Markedly enhanced absorption and direct radiative forcing of black carbon under polluted urban environments

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Carbon soot in the atmosphere

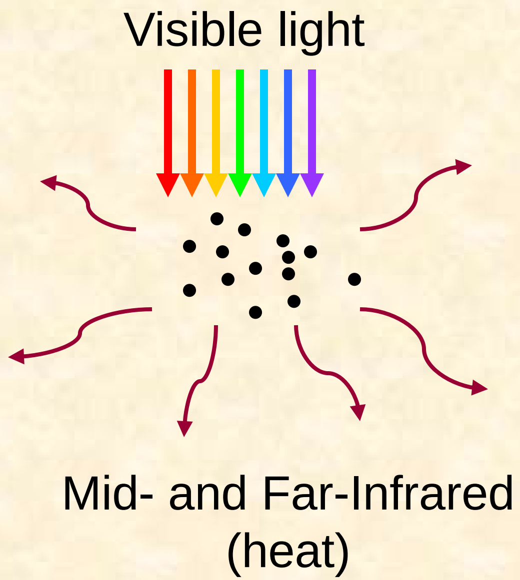
- Emitted from incomplete combustion of fossil fuels and biomass burning
- Average global emission rate of 12-24 Tg yr^{-1}
- Atmospheric processing affects soot morphology, hygroscopicity and optical properties
- Atmospheric aging

  - Condensation of pollutants, e.g. H_2SO_4, organics
  - Coagulation with existing aerosol particles
  - Oxidation by OH, O_3, etc.

Impacts on climate forcing

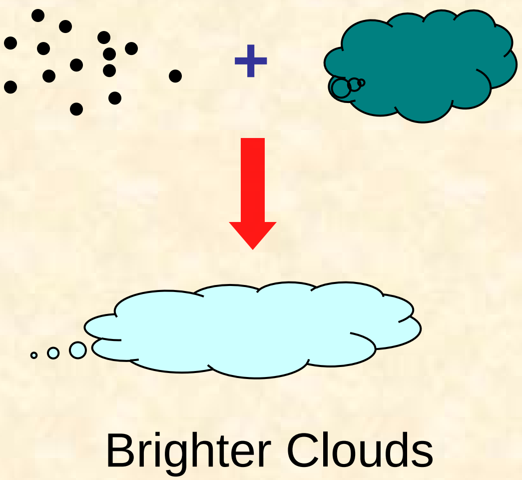
Direct effect

- Absorption and scattering of solar radiation
- High optical absorptivity
  The second most important climate-warming agent after carbon dioxide. Warming effect from soot nearly balances the net cooling effect of other anthropogenic aerosols
- Sulfate and organic coatings enhance absorption and scattering



Indirect effect

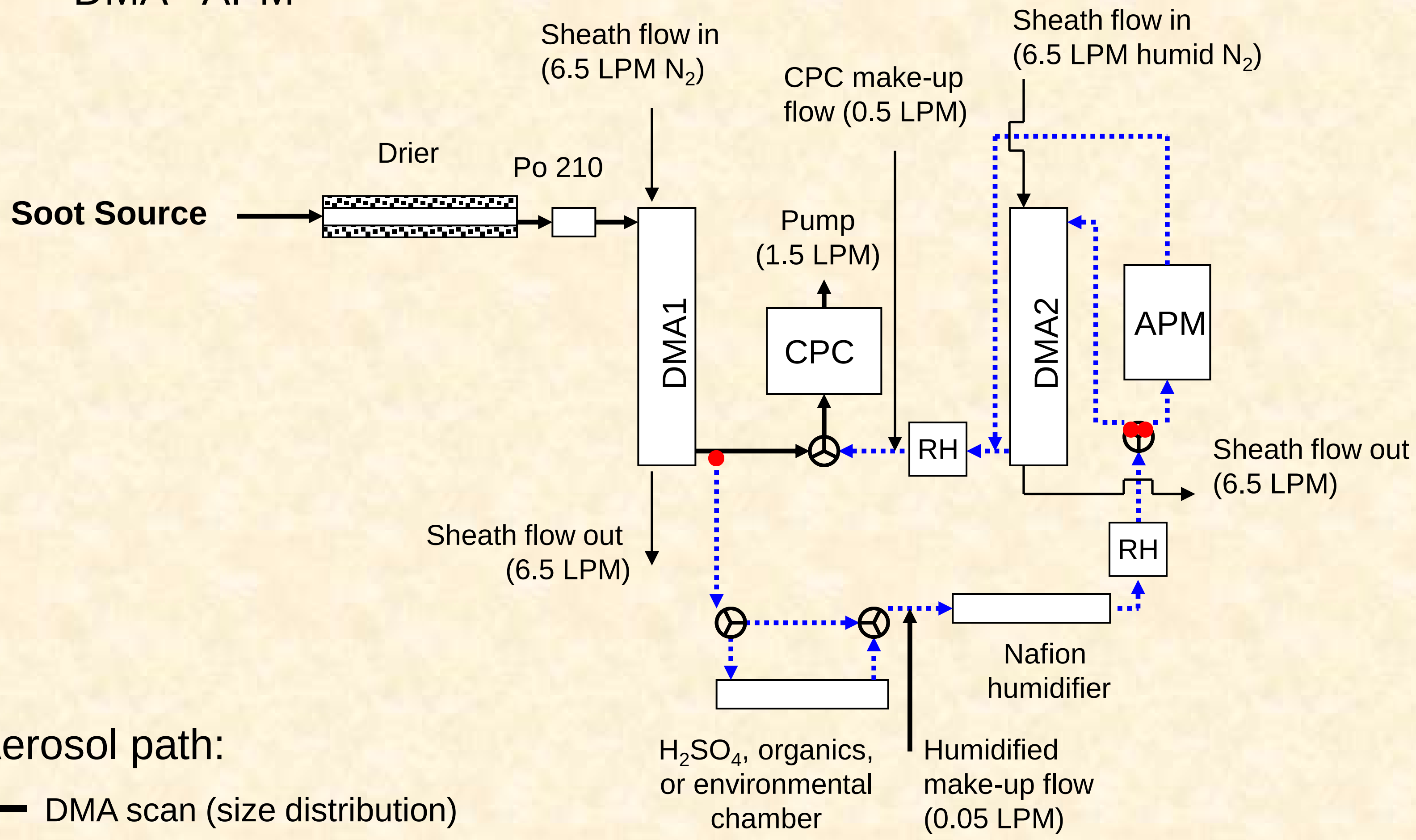
- Fresh soot is typically hydrophobic
- Aged soot can act as potential CCN
  - enhanced albedo (cooling)
  - suppressed or enhanced rain



TDMA/DMA-APM system

Operation modes:

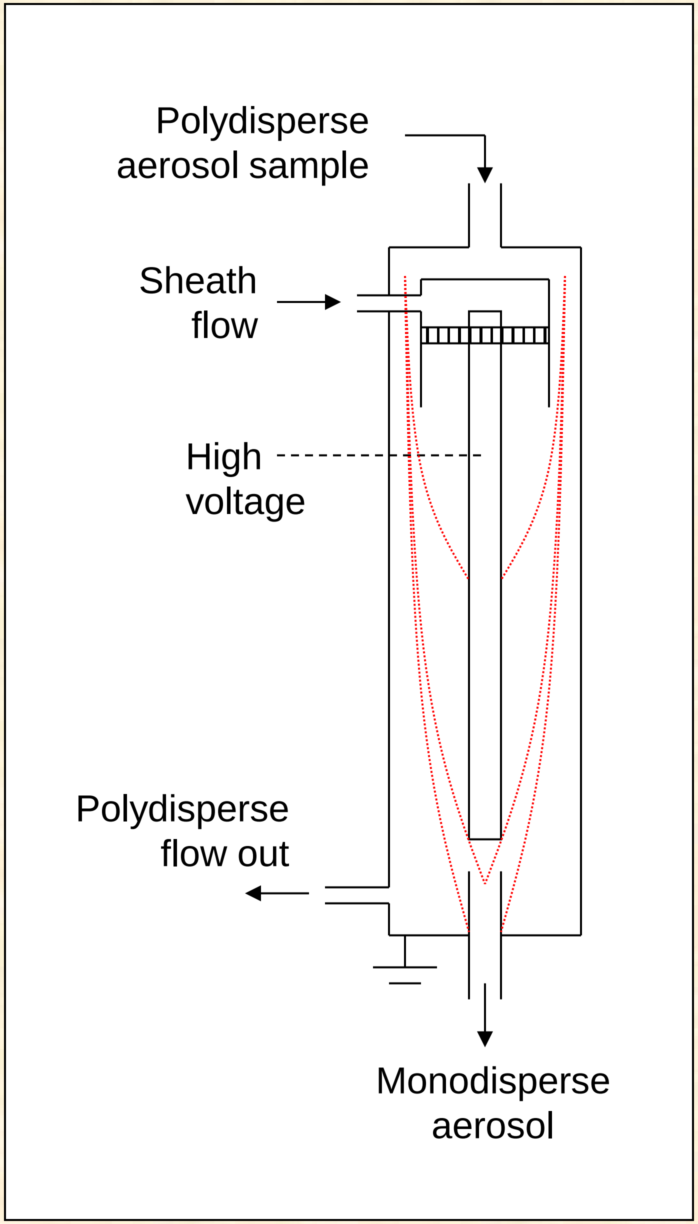
- single DMA (scanning mobility particle sizer)
- DMA - DMA
- DMA - APM

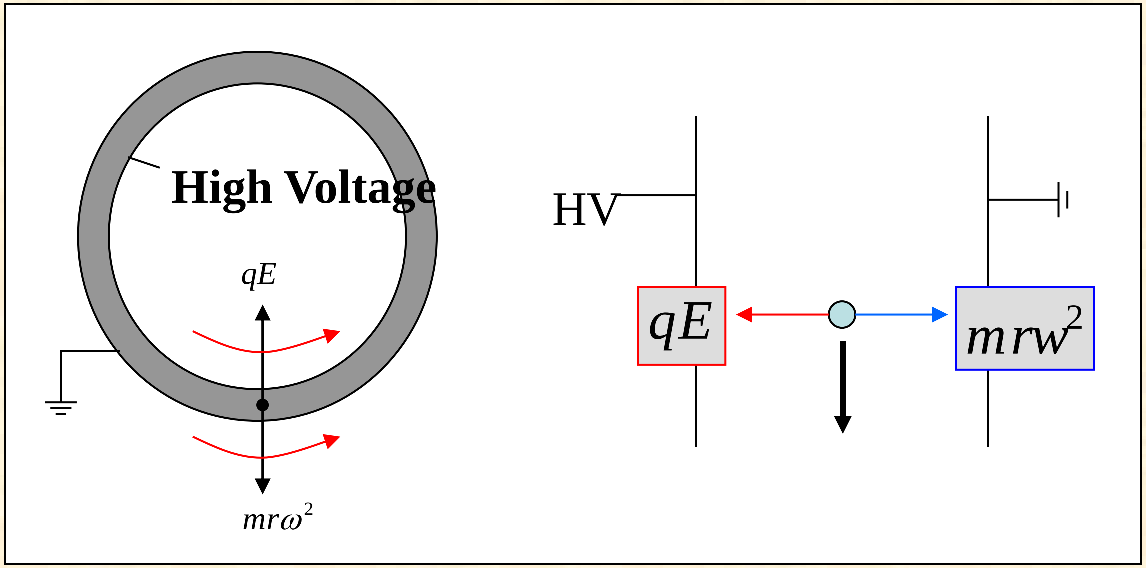


Aerosol path:

- DMA scan (size distribution)
- TDMA or DMA-APM scan

Differential Mobility Analyzer (DMA)   Aerosol Particle Mass (APM) Analyzer





DMA

- **Particles of a certain electrical mobility can penetrate through the DMA for the fixed sheath to sample flow ratio and voltage**

$$Z_p = \frac{neC}{3\pi\mu D_p}$$

APM

- **Particles of a certain mass can penetrate through the APM for the fixed rotational speed and voltage**
- **Electrostatic force = Centrifugal force**

$$mr\omega^2 = \frac{\pi d_{ve}^{\ 3}}{6}\rho_{true} r\omega^2 = neE_{APM}$$

Effective Density and Fractal Dimension of Soot

- **Effective density of soot calculated from the mass (DMA-APM) and mobility (DMA-DMA) measurements**

$$\rho_{eff} = \frac{m}{V} = \frac{6m}{d_B^3 \pi}$$

- **Fractal dimension, D_f, indicates how completely a fractal appears to fill space**

$$m \propto d_B^{Df}$$

(d_B is mobility diameter)

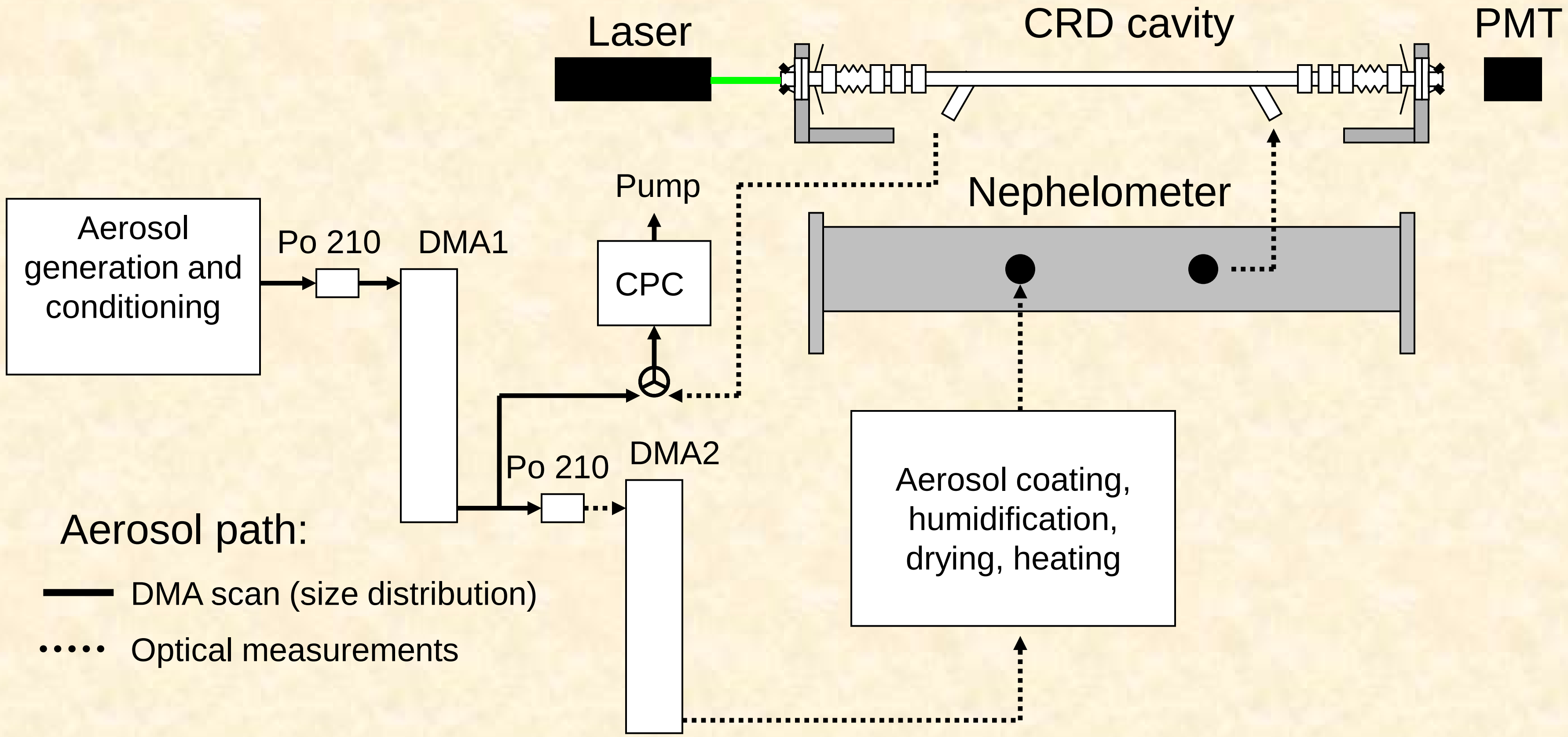
$$\rho_{eff} \propto d_B^{Df-3}$$

- **Fractal dimension of a plane is 2**

  a solid sphere is 3

$$D_{ve} = \sqrt[3]{\frac{6m_p}{\pi\rho_m}} \qquad \chi = \frac{D_p}{C_p}\frac{C_{ve}}{D_{ve}}$$

TDMA/Nephelometer/Cavity Ring-Down



Environmental Science & Technology
Article
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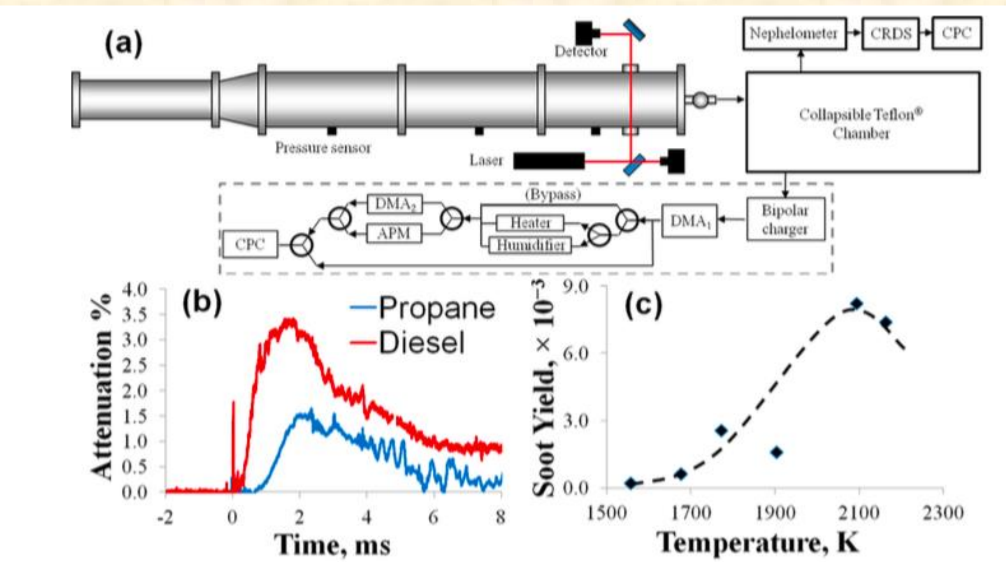
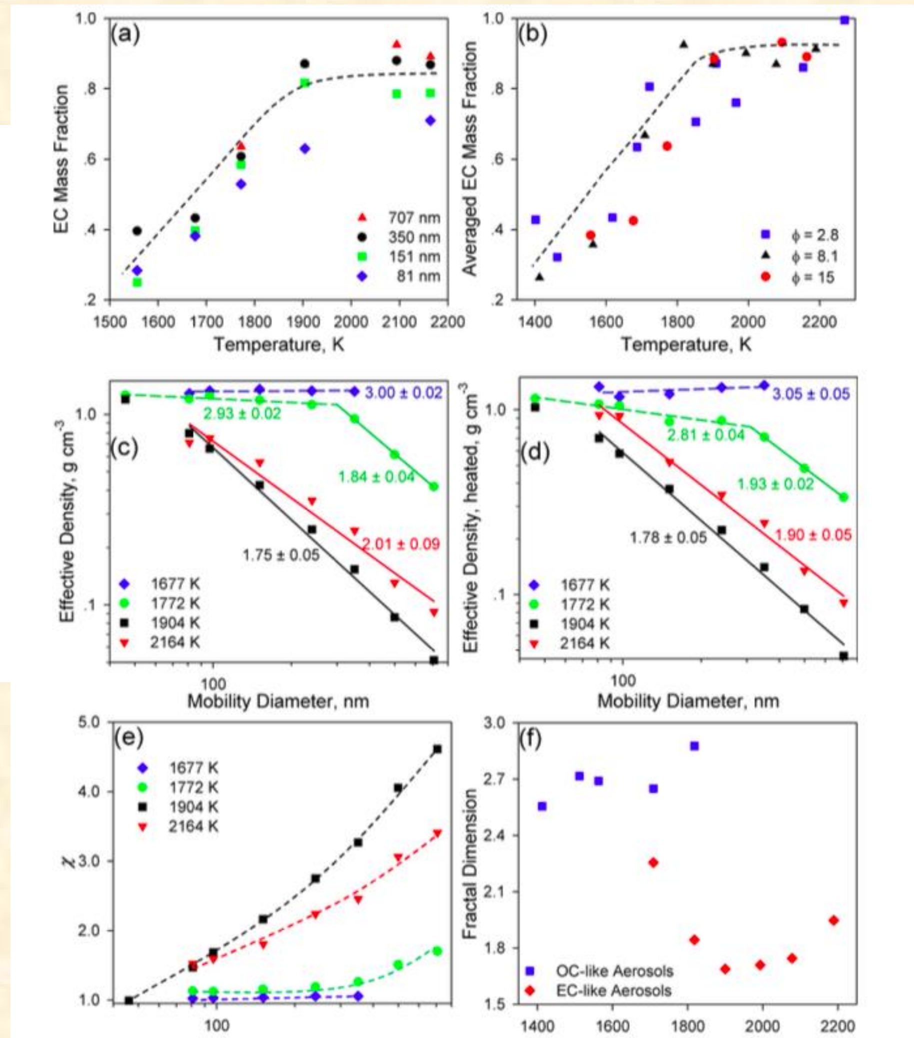
High Sensitivity of Diesel Soot Morphological and Optical Properties to Combustion Temperature in a Shock Tube

Chong Qiu,*[†,‡] Alexei F. Khalizov,[§,∥] Brian Hogan,[⊥] Eric L. Petersen,[⊥] and Renyi Zhang*[‡,§]





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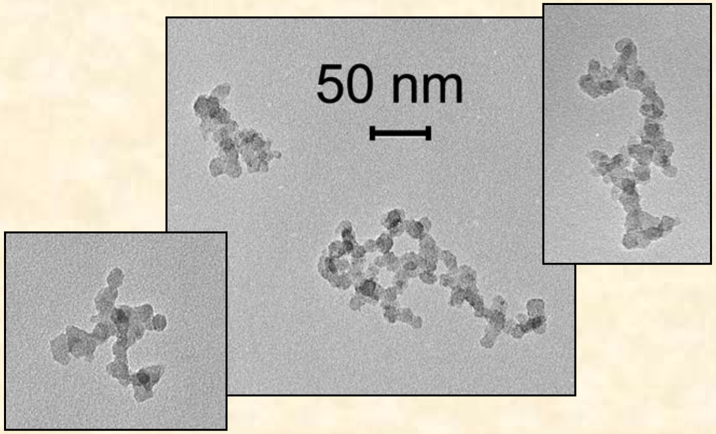
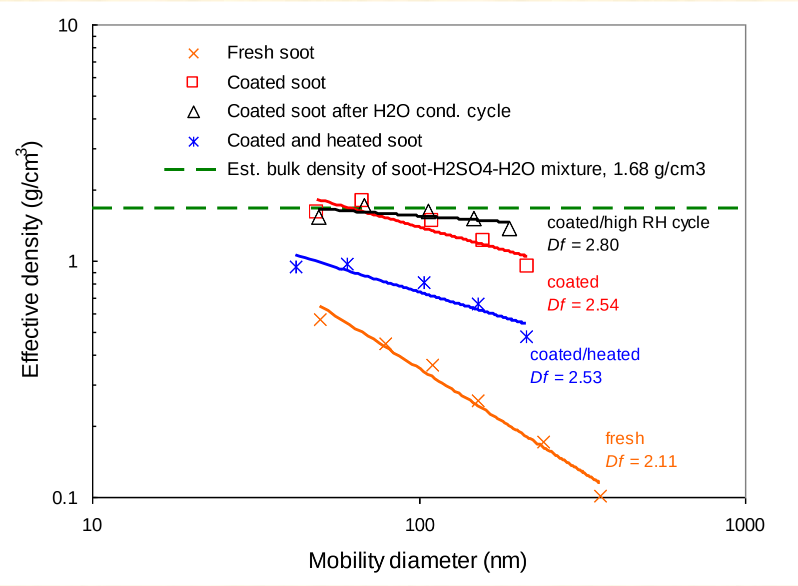
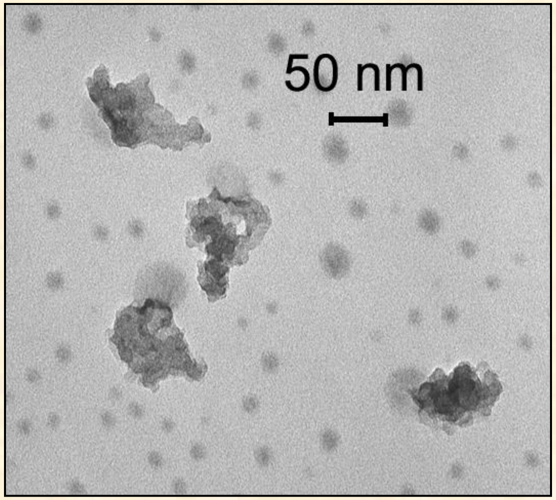
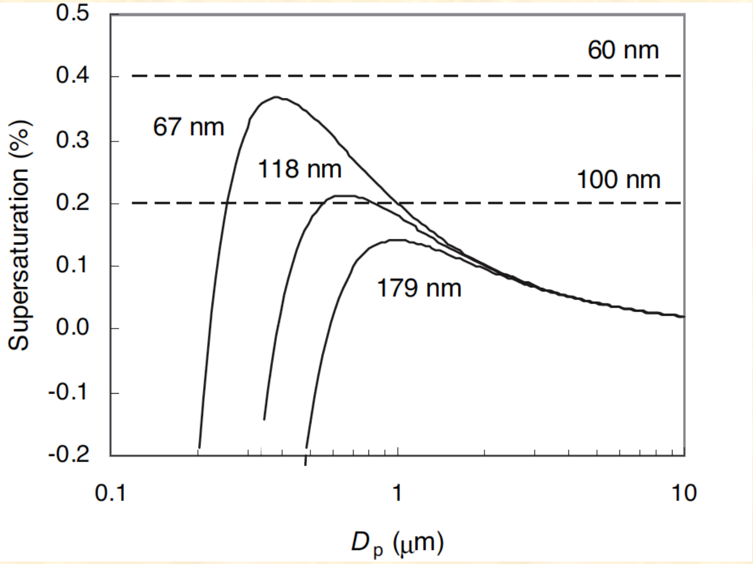


Fresh soot agglomerates

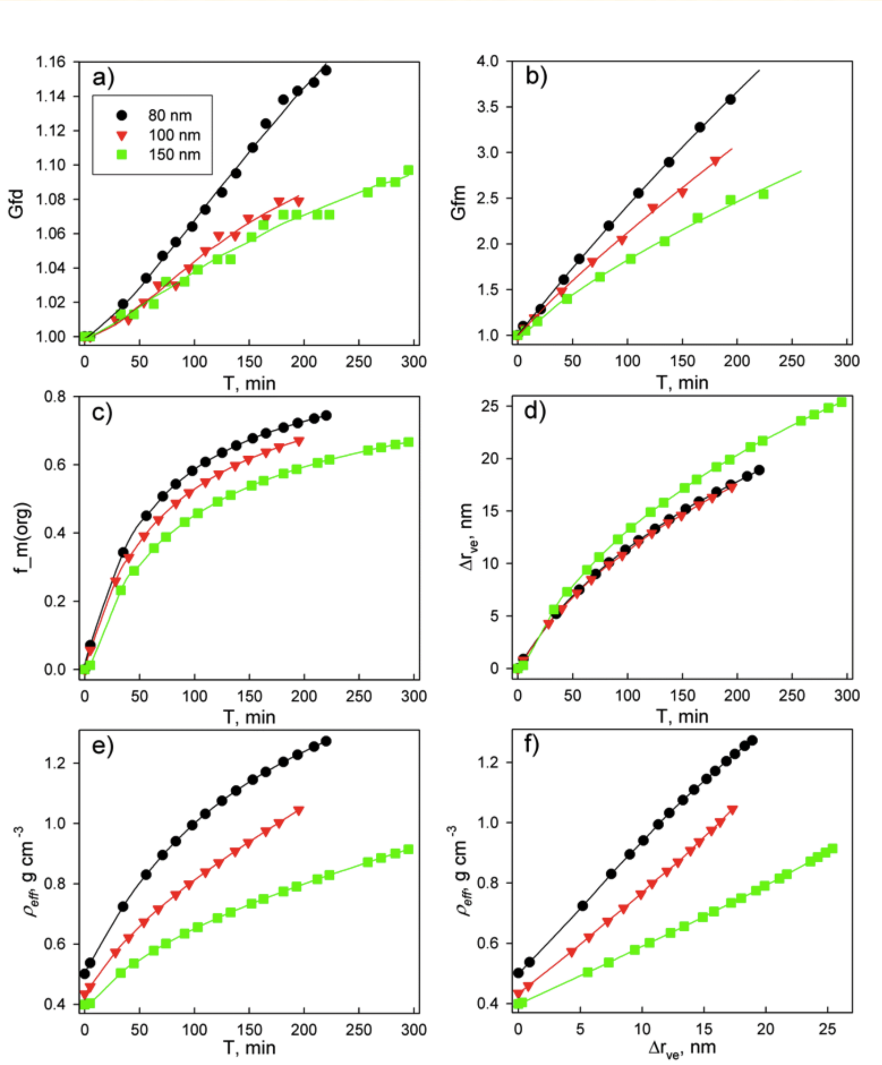


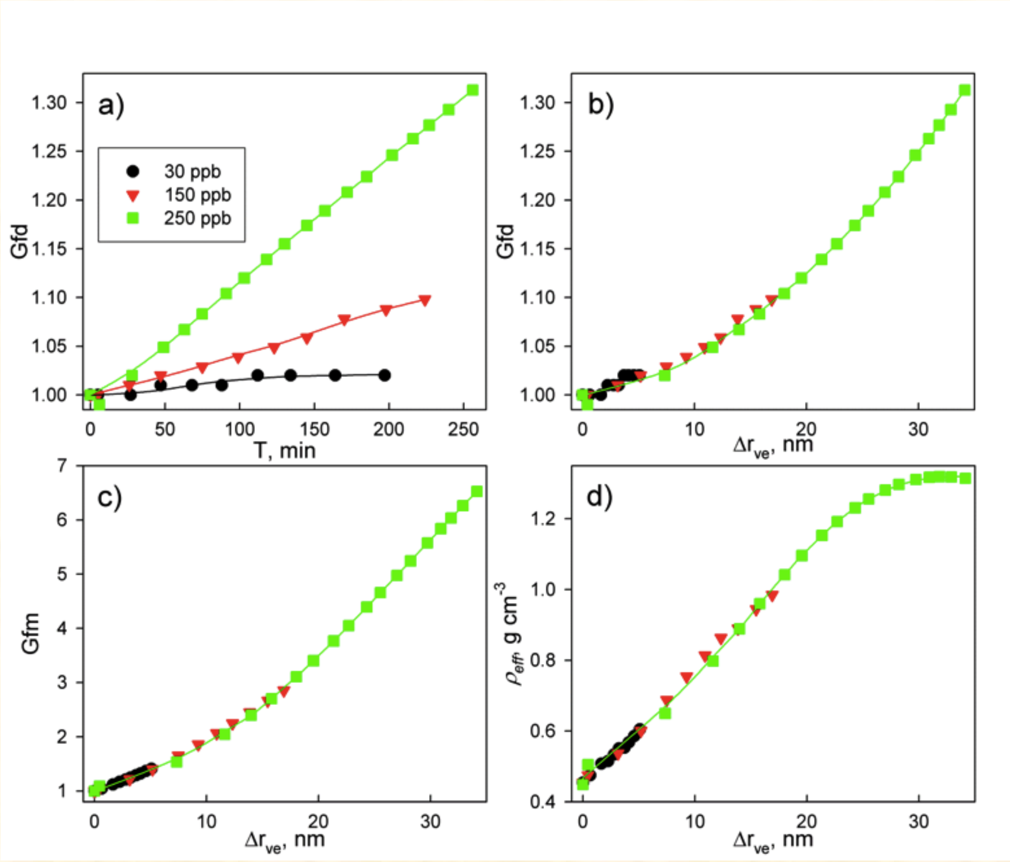


Soot exposed to 1.5×10^{10} cm^{-3} H_2SO_4 vapor for ~12 s

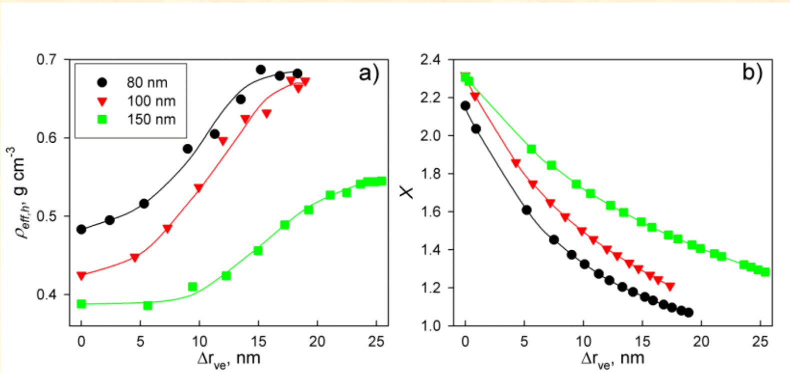


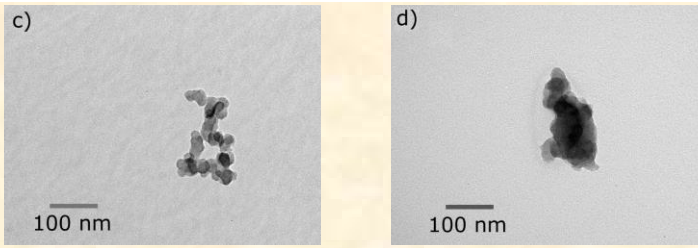
Qiu et al. Soot Aging from OH-Initiated Oxidation of Toluene, *Environ. Sci. Technol*. 46, 9464 (2012)

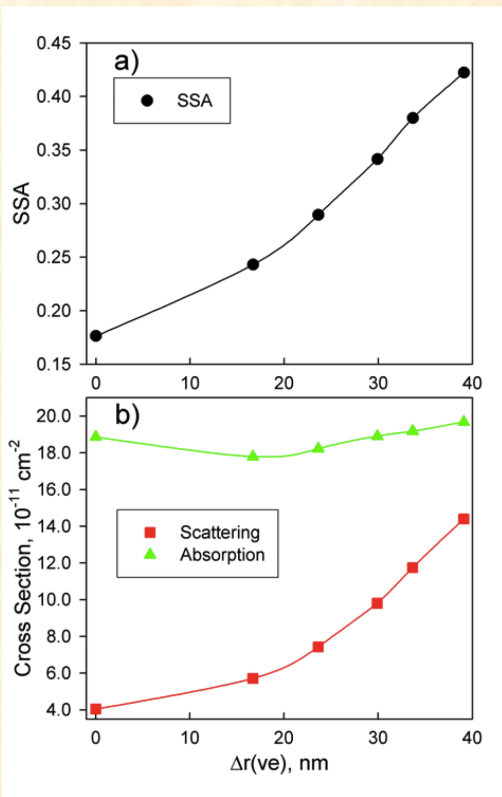


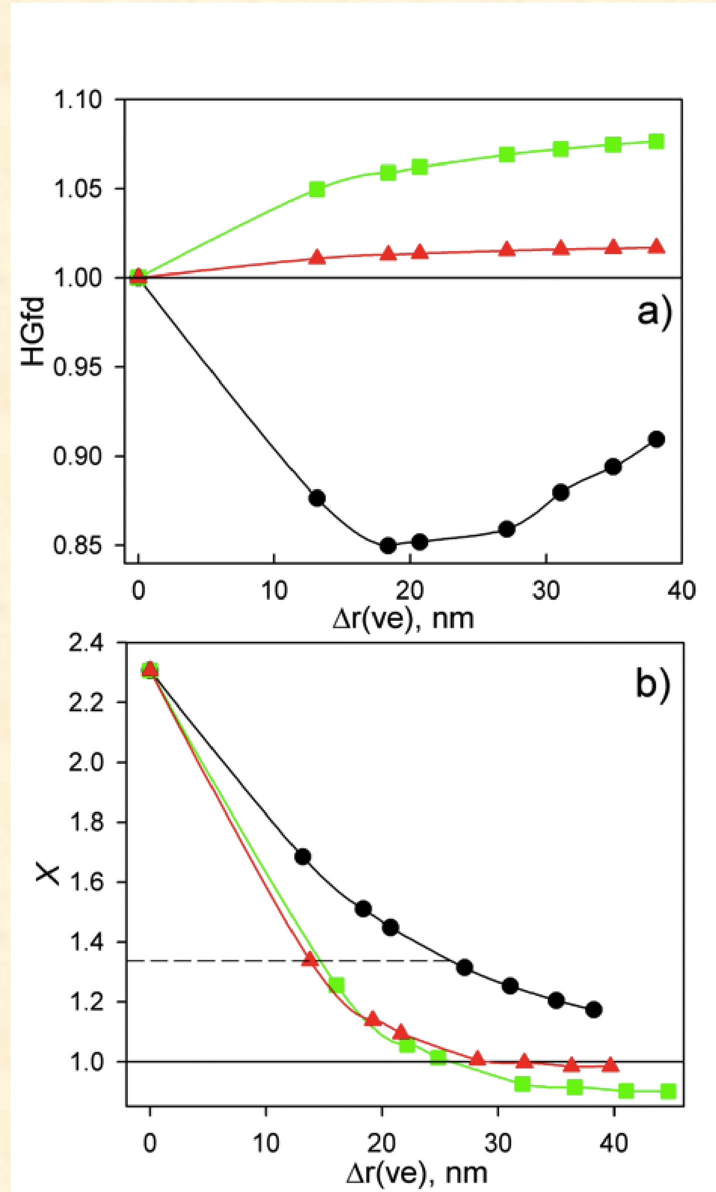


***Qiu et al*. Soot Aging from OH-Initiated Oxidation of Toluene, *Environ. Sci. Technol*. 46, 9464, 2012**









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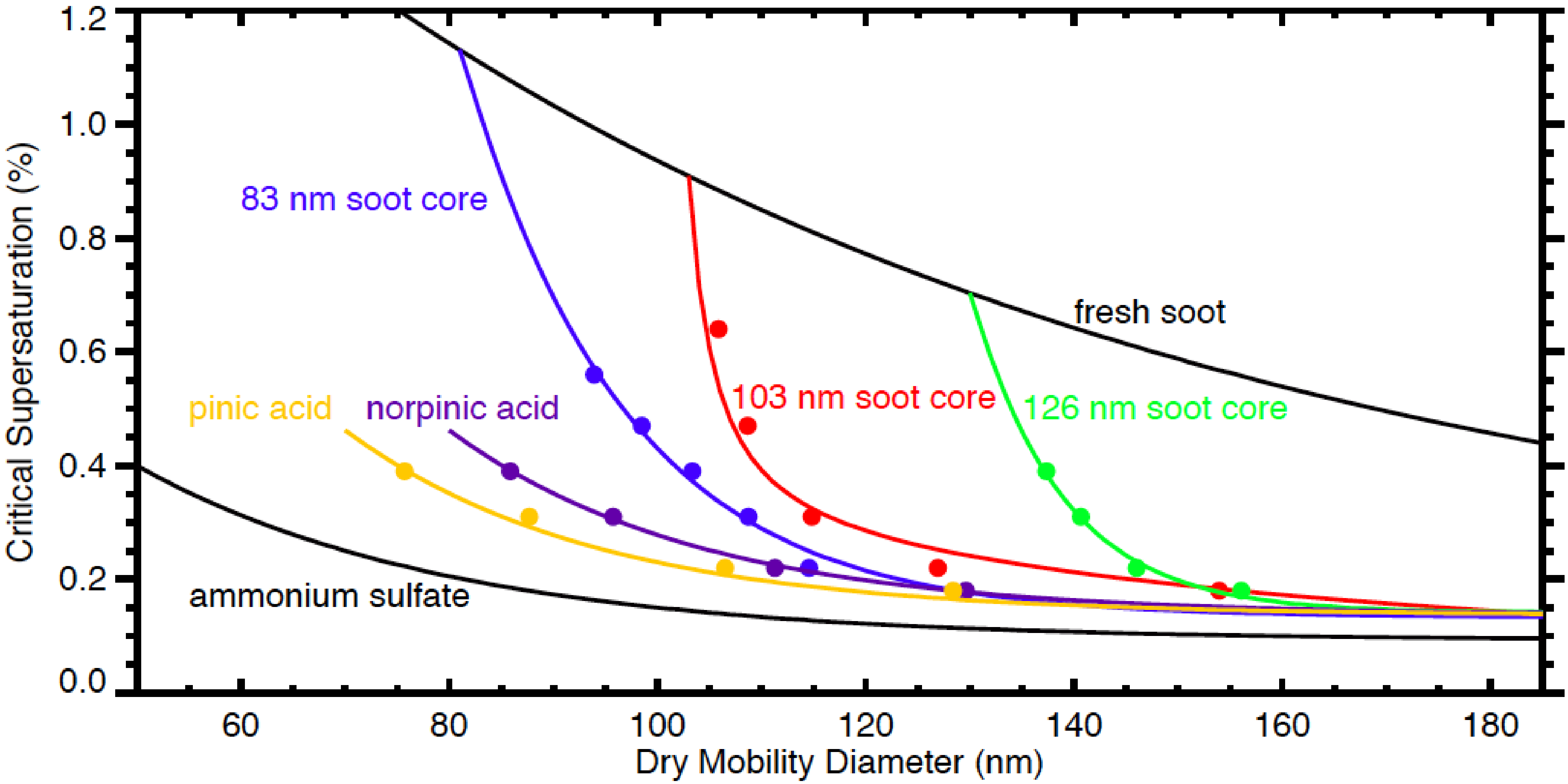
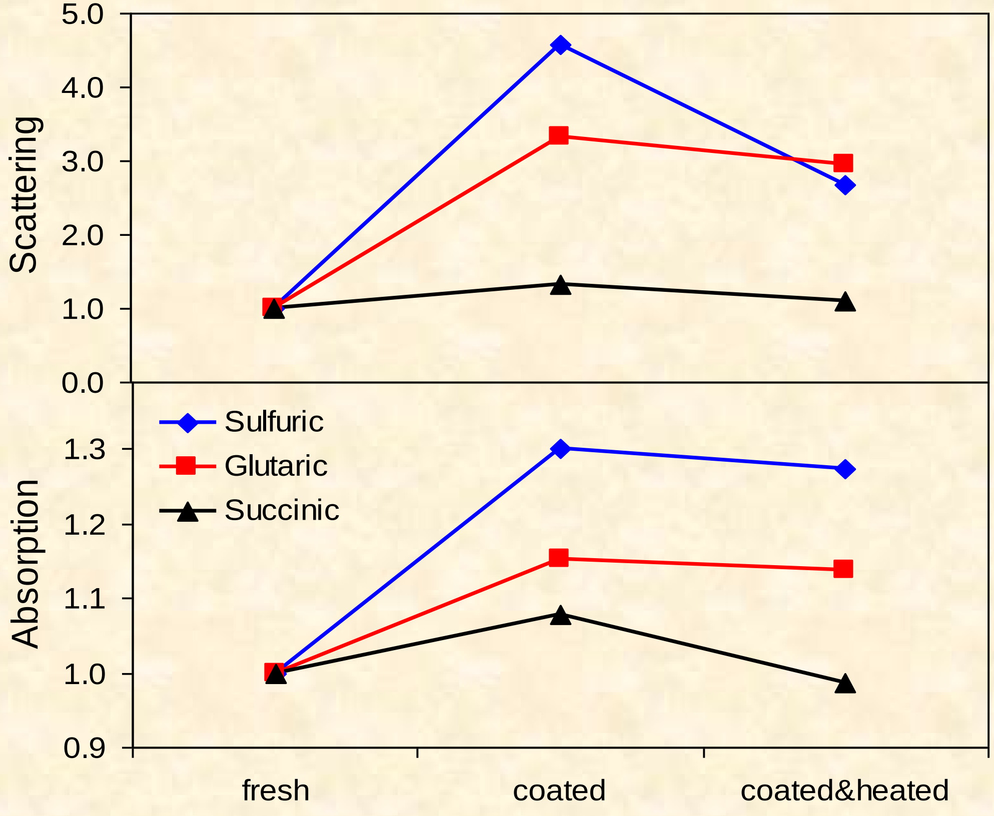
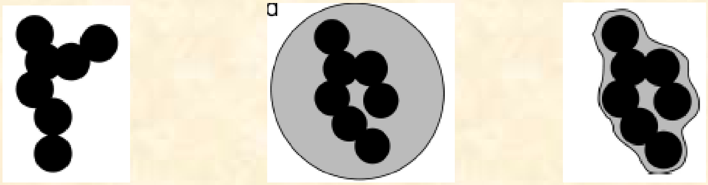


Lensing and restructuring for different coatings

Initial diameter 320 nm; coating fraction 30-40% by mass

Amplification = $\sigma_{processed}/\sigma_{fresh}$





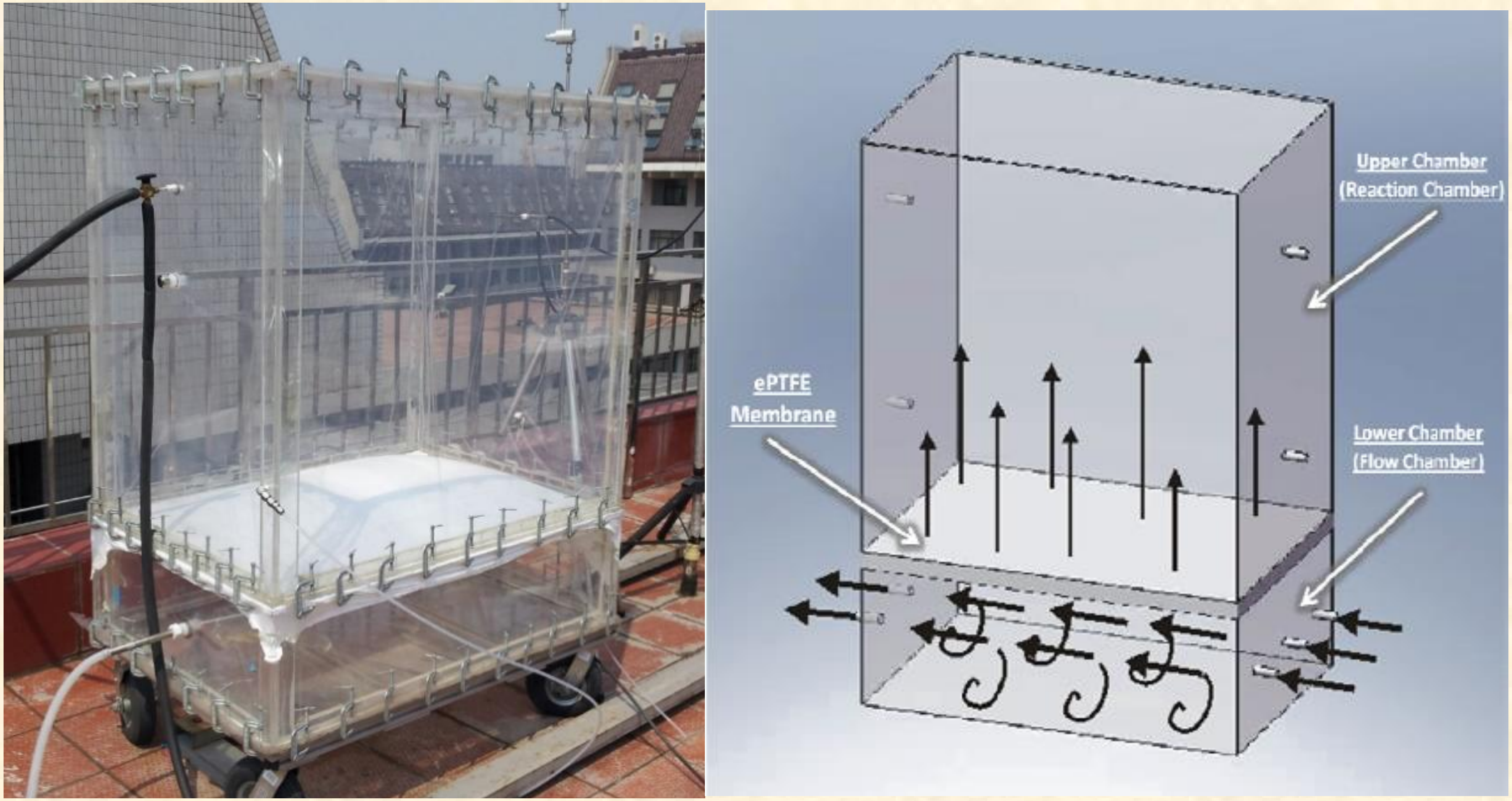
***(Xue et al., Phys. Chem. Chem. Phys*. 11, 7865, 2009)**

A Tales of Two Cities: US and China "Air Pollution Capitals"
Houston vs. Beijing

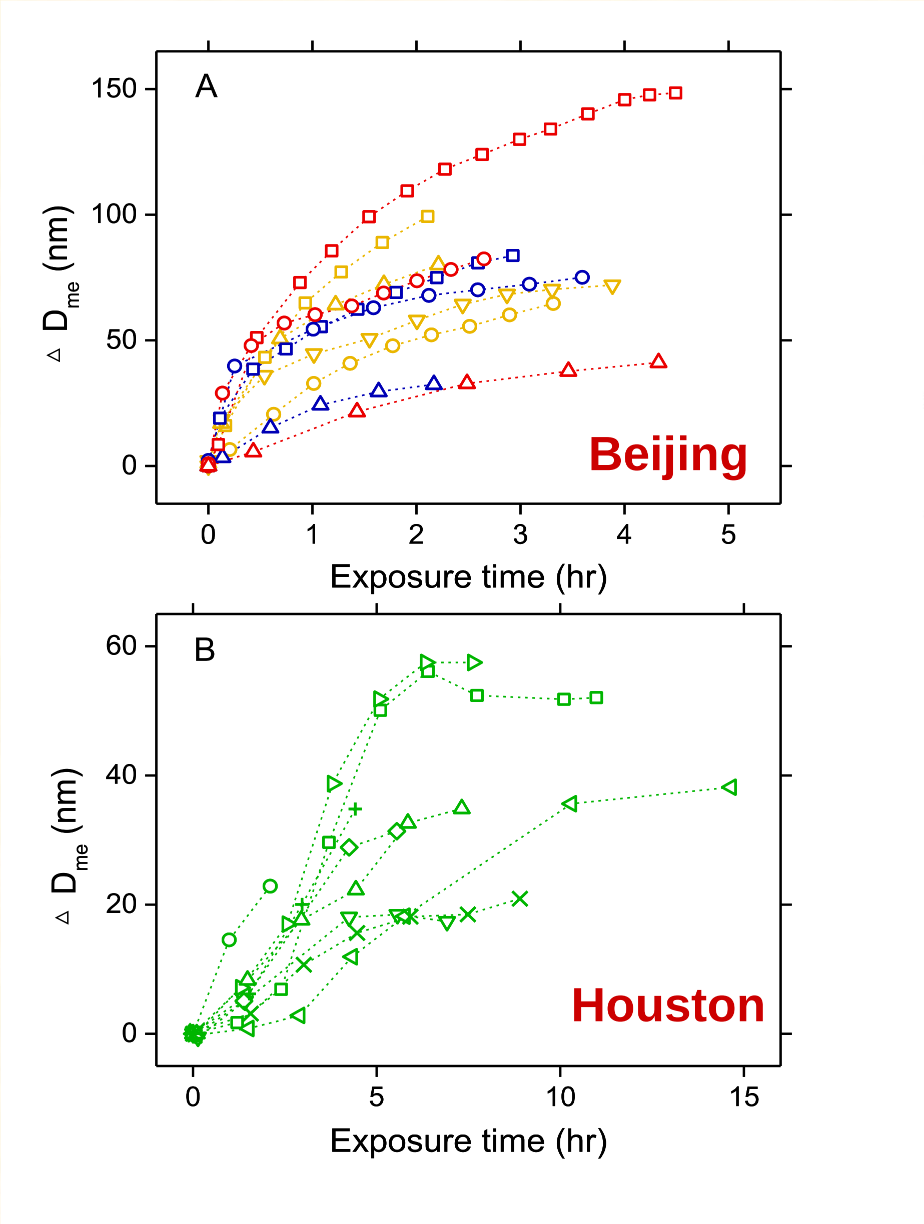
	Houston, Texas	Beijing, China
Population	6.2 million	21.2 million
Area	10,062 square miles ($26{,}060\ km^2$)	6,487 square miles ($16{,}801\ km^2$)
Number of Vehicles	3.3 million	5.4 million
Industry	Energy industry - oil and natural gas, biomedical research, aeronautics, top U.S. market for exports, petroleum products, chemicals, and oil and gas extraction equipment.	Services sector (76.9%) followed by manufacturing and construction (22.2%), agriculture and mining (0.8%).

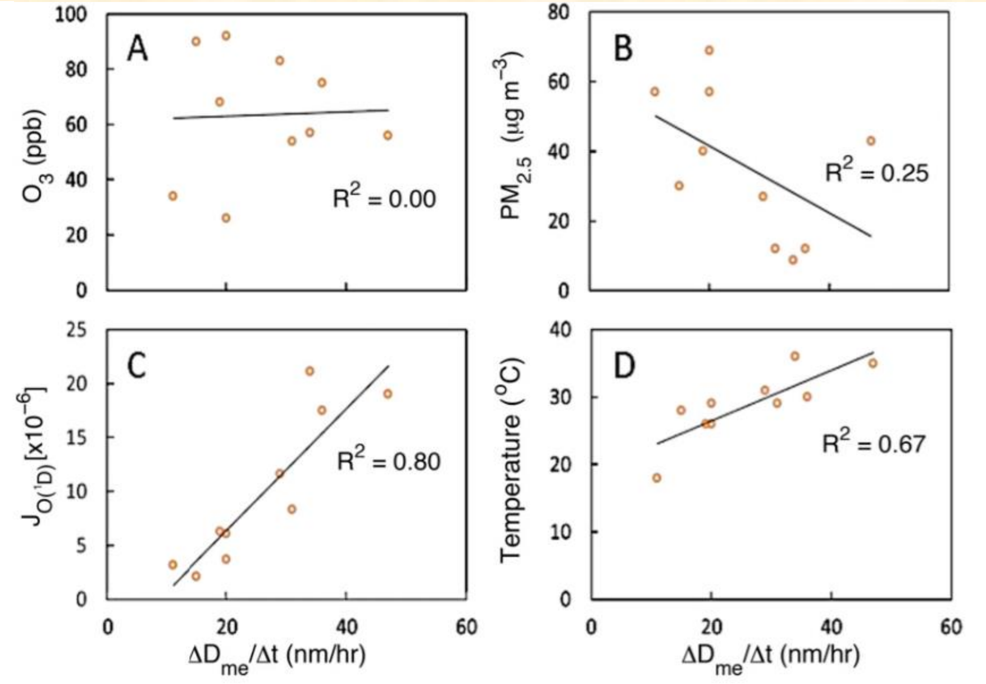
	Houston	Beijing
Primary Pollutants:	NOx, VOCs (light olefins)	NOx, SO_2, VOCs (aromatics)
Secondary Pollutants:	Ozone	PM

The QUALITY chamber. Photo (left) and schematic (right) of the quasi-atmospheric aerosol evolution study (QUALITY) chamber.



J. Peng et al., **Markedly enhanced absorption and direct radiative forcing of black carbon under polluted urban environments*, Proc. Natl. Acad. Sci. USA 113*, 4266–4271 (2016)**





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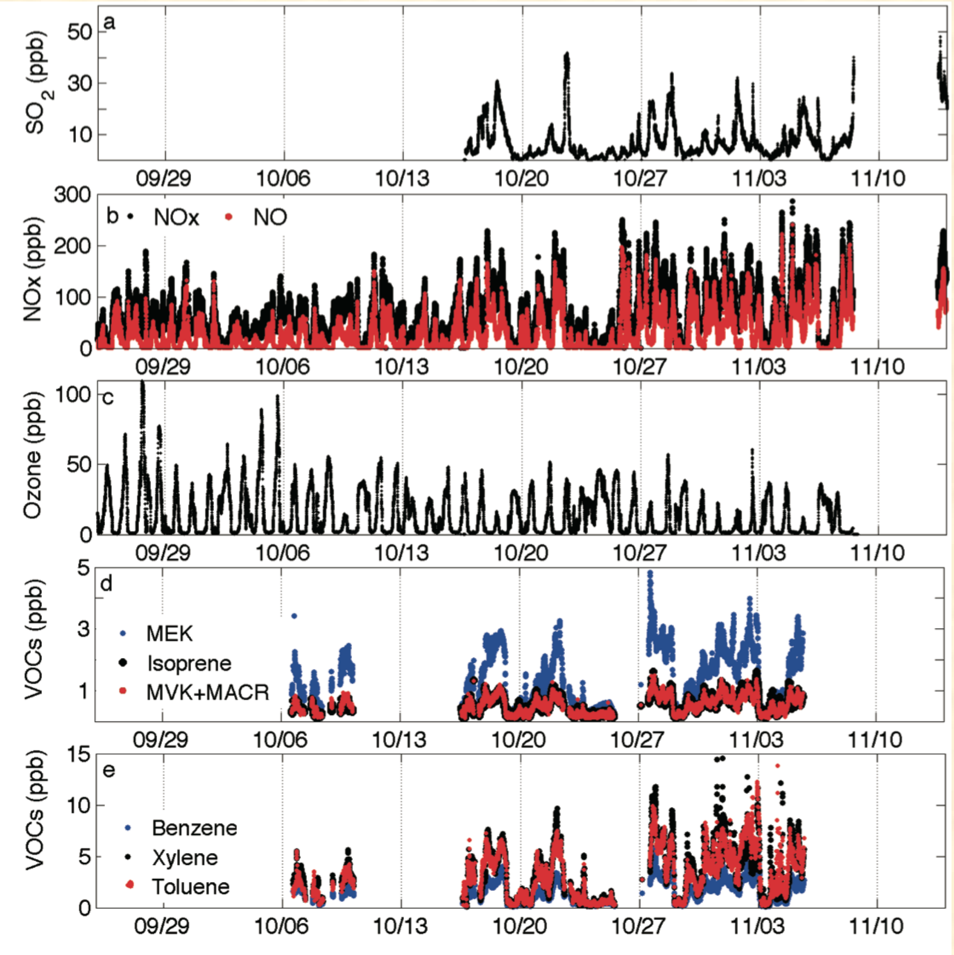
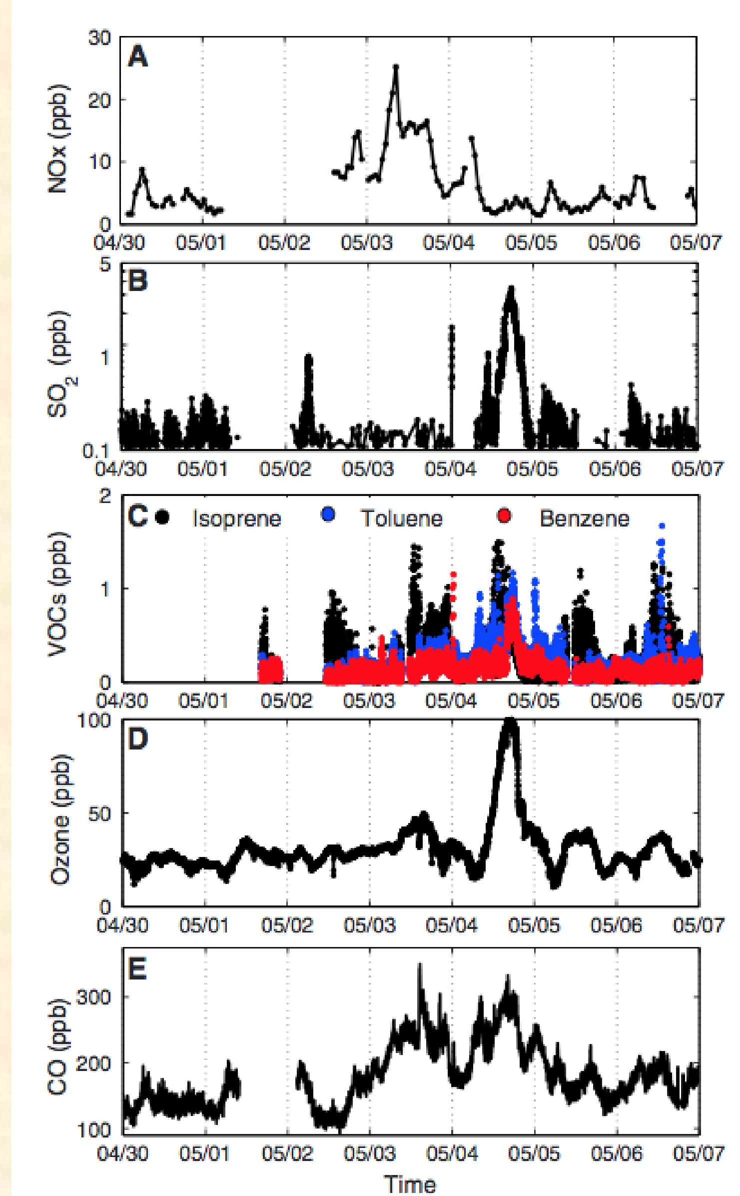


***S. Guo et al*., Elucidating severe urban haze formation in China, *Proc. Natl Acad. Sci. USA* 111, 17373, (2014)**



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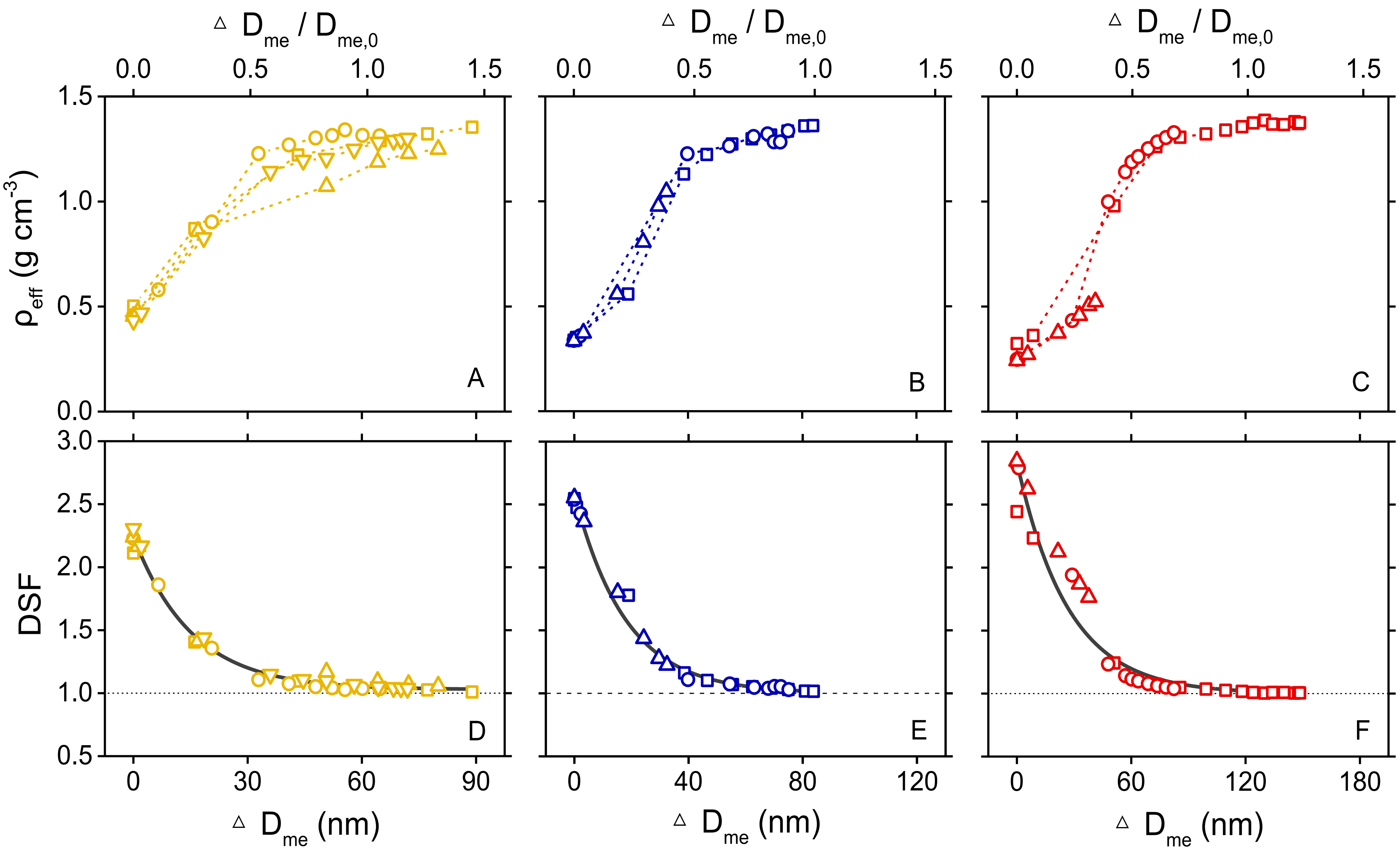

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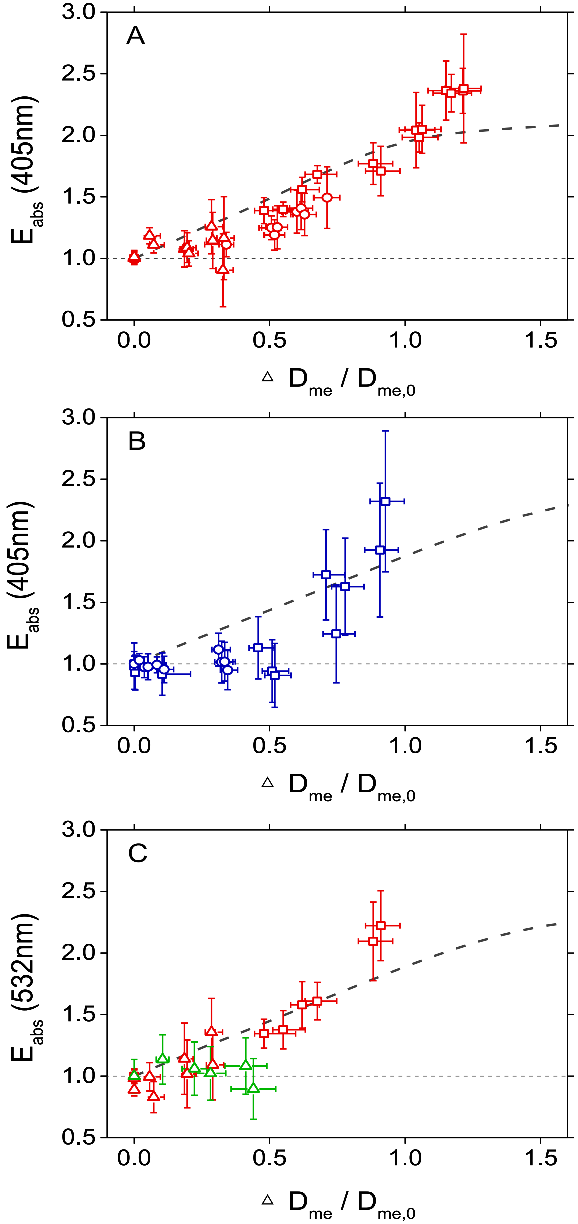

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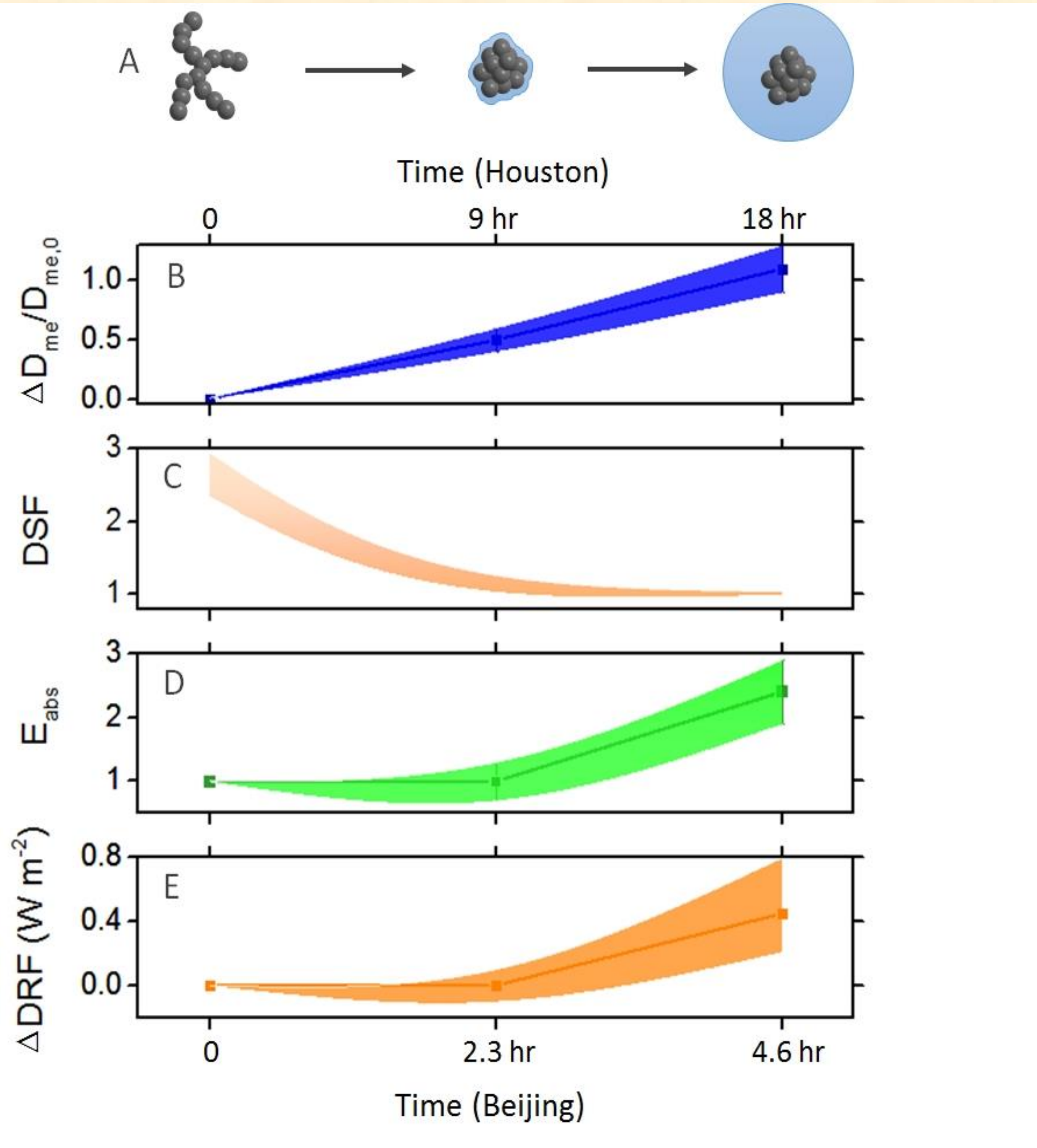

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COMMENTARY

Convergence on climate warming by black carbon aerosols

Örjan Gustafsson[a,1] and Veerabhadran Ramanathan[b]

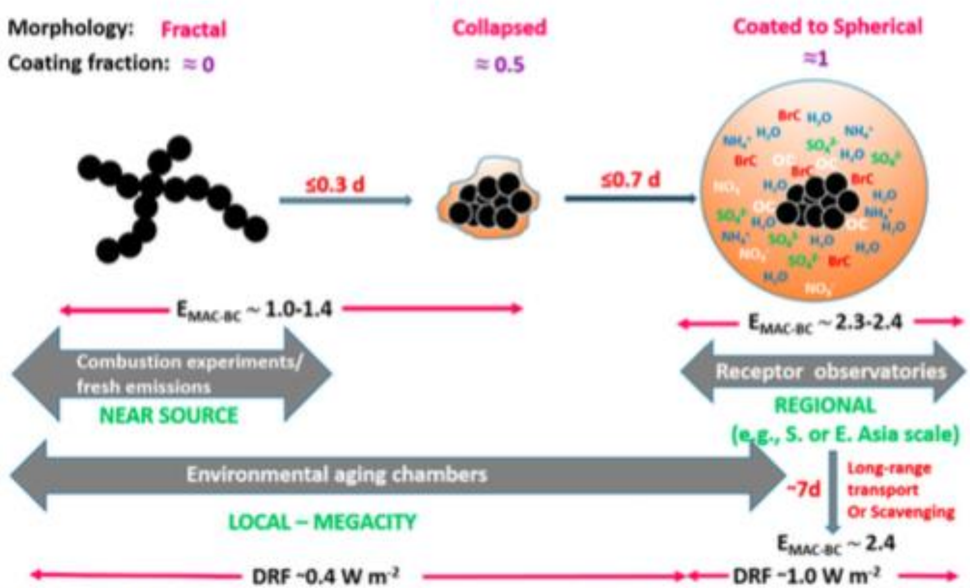

Fig. 2. Time-course evolution of BC aerosol composition, light absorption (where E_{MAC-BC} is the enhancement because of coatings), and associated climate effects (as DRF).

"BC solar absorption became a central issue in climate change research when a synthesis of satellite, in situ, and ground observations concluded that the global solar absorption (i.e., direct radiative forcing, DRF) by atmospheric BC is as much as 0.9 W·m^{-2}, second only to the CO_2 DRF"

"This will close the factor of 2–3 gap between model predictions and observations on the effect of BC aerosols on climate"

Atmospheric Environment 81 (2013) 713–715

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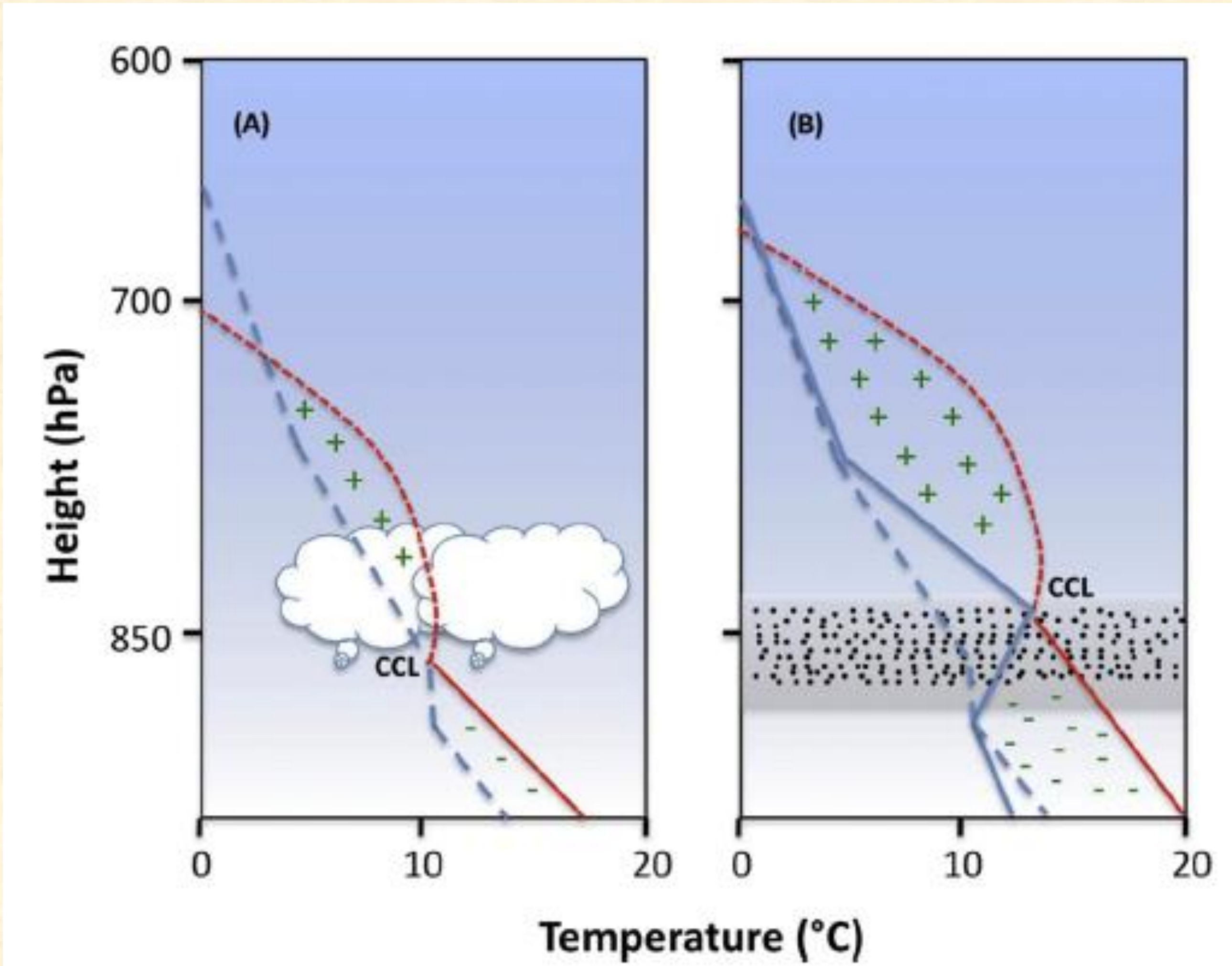
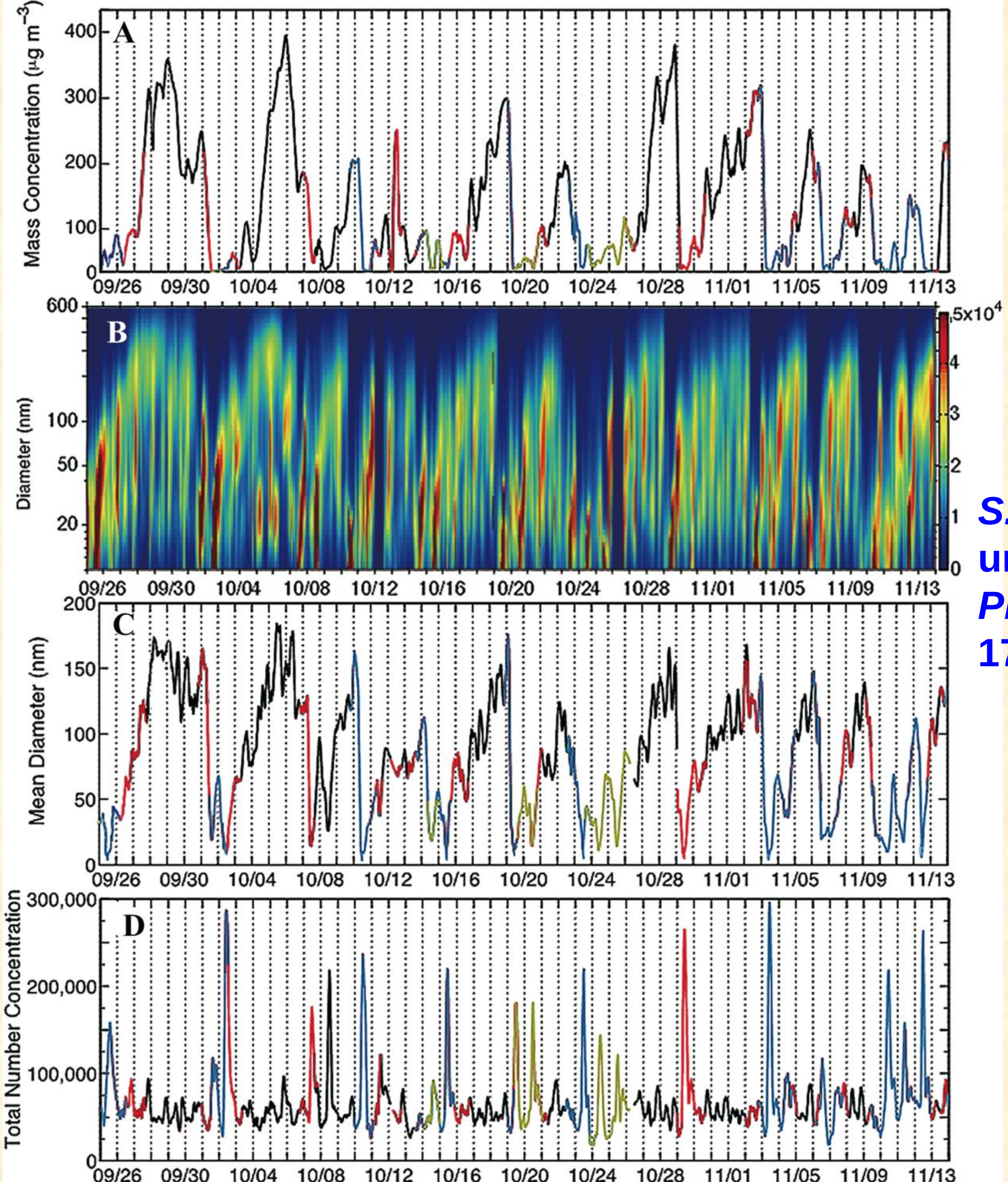
New Directions: Light absorbing aerosols and their atmospheric impacts☆





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Conclusions

- **Soot particles undergo rapidly aging under atmospheric conditions, considerably modifying their properties**
- **Atmospheric aging of soot particles leads to enhanced hygroscopicity**
- **Atmospheric aging of soot particles enhances absorption and scattering, because of changed mass and morphology**

Acknowledgements

Zhang's group members and colleagues from PKU (Prof. Min Hu's group)
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