Investigating the effects of atmospheric aging on the direct radiative properties and climate impacts of black- and brown- carbon aerosol

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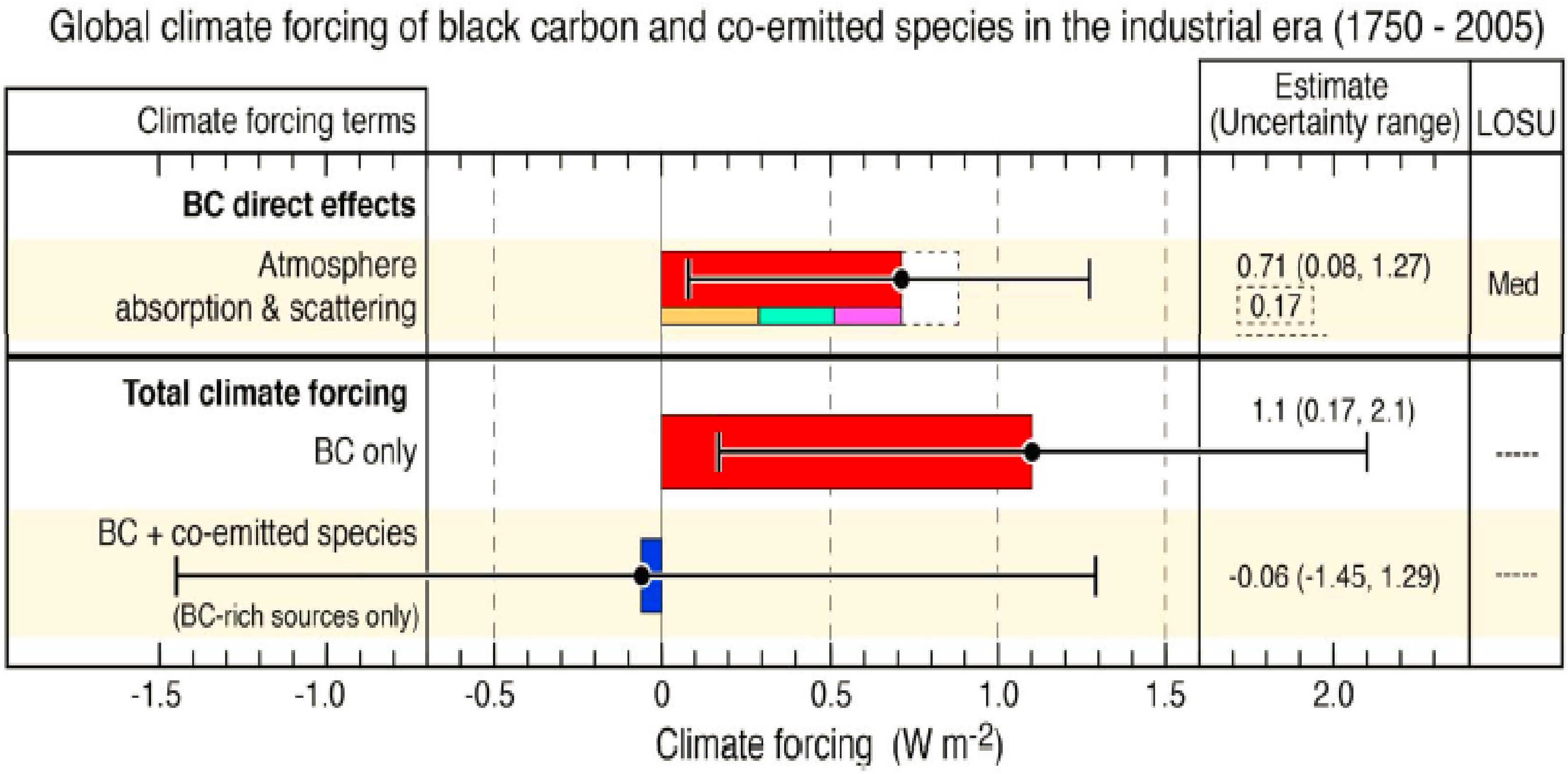
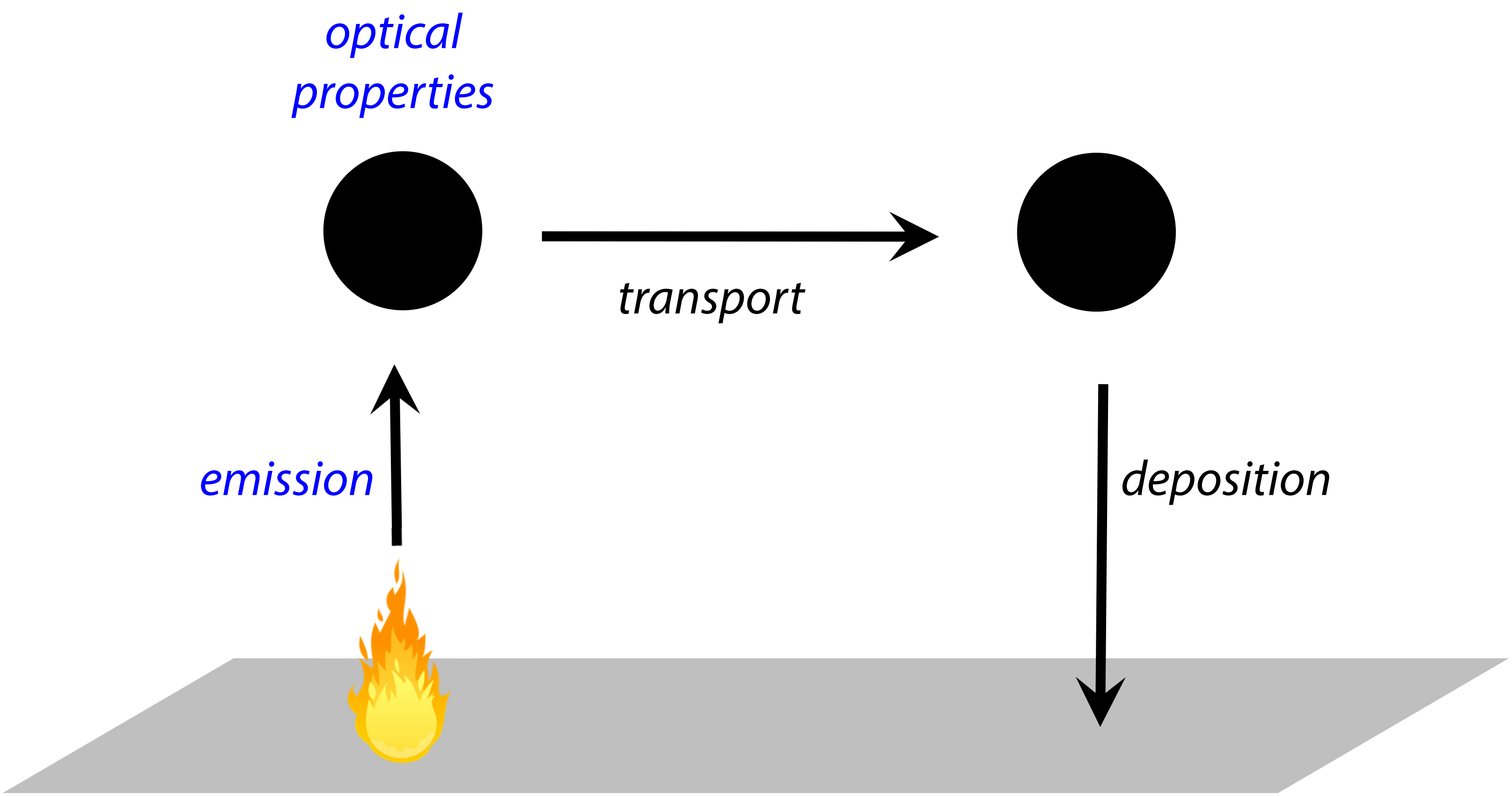
BC climate forcing: Large, complex, uncertain



[Bond et al., 2013]

Simple lifecycle of atmospheric BC



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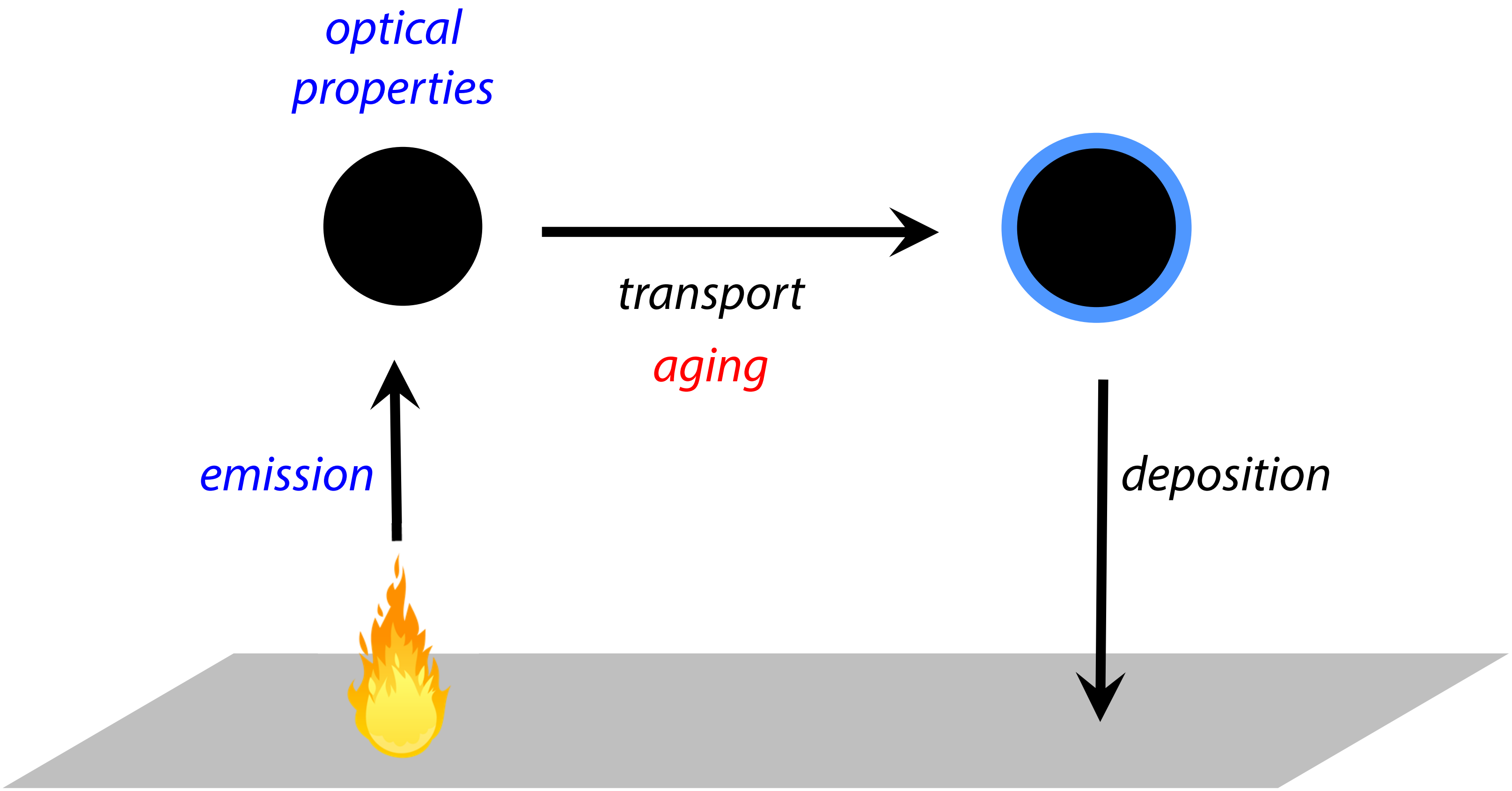
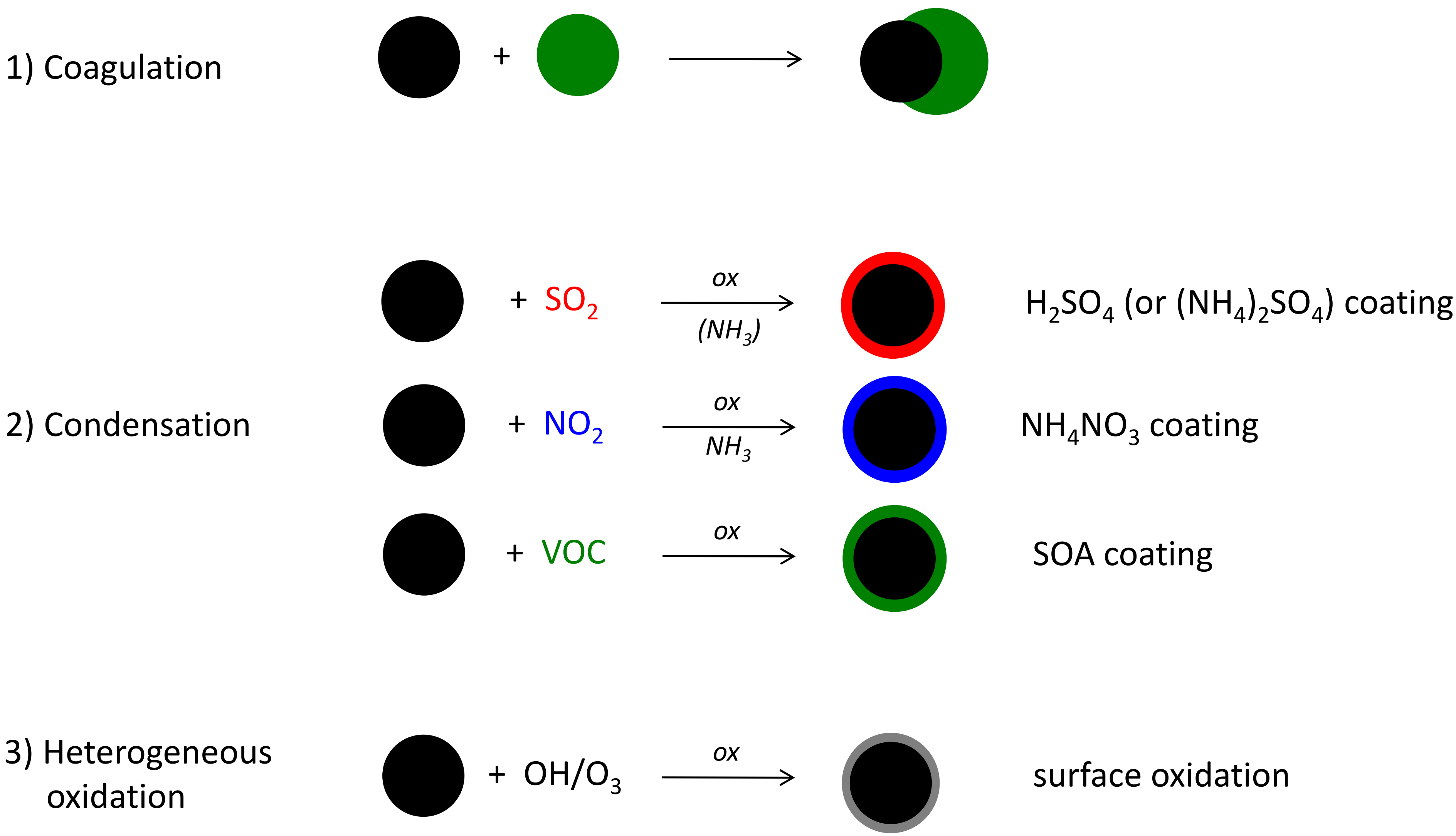


This project

Black/brown carbon aerosol is chemically dynamic, subject to atmospheric aging reactions; these can lead to dramatic changes in physicochemical properties, and therefore climate forcing effects.

A complete understanding of this aging, and representation of this aging within models, is necessary for the accurate simulation of global direct radiative forcing.

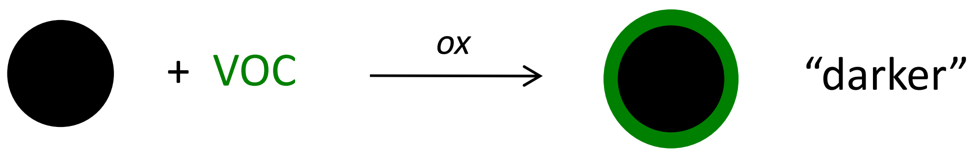
Key BC aging reactions



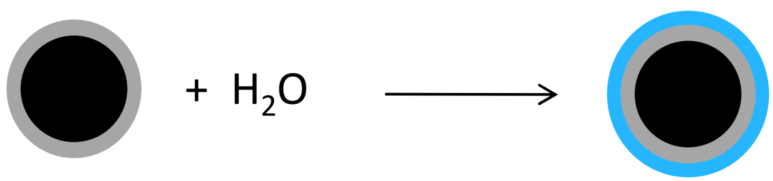
Effects of aging on BC properties

1) Enhancement of light absorption by coatings (“lensing effect”)

[*e.g., Schnaitner 2005, Bond et al. 2006, Schwarz et al. 2008, Lack et al. 2009, Cappa et al. 2012*]



2) Increased water-uptake ability by coated or oxidized BC

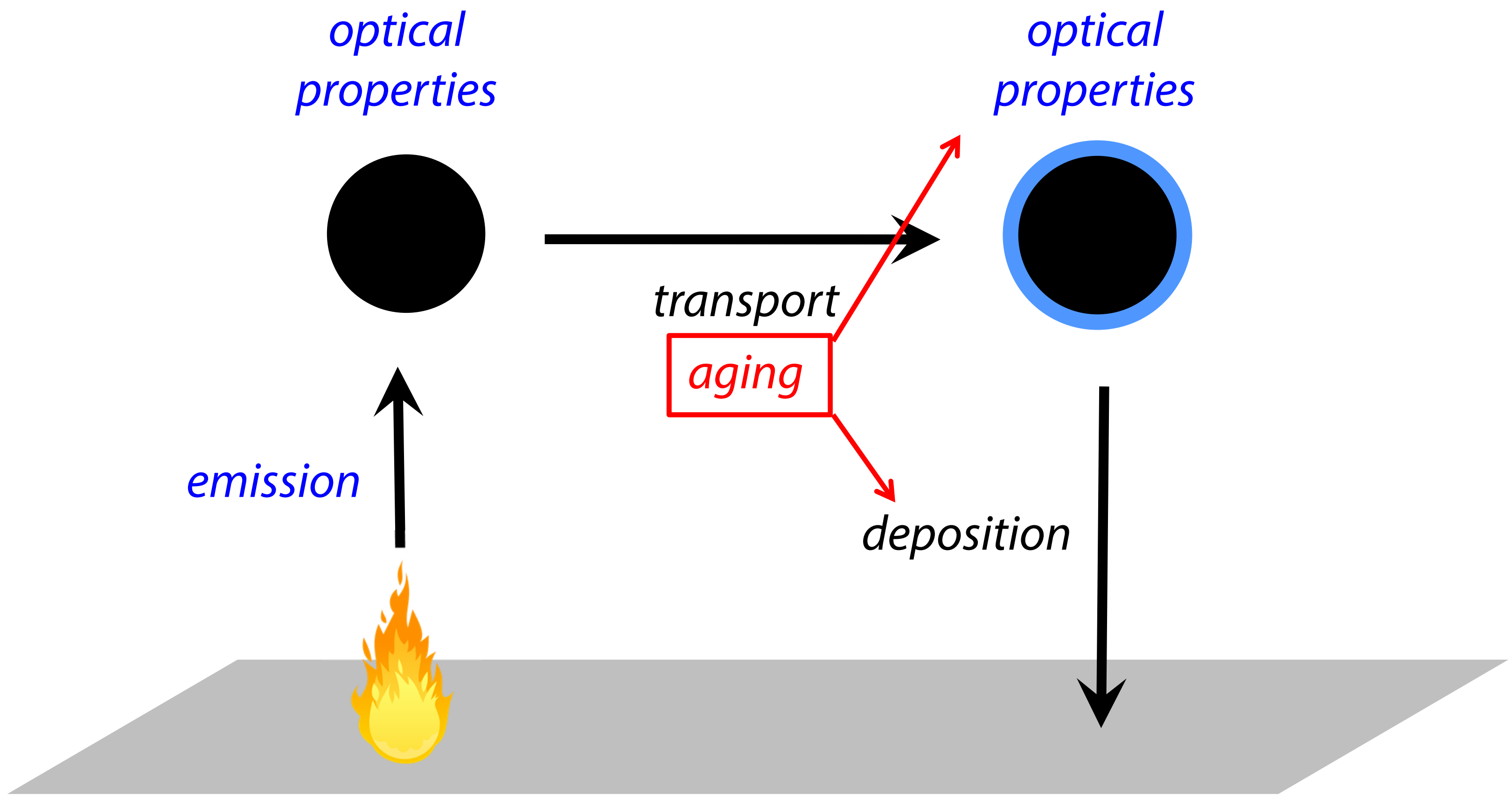


Higher hygroscopicity can lead to …

- more efficient light scattering (due to larger particles from water uptake)
- shorter atmospheric lifetimes due to increased wet deposition

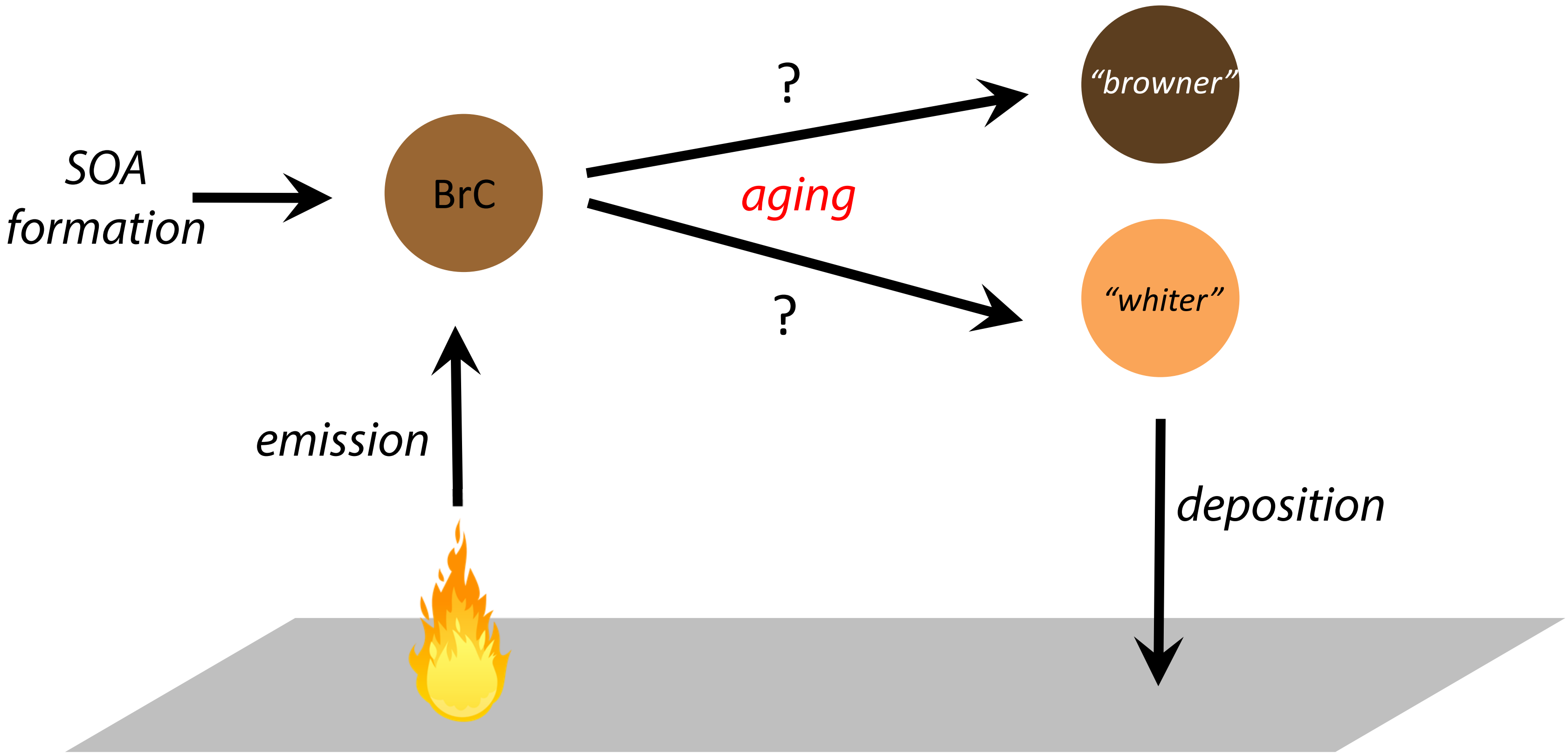
(- more facile activation to form cloud droplets)

Simple lifecycle of atmospheric BC



Simple lifecycle of atmospheric BrC

"Brown carbon": OC that absorbs in the UV/visible (distinct molecules)



This project

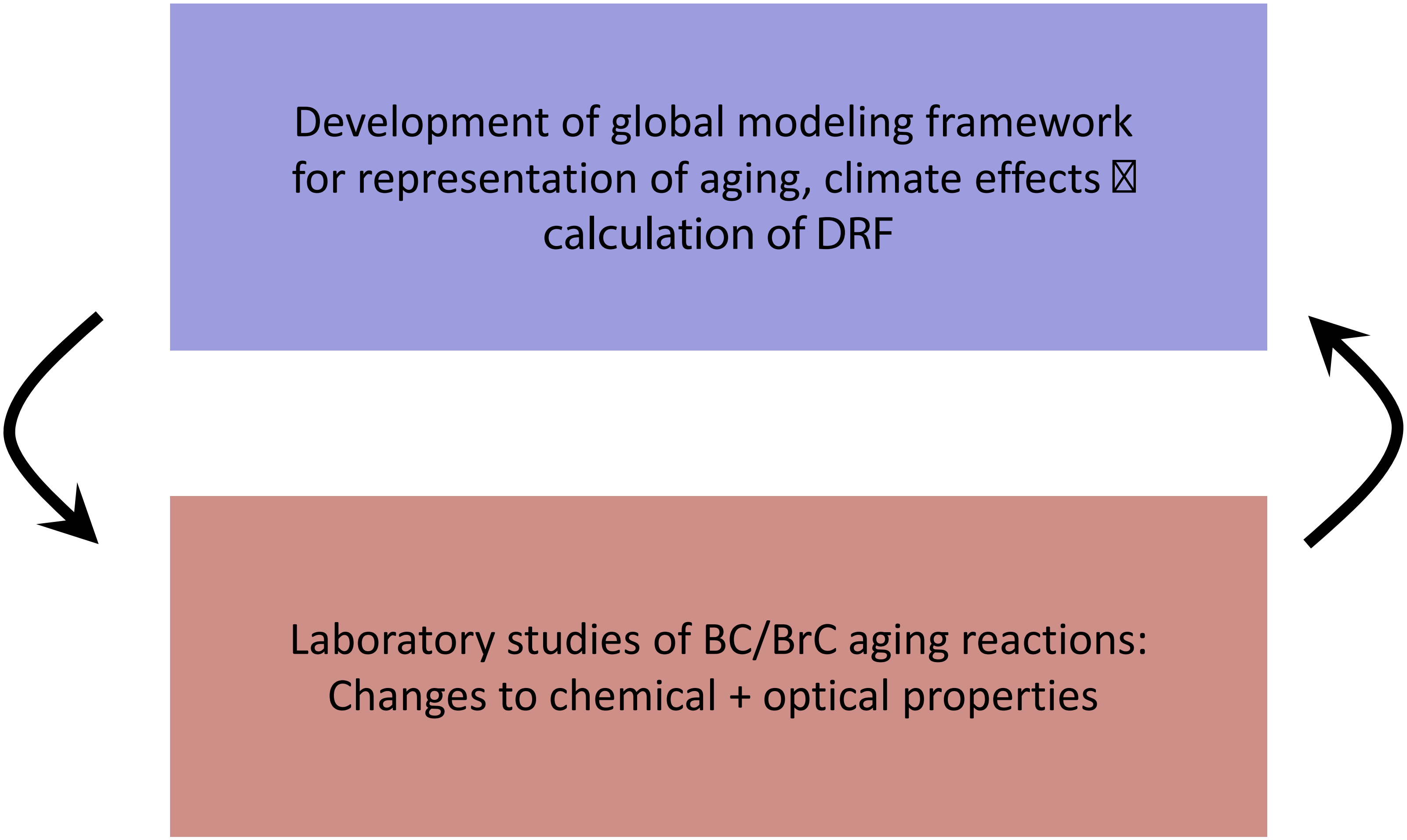
Black/brown carbon aerosol is chemically dynamic, subject to atmospheric aging reactions; these can lead to dramatic changes in physicochemical properties, and therefore climate forcing effects.

A complete understanding of this aging, and representation of this aging within models, is necessary for the accurate simulation of global direct radiative forcing.

Major questions:

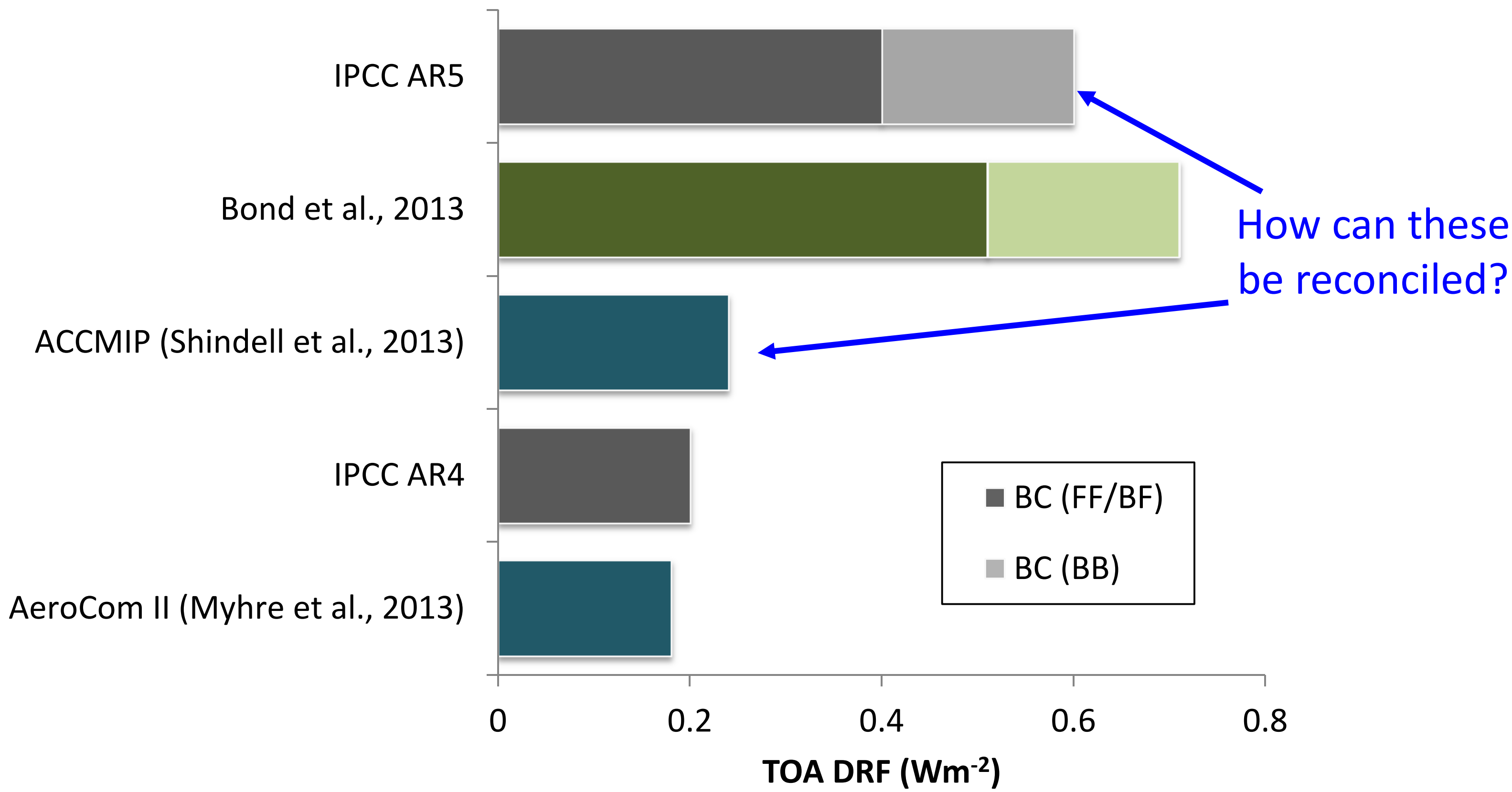
- what are the most important atmospheric aging transformations of BC/BrC?
- what effects do aging reactions have on climate-relevant properties of BC/BrC?
- how do these aging reactions impact BC/BrC direct radiative forcing?

Approach: Laboratory + Modeling

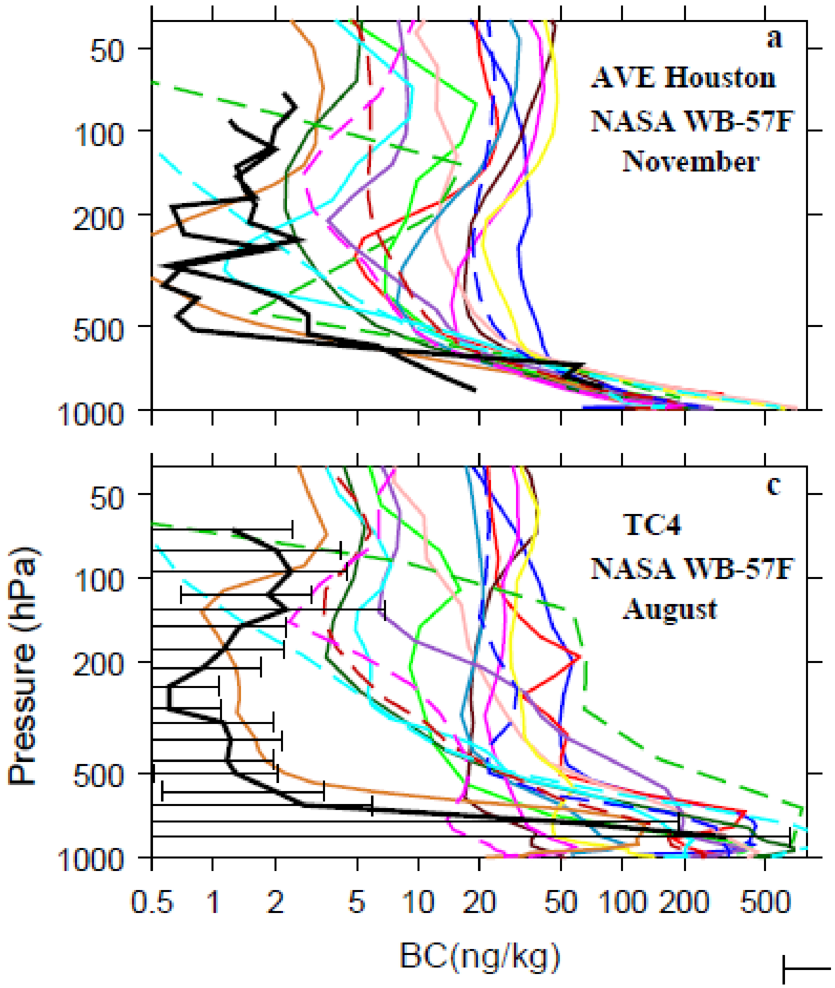


IPCC AR5 Estimates that Black Carbon is the 2nd Largest Warming Agent in the Atmosphere.

(but that's not what models say)

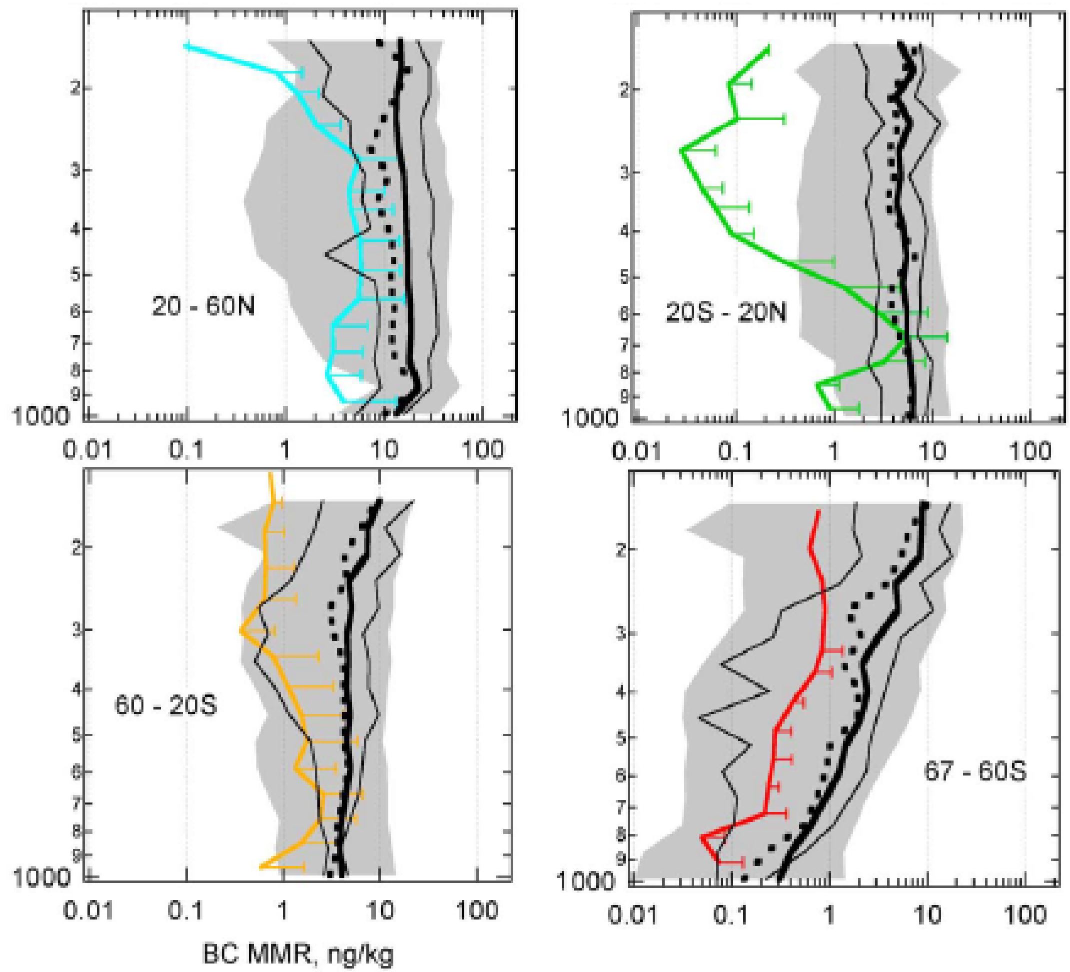


Observations Also Suggest That Models Overestimate BC



Obs in black, AeroCom models in colour

[Koch et al., 2009]

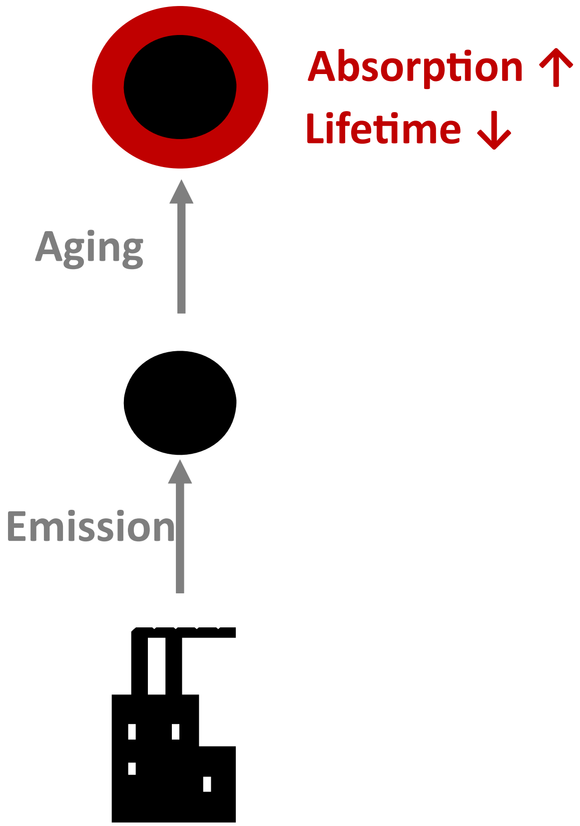


AeroCom means in black, HIPPO obs in colour

[Schwarz et al., 2010]

AeroCom models overestimate BC over Americas & remote Pacific by factor ~5-8.

Aging Processes may Reconcile Both Mass and Absorption Constraints

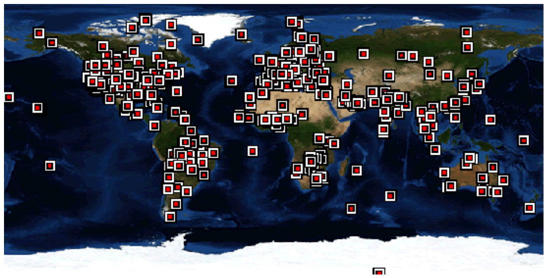


GEOS-Chem integrated with RRTMG (GC-RT)
(Heald et al., 2014)

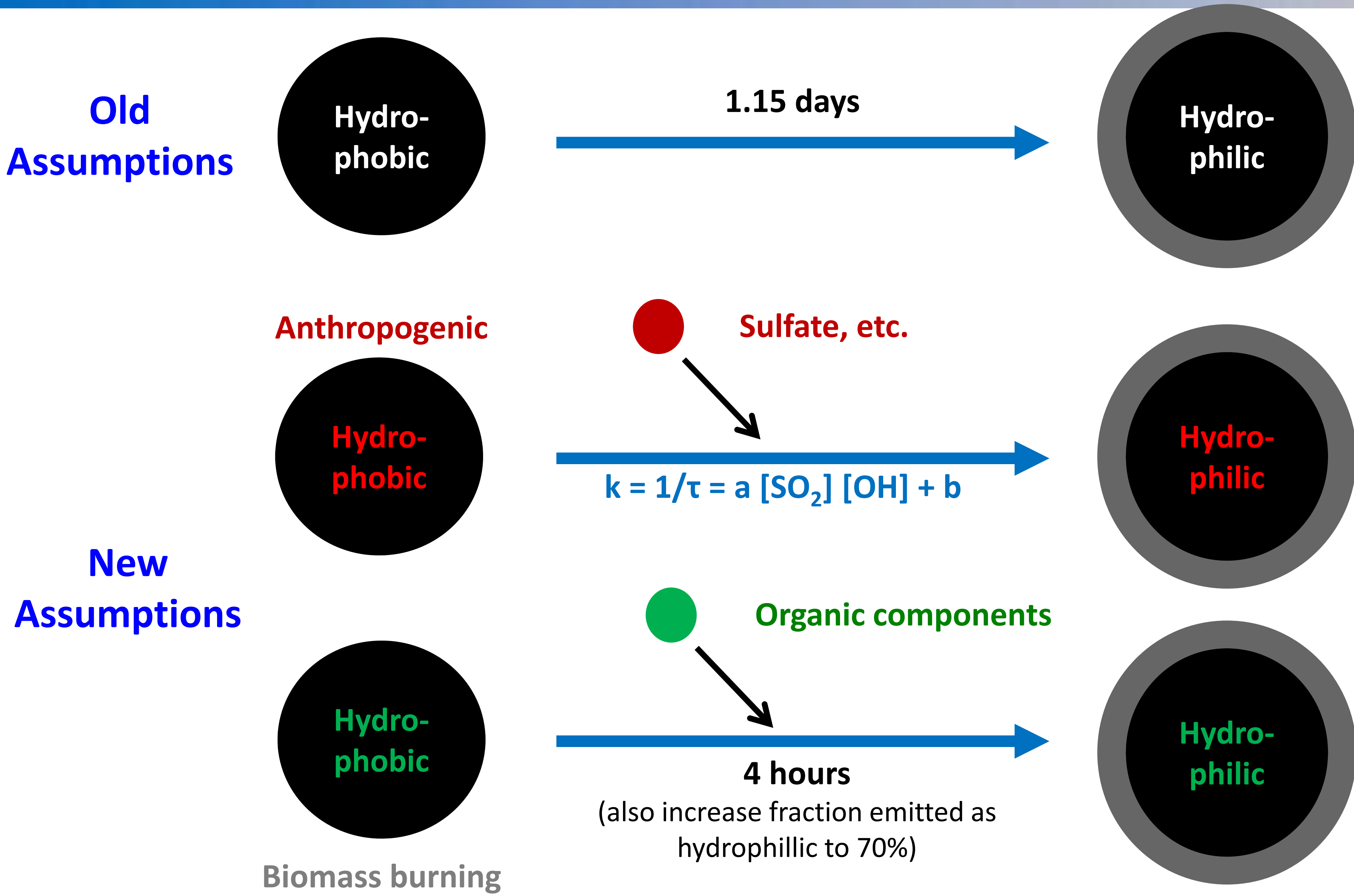
+

Mass Concentrations (ARCTAS, EUCAARI, HIPPO + surface)

Absorption (AERONET)



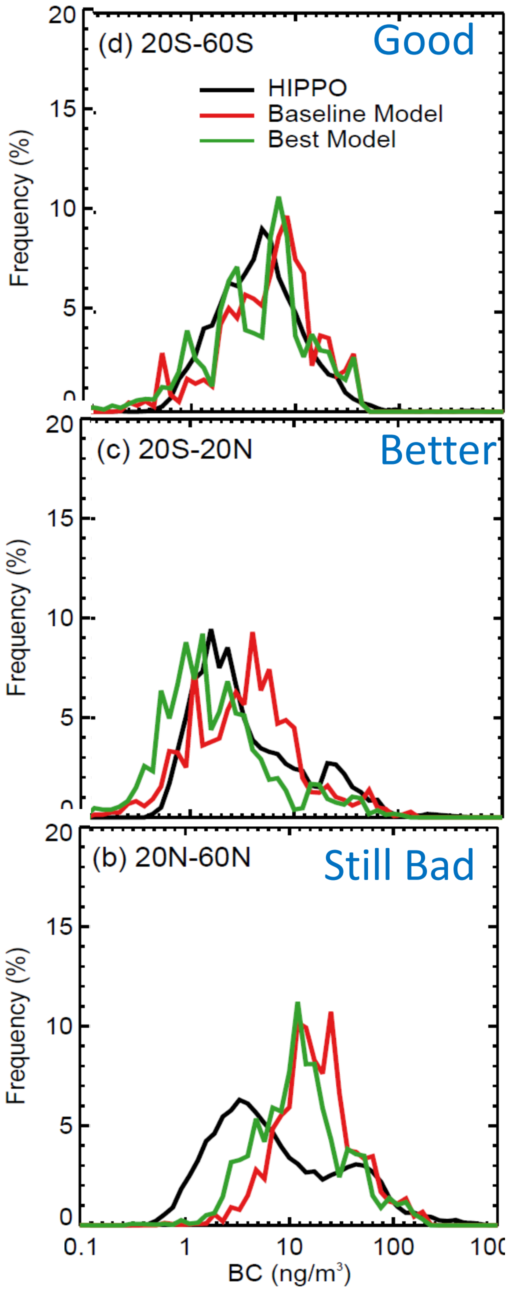
New Model Aging Processes for BC



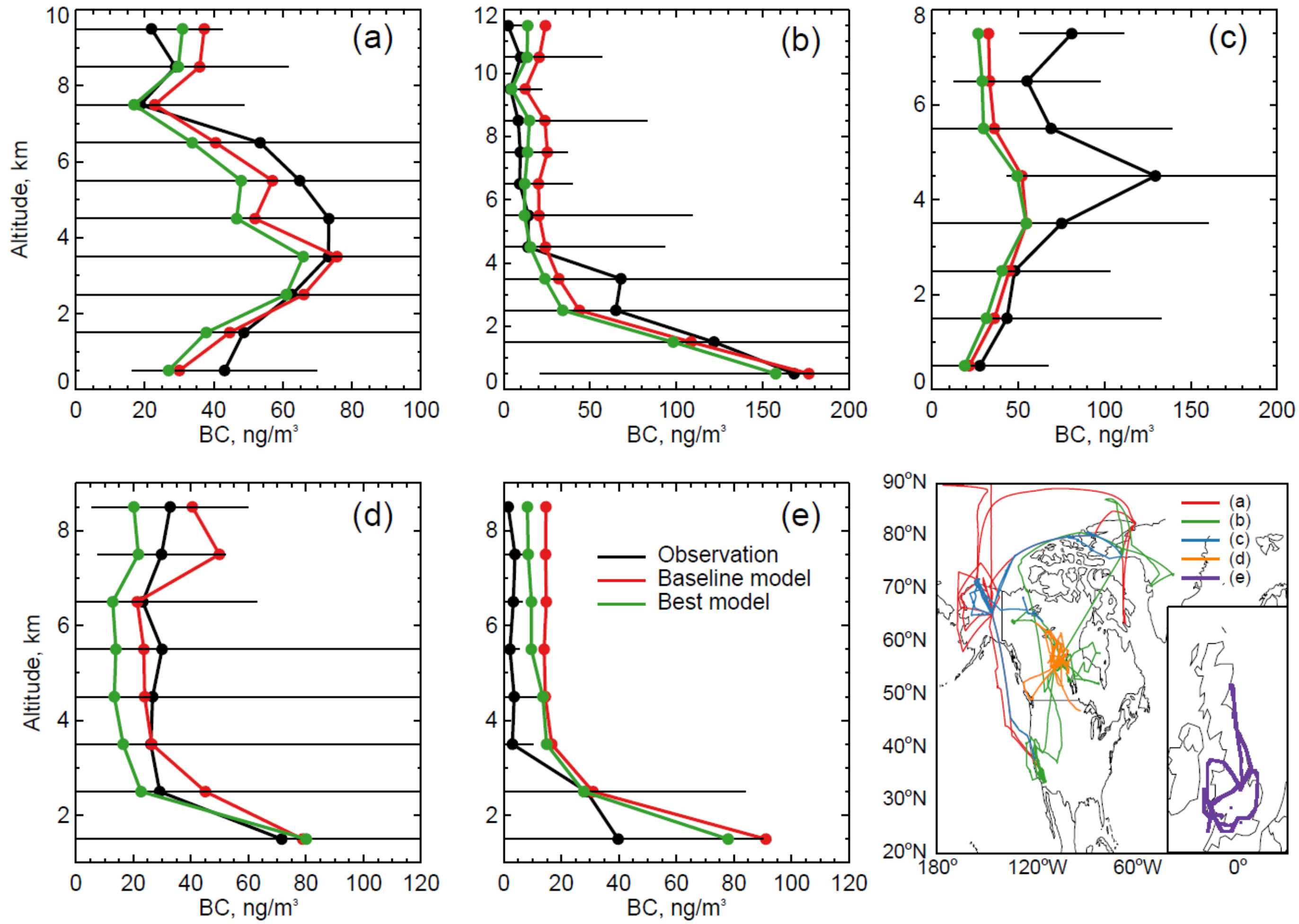
Based on several lab and field studies, esp Liu et al. (2010); Akage et al. (2012)

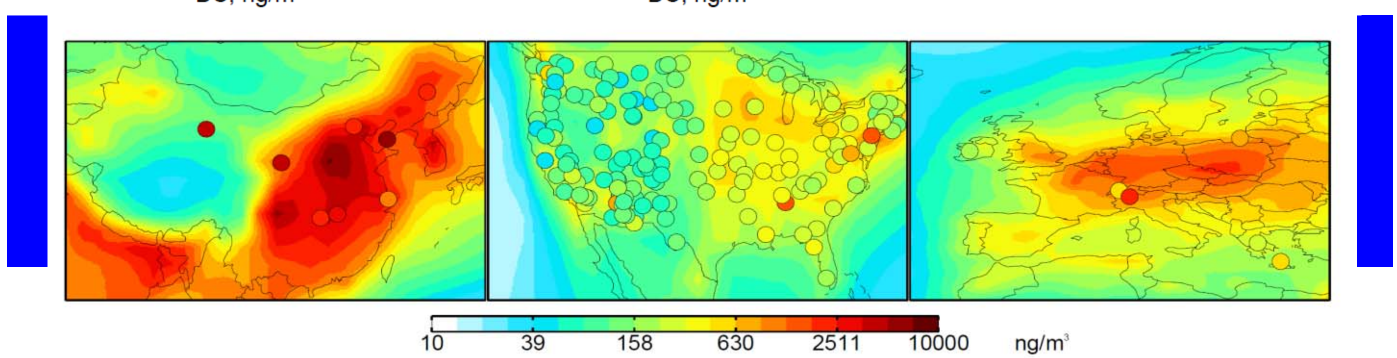
Impact of New Model Aging Processes on Simulation of BC

HIPPO

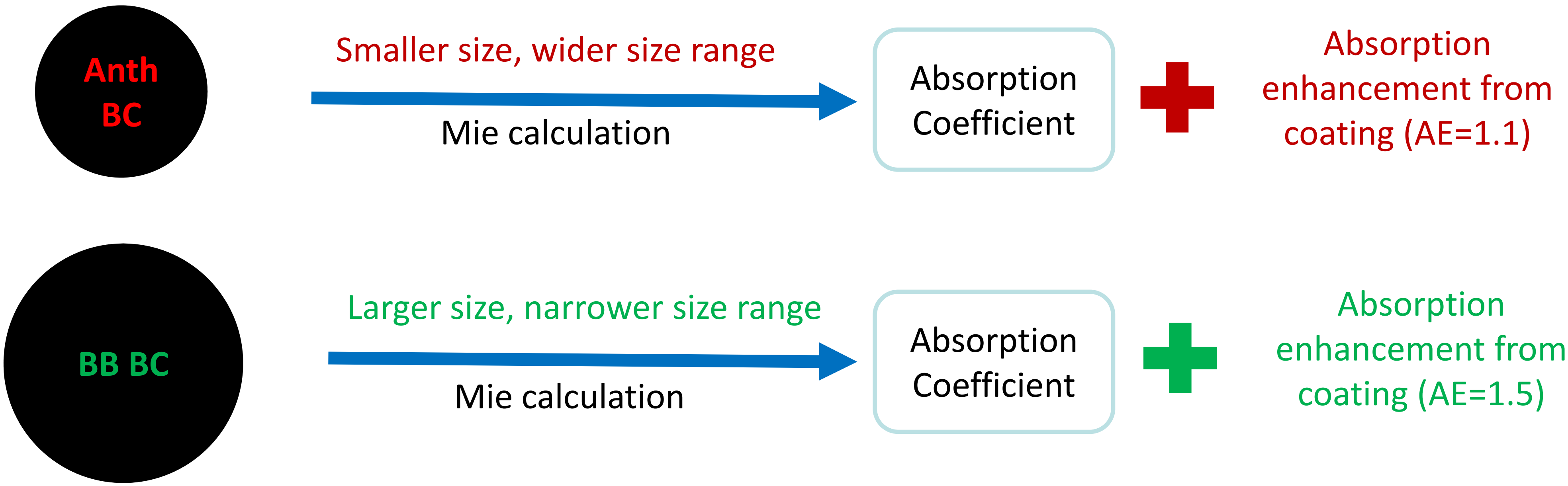


Continental (Near-Source)





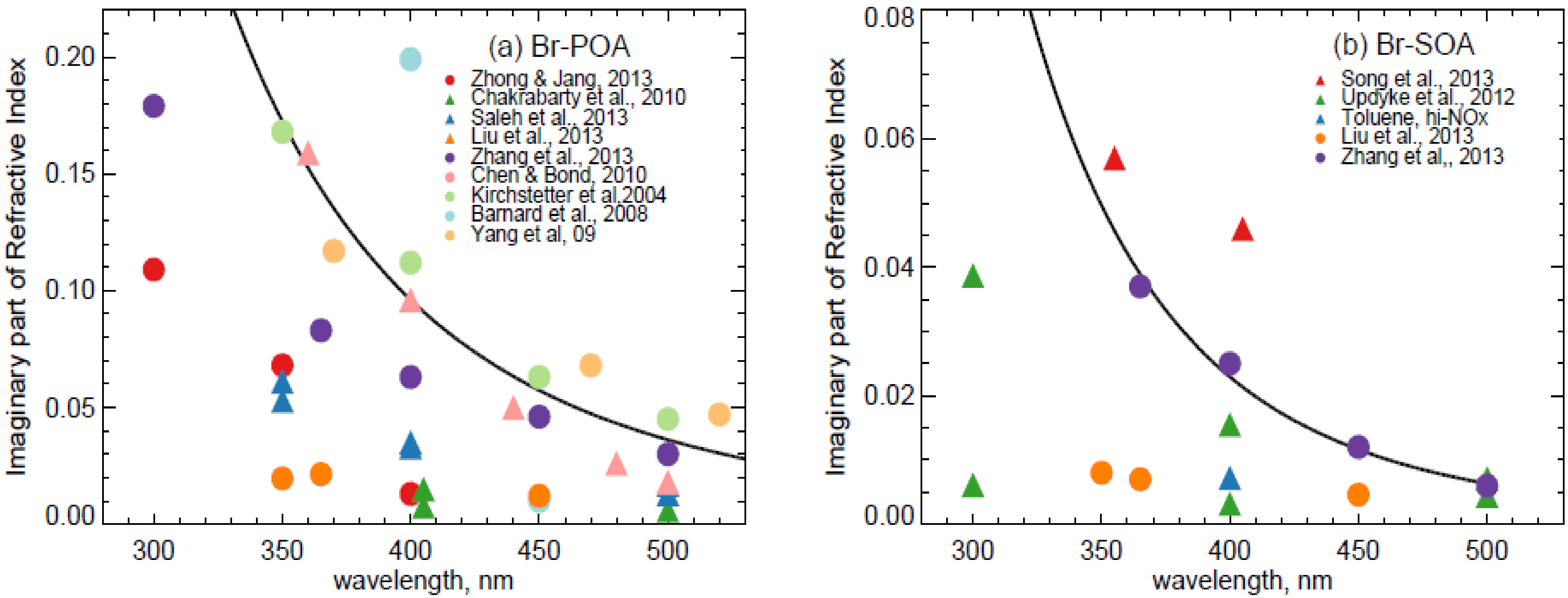
Considering Absorption Enhancement of BC



Also "Most Absorbing" Simulation : Set AE=2 and standard aging mechanism (longer lifetime)

(Akage et al., 2012; Schwarz et al., 2006; 2007; 2008; Lack et al., 2012; Dubovik et al., 2002; Shamjad et al., 2012; Moffet et al., 2009; Knox et al., 2009; Kondo et al., 2011; Lack et al., 2012; Moffet and Prather, 2009; Bond et al., 2006; Cappa et al., 2012)

Adding Brown Carbon to GEOS-Chem

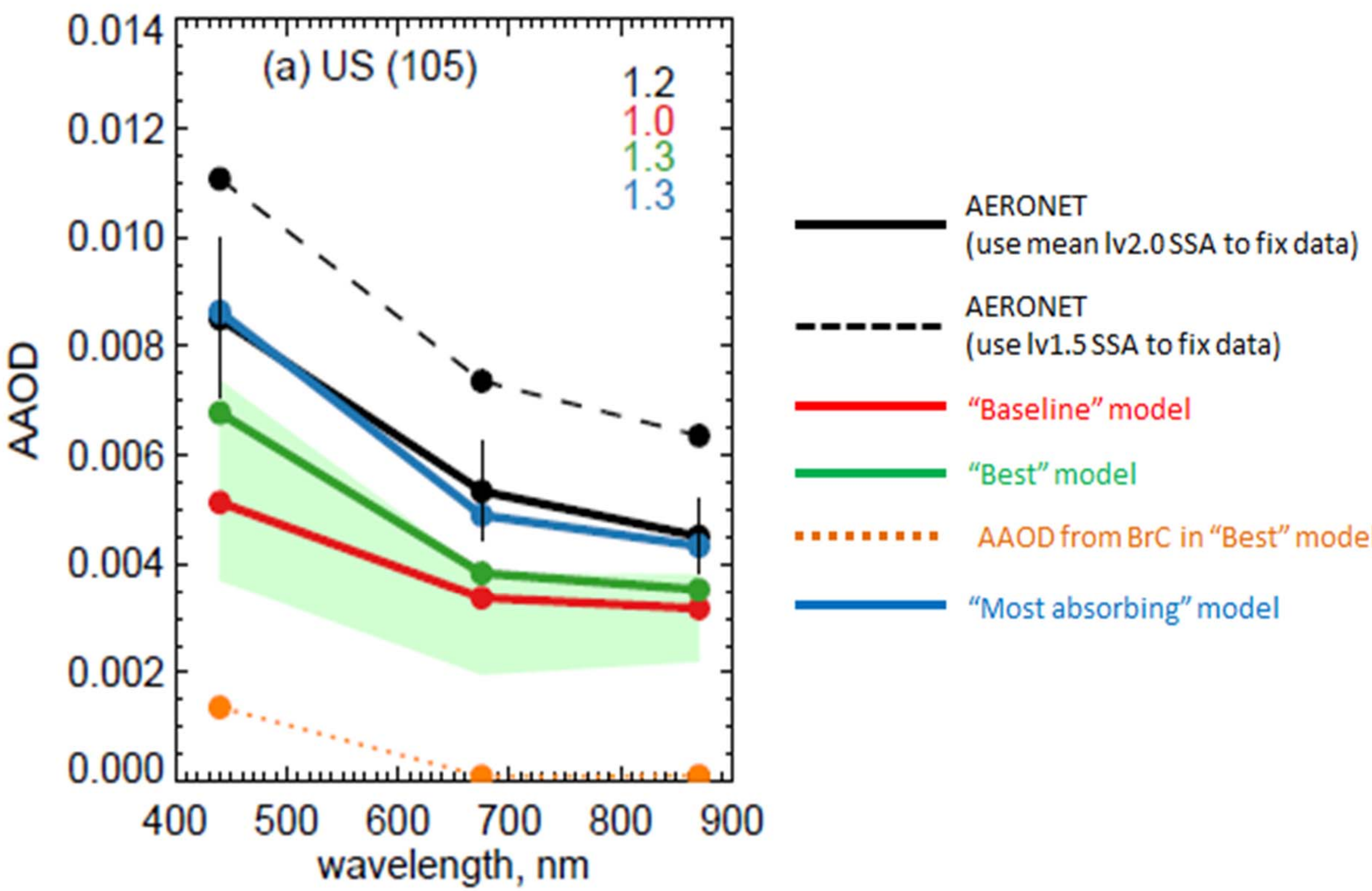


MAE=1 m^2/g MAE=0.3 m^2/g

Absorption of BrC is highly uncertain - we choose upper-range estimates

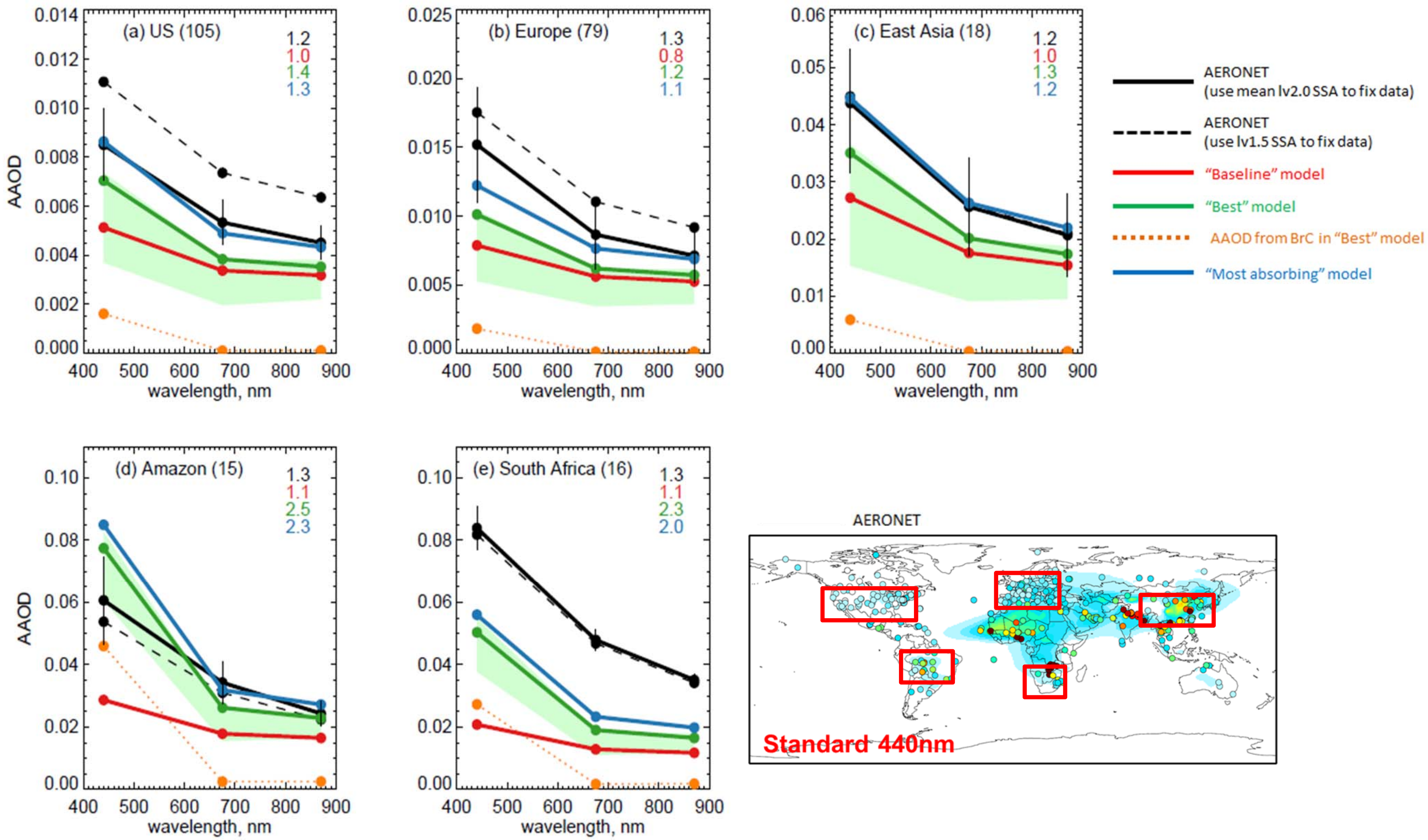
Including Brown Carbon is Critical to Capturing the Spectral Dependence of AERONET AAOD

*AAOD product here using lev2 SSA with lev1.5 AOD



Measurements Still Suggest Absorption is Underestimated

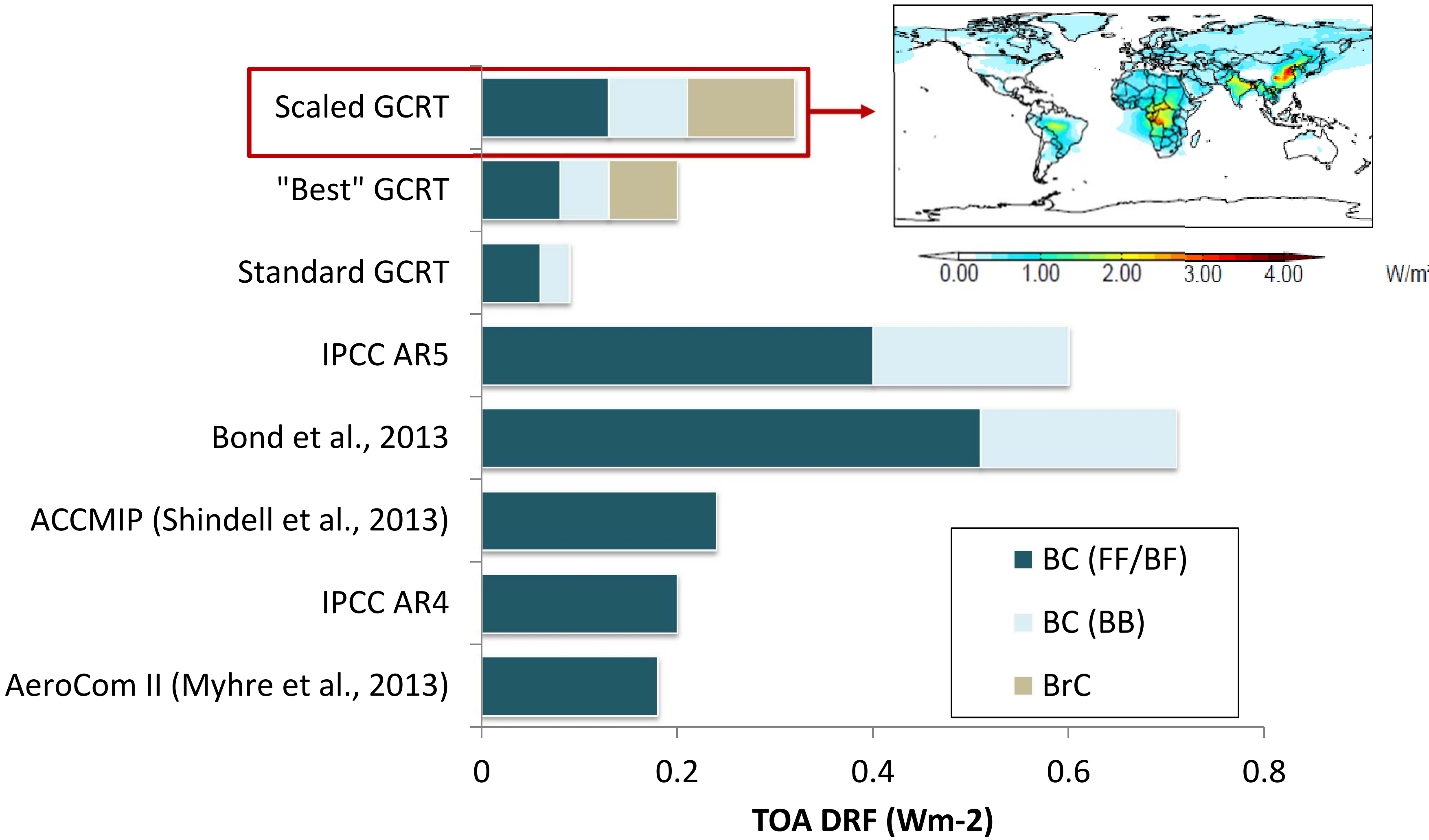
*AAOD product here using lev2 SSA with lev1.5 AOD



Better able to capture the spectral AAOD with our "best" simulation (including BrC), but still biased low (especially in some biomass burning regions).

Can "scale up" our model to match observations (Bond et al., 2013) – emissions or optics?

Our Work Suggests Smaller BC DRF Required to Match All Observational Constraints

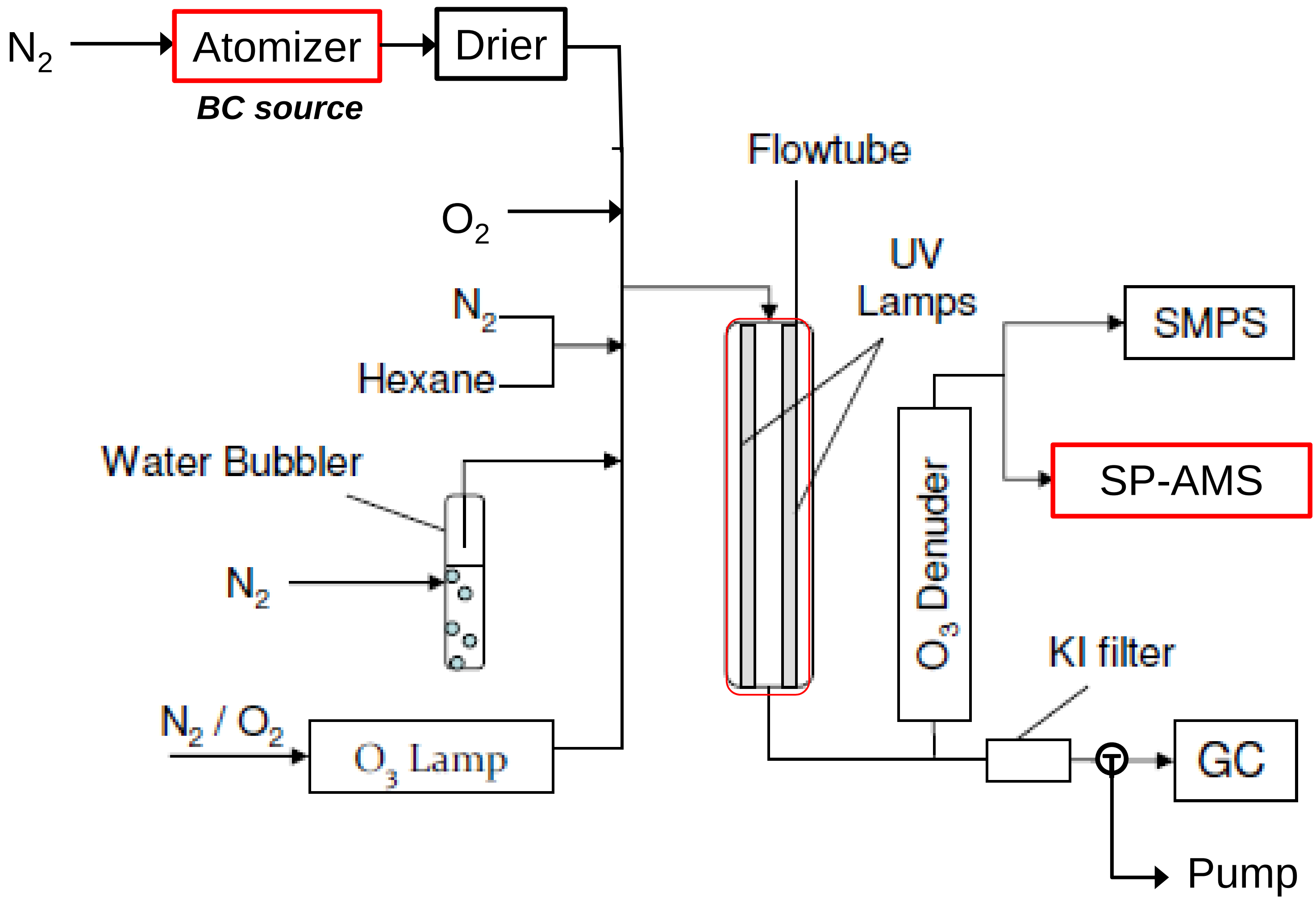


Brown Carbon contributes 35% of the warming from carbonaceous aerosols.
BC DRF is less than methane and tropospheric ozone.
Suggests that controlling BC is less effective for climate mitigation.

[Wang, et al., ACP, 2014]

Lab Study 1: Heteogeneous oxidation of BC

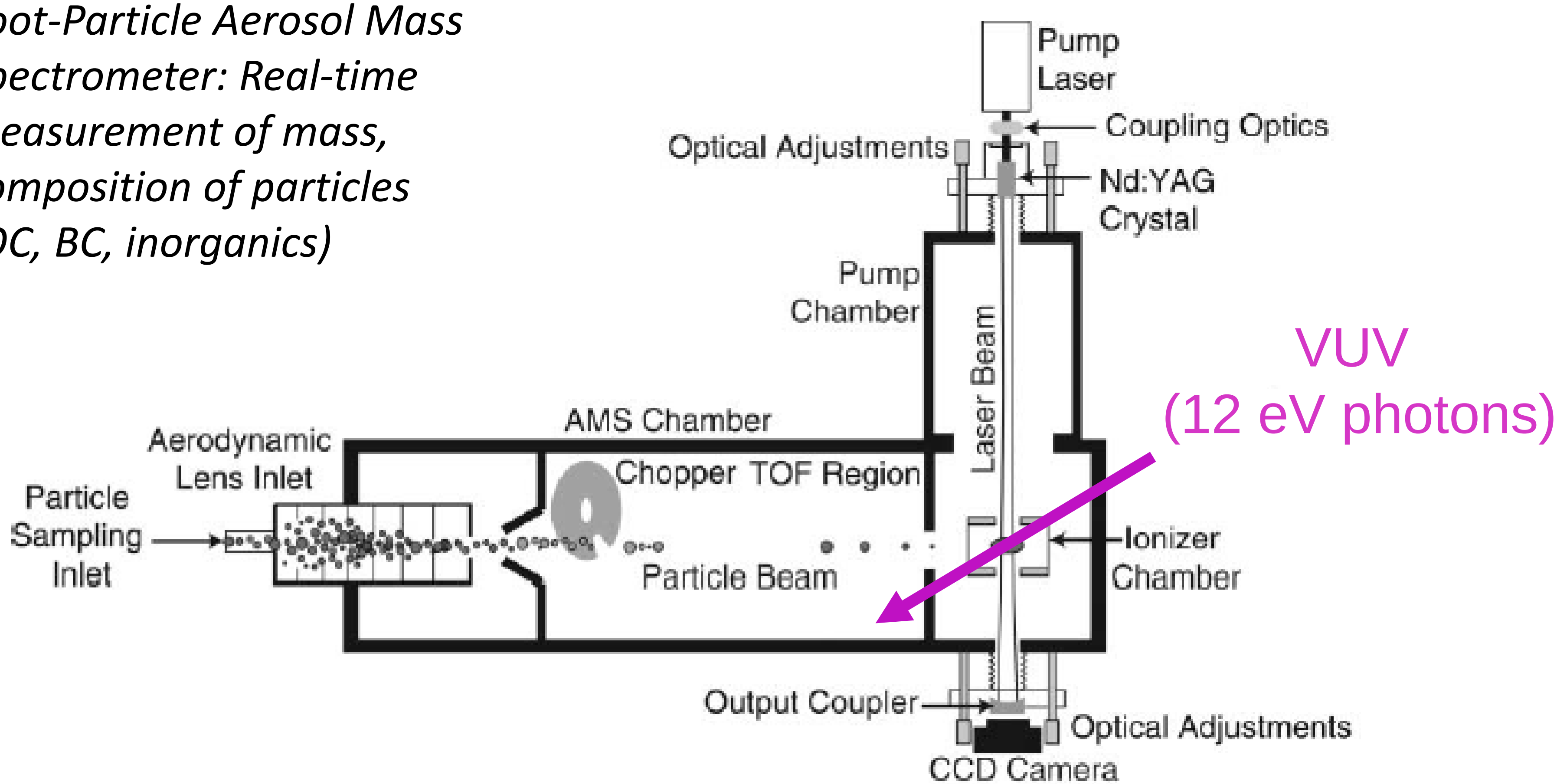
with Kevin Wilson (LBNL), Manjula Canagartna and Paola Massoli (ARI)



[Browne, et al., submitted]

SP-AMS with VUV ionization

Soot-Particle Aerosol Mass Spectrometer: Real-time measurement of mass, composition of particles (OC, BC, inorganics)



Ionization efficiencies

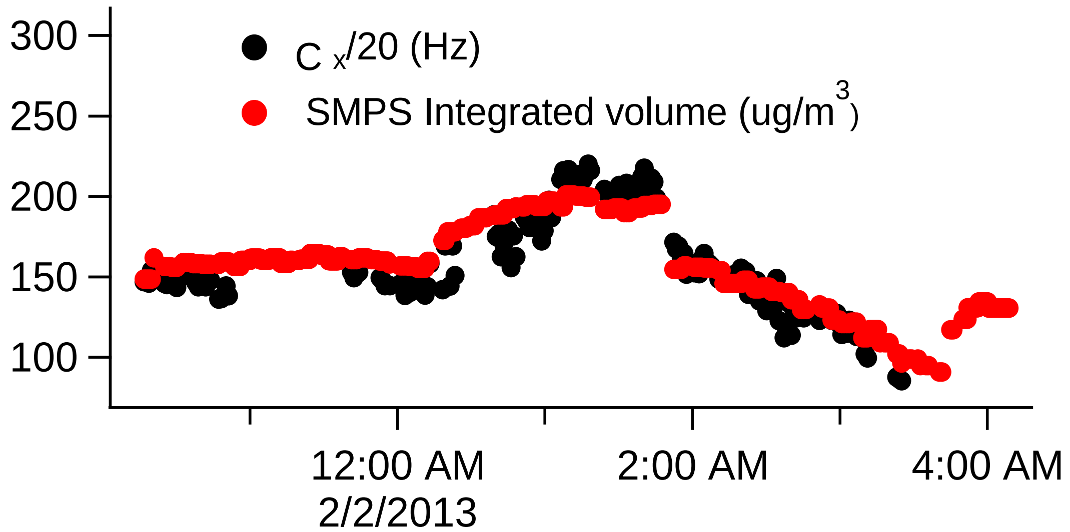
PAHs: 7-8 eV

Most organics: 9-10 eV

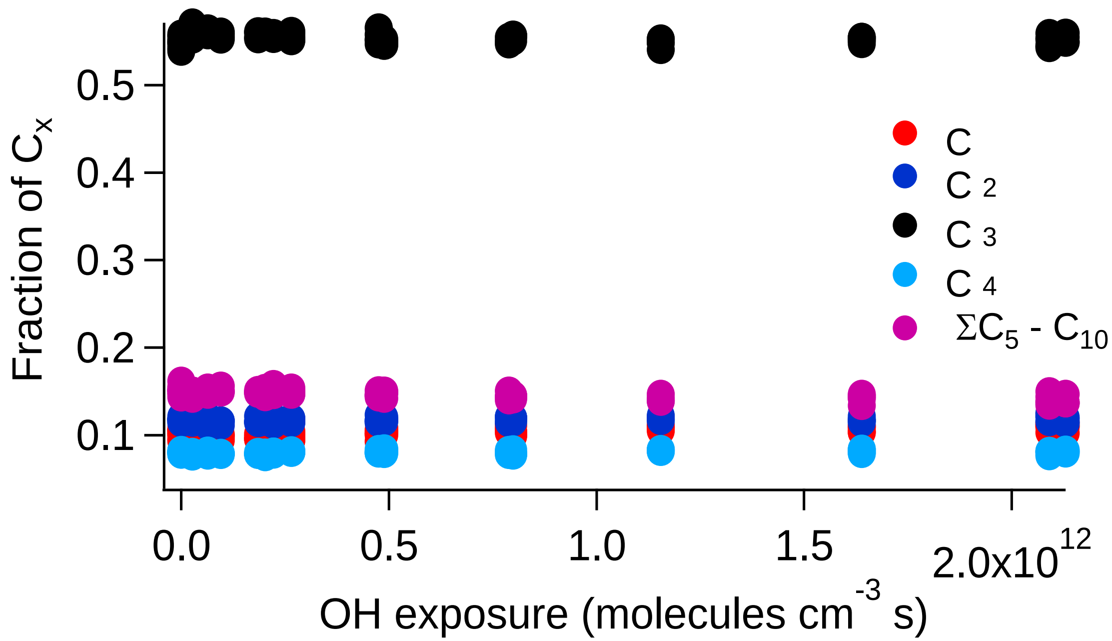
BC fragments (e.g. C_3): ~11.5 eV

[Onasch et al. 2012]

Heterogeneous aging: BC “cores” unaffected

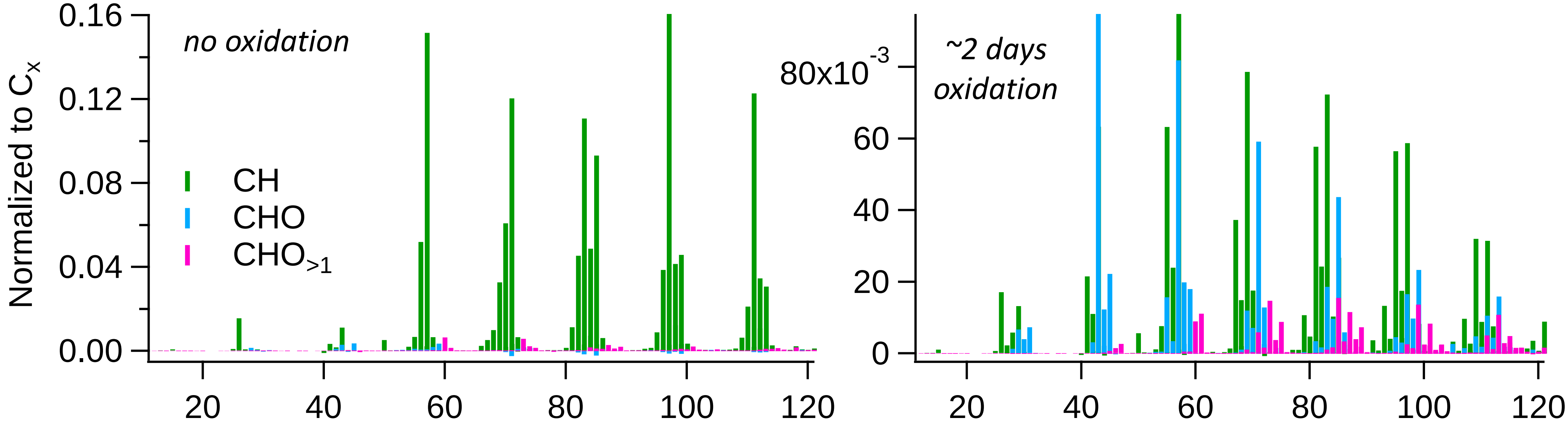


C_x accounts for the majority of the flame soot mass



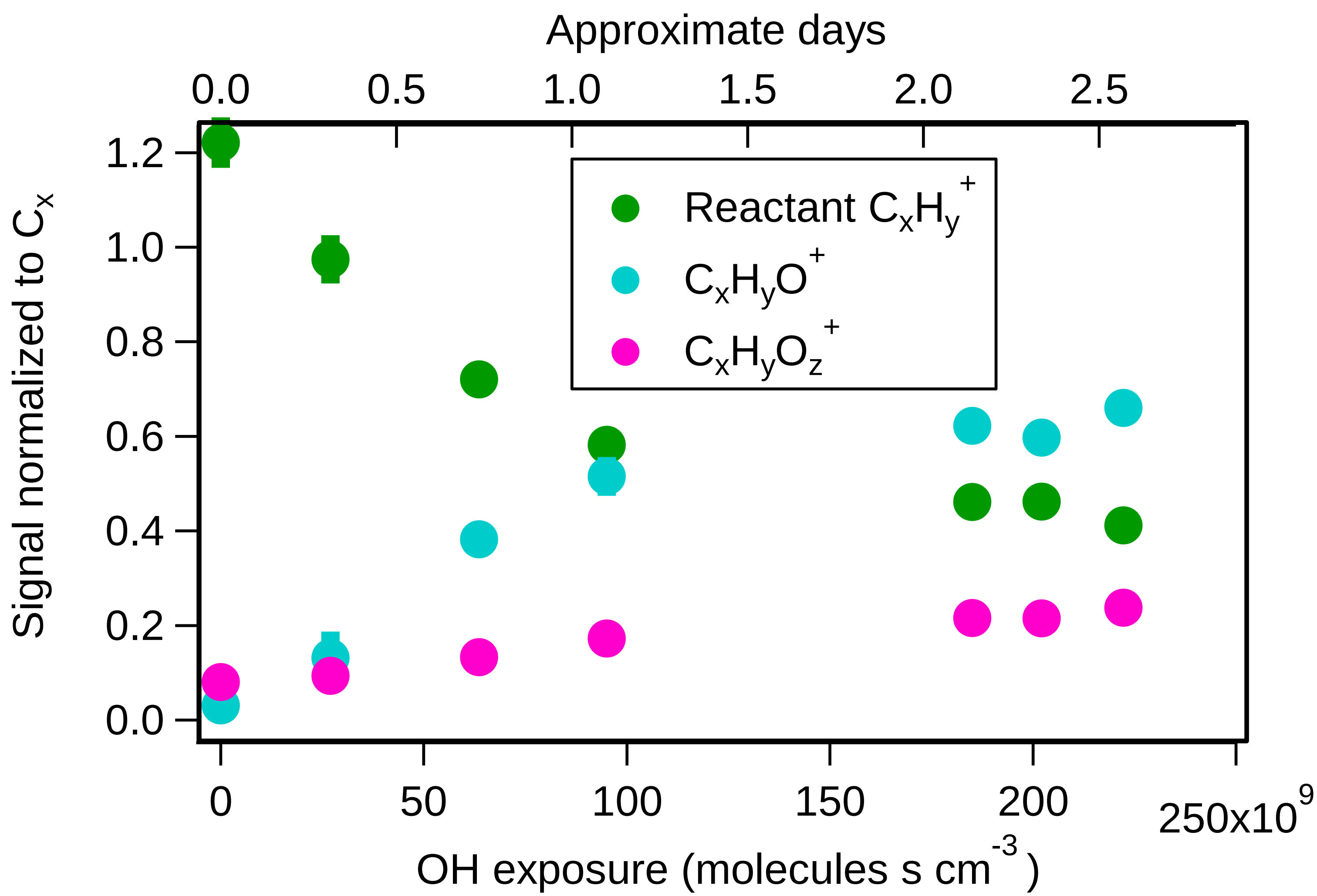
C_x spectrum remains constant with aging: BC “core” is inert on the timescale of atmospheric aging

Changes to adsorbed organic species

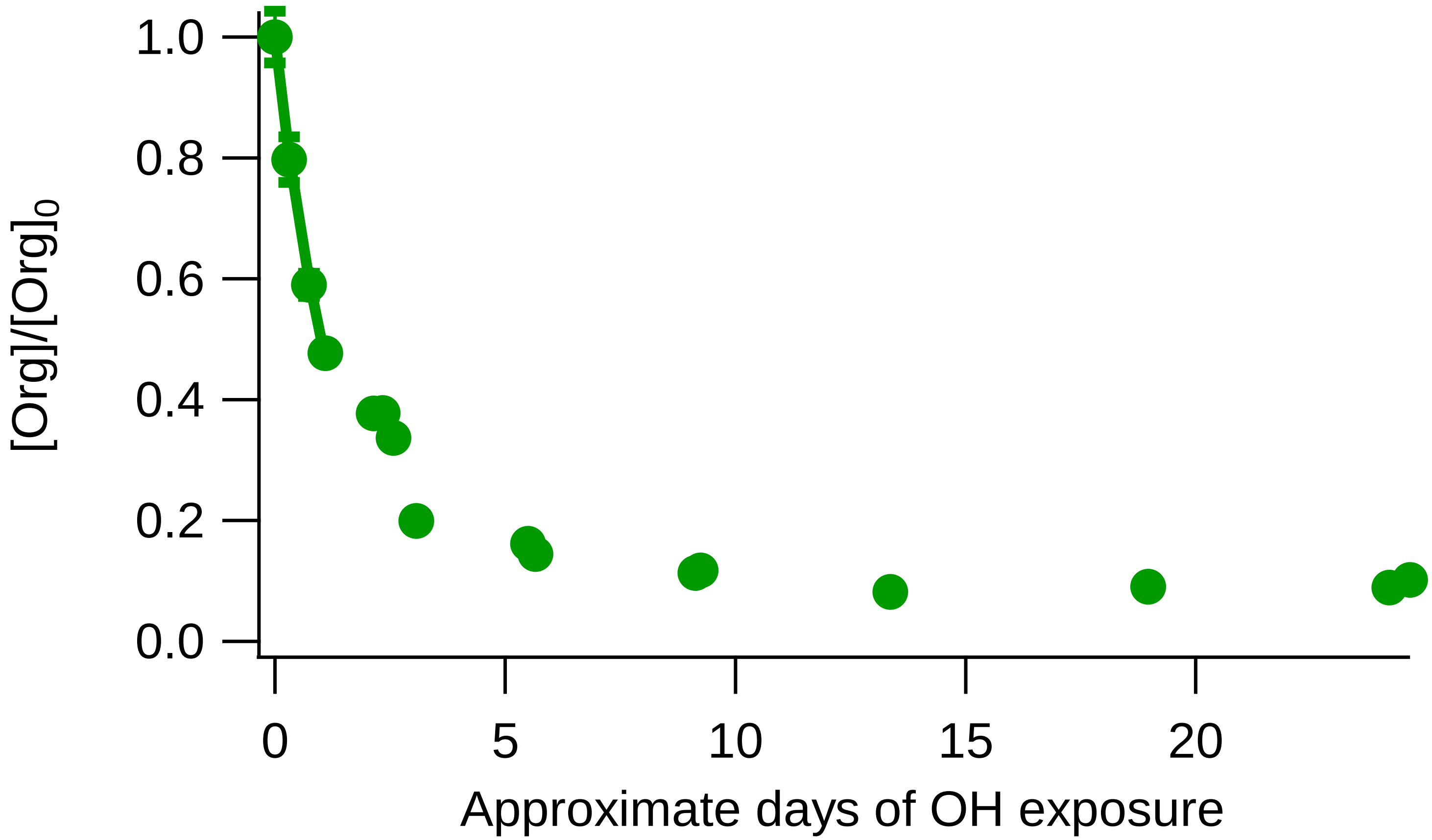


Organics associated with BC (representing small fraction of total particles) undergo dramatic chemical changes

Continual changes with oxidant exposure



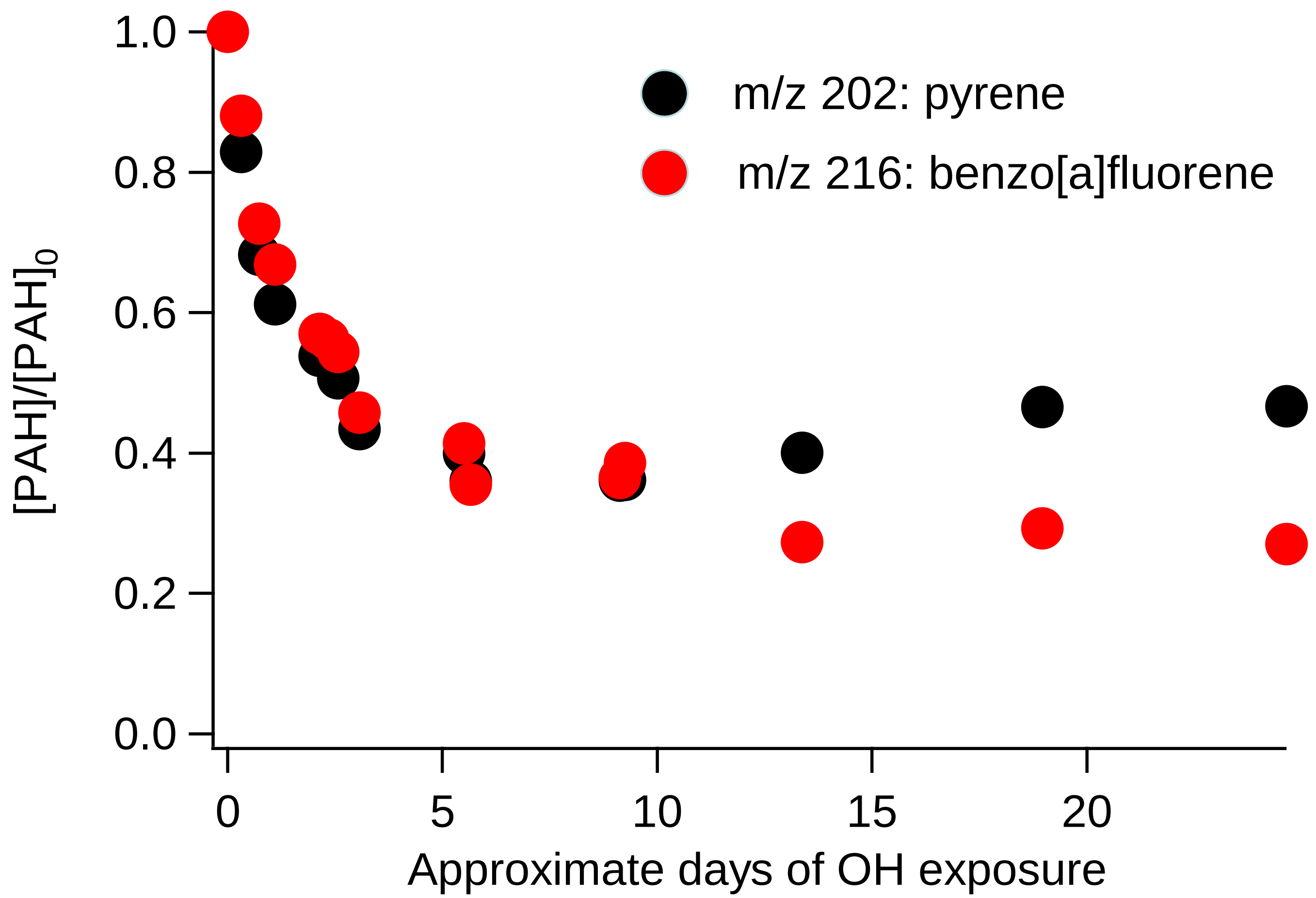
Rapid loss of organic species



Lifetime of adsorbed organics is short

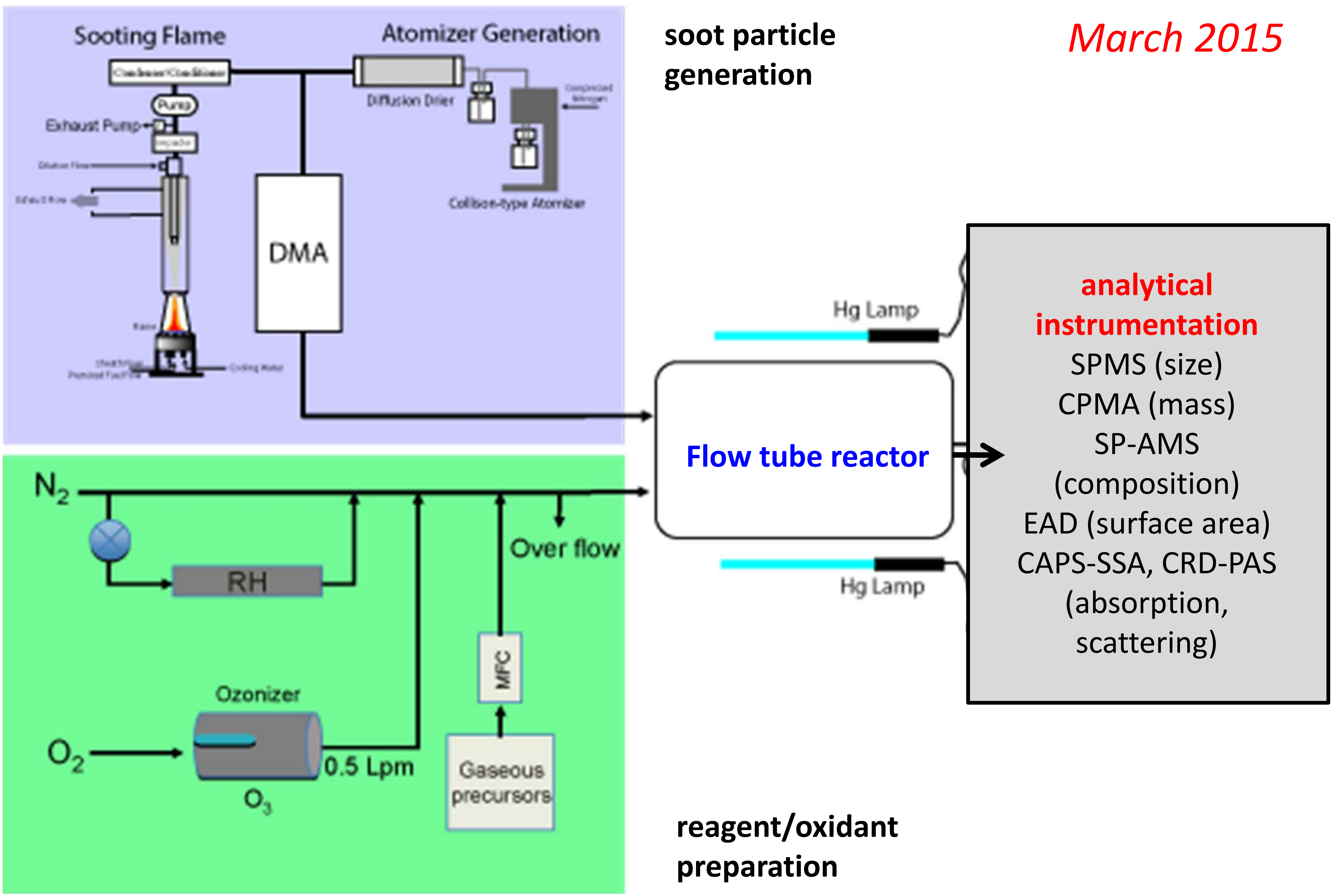
Follow-up experiments: Water uptake (hydrophobic → hydrophilic)

Loss of adsorbed PAHs



Long-timescale “survival” of PAHs (morphology)

Next: Coating + heterogeneous ox. experiments



Experimental matrix

BC source

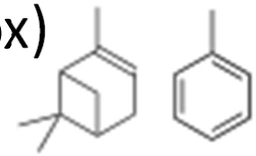
- fractal soot from McKenna burner (denuded at 300^{o}C)
- also atomized black carbon spheres

Particle size

- monodisperse, 30-300 nm

Aging type

- OH + SO_2 (sulfuric acid coating, het. ox)
- OH + NH_3 + SO_2 (ammonium sulfate coating, het. ox)
- OH/O_3 + VOCs (SOA coating, het. ox)
- mixed coatings



Parameterization

Changes to composition/hygroscopicity + optics as a function of atmospheric exposure (to match representation in Wang et al. 2014)

Key optical parameters determined, included in a “lookup table” (or interpolated function) based on experimental results

particle size

relative humidity

wavelength

mixing state (BC:org:SO_4)

→

mass extinction efficiency

single scattering albedo

asymmetry parameter

Summary/conclusions

- Modeling vs measurements of BC: models overestimate loadings, underestimate aerosol absorption

- Aging processes can affect both concentrations (via changes to deposition) and optical properties (via changes to coatings); need for an improved understanding, description of such processes

- Global modeling results: Improved agreement between predicted, measured BC loadings and properties (but AAOD still underestimated!)

- Laboratory results: Heterogeneous oxidation an efficient way to change organic components of soot; oxidation can dramatically decrease“brown”-ness of brown carbon

- Next step: Laboratory results $\rightarrow$ implementation in models

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Dantong Liu
Hugh Coe

Anthony Clarke

<u>Publications from this Project</u>:
Heald et al., ACP, 2014
Wang et al., ACP, 2014
Browne et al., JPCA, submitted
Lambe et al., ES&T, 2013

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