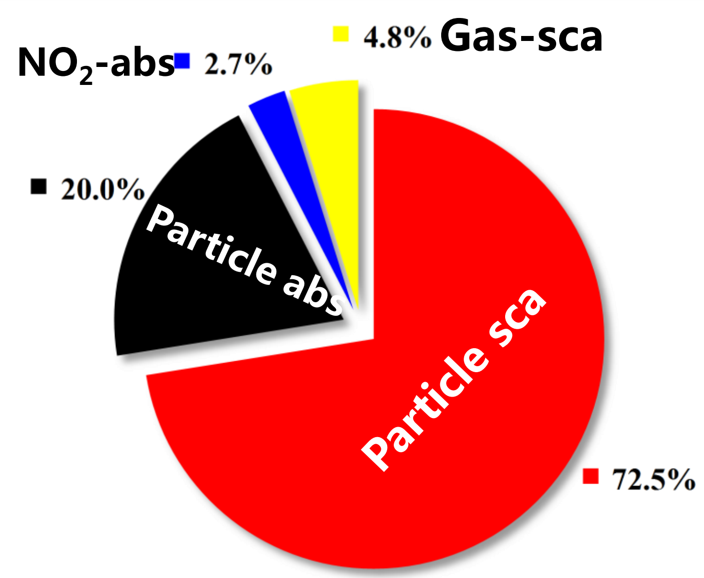
- **Size distribution shift to larger size during aging;**
- **There is limit of enhancement due to size.**
- **Theoretically, the smaller core with bigger coating has the largest absorption enhancement.**

Influence– air quality and visibility

(visibility comparison of Beijing, google image)

Influence– air quality and visibility

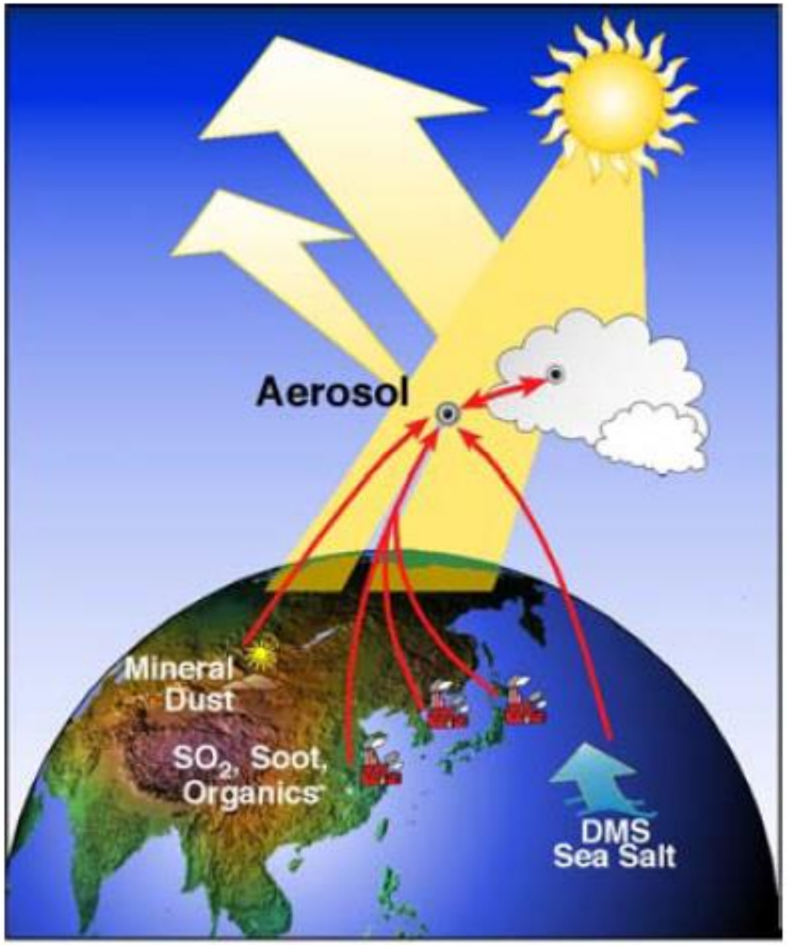




(example from a Chinese campaign, 2012)

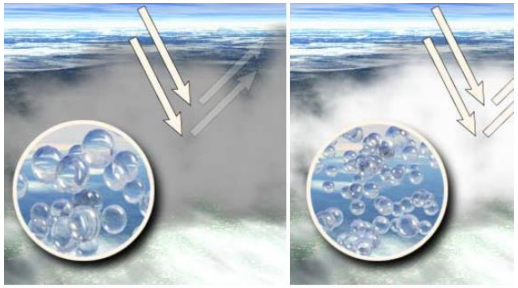
- **Particle absorption is not negligible**
- **Black carbon is the dominant absorbing component**

Influence on Climate



Direct effect:
Scattering and absorption of incoming sunlight by aerosol particles

Indirect effect:
The number concentration of cloud condensation nuclei (CCN) influences the cloud droplet size and thereby changes the cloud albedo and lifetime

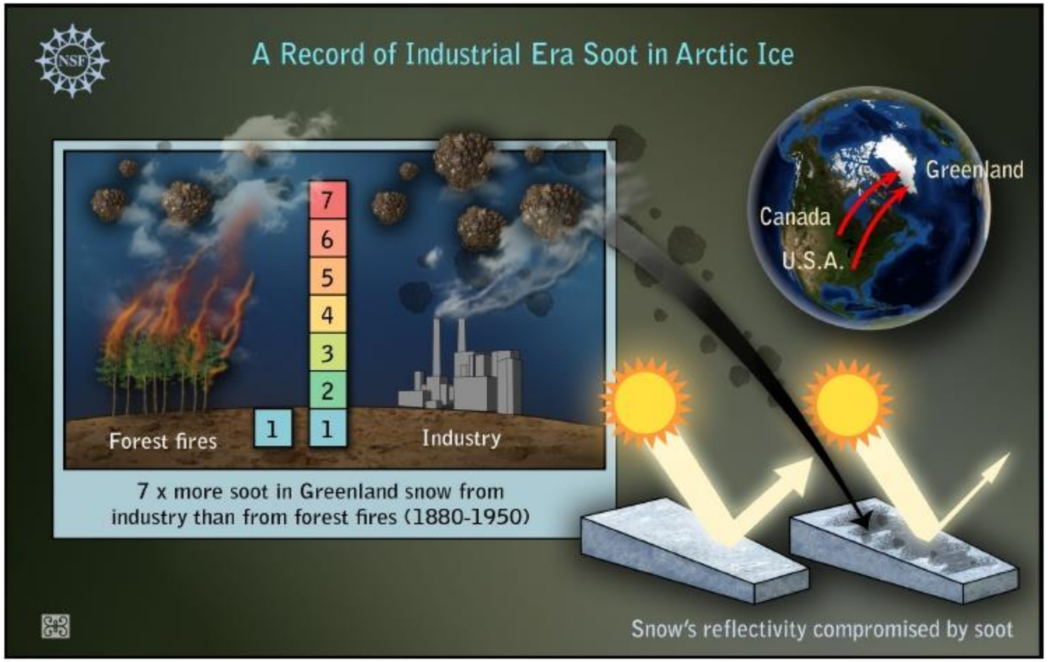


Large droplets
→ Weak reflection

Small droplets
→ Strong reflection

(Urs, ETH lecture)

Influence on polar climate: direct



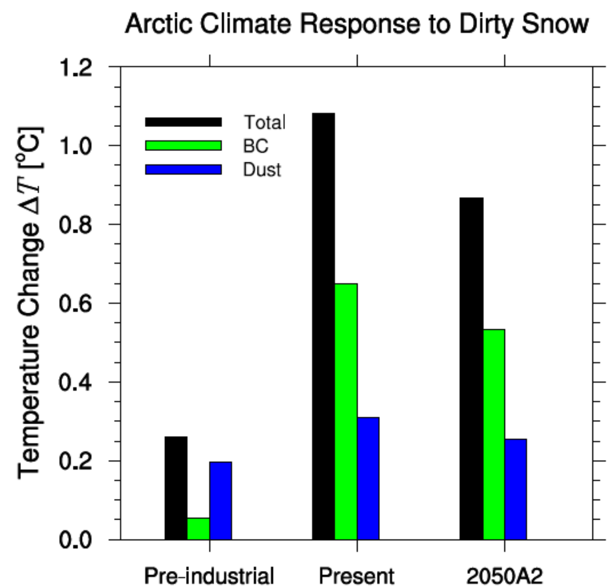
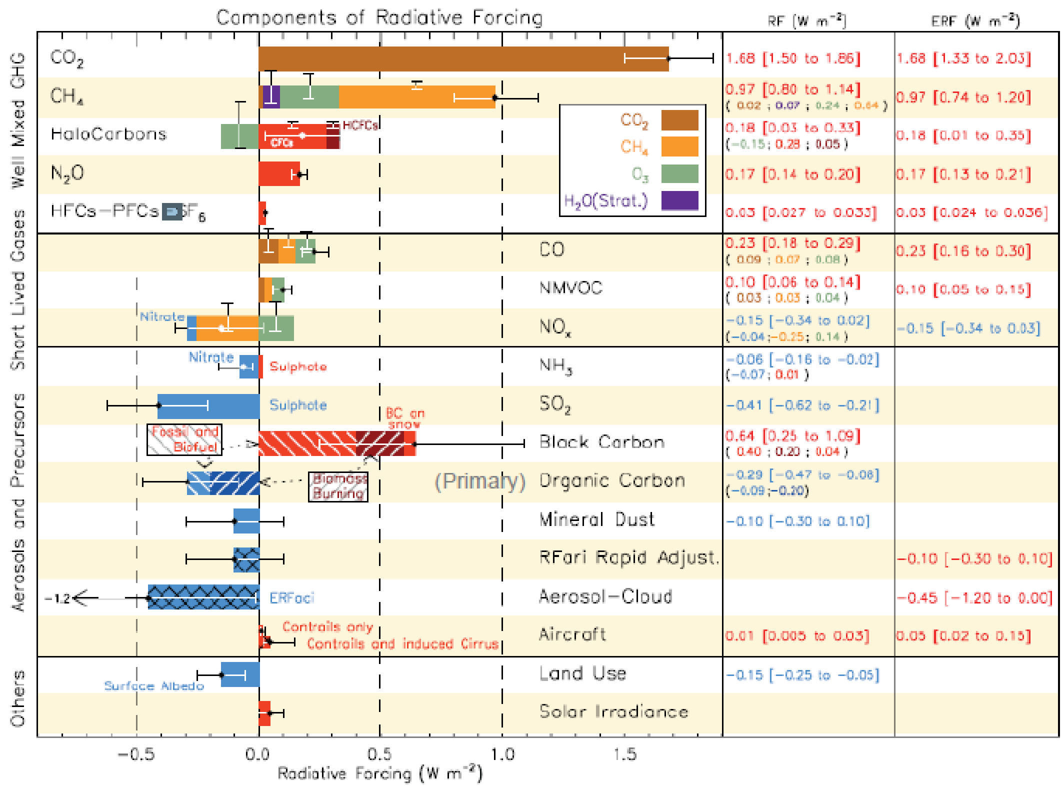


Figure 4: Predicted Arctic-mean temperature response [°C] to snowpack heating by black carbon and dust during Pre-Industrial, Present Day, and 2050 IPCC A2 climates. (*Zender and Flanner*, Manuscript in Preparation)

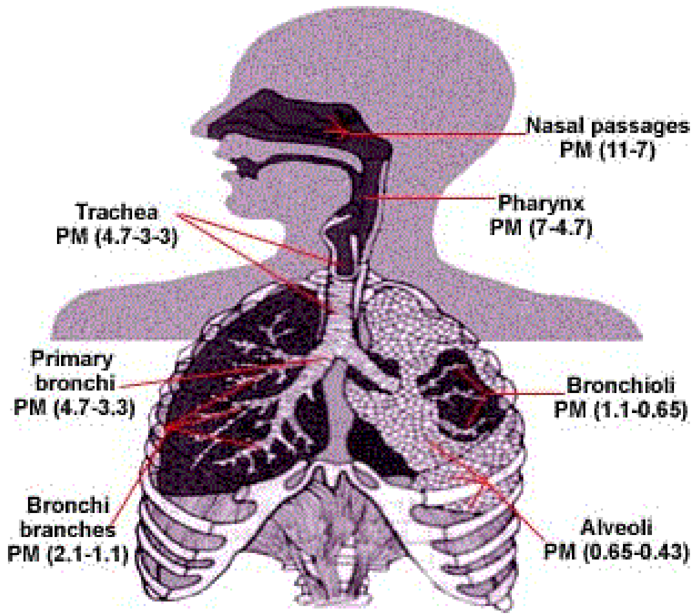
Radiative forcing and uncertainty

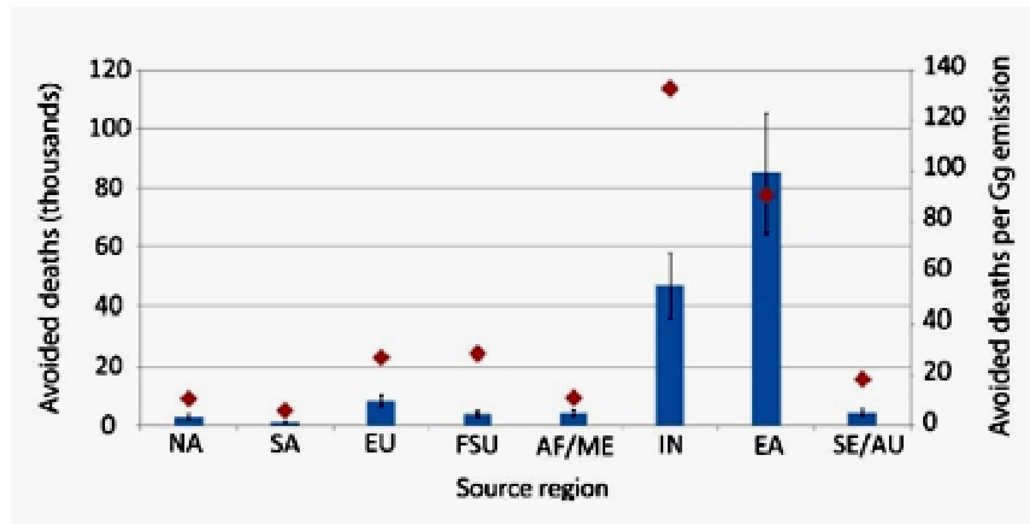


IPCC, 2013

- **BC has been the second contributor for climate warming after CO2.**
- **Reducing BC is promising for the mitigation of climate warming since the life time of BC is much shorter than CO2.**
- **Big uncertainties remains due to complicated optical properties of BC and measurement.**

Other influence: health





(Pope, 2006)

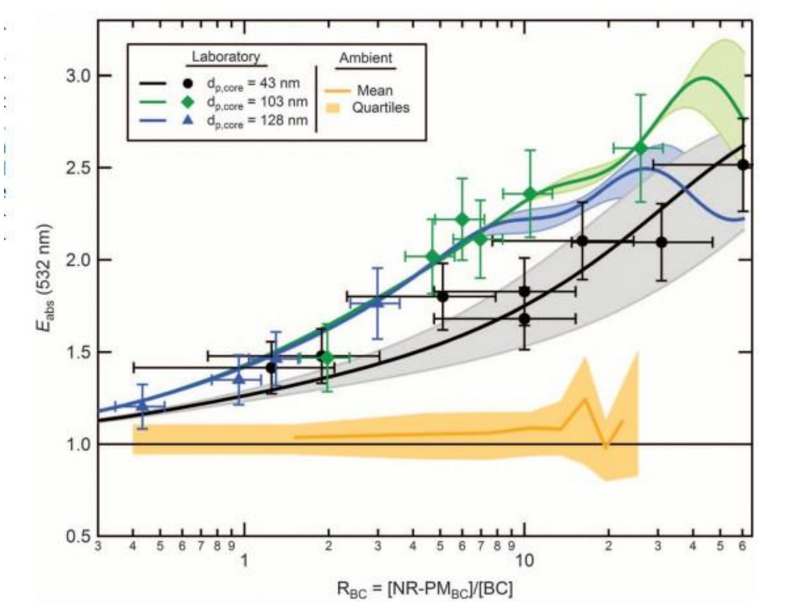
- **Small particles can go through alveoli and into flood;**
- **BC adsorb unhealthy component due to its large surface to volume ratio.**

Research of black carbon in terms of optical properties

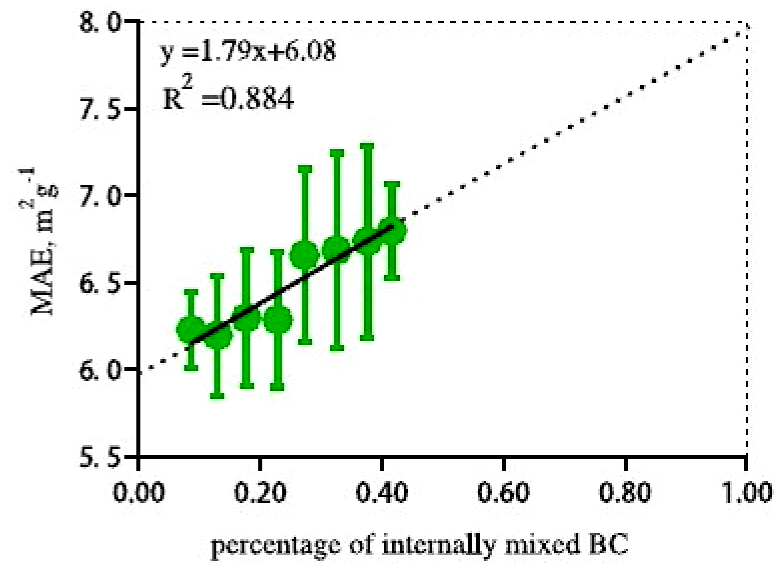
- Ambient measurement: real atmospheric condition, constraining the model;
- Lab experiment: explore the mechanism
- Model: large scale estimation combing emission inventory and BC mass absorption cross section

Cutting edge: How much is the absorption enhancement by BC?

Theoretical core-shell modelling study (Jacobson, Nature, 2001) shows significant increase (50%) of absorption due to coating. However, some ambient studies show only 6-7% enhancement as below

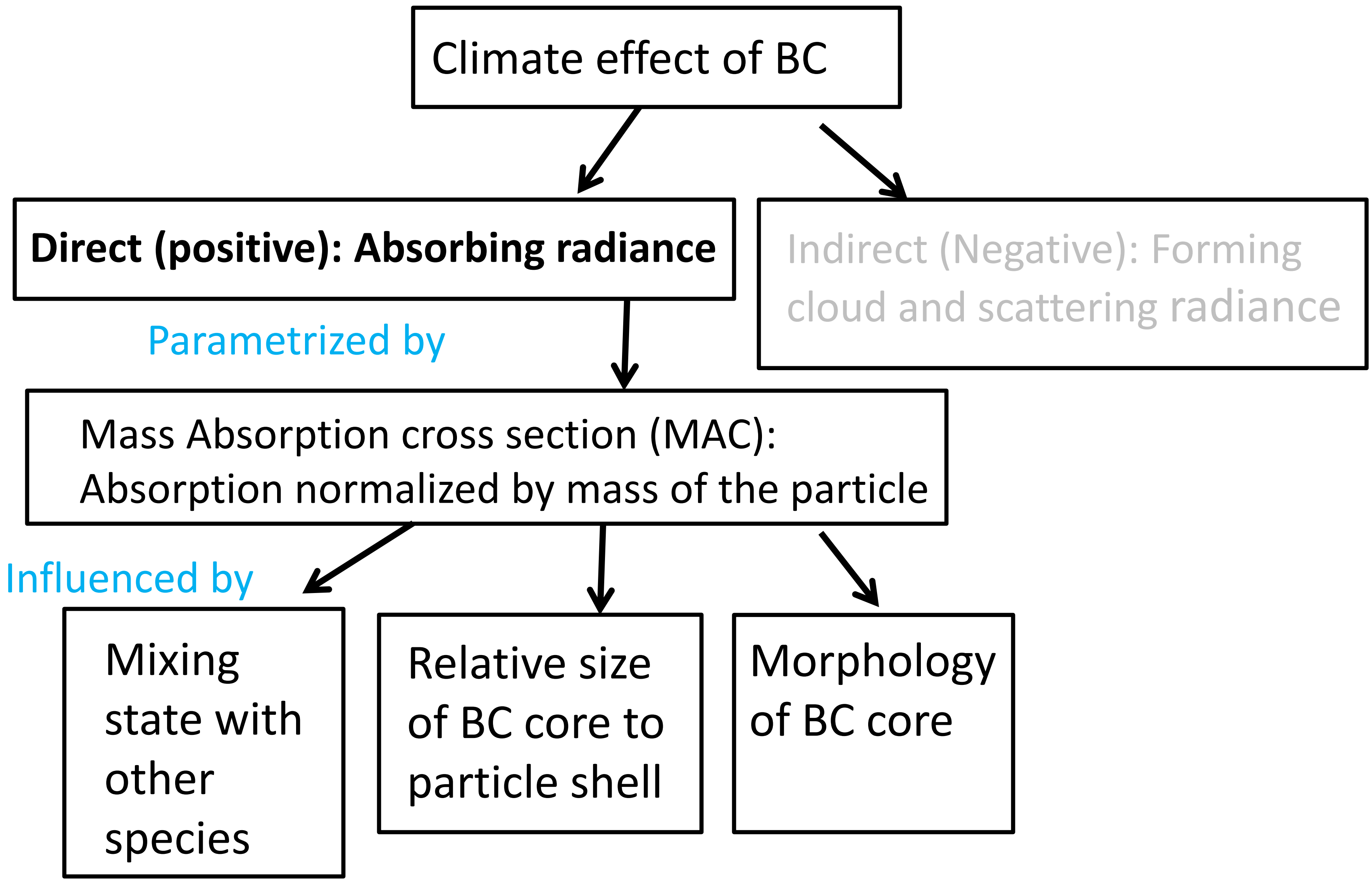


Cappa et al,2014 （Nature）



Lan et al,2013 （AE）

My ongoing projects: big picture



Explore through lab and ambient experiment

On going Projects and goals

- 1. Testing the performance of an laser induced incandescent instrument measuring BC mass concentration----- technical work;
- 2. Lab experiment: exploring the influencing factors of BC absorption enhancement----scientific study
- 3. Ambient campaign: combing with lab experiments to study the BC absorption enhancement factors.

Data analysis is under process and the results will be showed in EAC (2017) conference in ETHZ

Brown carbon

Light absorption by brown carbon aerosol in the PRD region of China

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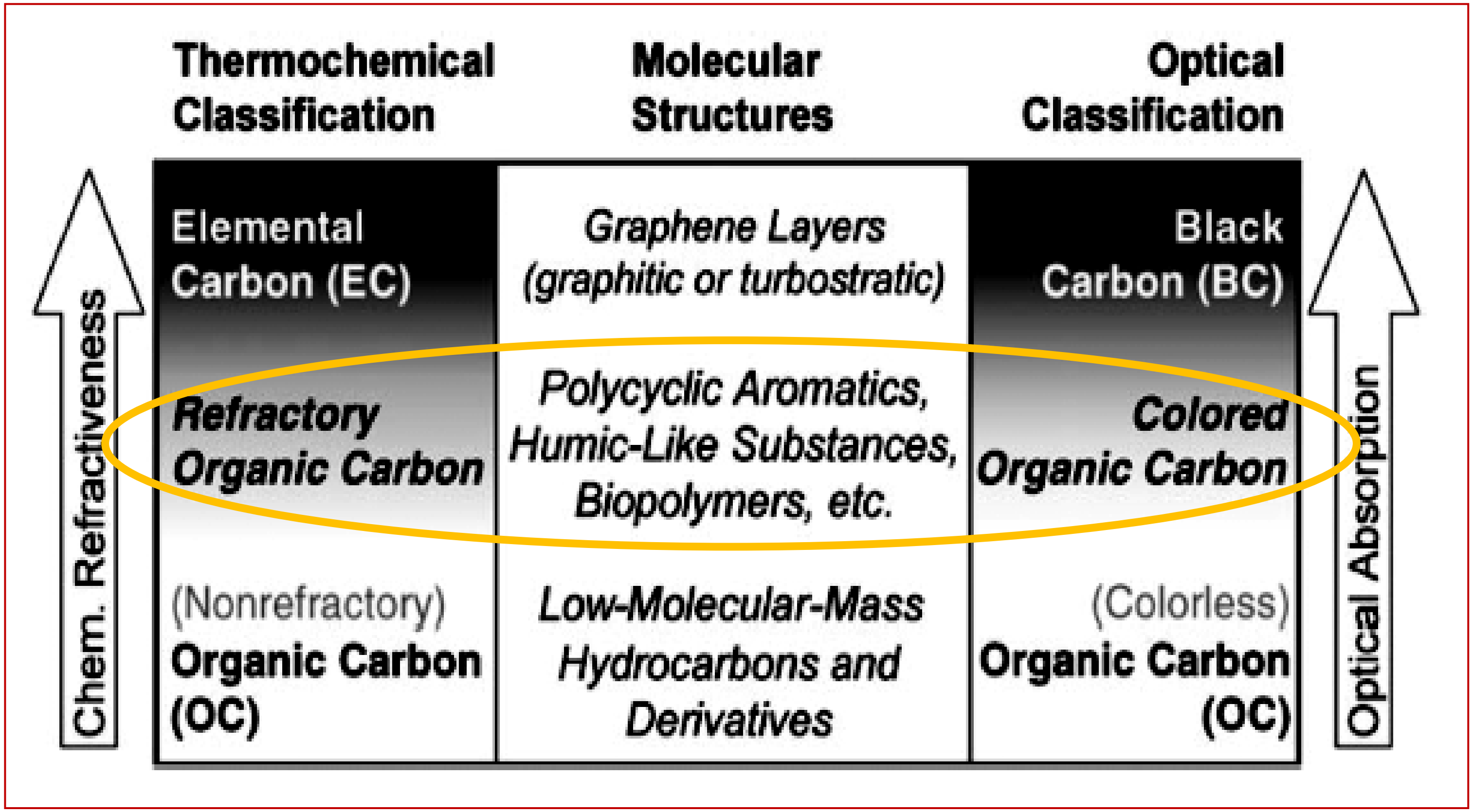
Peking University, China

2015-12-18

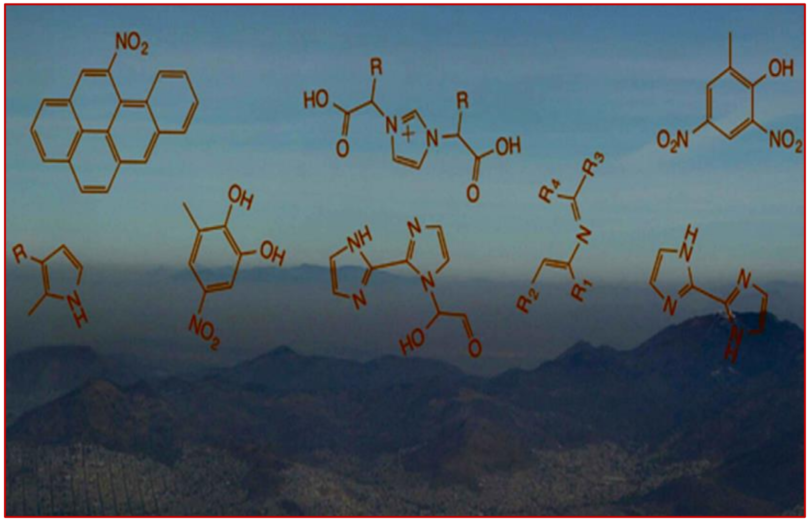
Classification of carbonaceous aerosol

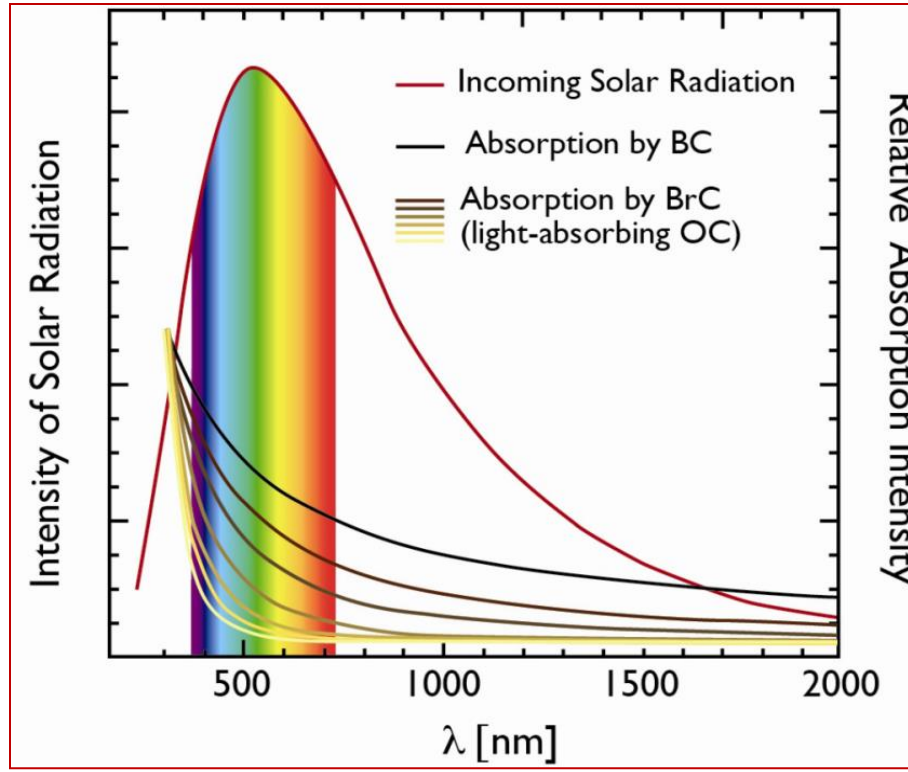


(Ulrich Poschl, AC, 2005)

Definition of Brown Carbon (BrC): light absorbing organic carbon

Chemical and optical properties of BrC

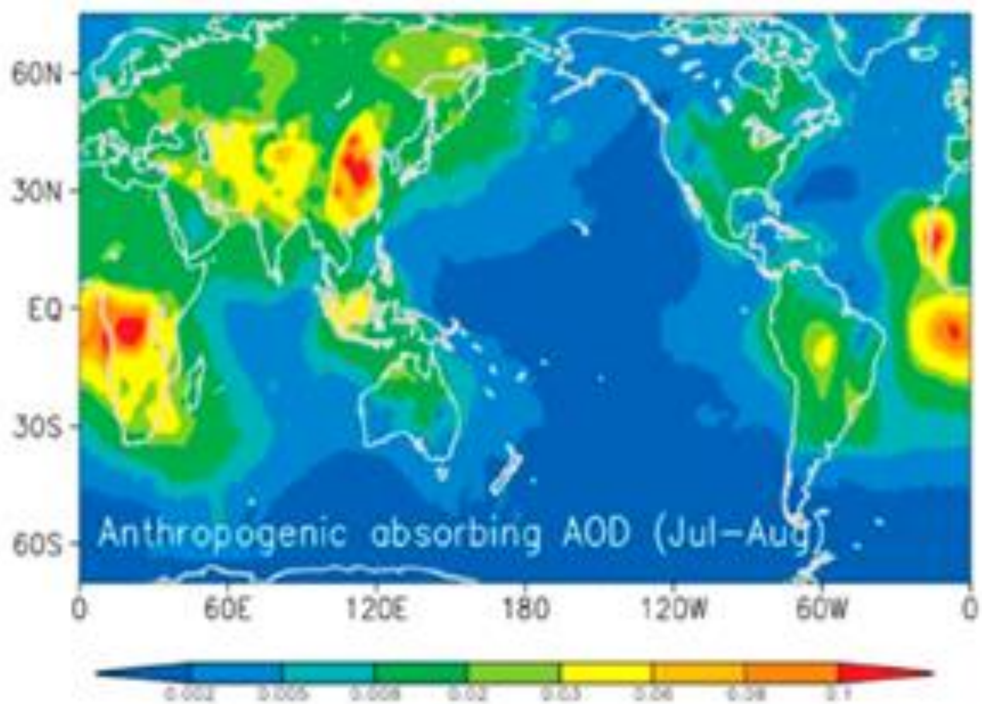




- Complexity and variety of molecular composition
- Strong wavelength-dependence of light absorption

Source , distribution and climate effect



The modeled anthropogenic AAOD, to which BrC contributed significantly

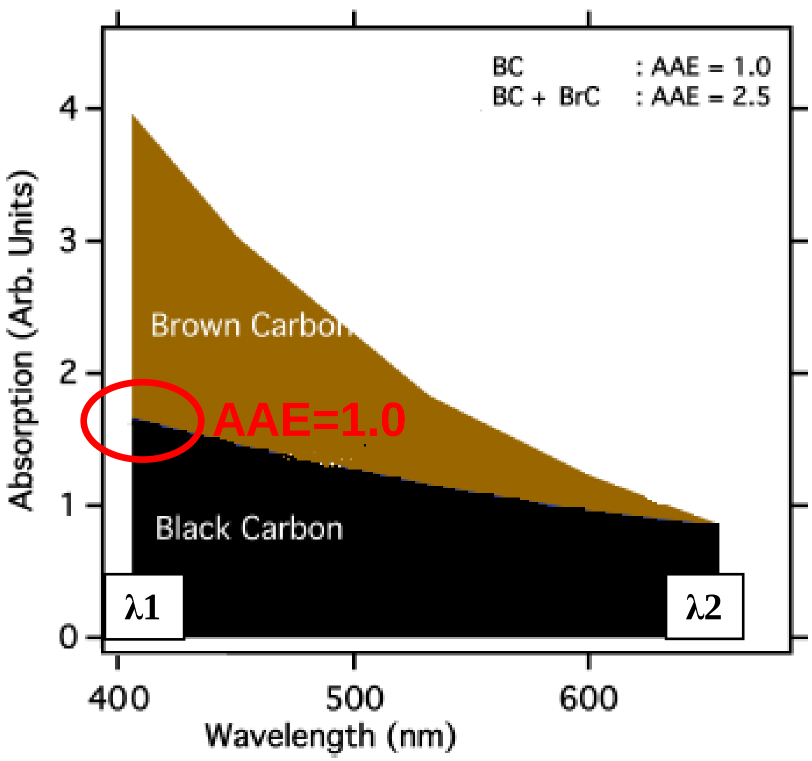
- Absorbing light
- Radiative forcing
- BrC in East Asia

(Ramanathan et al, 2007)

Identification of BrC's role in light absorption

- Absorption Angstrom Exponent (AAE) method

$$AAE\ (\lambda_1, \lambda_2) = \left(\frac{\ln(Abs_{\lambda_1}/Abs_{\lambda_2})}{\ln(\lambda_1 / \lambda_2)} \right)$$

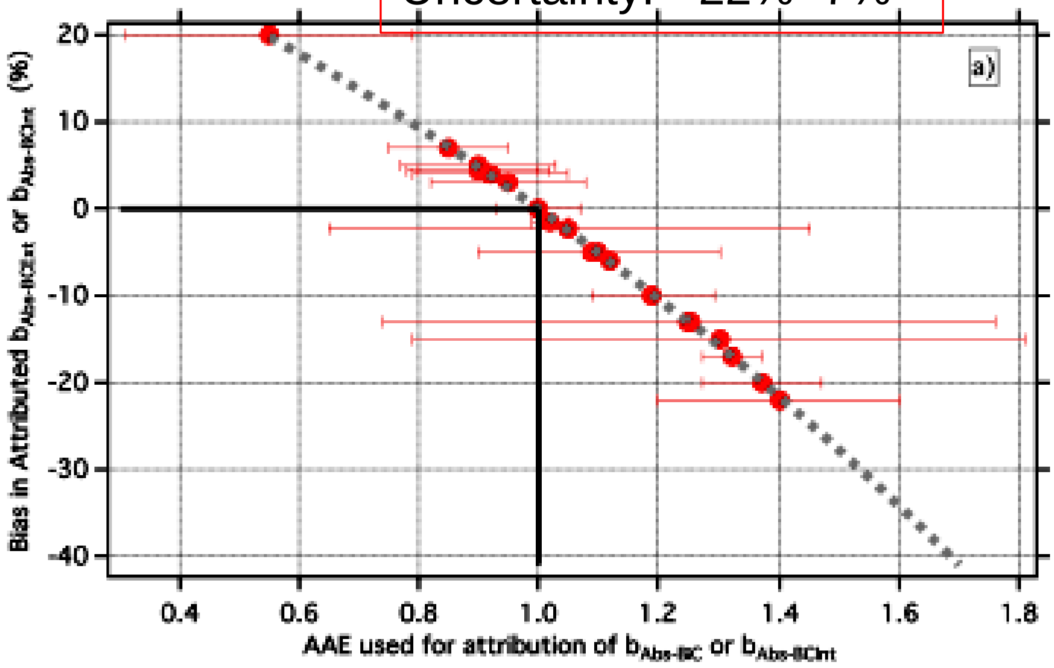


- **BC's AAE is 1.0 in theory**
- Total aerosol's AAE would be higher due to BrC existing

Is it reliable to use AAE_bc =1.0 in real atmosphere?

AAE_bc=1.0

Uncertainty: -22%~7%



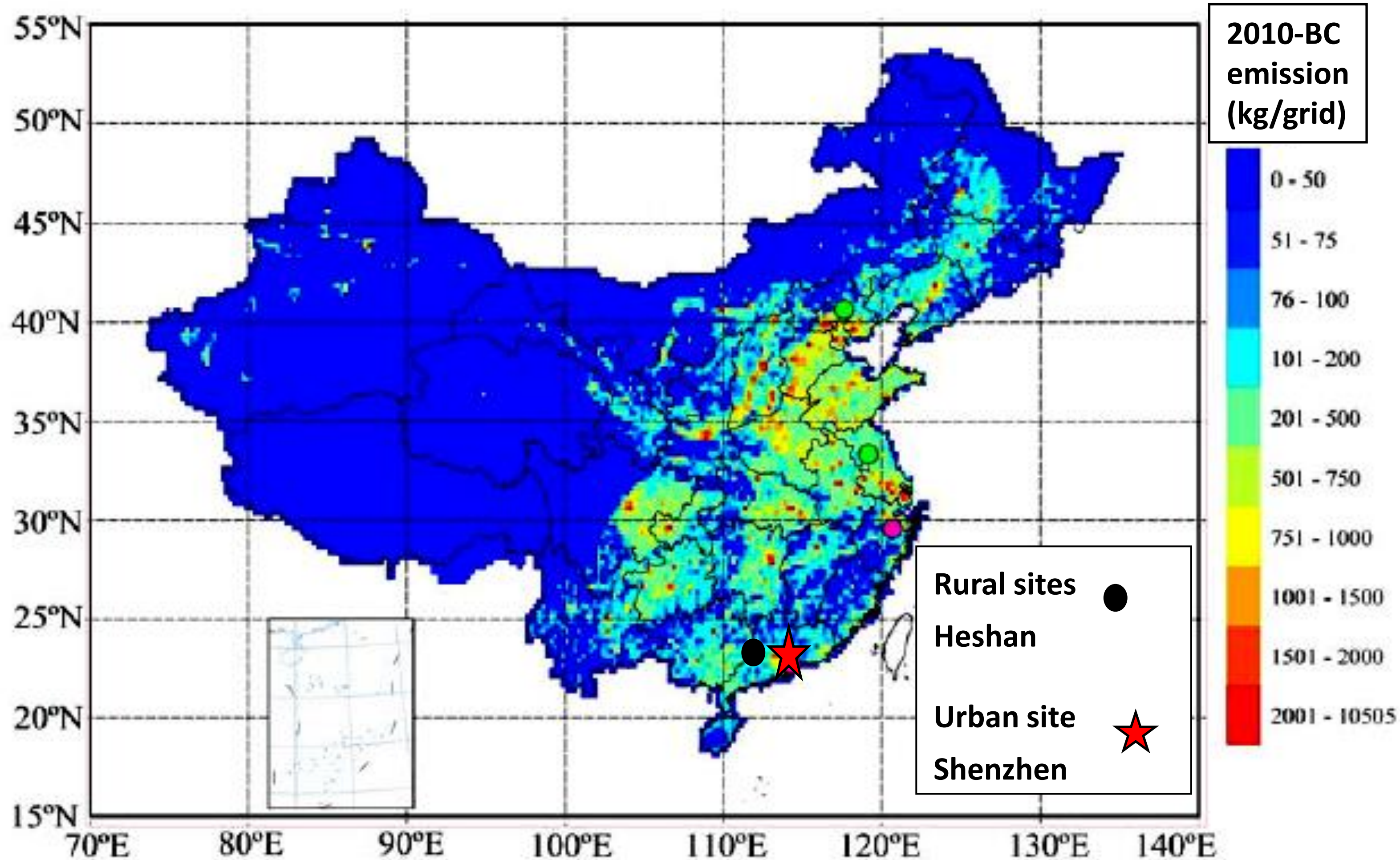
(D.A. Lack et al, ACP, 2013)

- More realistic AAE_bc with less uncertainty should be explored

Highlights of this study

- Light extinction of $PM_{2.5}$ in the PRD region of China.
- Finding real AAE_bc based on local ambient measurement.
- Quantitative calculation of light absorption of brown carbon with uncertainty evaluation at short-wave wavelengths.

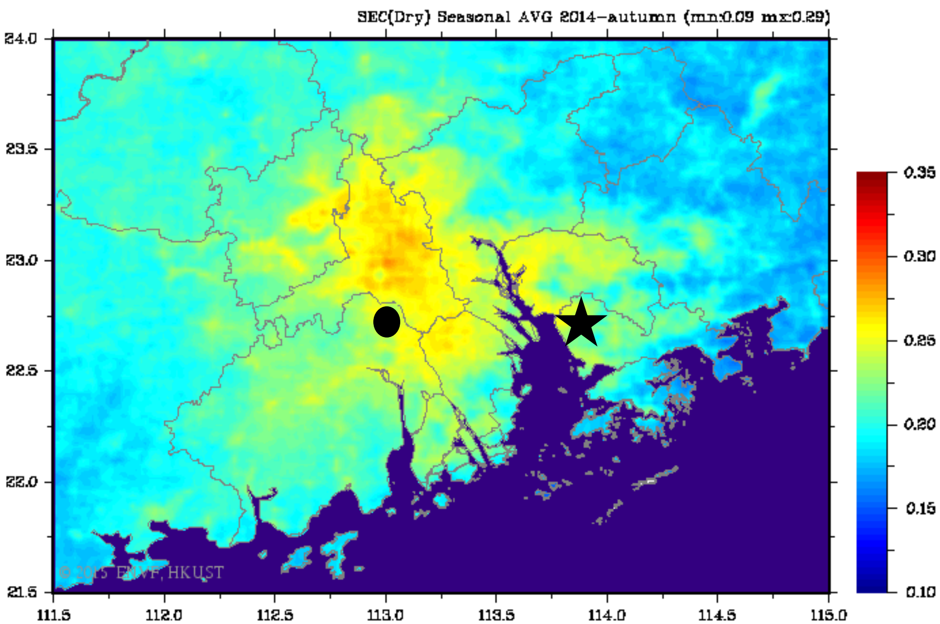
Sampling sites



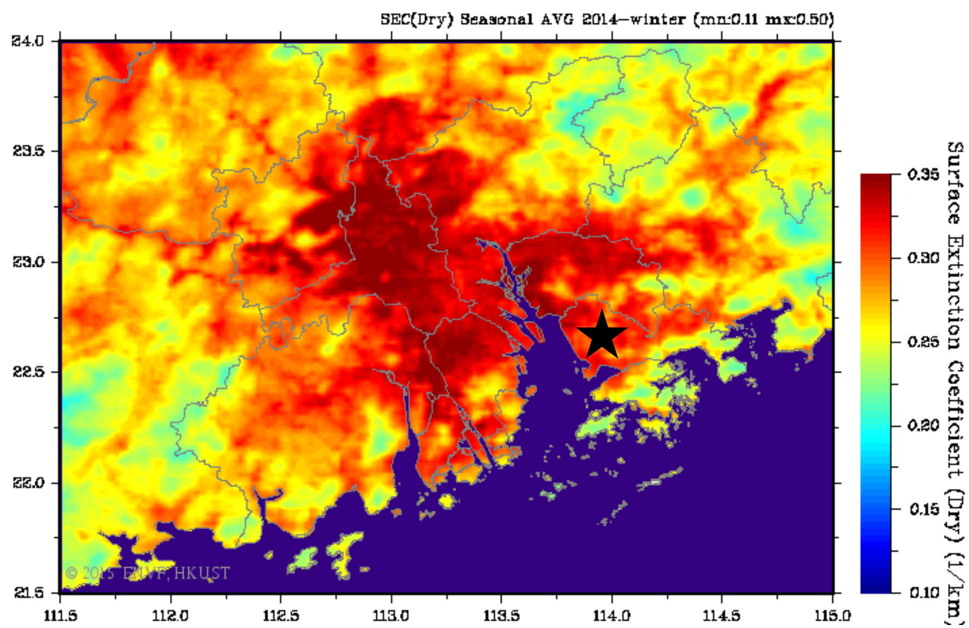
Shenzhen (22.60 ° N, 113.97 ° E), Heshan (22.71 ° N, 112.93 ° E)

Sampling periods

2014- Fall



2014-Winter



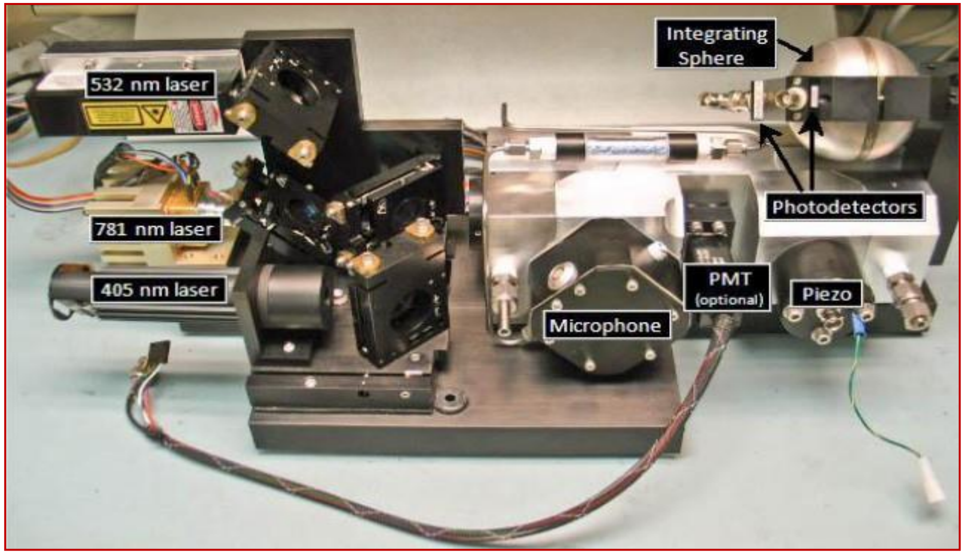
Seasonal average surface extinction coefficient in PRD (Dry) (1/km)

Shenzhen (2014/01.15-02.19, 2014/09.12-10.09) : SZ_winter & SZ_fall

Heshan (2014/11.01-11.21): HS_fall

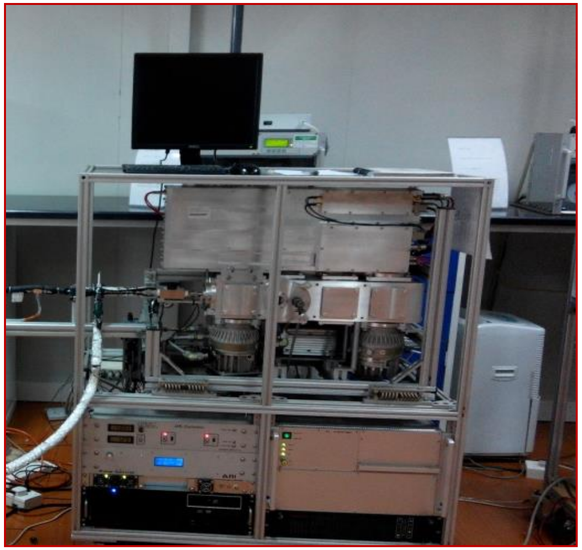
Instrumentation

Light absorption of $PM_{2.5}$



PASS-3 (@405, 532, 781 nm)

Mass concentration of Org



AMS

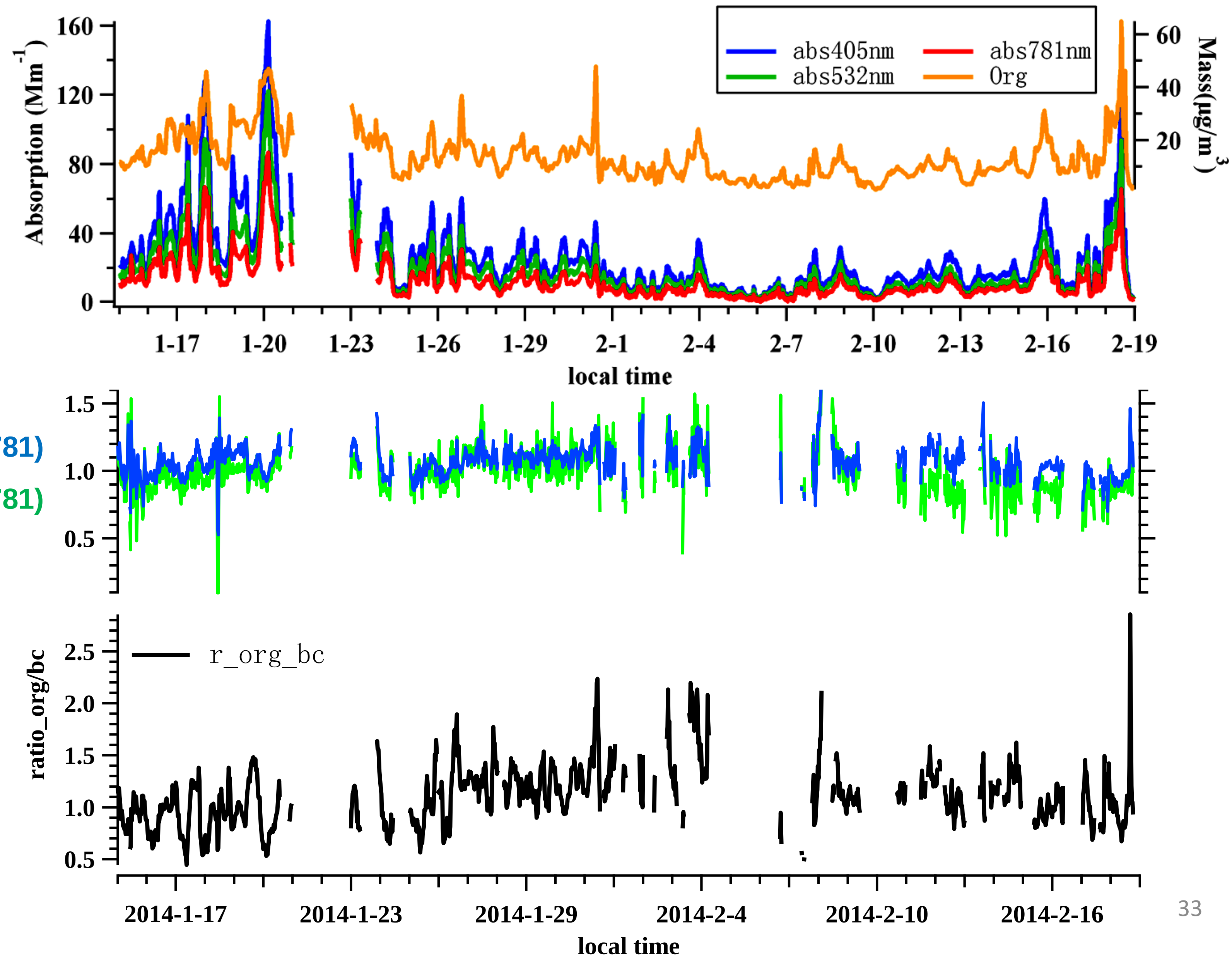
- Advantages of PASS-3 & AMS: online, in situ, high time resolution
- Principle of PASS-3: photo-acoustic method

Time series of AAE of ambient aerosols

eg: Shenzhen (winter) campaign

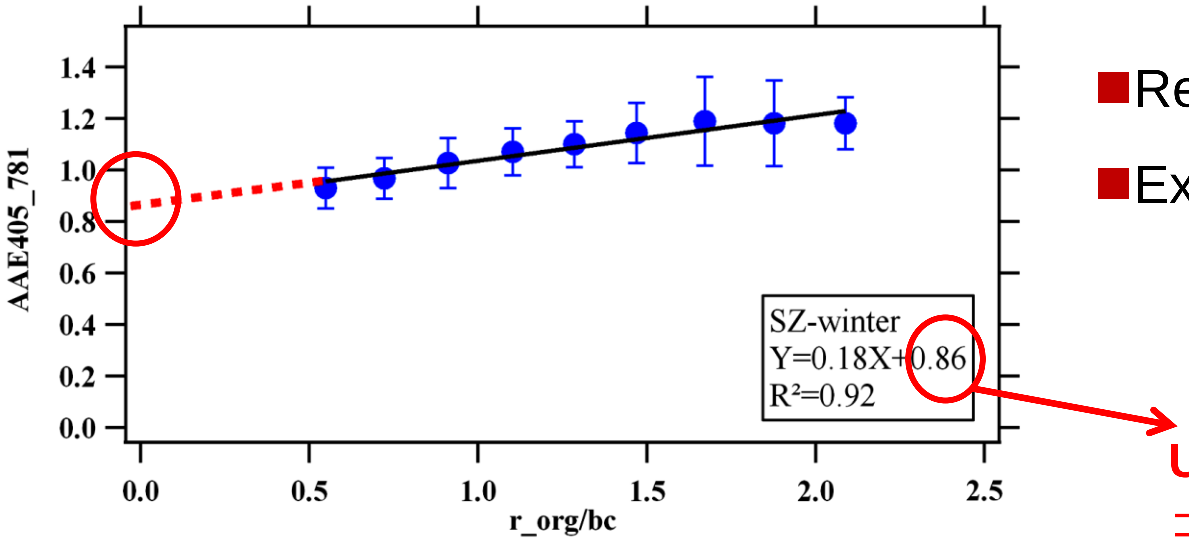
AAE(405, 781)

AAE(532, 781)



Determination of AAE baseline (AAE_bc)

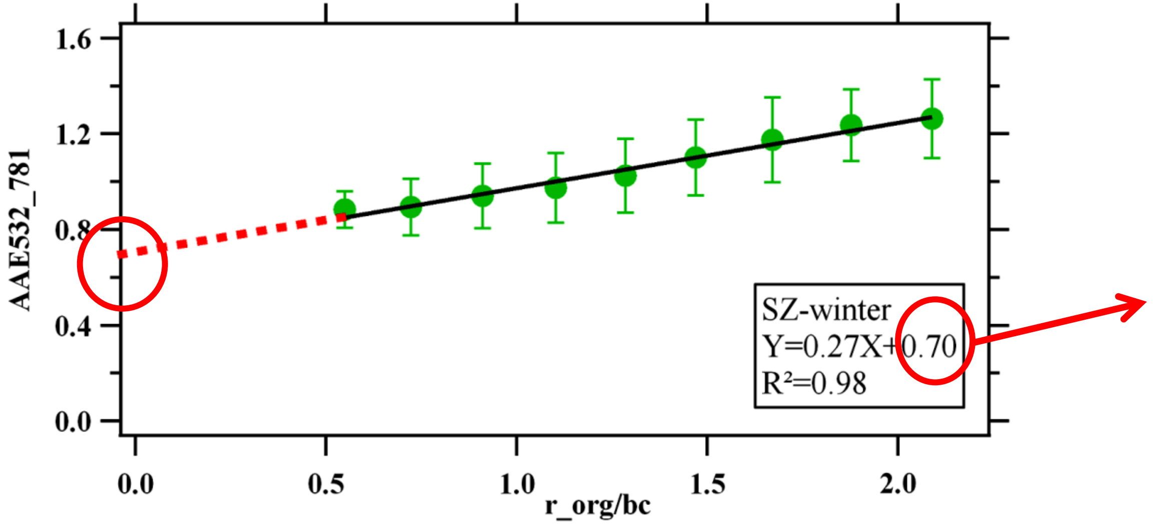
eg: Shenzhen (winter) campaign



- Regression analysis
- Extrapolation of the AAE_bc

Uncertainty: ±6%

$$U_{AAE\_bc} = t_p \times S(a)$$



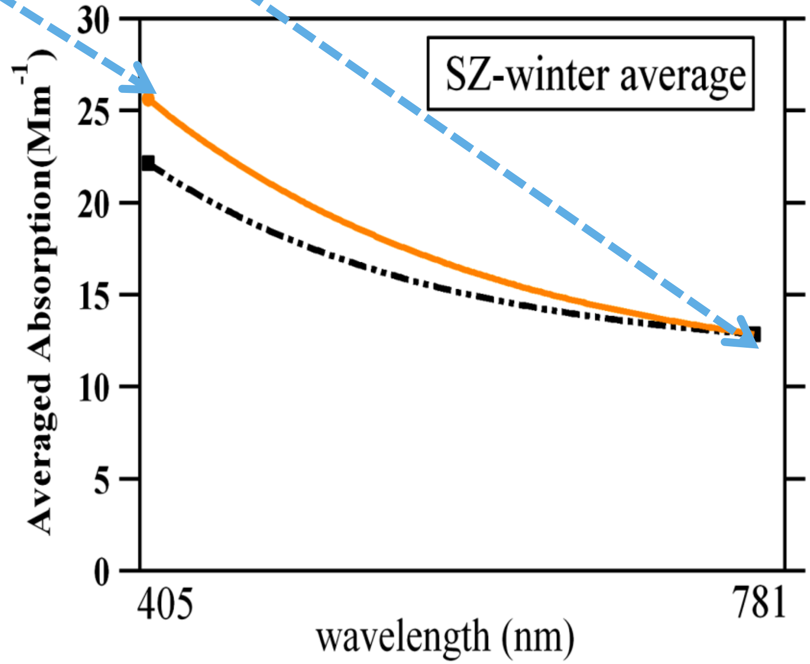
Uncertainty: ±5%

BrC absorption total uncertainty

$$U_{total} = \sqrt{(U_{abs\lambda 1})^2 + (U_{abs\lambda 2})^2 + (\ln(\lambda_2/\lambda_1) \times U_{AAE\_bc})^2}$$

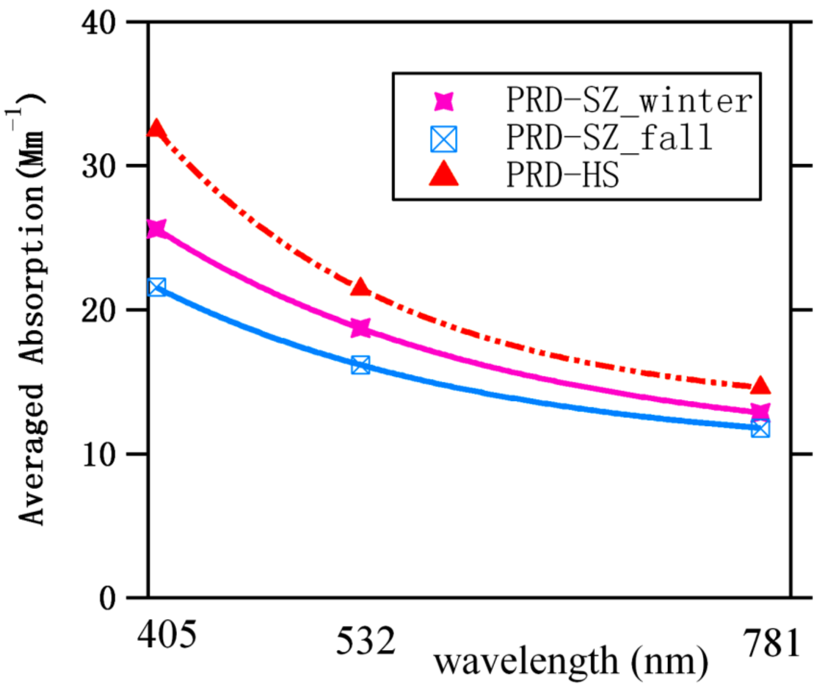
$$U_{Abs\lambda} = \frac{1}{n} \times \sqrt{\sum (U_{2\min})^2}$$

$$U_{2\min} = NoiseEq\_Abs / Abs$$



Wavelength-dependence and BrC absorption

Total aerosol AAE

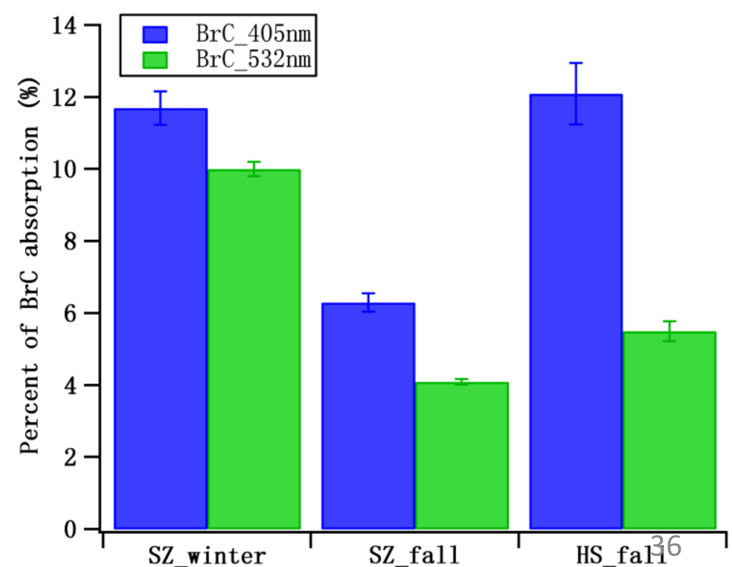


AAE_bc

	AAE_bc, 405_781	AAE_bc, 532_781
SZ_winter	0.86	0.70
SZ_fall	0.82	0.71
HS	1.02	0.86

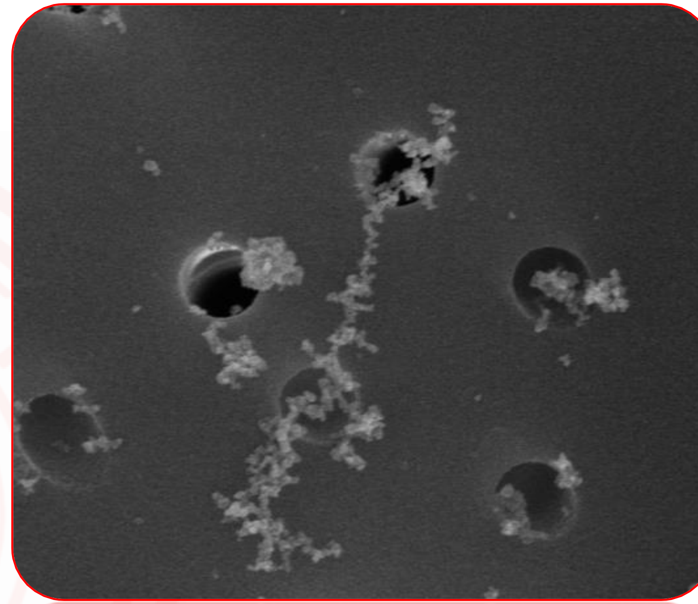
BrC

Absorption contribution by BrC



	Abs_BrC (%), 405nm	Abs_BrC (%), 532nm
SZ_winter	11.7	10.0
SZ_fall	6.3	4.1
HS	12.1	5.5

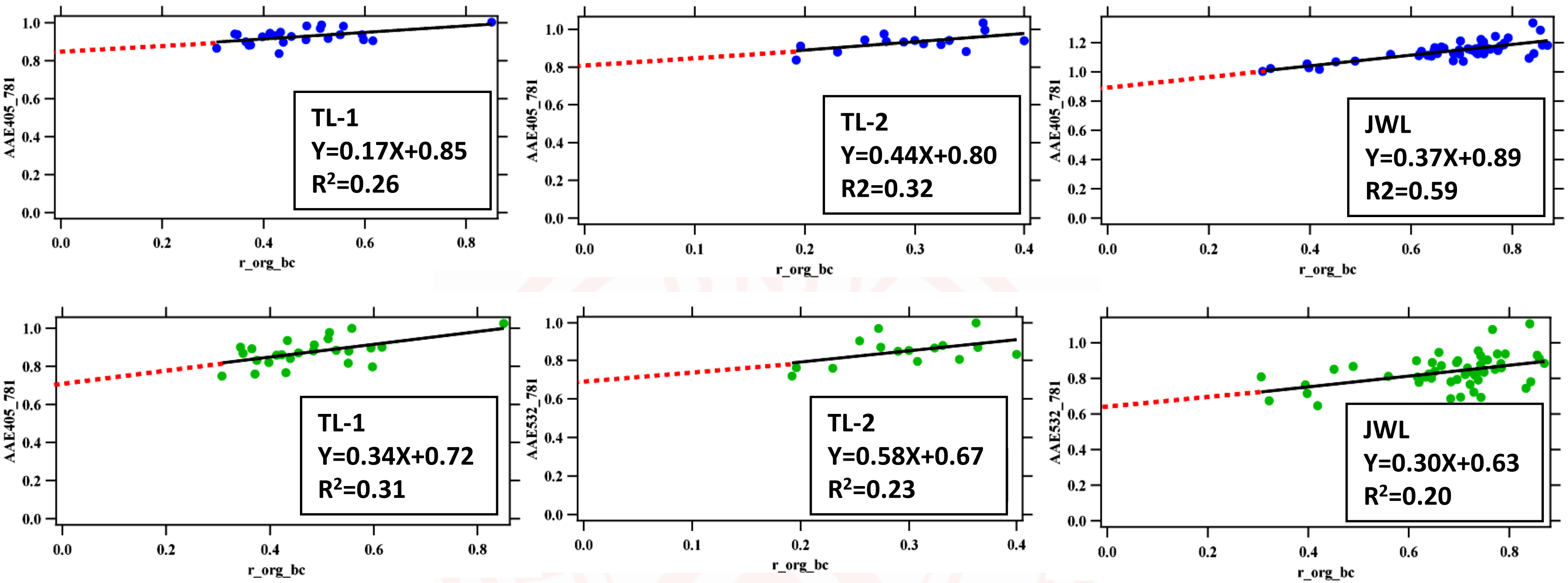
Validation of AAE_bc by Roadway Tunnel Experiments in Shenzhen



- Vehicular emissions are dominant
- Fresh vehicular emissions
- More close to “pure BC”.
- 2 tunnels were measured

BC (μg/m³)	ave	max	min
TL-1	16.7	35.4	5.6
TL-2	19.7	122.6	42.3
JWL	8.9	42.3	2.3

Extrapolation of AAE_bc in Tunnels



Ambient	AAE_{405_781}	AAE_{532_781}	Tunnels	AAE_{405_781}	AAE_{532_781}
SZ_winter	0.86	0.70	TL-1	0.85	0.72
SZ_fall	0.82	0.71	TL-2	0.80	0.67
HS	1.02	0.84	JWL	0.89	0.63

- The AAE_bc obtained are in good accordance with the Shenzhen ambient air measurements.

Validation of BrC source by Biomass burning Simulation Experiments

Biomass type	Burning modes	AAE_{405_781}	AAE_{532_781}	AAE_{405_532}
Ficus microcarpa leaf		4.46 ± 1.20	3.46 ± 0.96	5.85 ± 1.69
Lychee leaf		3.48 ± 1.20	2.52 ± 1.07	4.90 ± 1.61
Corn stalk		2.97 ± 1.16	2.39 ± 1.06	3.83 ± 1.49
Peanut stalk	Stove burning	1.99 ± 0.50	2.08 ± 0.86	2.13 ± 1.01
Litchi wood		2.61 ± 1.00	1.95 ± 0.85	3.55 ± 1.50
Eucalyptus wood		1.71 ± 0.50	1.30 ± 0.53	2.34 ± 0.85
Short straw		1.76 ± 0.25	1.39 ± 0.47	2.32 ± 0.65
Short straw	Open burning	6.20 ± 1.33	4.96 ± 1.15	8.27 ± 1.34

- The higher absorption spectral dependence between 405~532nm is in good accordance with the Heshan observation influenced by biomass burning.

Summary of brown carbon study in China

- BC absorption still dominates the total absorption, while BrC absorption cannot be neglected, contributing ~10% (@ 405 &532nm) in the PRD region of China.

- The tunnel experiments further supported the effectiveness of AAE_bc values extrapolated from ambient measurements.

- The biomass burning simulation experiments further proved the higher absorption spectral dependence between 405 and 532nm as a result of biomass burning in rural site of PRD region

Thank you !