Investigating the effects of atmospheric aging on the radiative properties and climate impacts of black (+ brown) carbon aerosol

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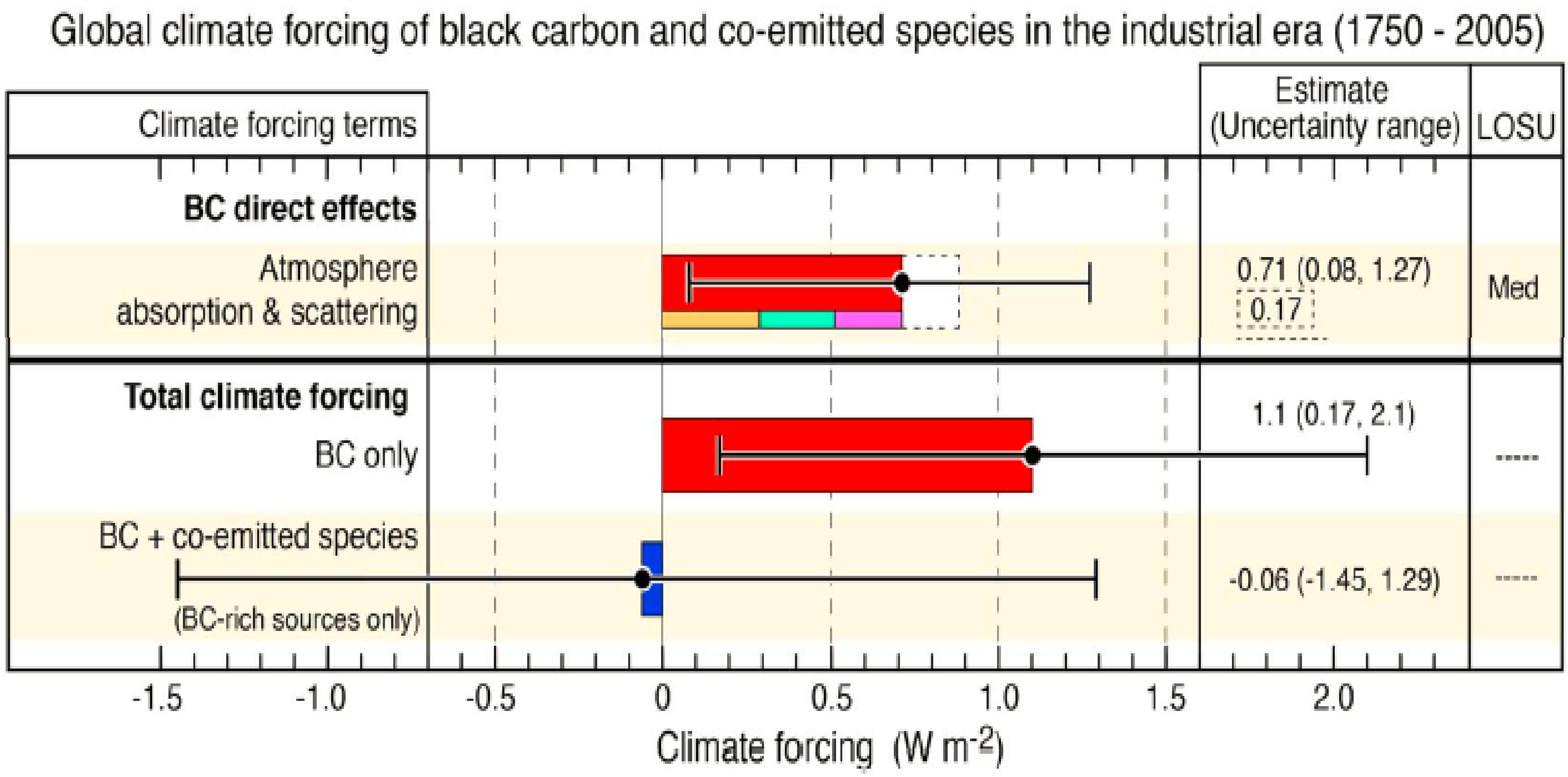
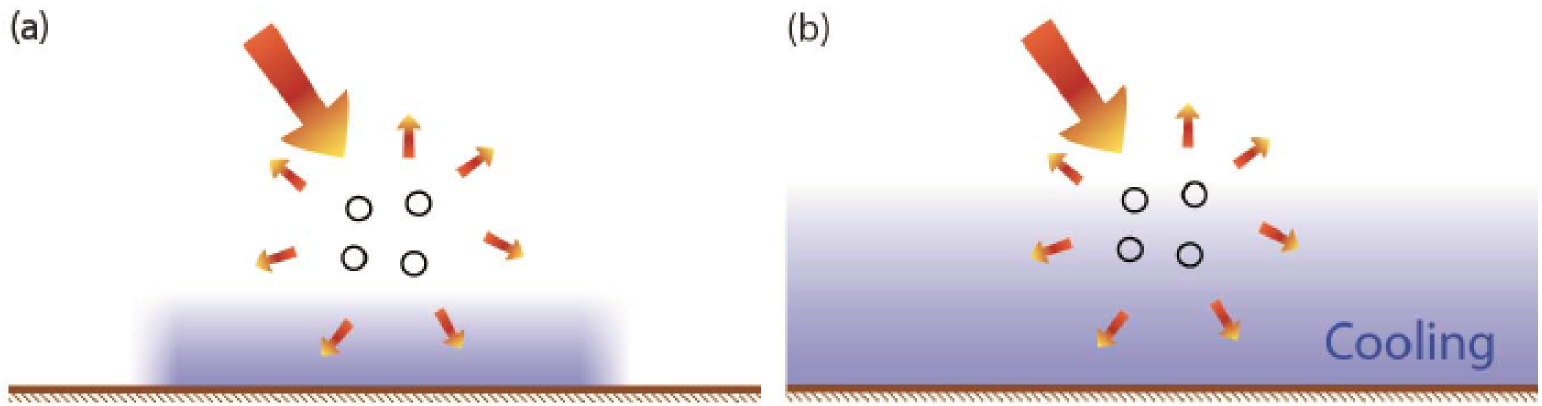
BC climate forcing: Large, complex, uncertain



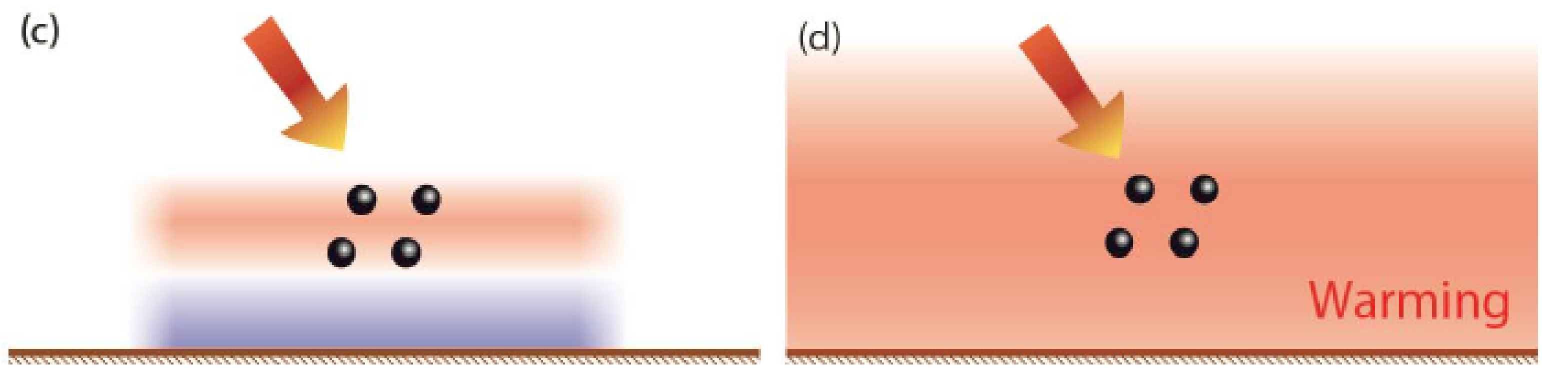
[Bond et al., 2013]

Focus on direct radiative effect

Scattering particles



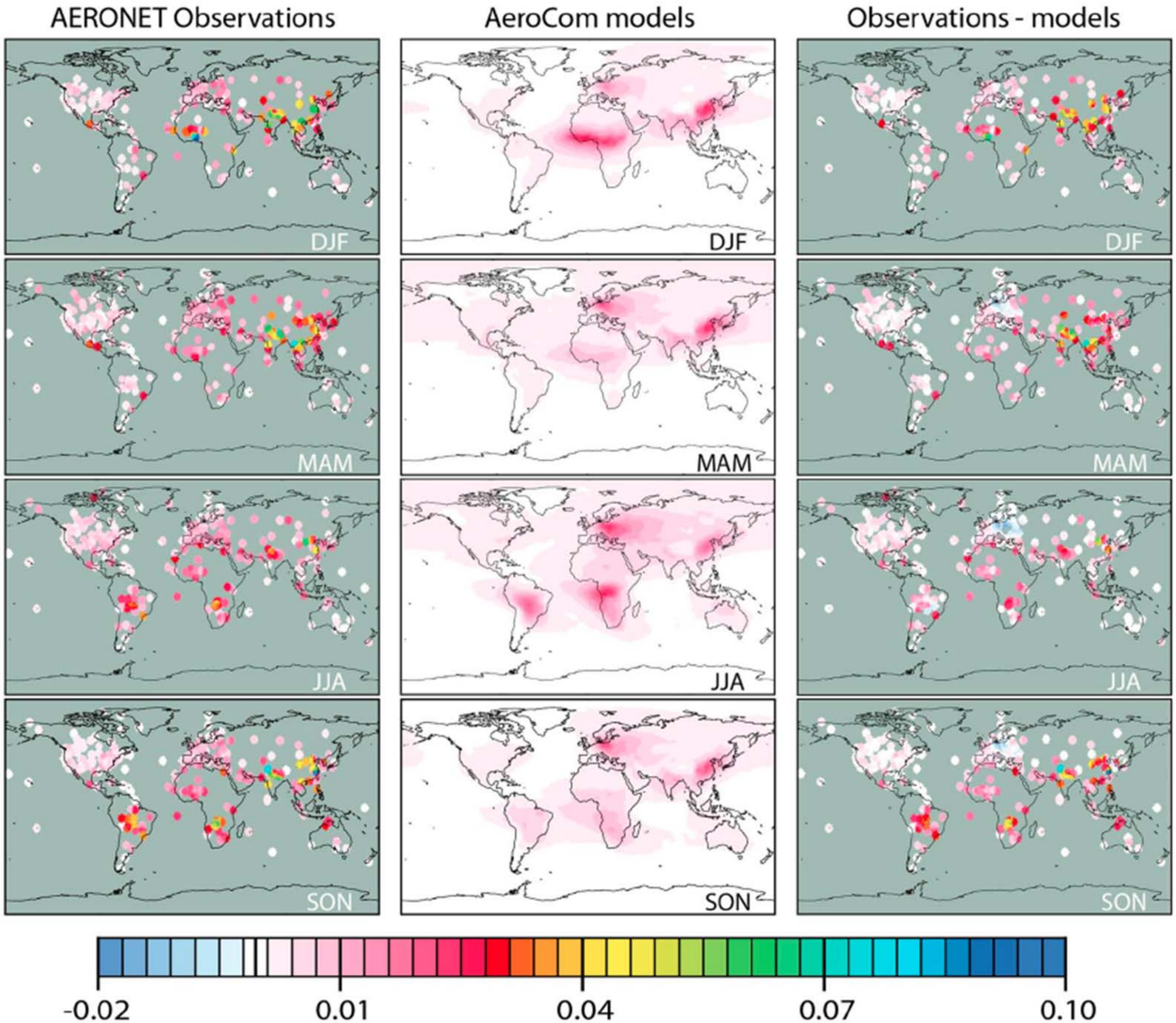
Absorbing particles



Local effect *Distributed effect*

IPCC, Fifth Assessment Report, 2013.

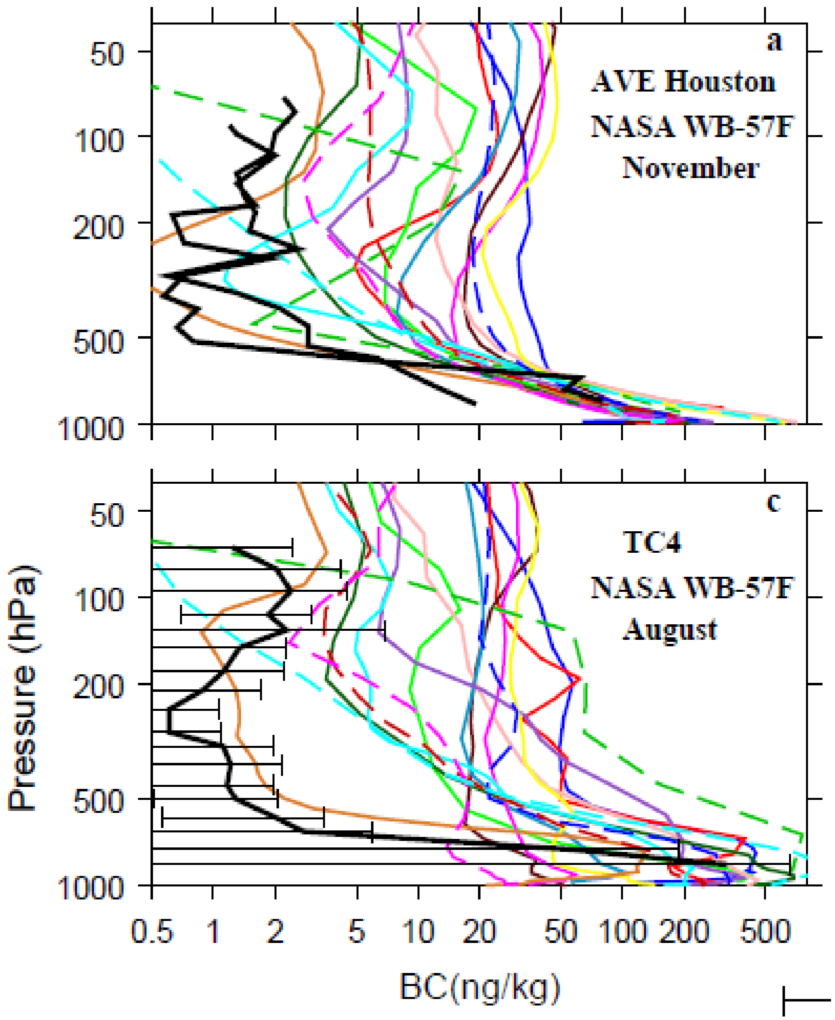
(1) Global models *underestimate* absorption AOD



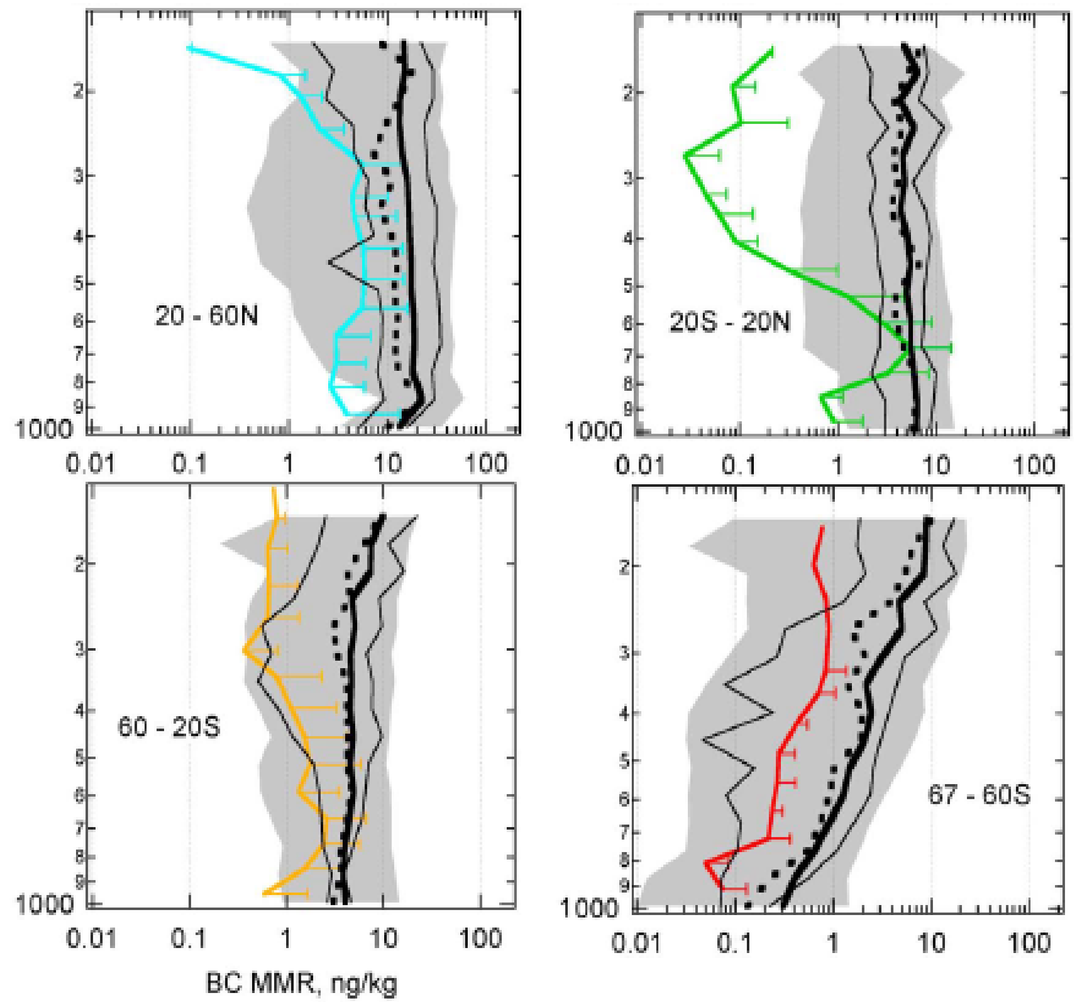
AeroCom models underestimate AAOD, often by a lot

[Bond et al., 2013]

(2) Global models *overestimate* BC loading



Obs in black, AeroCom models in color



AeroCom means in black, observations in color

AeroCom models overestimate BC over Americas by factor of ~8, overestimate remote BC by factor of ~5.

[Koch et al., 2009]

[Schwarz et al., 2010]

Our project

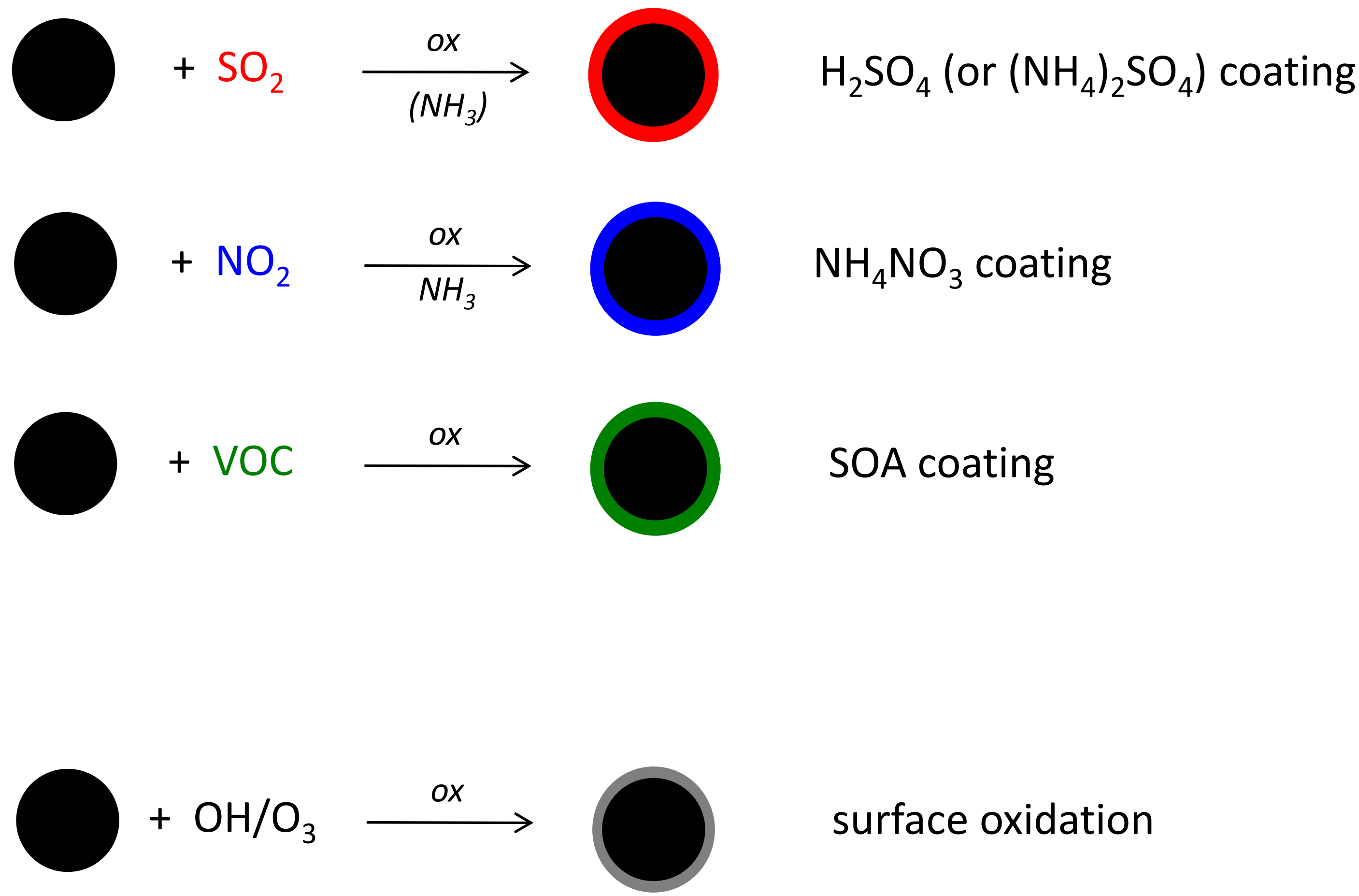
Black carbon aerosol is a chemically dynamic system, subject to atmospheric aging reactions; these can lead to dramatic changes in physicochemical properties, and therefore climate forcing effects.

An incomplete understanding of this aging, and/or representation of this aging within models, may explain some fraction of the model-measurement discrepancies.

Key BC aging reactions

1) Coatings

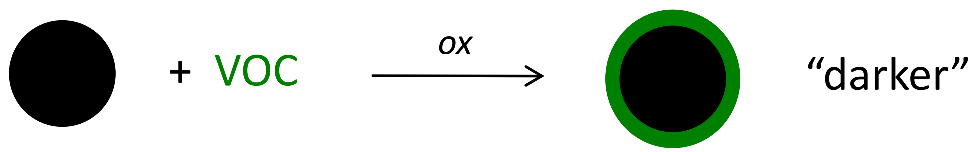
2) Heterogeneous oxidation



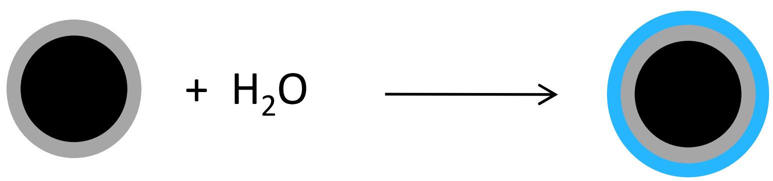
Effects of aging on BC properties

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

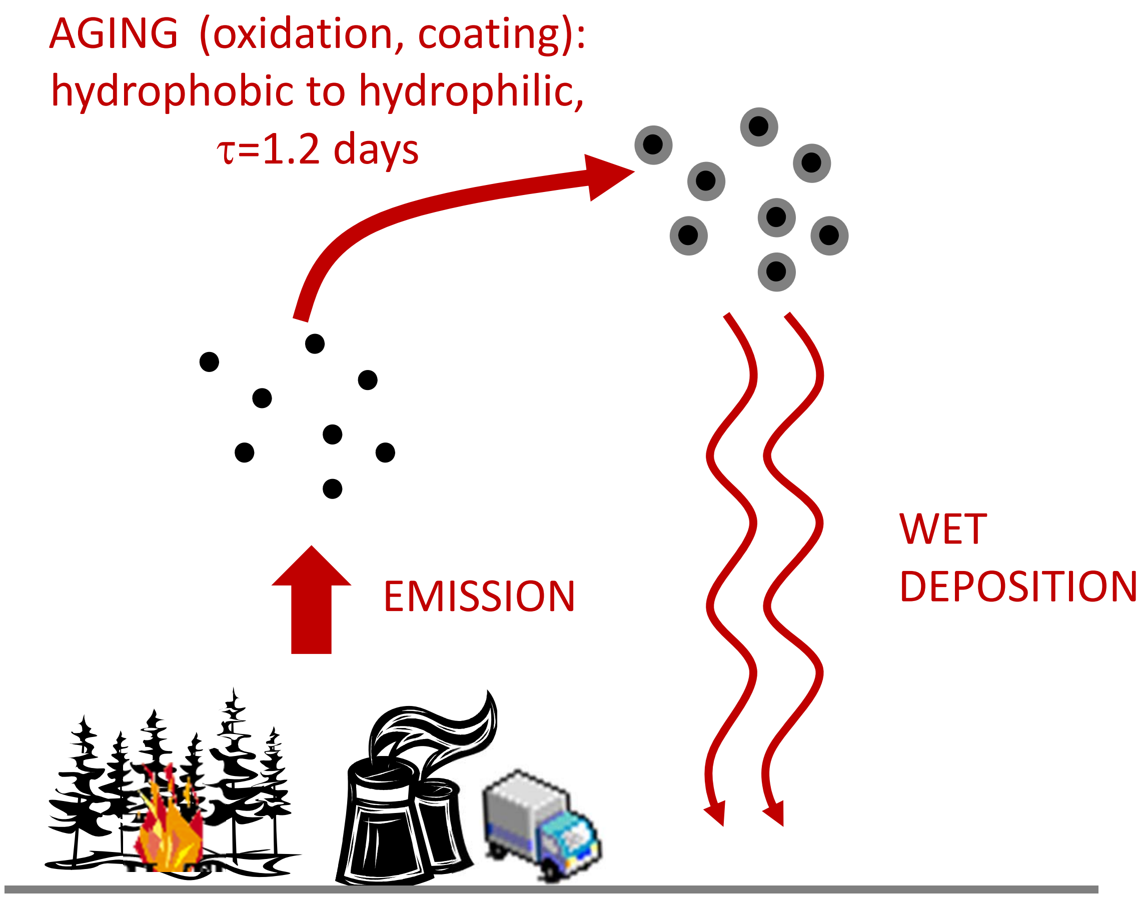


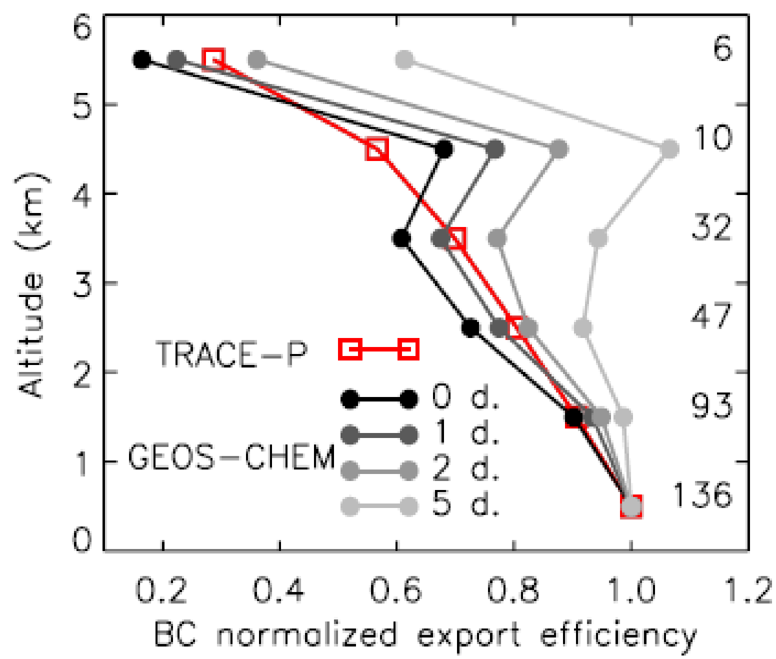
Higher hygroscopicity can lead to …

- 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)

Generic BC aging in global models*





Park et al. [2005]: ~1 day conversion gave best agreement with measured BC export efficiency

*including GEOS-Chem mass-based scheme

Aging in global models

AeroCom models	Energy Emis[1]	BB Emis[1]	Aging[2]	BC lifetime days	Ice/snow removal[3]	Mass median diameter of emitted particle[4]	BC density $g\,cm^{-3}$[6]	Refractive index at 550 nm[6]	MABS $m^2\,g^{-1}$[5,6]	References for aerosol module
GISS 99	B04	GFED	A	7.2	12%	0.08	1.6	1.56–0.5i	8.4	Koch et al. (2006, 2007), Miller et al. (2006)
ARQM 99	C99	L00, L96	I	6.7	T	0.1	1.5		4.1	Zhang et al. (2001); Gong et al. (2003)
CAM	C99	L96	A		L		X	X	X	Collins et al. (2006)
DLR	CW96	CW96	I		5% accum, strat	0.08, 0.75 FF 0.02, 0.37 BB	X	X	X	Ackermann et al. (1998)
GOCART	C99	GFED, D03	A	6.6	T	0.078	1.0	1.75–0.45i	10.0	Chin et al. (2000, 2002), Ginoux et al. (2001)
SPRINTARS	NK06	NK06	BCOC		L	0.0695 FF, 0.1 others	1.25	1.75–0.44i	2.3	Takemura et al. (2000, 2002, 2005)
LOA B	B04	GFED	A	7.3	LI	0.0118	1.0	1.75–0.45i	8.0 #	Boucher and Anderson (1995); Boucher et al. (2002); Reddy and Boucher (2004); Guibert et al. (2005)
LSCE	G03	G03	A	7.5	L	0.14	1.6	1.75-0.44i	3.5 (4.4 #)	Claquin et al. (1998, 1999); Guelle et al. (1998a, b, 2000); Smith and Harrison (1998); Balkanski et al. (2003); Bauer et al. (2004); Schulz et al. (2006)
MATCH	L96	L96	A		L	0.1	X	X	X	Barth et al. (2000); Rasch et al. (2000, 2001)
MOZGN	C99, O96	M92	A		L	0.1	1.0	1.75–0.44i	8.7	Tie et al. (2001, 2005)
MPIHAM	D06	D06	I #	4.9	S	0.069 (FF, BF) 0.172 (BB)	2.0	1.75–0.44i	7.7 #	Stier et al. (2005)
MIRAGE	C99	CW96, L00	I #	6.1	L	0.19, 0.025	1.7	1.9-0.6i	3 aitk, 6 acc	Ghan et al. (2001); Easter et al. (2004); Ghan and Easter (2006)
TM5	D06	D06	A	5.7	20%	0.034	1.6	1.75-0.44i	4.3	Metzger et al. (2002a, b)
UIOCTM	C99	CW96	A	5.5	L	0.1 (FF), 0.295, 0.852 (BB)	1.0	1.55-0.44i	7.2 #	Grini et al. (2005); Myhre et al. (2003); Berglen et al. (2004); Berntsen et al. (2006)
UIOGCM 99	IPCC	IPCC	I #	5.5	none	0.0236–0.4	2.0	2.0–1.0i	10.5 #	Iversen and Seland (2002); Kirkevag and Iversen (2002); Kirkevag et al. (2005)
UMI	L96	P93	N	5.8	L	0.1452 (FF), 0.137 (BB)	1.5	1.80–0.5i	6.8 #	Liu and Penner (2002)
ULAQ 99	IPCC	IPCC	A	11.4	L	0.02–0.32	1.0	2.07–0.6i	7.5 #	Pitari et al. (1993, 2002)

Koch et al. 2009

N = no aging

A = aging at a fixed lifetime

I = aging with coagulation and condensation

= aging affects optical properties

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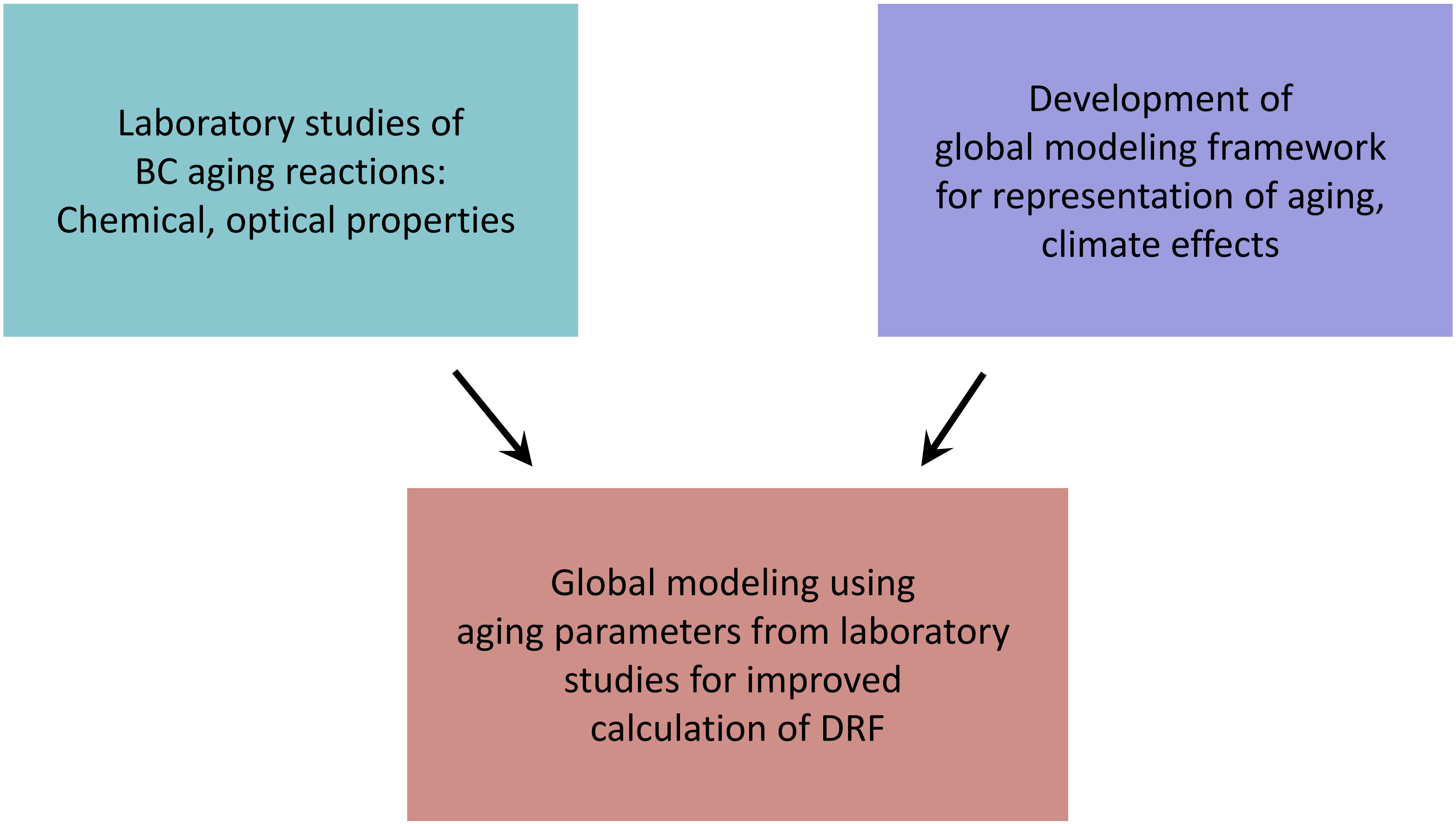
An incomplete understanding of this aging, and/or representation of this aging within models, may explain some fraction of the model-measurement discrepancies.

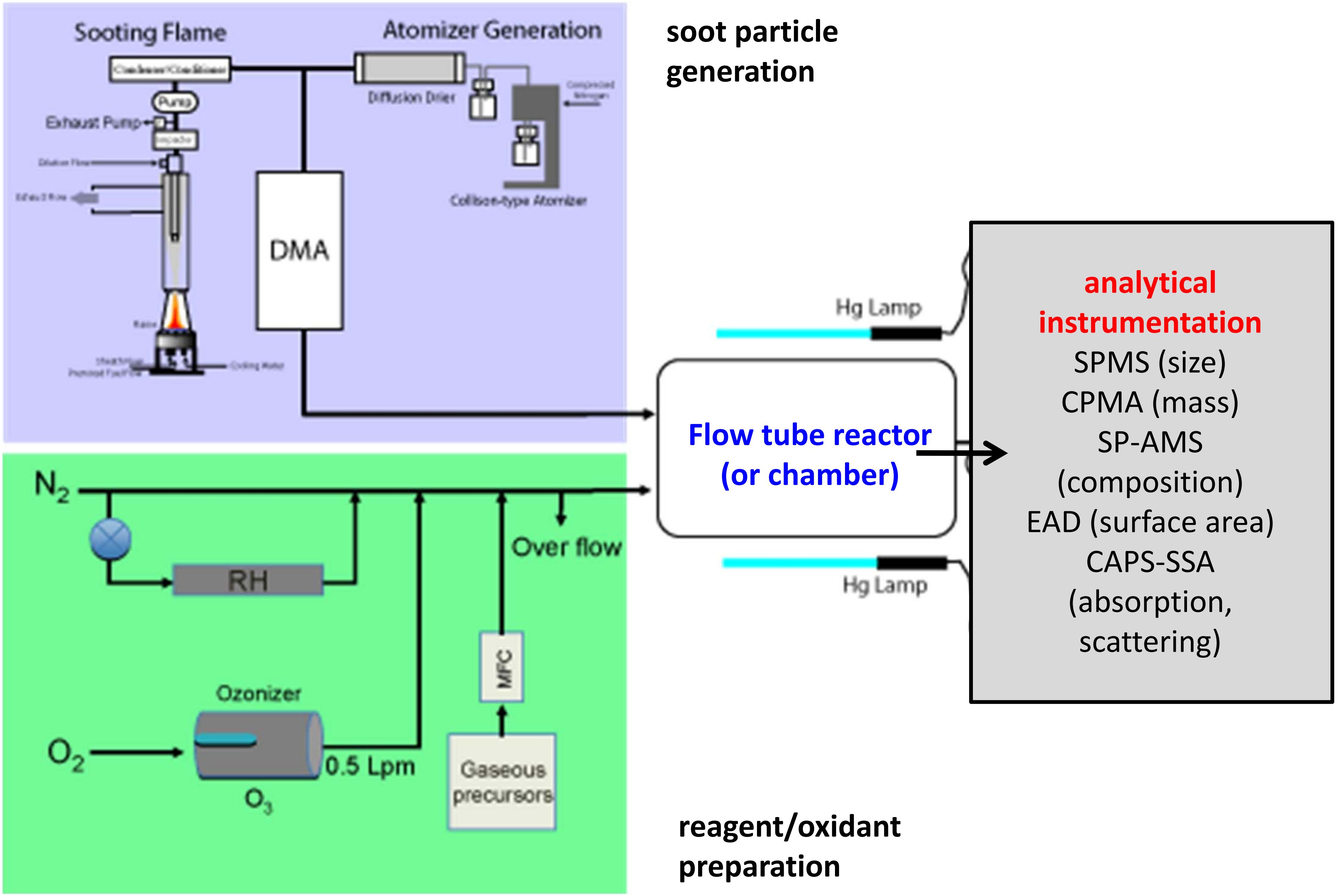
Major questions:

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

Approach: Laboratory + Modeling



Next steps



soot particle generation

reagent/oxidant preparation

Experimental matrix

Based upon "BC²" intercomparison study, 318 runs in 1 month *[Cross et al. 2010]*

BC source

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

Particle size

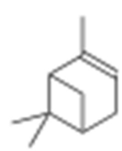
- monodisperse, 30-300 nm

Relative humidity

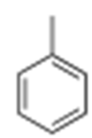
- controlled after reactor, but before instruments (multiplex of 0%, 30%, 60%, 90%)

Aging type

- heterogeneous oxidation (OH, O_3)
- coated with sulfuric acid (SO_2)
- coated with SOA (fresh, aged)
- ammonium sulfate/nitrate
- mixed coatings



α-pinene
SOA



toluene
SOA

Parameterization

- Calculation of radiative forcing in models requires knowledge of key optical parameters as a function of particle properties
- This will be done by construction of 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

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

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

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

- Next steps: Laboratory results → implementation in models

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