



# **Enhanced light absorption and direct radiative forcing by Black Carbon agglomerates**

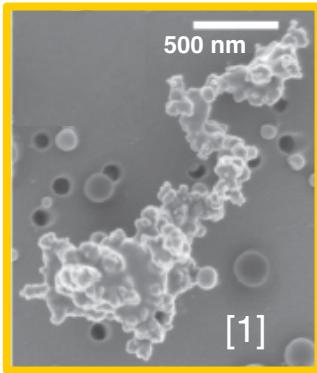
Georgios A. Kelesidis<sup>1</sup>, David Neubauer<sup>2</sup>, Liang-Shih Fan<sup>3</sup>,  
Ulrike Lohmann<sup>2</sup> & Sotiris E. Pratsinis<sup>1</sup>

<sup>1</sup>Particle Technology Laboratory, ETH Zürich, Switzerland

<sup>2</sup>Institute of Atmospheric and Climate Science, ETH Zürich, Switzerland

<sup>3</sup>The Ohio State University, Ohio, USA

# Climate models vs. AERONET Observations



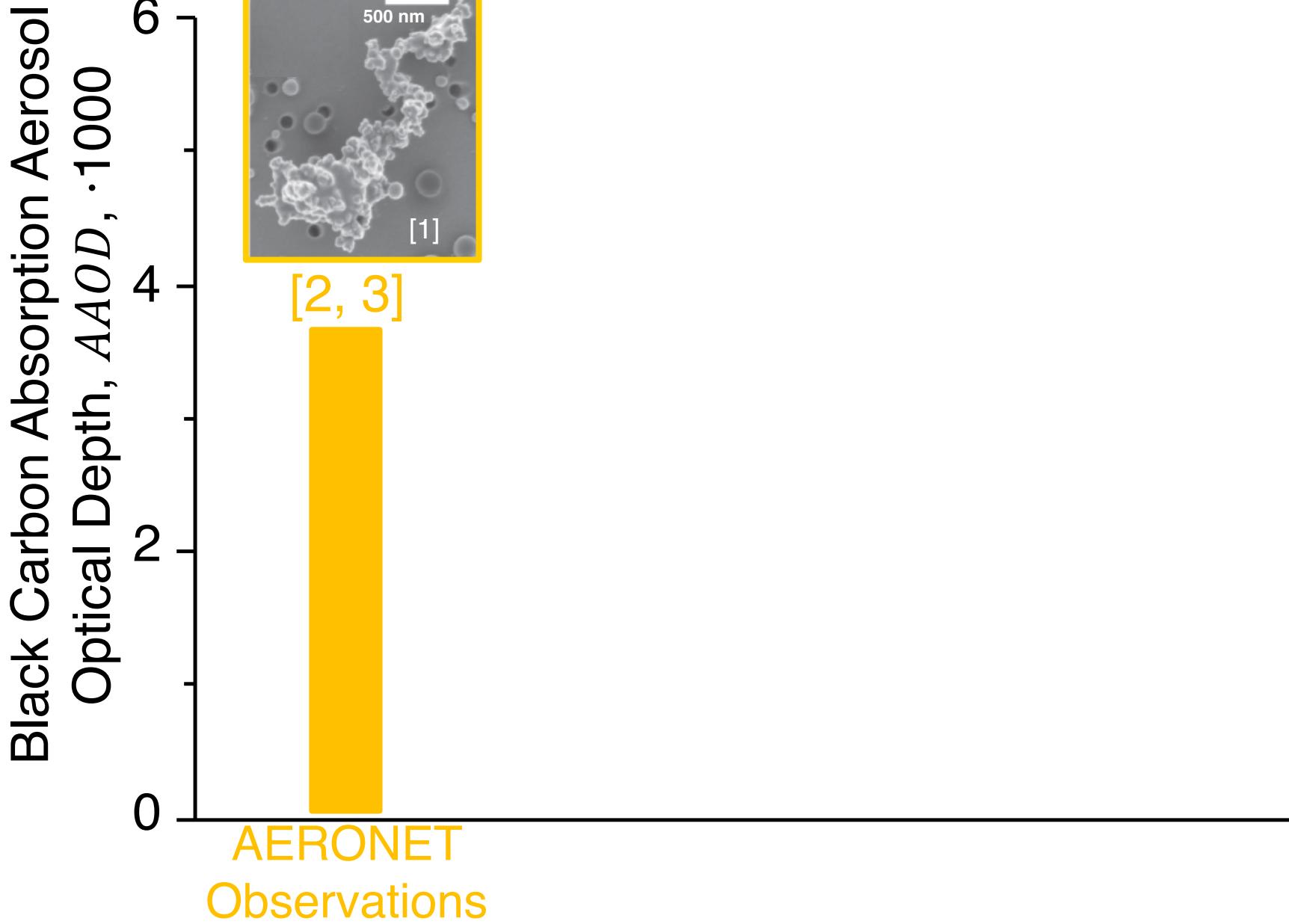
Direct Radiative forcing,  $RF$ :

$$RF = AAOD \square RFE$$

Absorption Aerosol  
Optical Depth

$RF$  efficiency

# Climate models vs. AERONET Observations

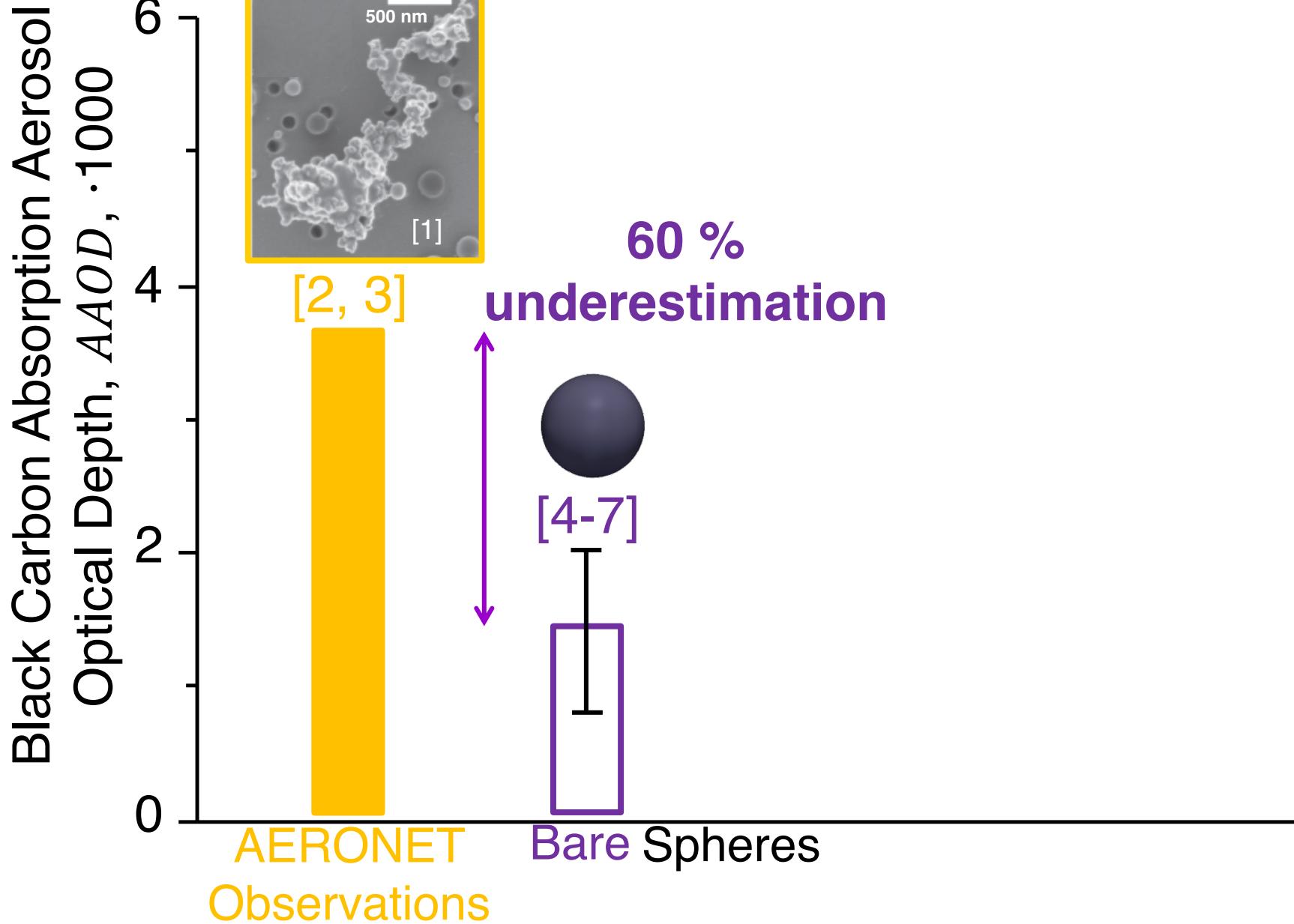


[1] S. China, et al., *Nature Comm.* 4 (2013) 2122-1-7.

[2] C. Chen, et al., *Atmos. Chem. Phys.* 19 (2019) 14585-14606.

[3] M. Sand, et al., *Atmos. Chem. Phys. Diss.* (2021) doi.org/10.5194/acp-2021-51.

# Climate models vs. AERONET Observations



[1] S. China, et al., *Nature Comm.* 4 (2013) 2122-1-7.

[2] C. Chen, et al., *Atmos. Chem. Phys.* 19 (2019) 14585-14606.

[3] M. Sand, et al., *Atmos. Chem. Phys. Diss.* (2021) doi.org/10.5194/acp-2021-51.

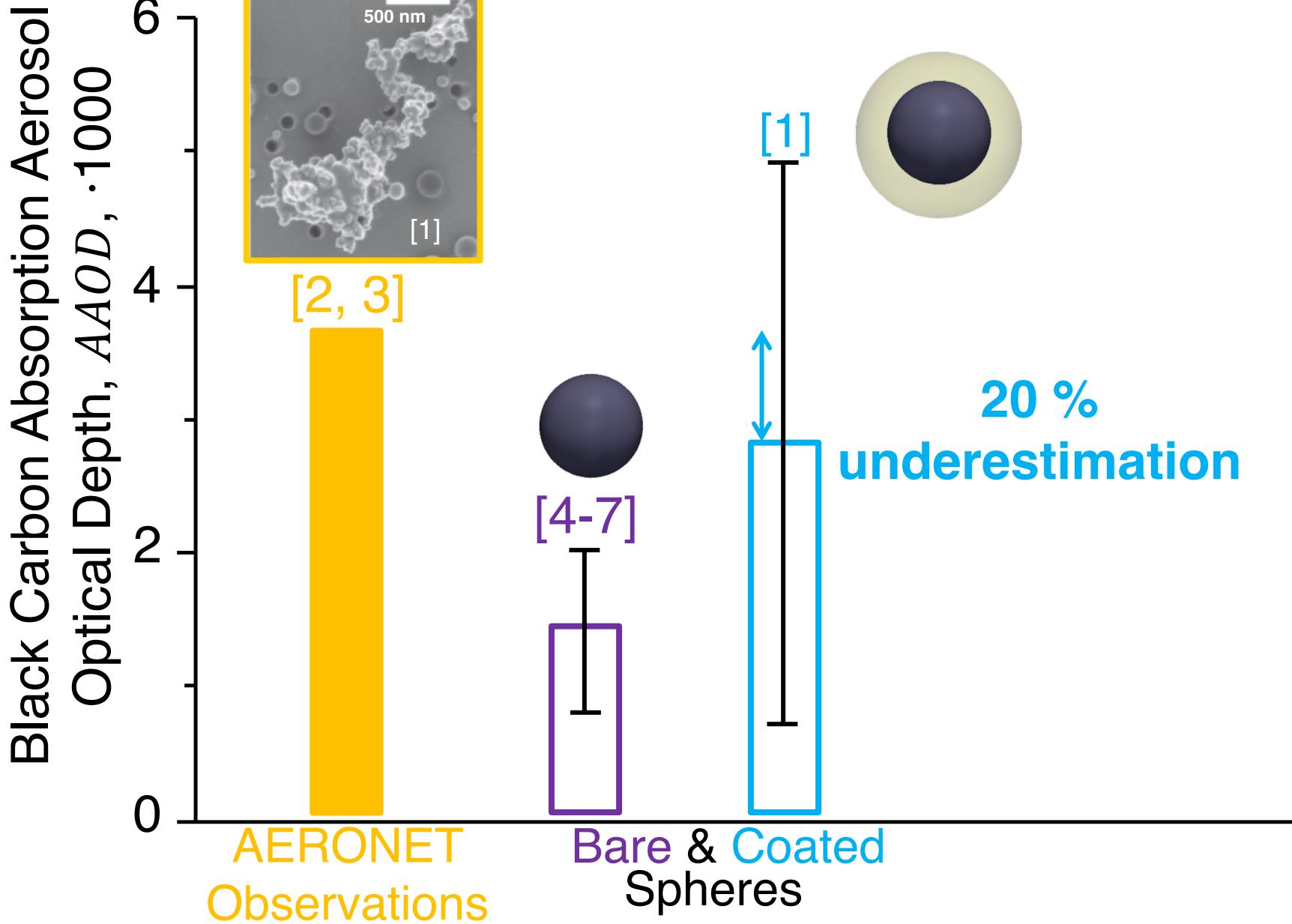
[4] D. Kim, et al., *J. Geophys. Res.* 113 (2008) D16309-1-36.

[5] S.H. Chung, J.H. Seinfeld, *J. Geophys. Res.* 107 (2002) 4407-1-33.

[6] M. Schulz, et al., *Atmos. Chem. Phys.* 6 (2006) 5225-5246.

[7] C. Textor, et al., *Atmos. Chem. Phys.* 6 (2006) 1777-1813.

# Climate models vs. AERONET Observations



[1] S. China, et al., *Nature Comm.* 4 (2013) 2122-1-7.

[2] C. Chen, et al., *Atmos. Chem. Phys.* 19 (2019) 14585-14606.

[3] M. Sand, et al., *Atmos. Chem. Phys. Diss.* (2021) doi.org/10.5194/acp-2021-51.

[4] D. Kim, et al., *J. Geophys. Res.* 113 (2008) D16309-1-36.

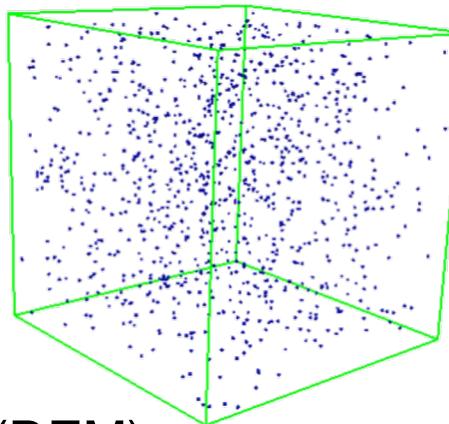
[5] S.H. Chung, J.H. Seinfeld, *J. Geophys. Res.* 107 (2002) 4407-1-33.

[6] M. Schulz, et al., *Atmos. Chem. Phys.* 6 (2006) 5225-5246.

[7] C. Textor, et al., *Atmos. Chem. Phys.* 6 (2006) 1777-1813.

# Discrete Element Modeling (DEM)

i) Initial configuration inception has largely ended.

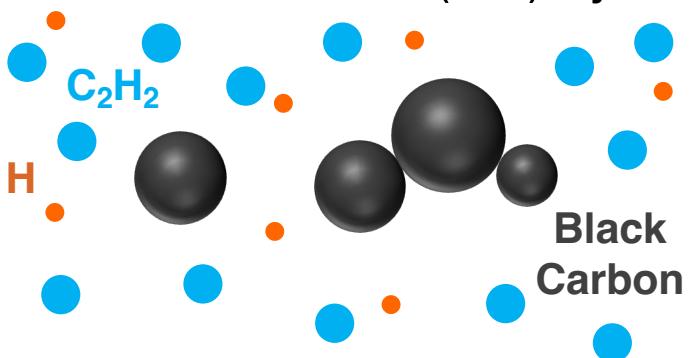


$$T = 1830 \text{ K}$$
$$d_{m,o} = 2 \text{ nm}$$
$$N_{tot,o} = 4.5 \cdot 10^{16} \text{ m}^{-3}$$

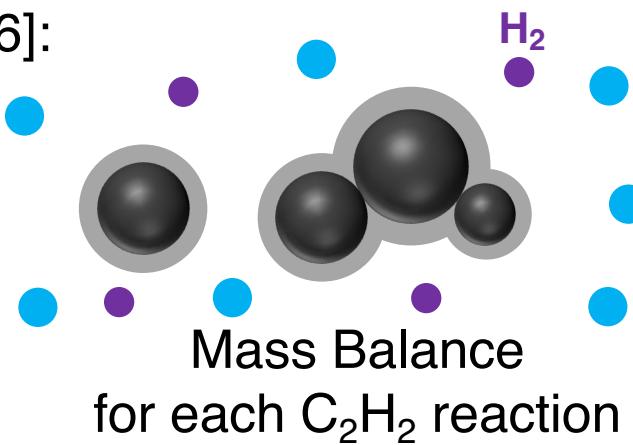
[1,2]

ii) Discrete Element Modeling (DEM) of **Particle Motion** and **Coagulation** during Combustion [3]

iii) **Surface Growth (SG)** by HACA mechanism [4-6]:

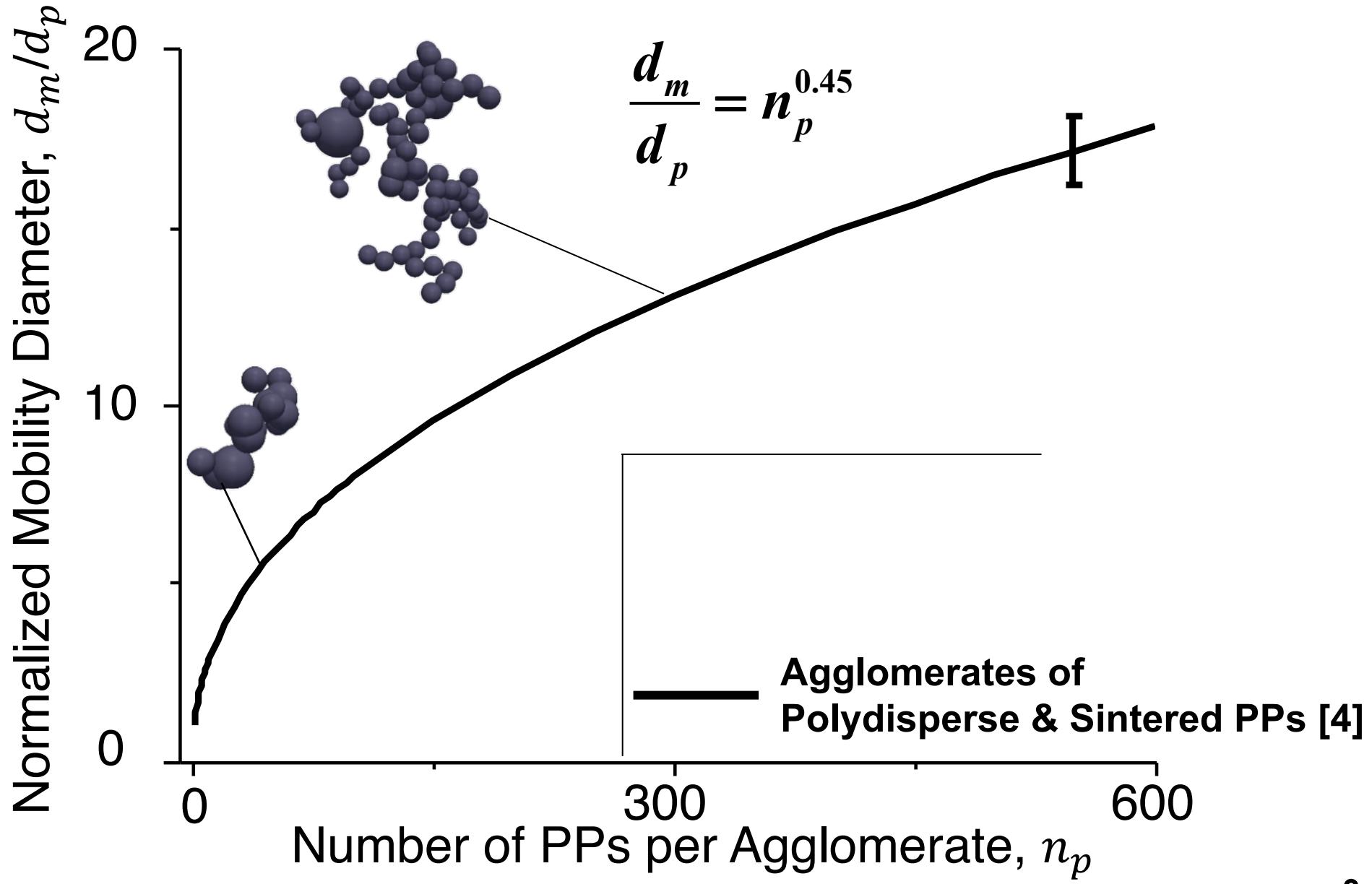


$$r_{HACA} [4]$$
$$\beta_{C_2H_2} [6]$$

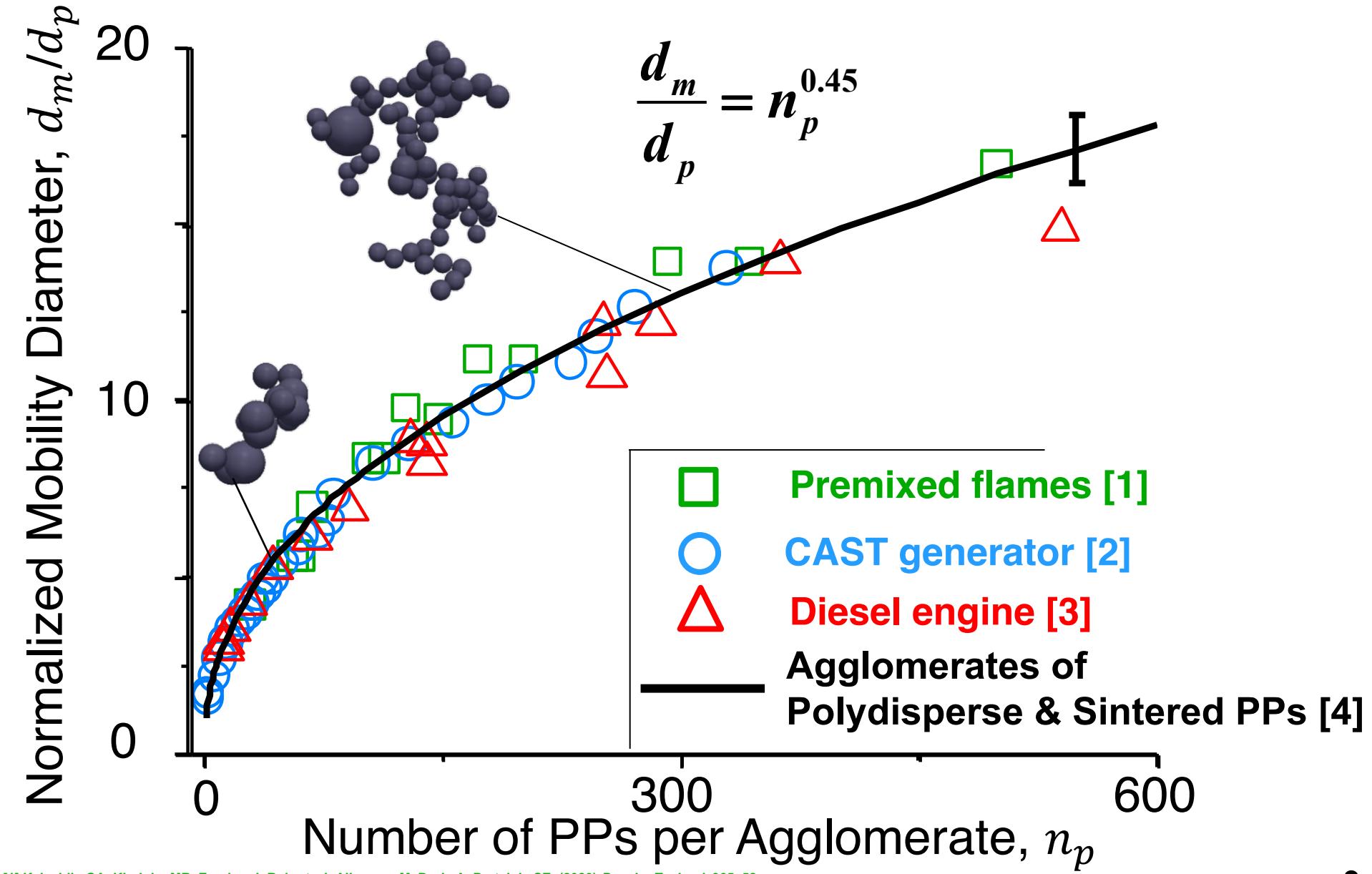


- [1] Abid, A. D.; Heinz, N.; Tolmachoff, E.D.; Phares, D.J.; Campbell, C.S.; Wang, H. *Combust Flame* 2008, 154, 775.
- [2] Camacho ,J.; Liu, C.; Gu, C.; Lin, H.; Huang, Z.; Tang, Q.; You, X.; Saggese, C.; Li, Y.; Jung, H.; Deng, L.; Wlokas, I.; Wang, H. *Combust Flame* 2015, 162, 3810.
- [3] Goudeli, E.; Eggersdorfer, M. L.; Pratsinis, S. E. *Langmuir* 2015, 31,1320.
- [4] Appel, J.; Bockhorn, H.; Frenklach, M. *Combust Flame* 2000, 121, 122.
- [5] Saggese, C.; Ferrario, S.; Camacho, J.; Cuoci, A.; Frassoldati, A.; Ranzi, E.; Wang, H.; Faravelli, T. *Combust Flame* 2015, 162, 3356.
- [6] Kelesidis, G. A.; Goudeli, E.; Pratsinis, S. E. *Proc Combust Inst* 2017 36, 29.

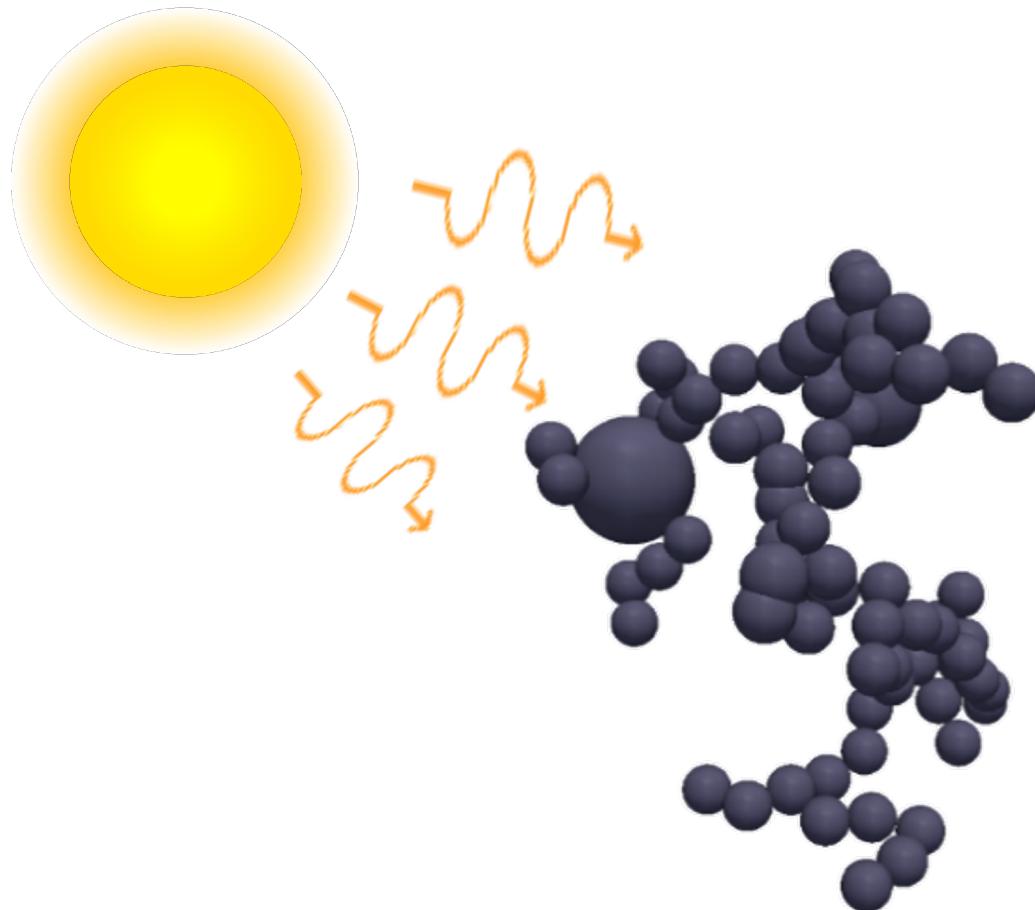
# Black Carbon Morphology



# Black Carbon Morphology



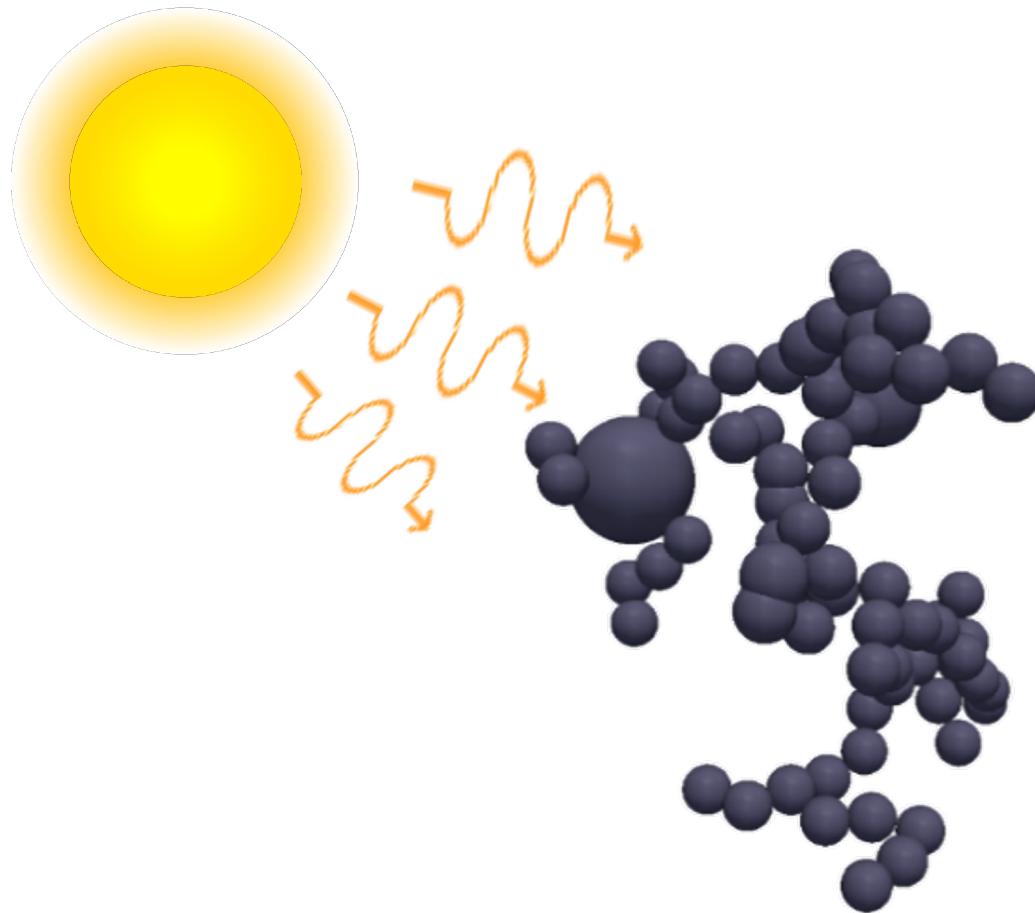
# Rayleigh Debye Gans (RDG) theory



C-rich Black Carbon:  
 $RI = 1.66 - 0.76i$  [1]

[1] Yon, J.; Bescond, A.; Liu, F., J Quant Spectrosc Radiat Transf 2015, 197.

# Rayleigh Debye Gans (RDG) theory



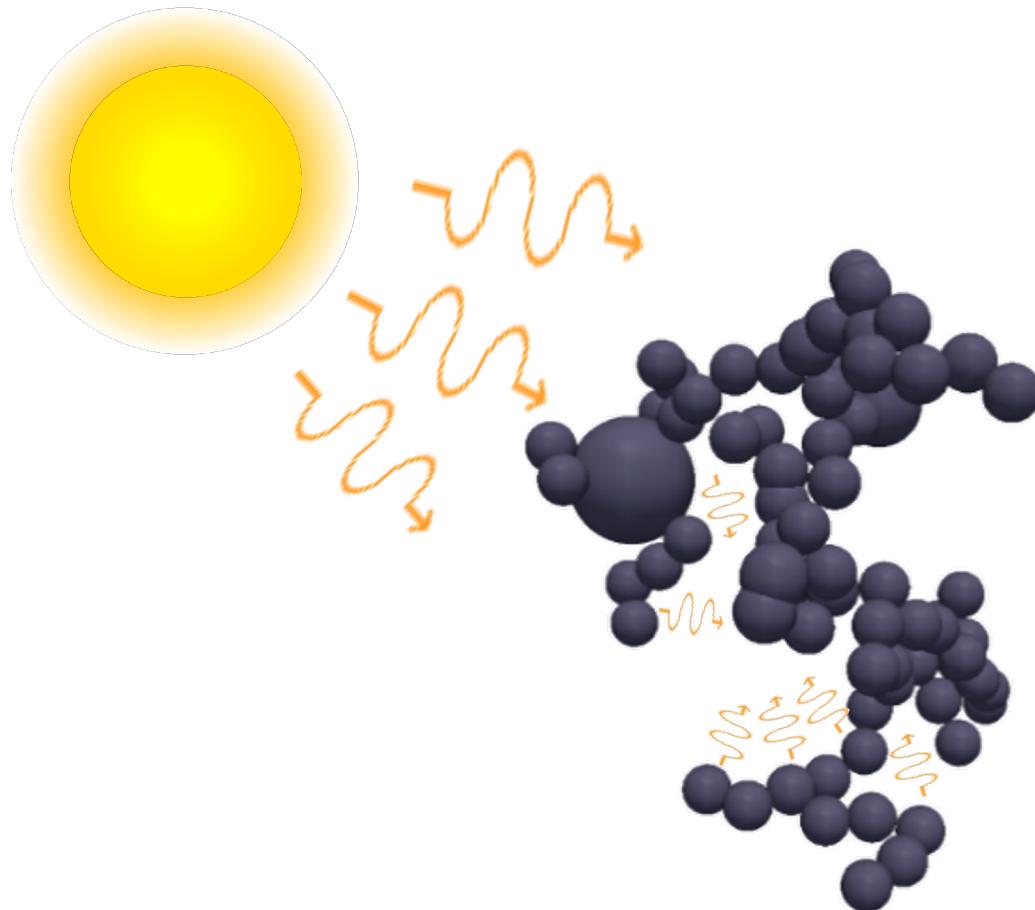
C-rich Black Carbon:  
 $RI = 1.66 - 0.76i$  [1]

H-rich Black Carbon:  
 $RI = 1.75 - 0.45i$  [2]

[1] Yon, J.; Bescond, A.; Liu, F., J Quant Spectrosc Radiat Transf 2015, 197.

[2] Textor, C.; Schulz, S.; Pratsinis, Guibert, S.; Kinne, S.; Balkanski, Y.; Bauer, S., Berntsen, T., et al. Atmos Chem Phys 2006 6, 1777.

# Rayleigh Debye Gans (RDG) theory



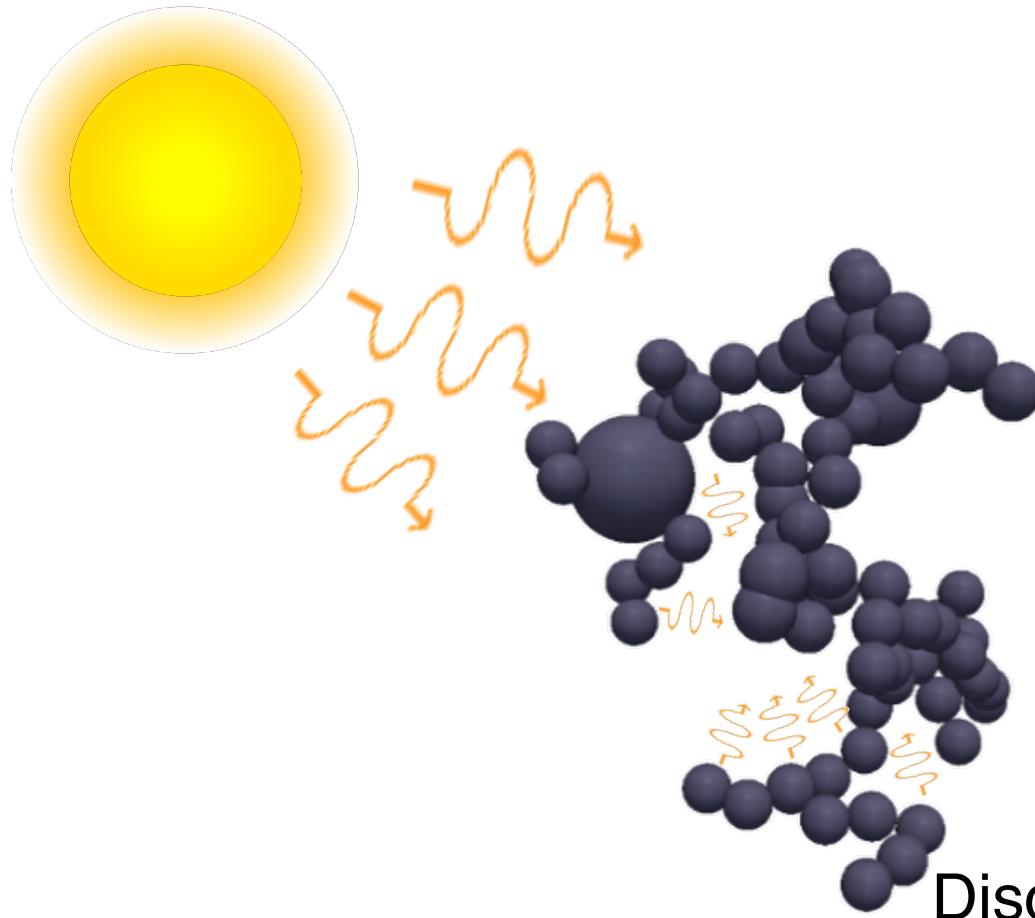
C-rich Black Carbon:  
 $RI = 1.66 - 0.76i$  [1]

H-rich Black Carbon:  
 $RI = 1.75 - 0.45i$  [2]

[1] Yon, J.; Bescond, A.; Liu, F., J Quant Spectrosc Radiat Transf 2015, 197.

[2] Textor, C.; Schulz, S.; Pratsinis, Guibert, S.; Kinne, S.; Balkanski, Y.; Bauer, S., Berntsen, T., et al. Atmos Chem Phys 2006 6, 1777.

# Rayleigh Debye Gans (RDG) theory



C-rich Black Carbon:  
 $RI = 1.66 - 0.76i$  [1]

H-rich Black Carbon:  
 $RI = 1.75 - 0.45i$  [2]

Discrete Element Modeling [3] &  
Discrete Dipole Approximation [4]  
 $d_m = 50 - 250 \text{ nm}; d_p = 7.5 - 40 \text{ nm}$

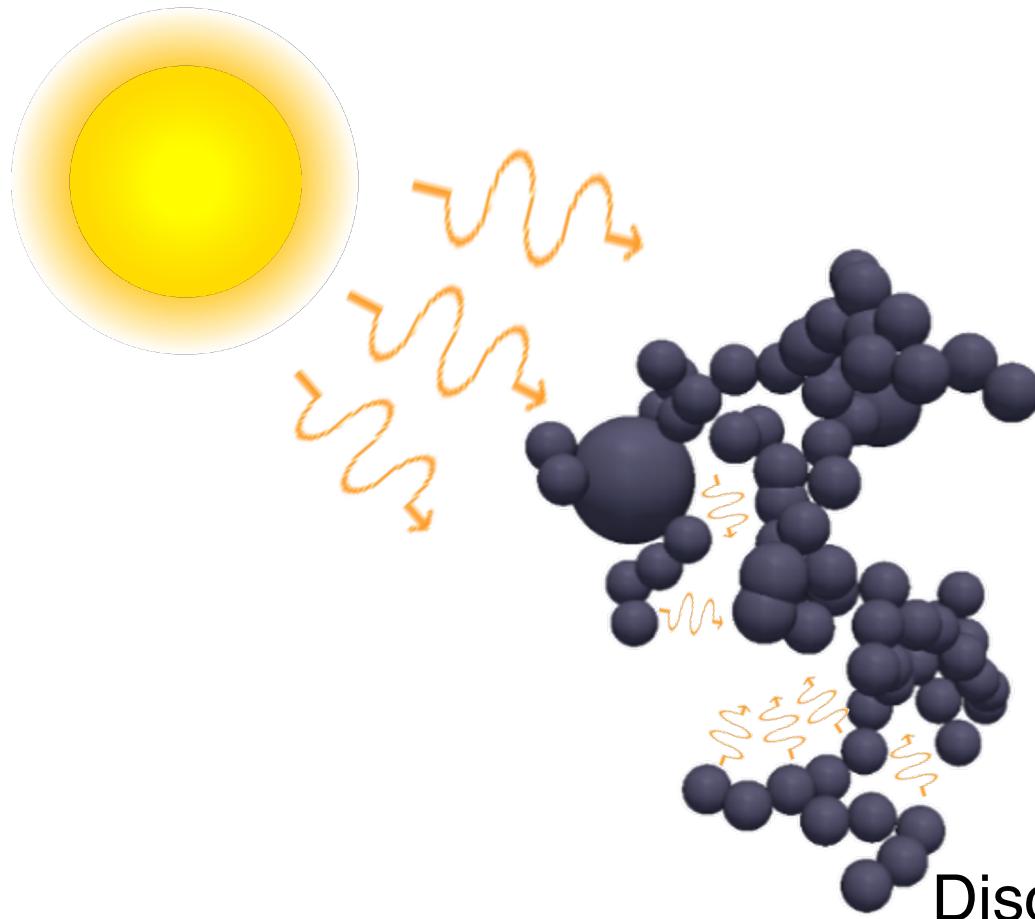
[1] Yon, J.; Bescond, A.; Liu, F., J Quant Spectrosc Radiat Transf 2015, 197.

[2] Textor, C.; Schulz, S.; Pratsinis, Guibert, S.; Kinne, S.; Balkanski, Y.; Bauer, S., Berntsen, T., et al. Atmos Chem Phys 2006 6, 1777.

[3] Kelesidis, G. A.; Goudeli, E.; Pratsinis, S. E. Proc Combust Inst 2017 36, 29.

[4] Kelesidis, G.A.; Kholghy, M.R.; Zurcher, J.; Robertz, J.; Allemann, M.; Duric, A.; Pratsinis, S.E. Powder Technol 2020, 365, 52.

# Rayleigh Debye Gans (RDG) theory



C-rich Black Carbon:

$$RI = 1.66 - 0.76i \quad [1]$$

H-rich Black Carbon:

$$RI = 1.75 - 0.45i \quad [2]$$

Light absorption by  
intra-particle scattering:

$$h = 1.18 \pm 0.03$$

Discrete Element Modeling [3] &  
Discrete Dipole Approximation [4]  
 $d_m = 50 - 250 \text{ nm}; d_p = 7.5 - 40 \text{ nm}$

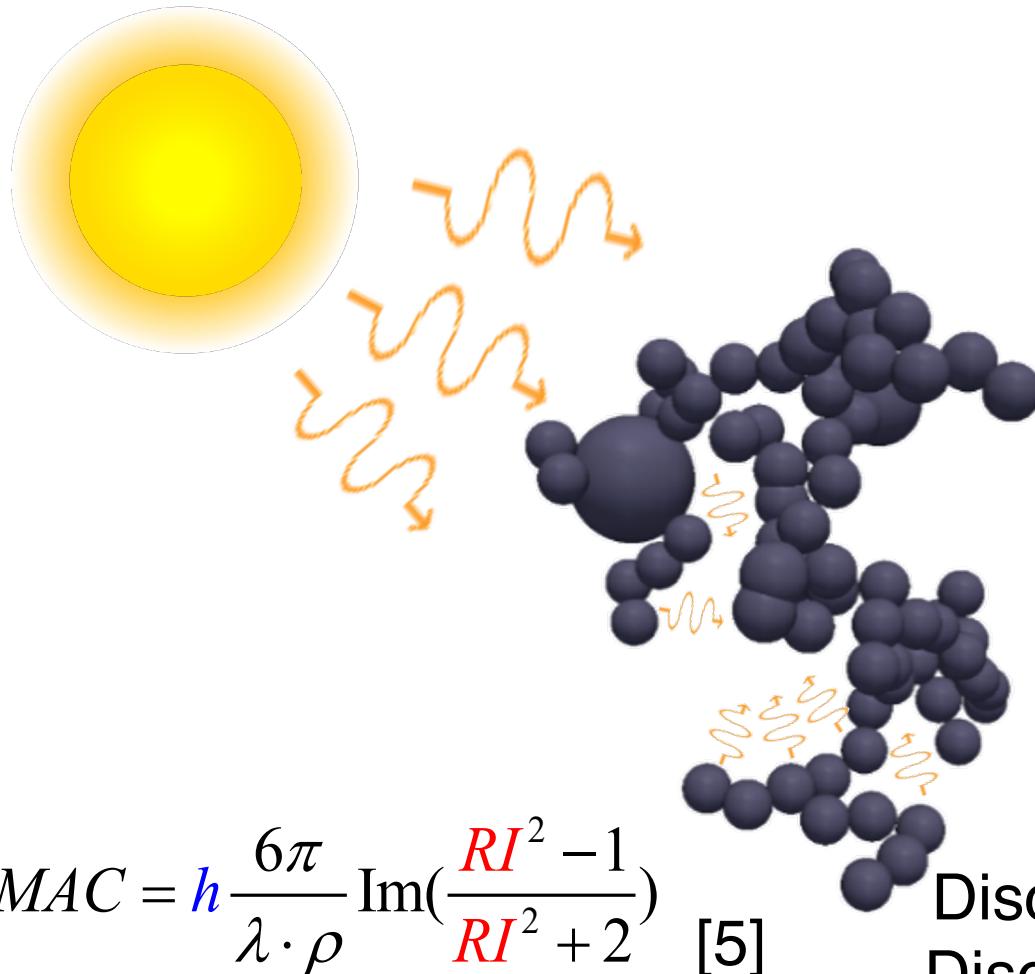
[1] Yon, J.; Bescond, A.; Liu, F., J Quant Spectrosc Radiat Transf 2015, 197.

[2] Textor, C.; Schulz, S.; Pratsinis, Guibert, S.; Kinne, S.; Balkanski, Y.; Bauer, S., Berntsen, T., et al. Atmos Chem Phys 2006 6, 1777.

[3] Kelesidis, G. A.; Goudeli, E.; Pratsinis, S. E. Proc Combust Inst 2017 36, 29.

[4] Kelesidis, G.A.; Kholghy, M.R.; Zurcher, J.; Robertz, J.; Allemann, M.; Duric, A.; Pratsinis, S.E. Powder Technol 2020, 365, 52.

# Rayleigh Debye Gans (RDG) theory



C-rich Black Carbon:  
 $RI = 1.66 - 0.76i$  [1]

H-rich Black Carbon:  
 $RI = 1.75 - 0.45i$  [2]

Light absorption by  
intra-particle scattering:

$$h = 1.18 \pm 0.03$$

Discrete Element Modeling [3] &  
Discrete Dipole Approximation [4]

$$d_m = 50 - 250 \text{ nm}; d_p = 7.5 - 40 \text{ nm}$$

[1] Yon, J.; Bescond, A.; Liu, F., J Quant Spectrosc Radiat Transf 2015, 197.

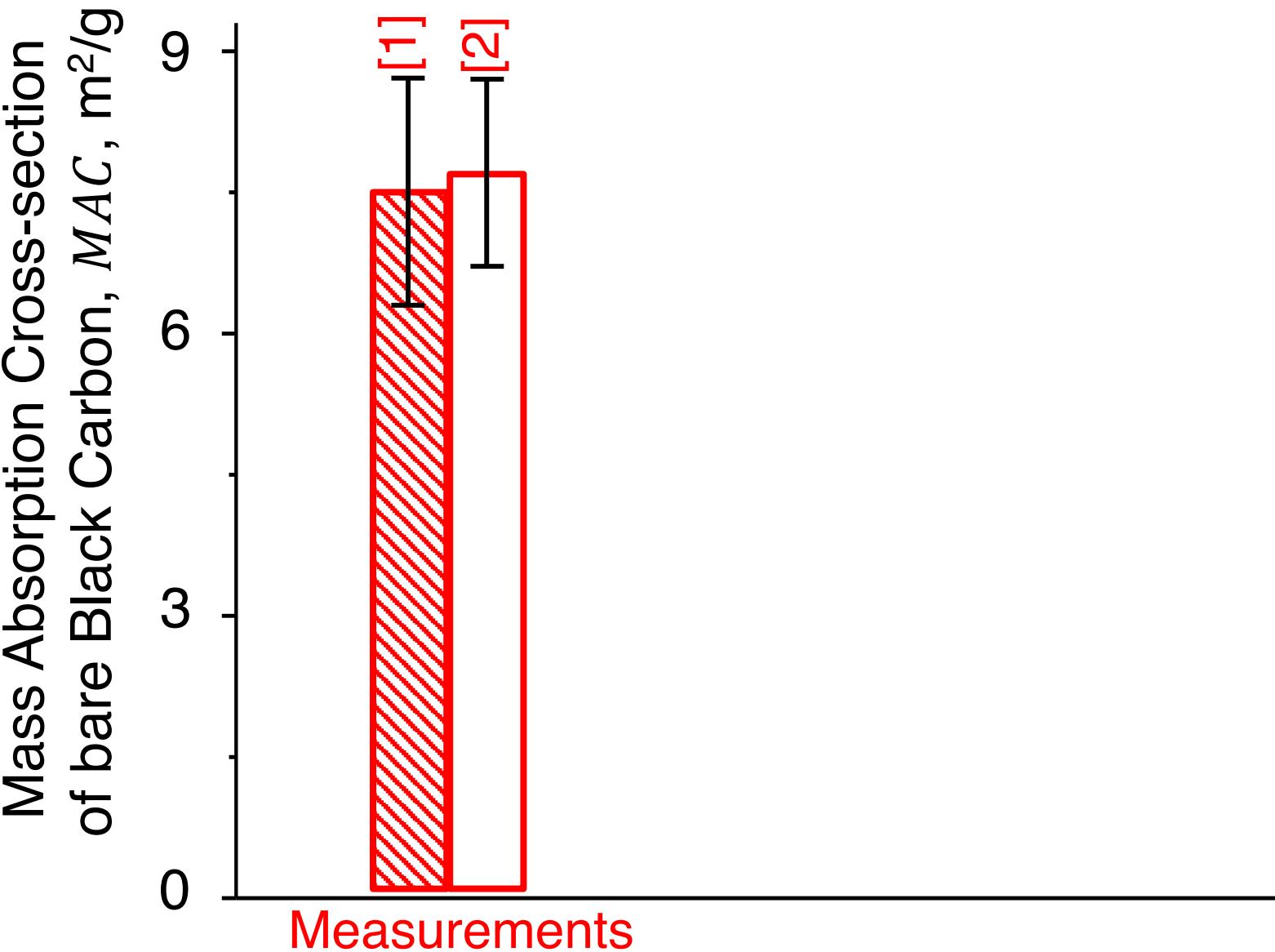
[2] Textor, C.; Schulz, S.; Pratsinis, Guibert, S.; Kinne, S.; Balkanski, Y.; Bauer, S., Berntsen, T., et al. Atmos Chem Phys 2006 6, 1777.

[3] Kelesidis, G. A.; Goudeli, E.; Pratsinis, S. E. Proc Combust Inst 2017 36, 29.

[4] Kelesidis, G.A.; Kholghy, M.R.; Zurcher, J.; Robertz, J.; Allemann, M.; Duric, A.; Pratsinis, S.E. Powder Technol 2020, 365, 52.

[5] Dobbins, R.A.; Megaridis, C.M., Appl Optics 1991, 30, 4747.

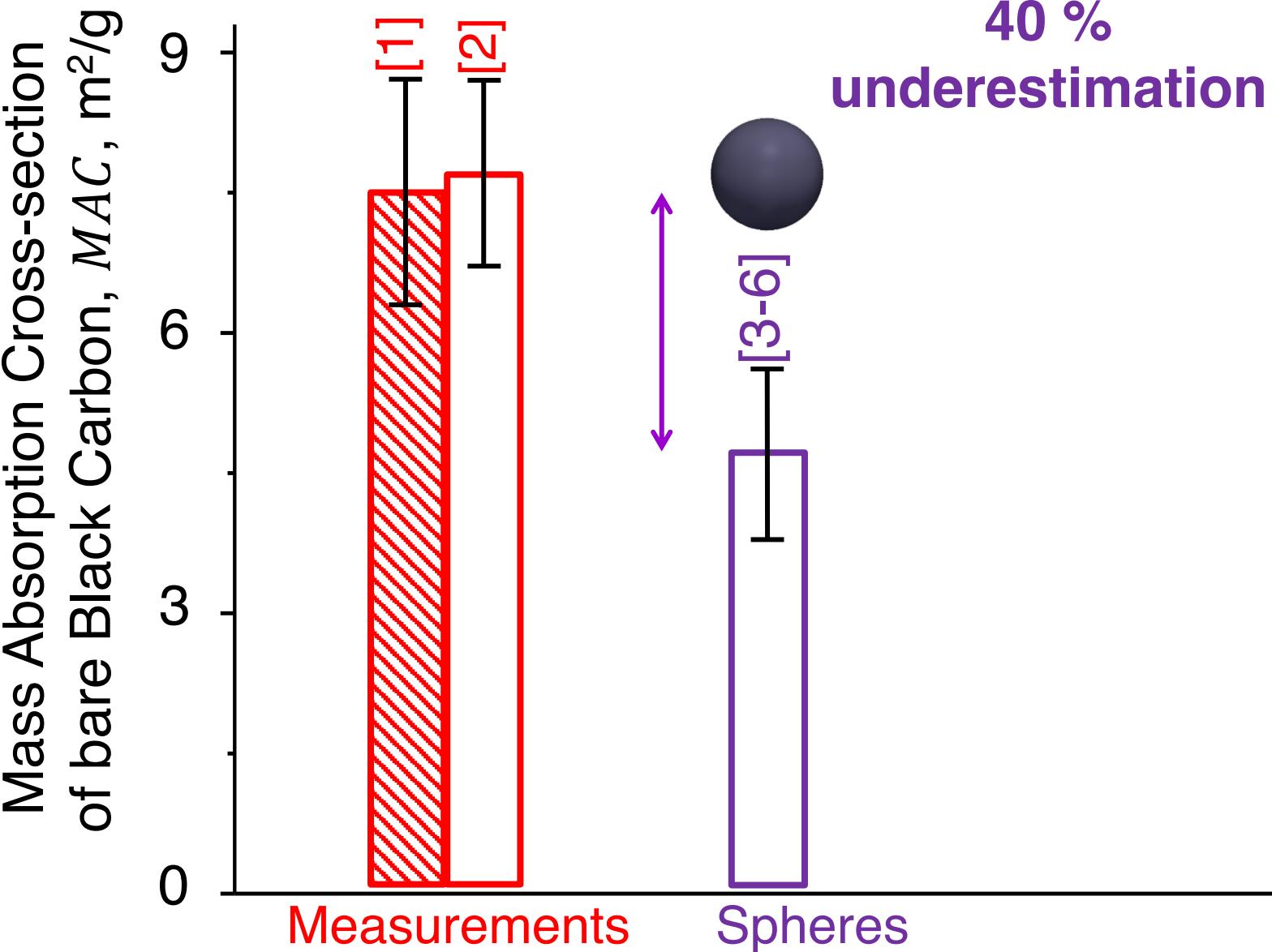
# Light absorption by bare Black Carbon



[1] T.C. Bond, R. W. Bergstrom, *Aerosol Sci. Technol.* 40 (2006) 27-67.

[2] F. Liu, et al., *Aerosol Sci. Technol.* 54 (2019) 33-51.

# Light absorption by bare Black Carbon



[1] T.C. Bond, R. W. Bergstrom, *Aerosol Sci. Technol.* 40 (2006) 27-67.

[2] F. Liu, et al., *Aerosol Sci. Technol.* 54 (2019) 33-51.

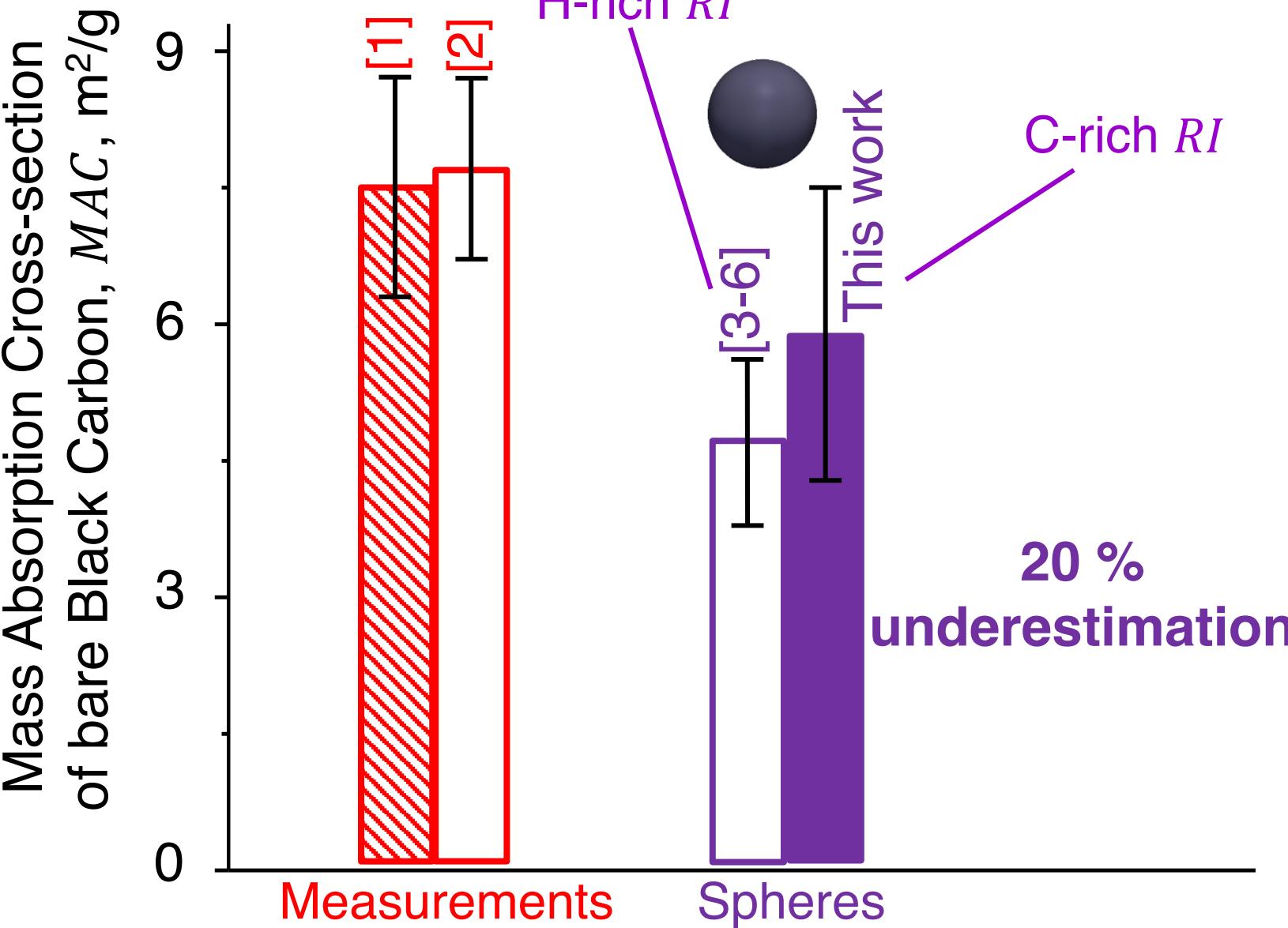
[3] D. Kim, et al., *J. Geophys. Res.* 113 (2008) D16309-1-36.

[4] S.H. Chung, J.H. Seinfeld, *J. Geophys. Res.* 107 (2002) 4407-1-33.

[5] M. Schulz, et al., *Atmos. Chem. Phys.* 6 (2006) 5225-5246.

[6] C. Textor, et al., *Atmos. Chem. Phys.* 6 (2006) 1777-1813.

# Light absorption by bare Black Carbon



[1] T.C. Bond, R. W. Bergstrom, *Aerosol Sci. Technol.* 40 (2006) 27-67.

[2] F. Liu, et al., *Aerosol Sci. Technol.* 54 (2019) 33-51.

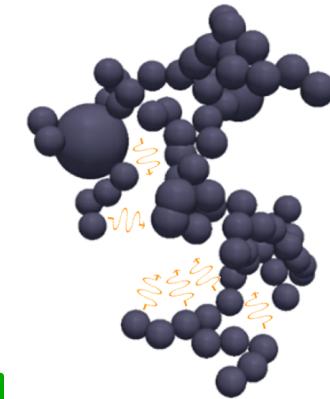
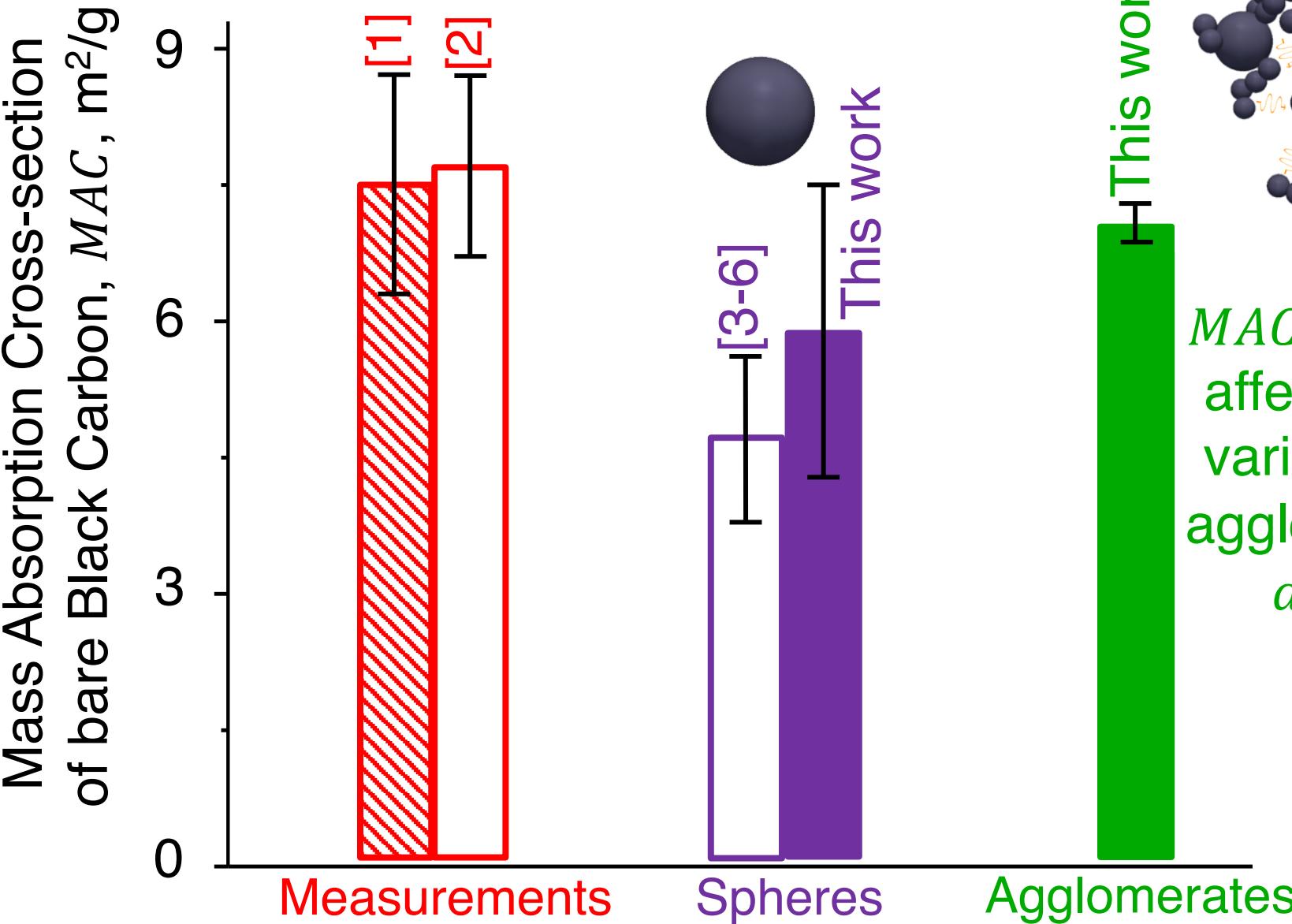
[3] D. Kim, et al., *J. Geophys. Res.* 113 (2008) D16309-1-36.

[4] S.H. Chung, J.H. Seinfeld, *J. Geophys. Res.* 107 (2002) 4407-1-33.

[5] M. Schulz, et al., *Atmos. Chem. Phys.* 6 (2006) 5225-5246.

[6] C. Textor, et al., *Atmos. Chem. Phys.* 6 (2006) 1777-1813.

# Light absorption by bare Black Carbon



MAC is NOT  
affected by  
variation of  
agglomerate  
 $d_m, d_p$

[1] T.C. Bond, R. W. Bergstrom, *Aerosol Sci. Technol.* 40 (2006) 27-67.

[2] F. Liu, et al., *Aerosol Sci. Technol.* 54 (2019) 33-51.

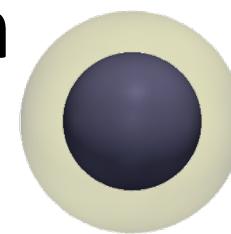
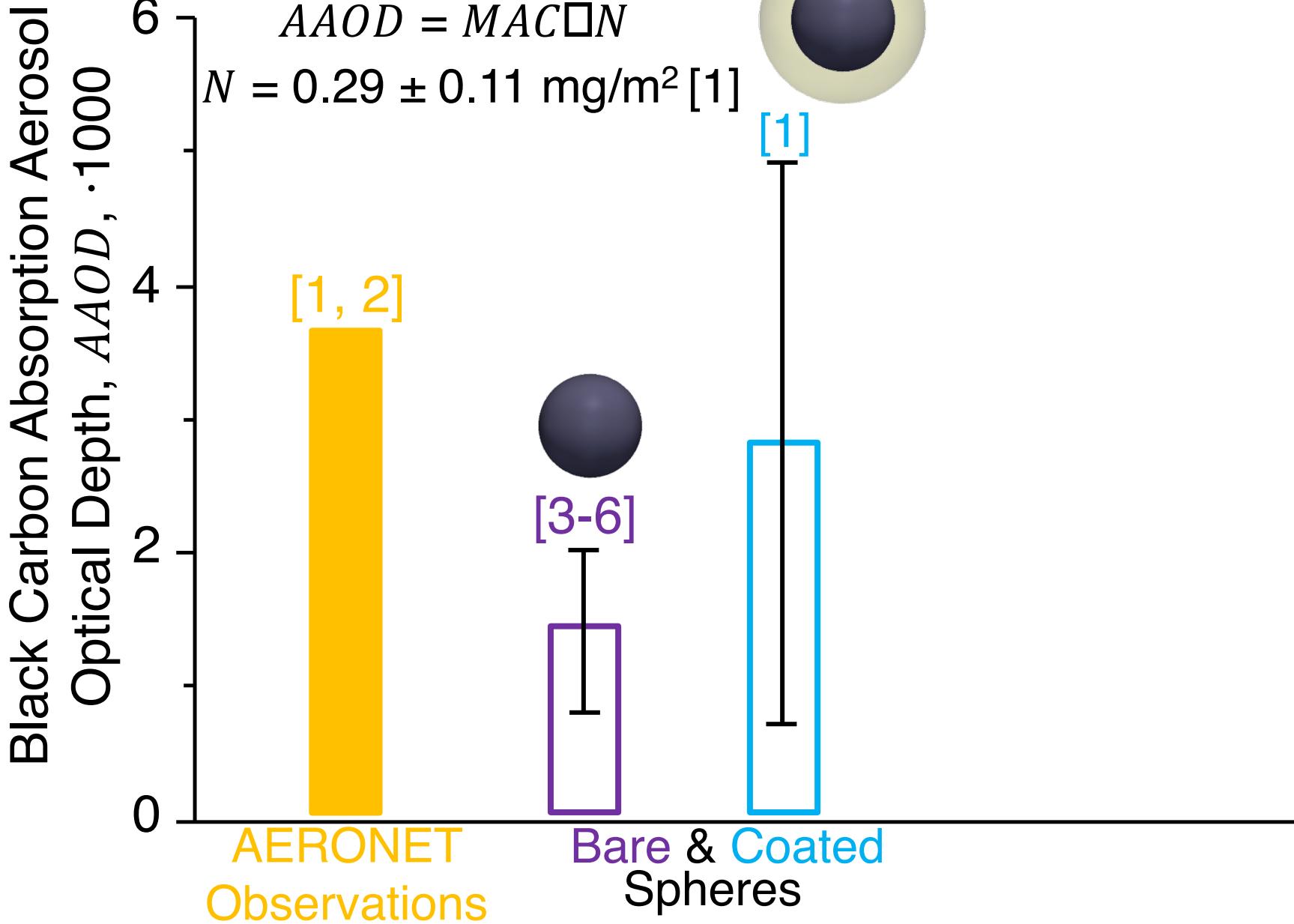
[3] D. Kim, et al., *J. Geophys. Res.* 113 (2008) D16309-1-36.

[4] S.H. Chung, J.H. Seinfeld, *J. Geophys. Res.* 107 (2002) 4407-1-33.

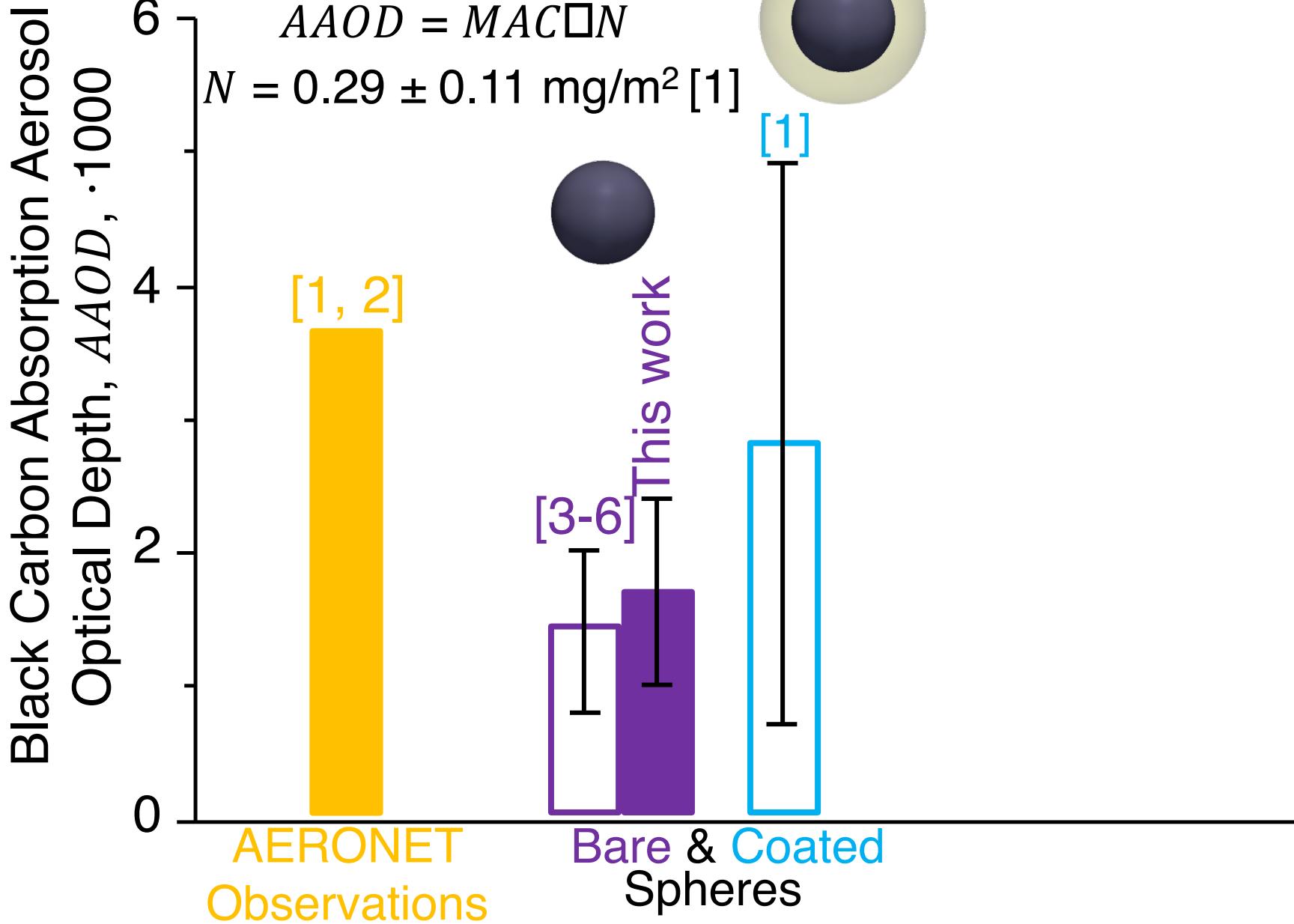
[5] M. Schulz, et al., *Atmos. Chem. Phys.* 6 (2006) 5225-5246.

[6] C. Textor, et al., *Atmos. Chem. Phys.* 6 (2006) 1777-1813.

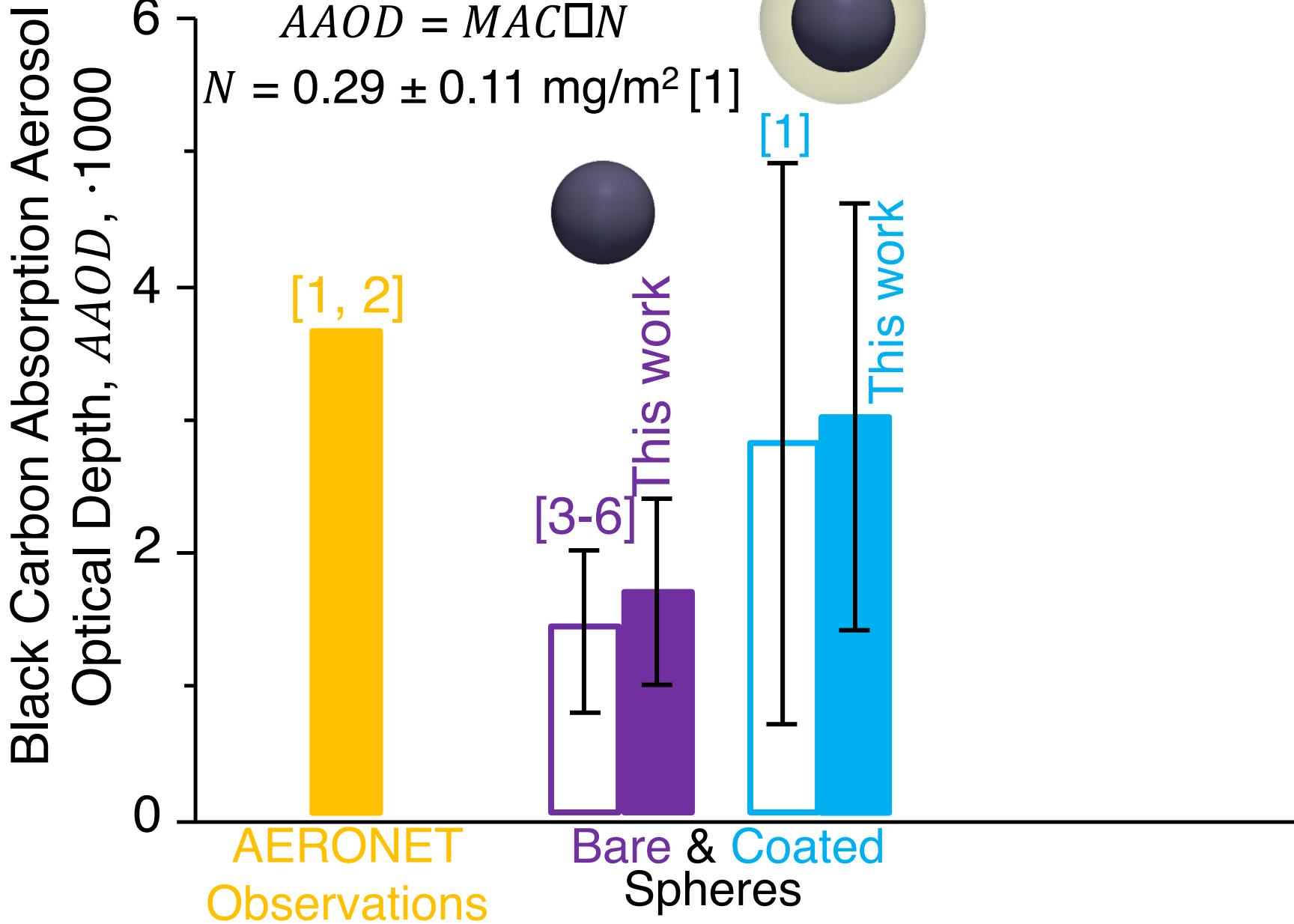
# AAOD by Black Carbon



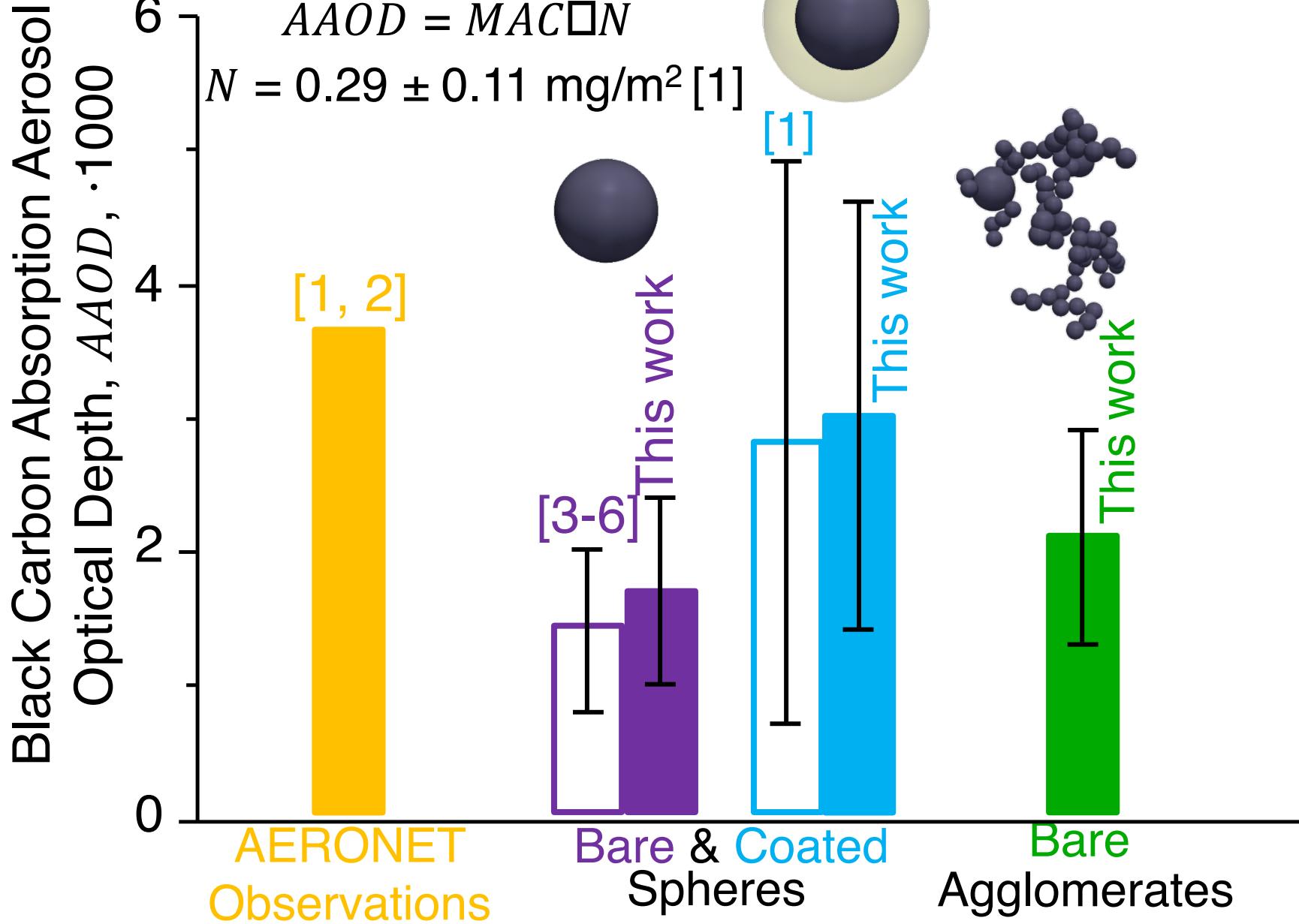
# AAOD by Black Carbon



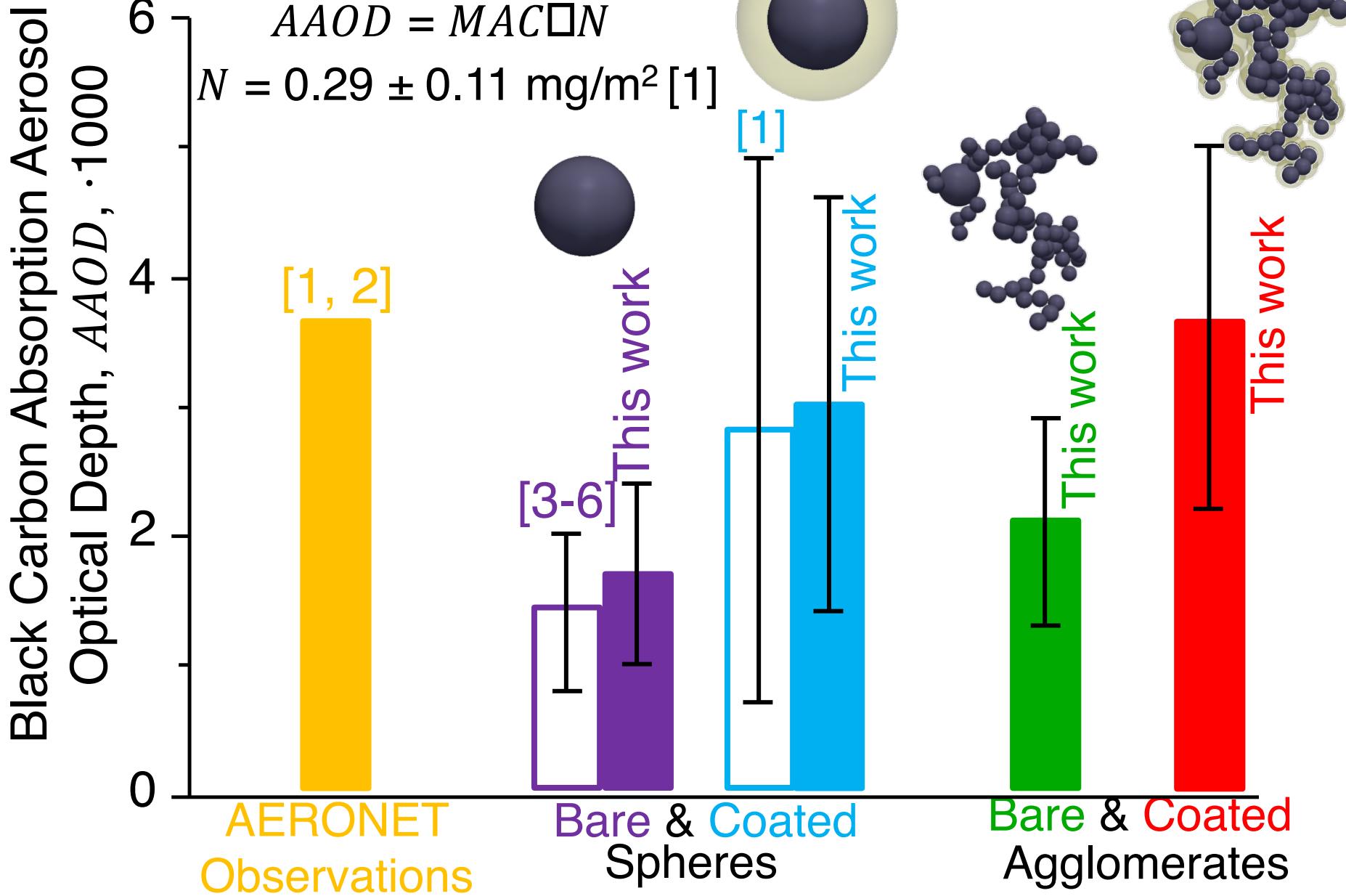
# AAOD by Black Carbon



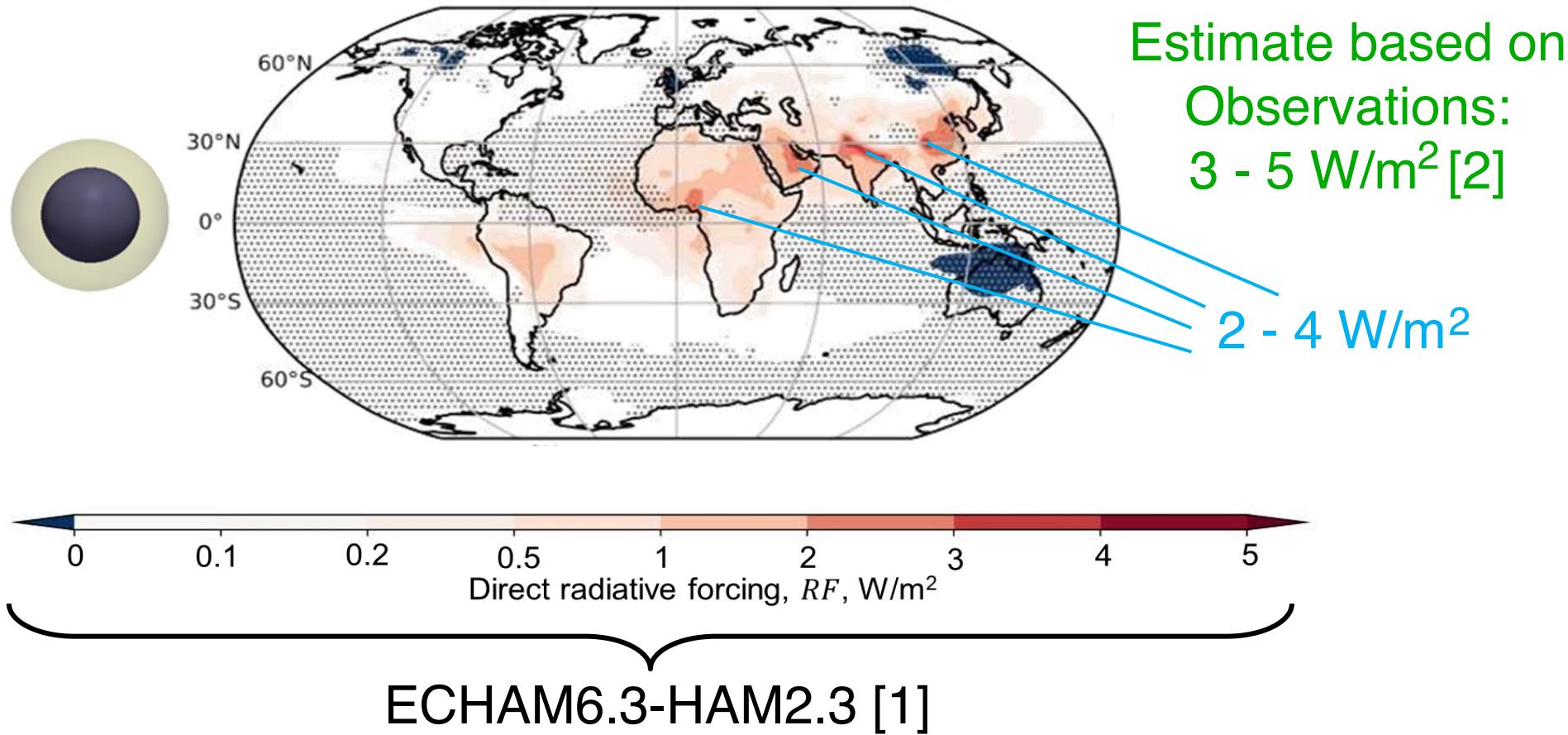
# AAOD by Black Carbon



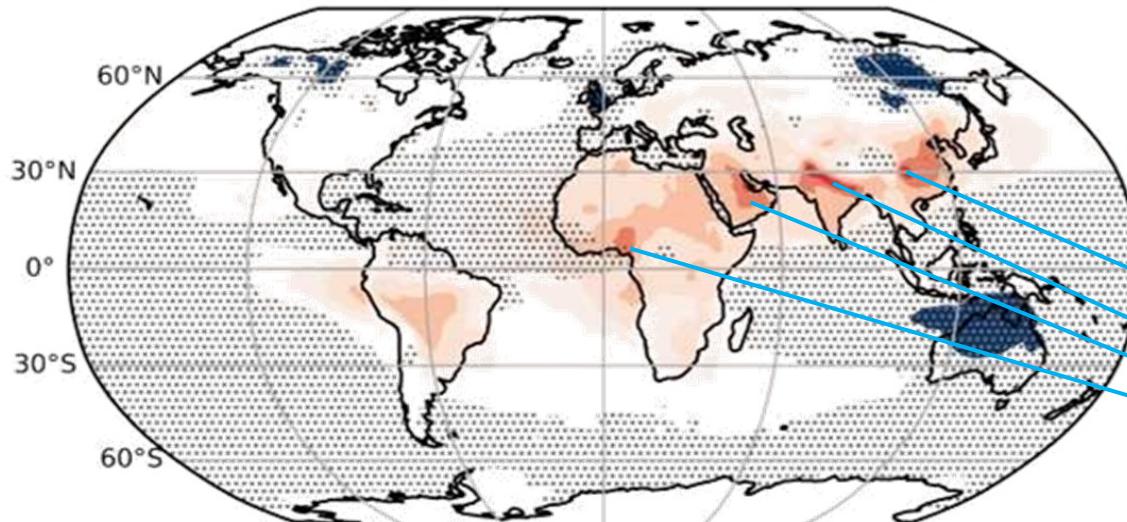
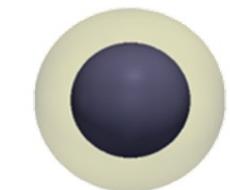
# AAOD by Black Carbon



# Climate impact of Black Carbon

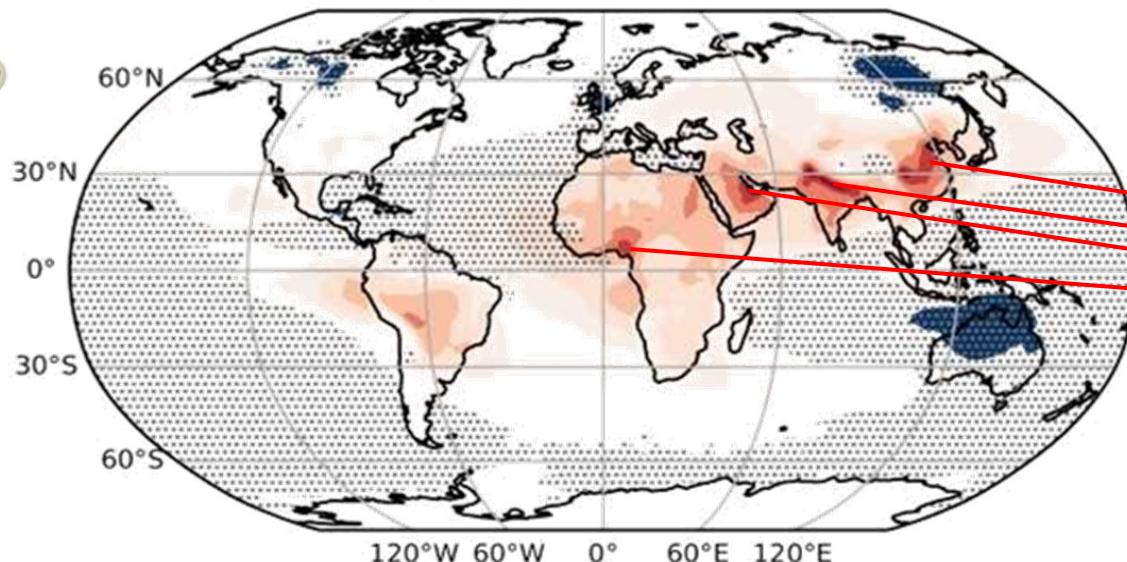


# Climate impact of Black Carbon



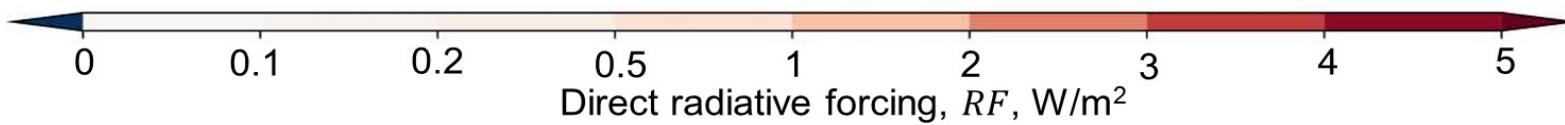
Estimate based on  
Observations:  
3 - 5 W/m<sup>2</sup> [2]

2 - 4 W/m<sup>2</sup>

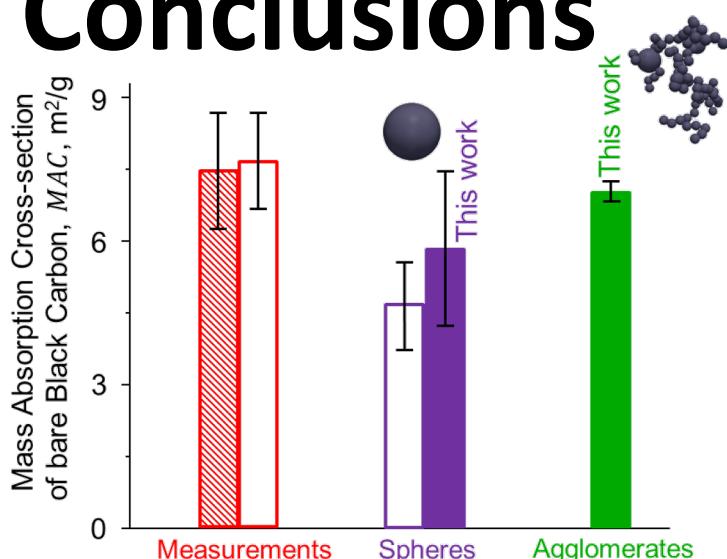


3 - 5 W/m<sup>2</sup>

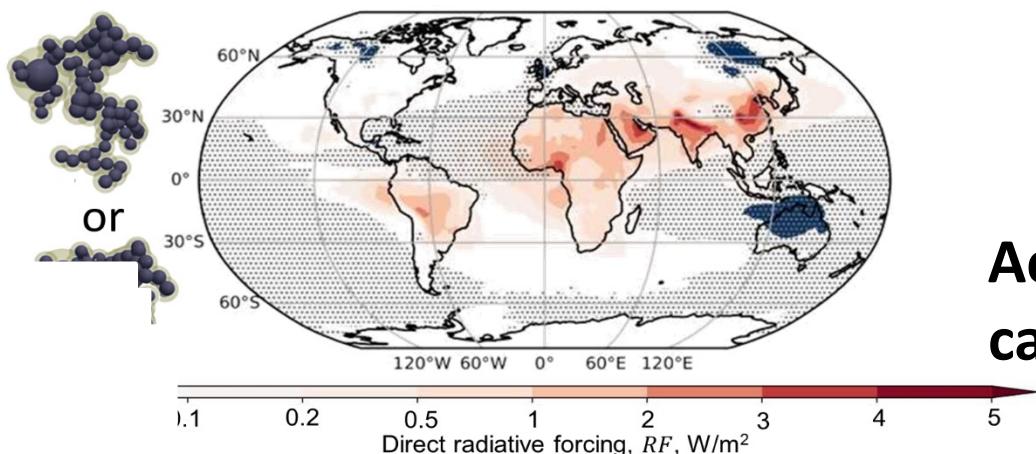
0.75 - 1.25 °C  
Increase!



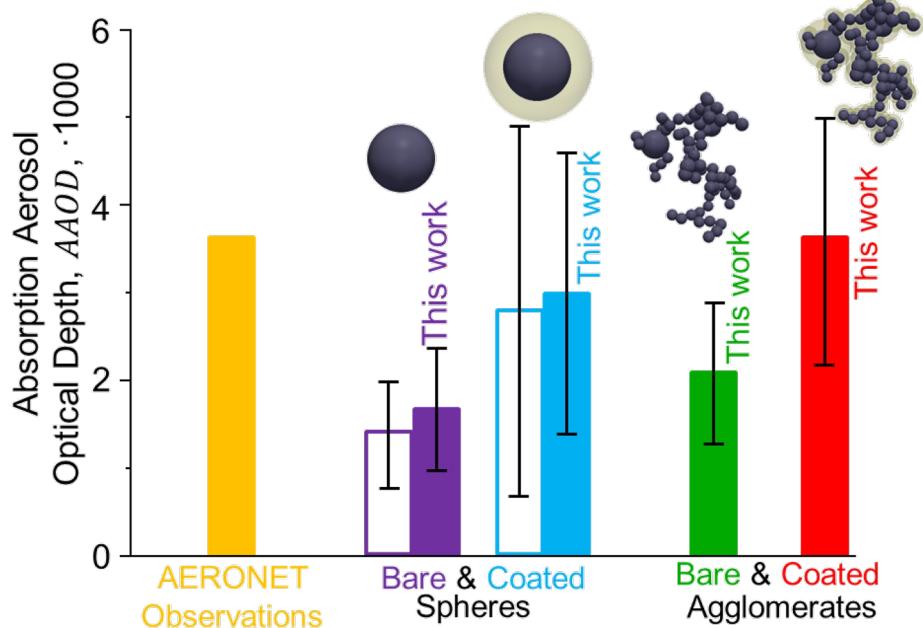
# Conclusions



Black Carbon agglomerate  $AAOD$   
in excellent agreement with  
AERONET observations.



Light absorption of Black Carbon is  
enhanced by PP light scattering  
due to the agglomerate structure  
and NOT  $d_m$ ,  $d_p$ !



Accounting for the realistic black  
carbon morphology enhances its  
direct  $RF$  by 25-50 %!

**Thank you for listening**