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

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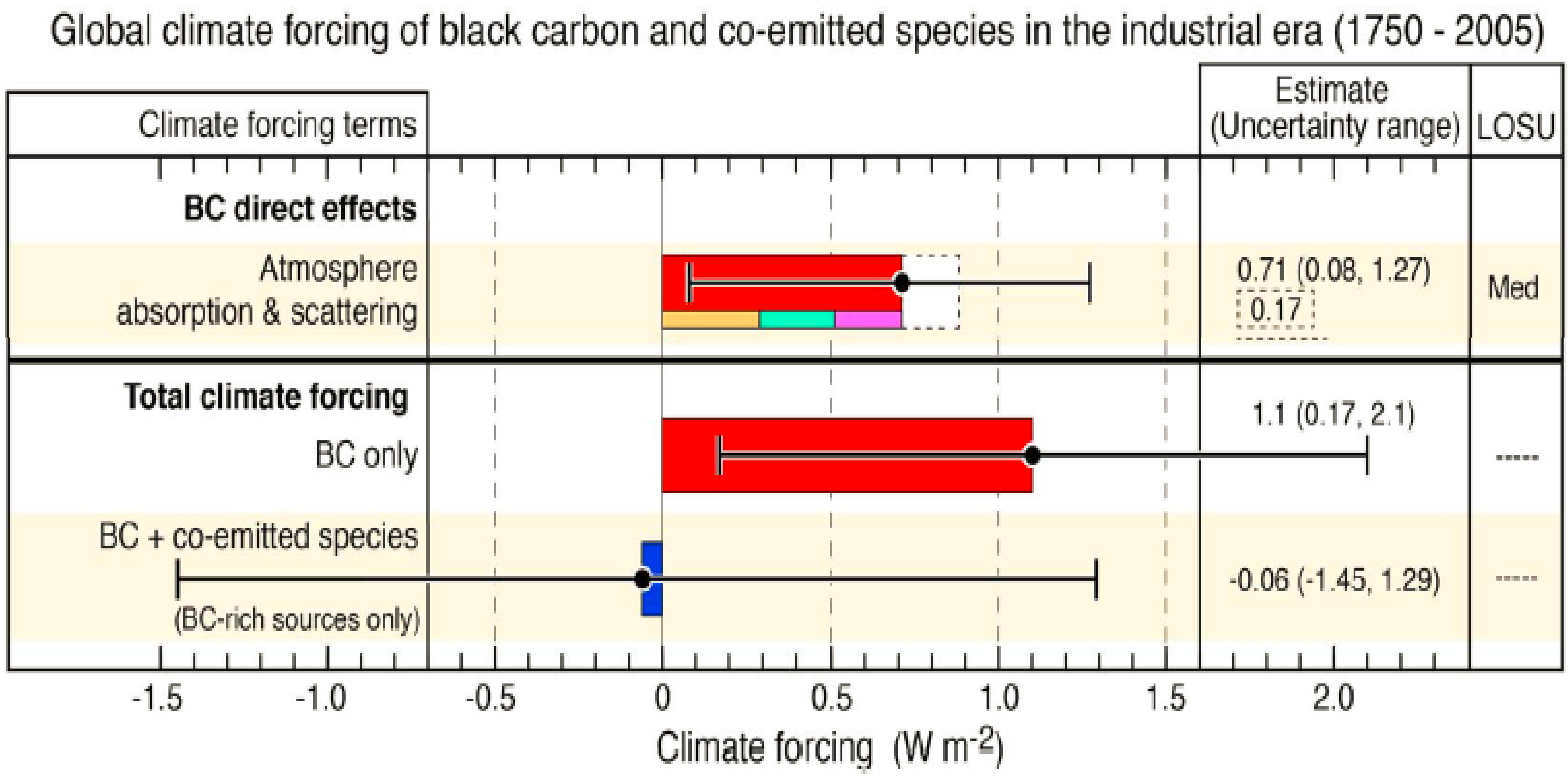
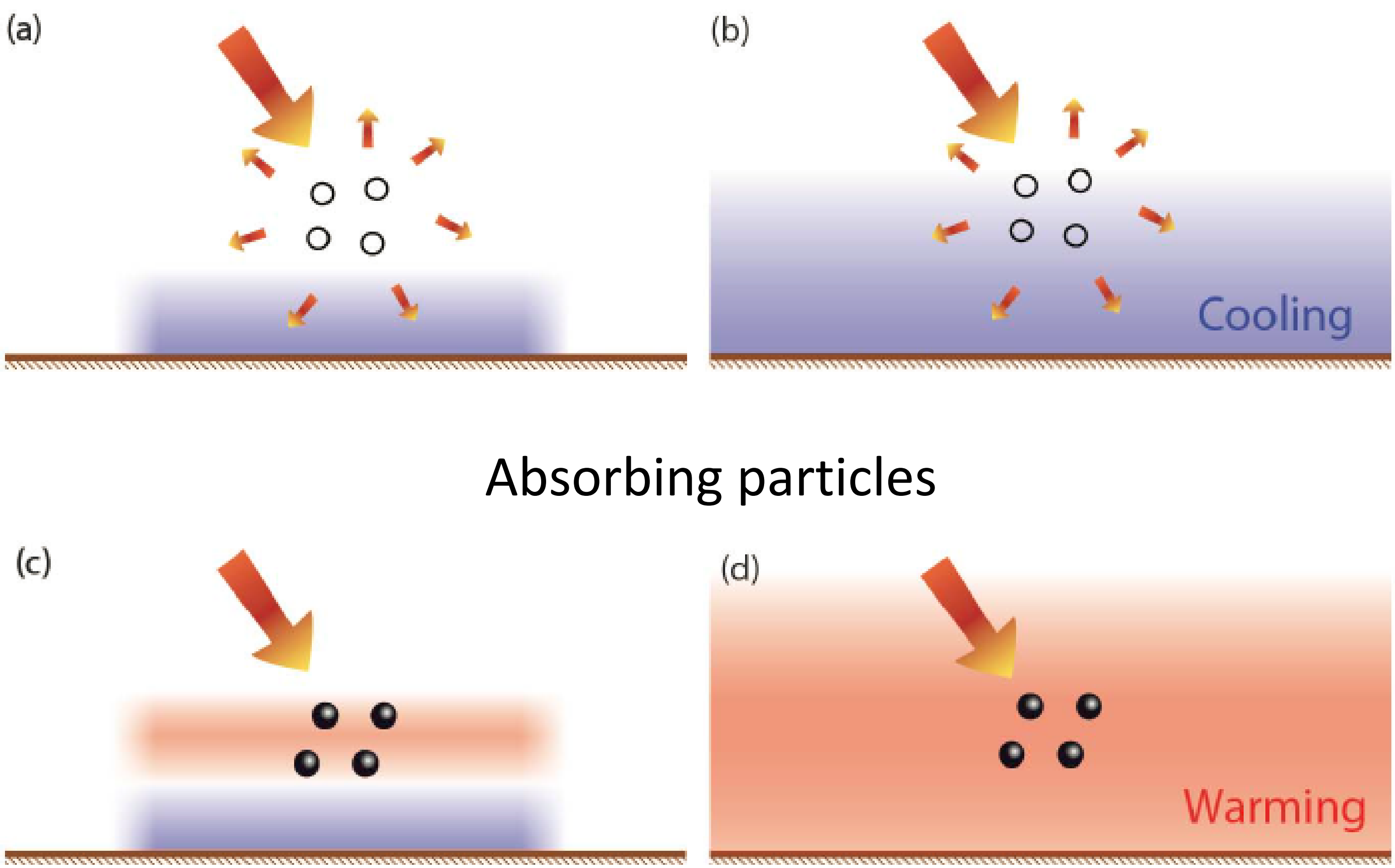
BC climate forcing: Large, complex, uncertain



[Bond et al., 2013]

Focus on direct radiative effect

Scattering particles

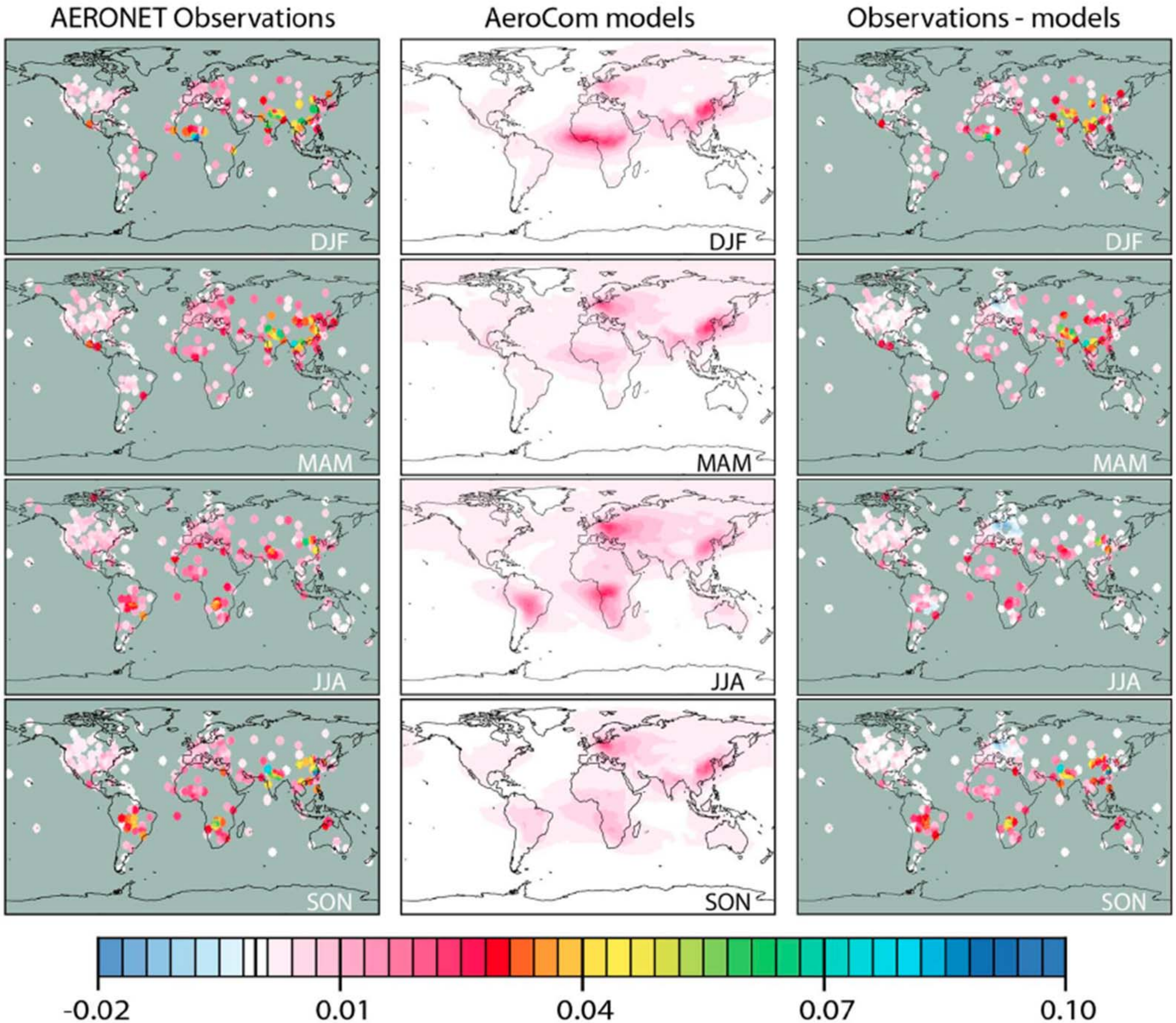


Absorbing particles

Local effect *Distributed effect*

IPCC, Fifth Assessment Report, 2013.

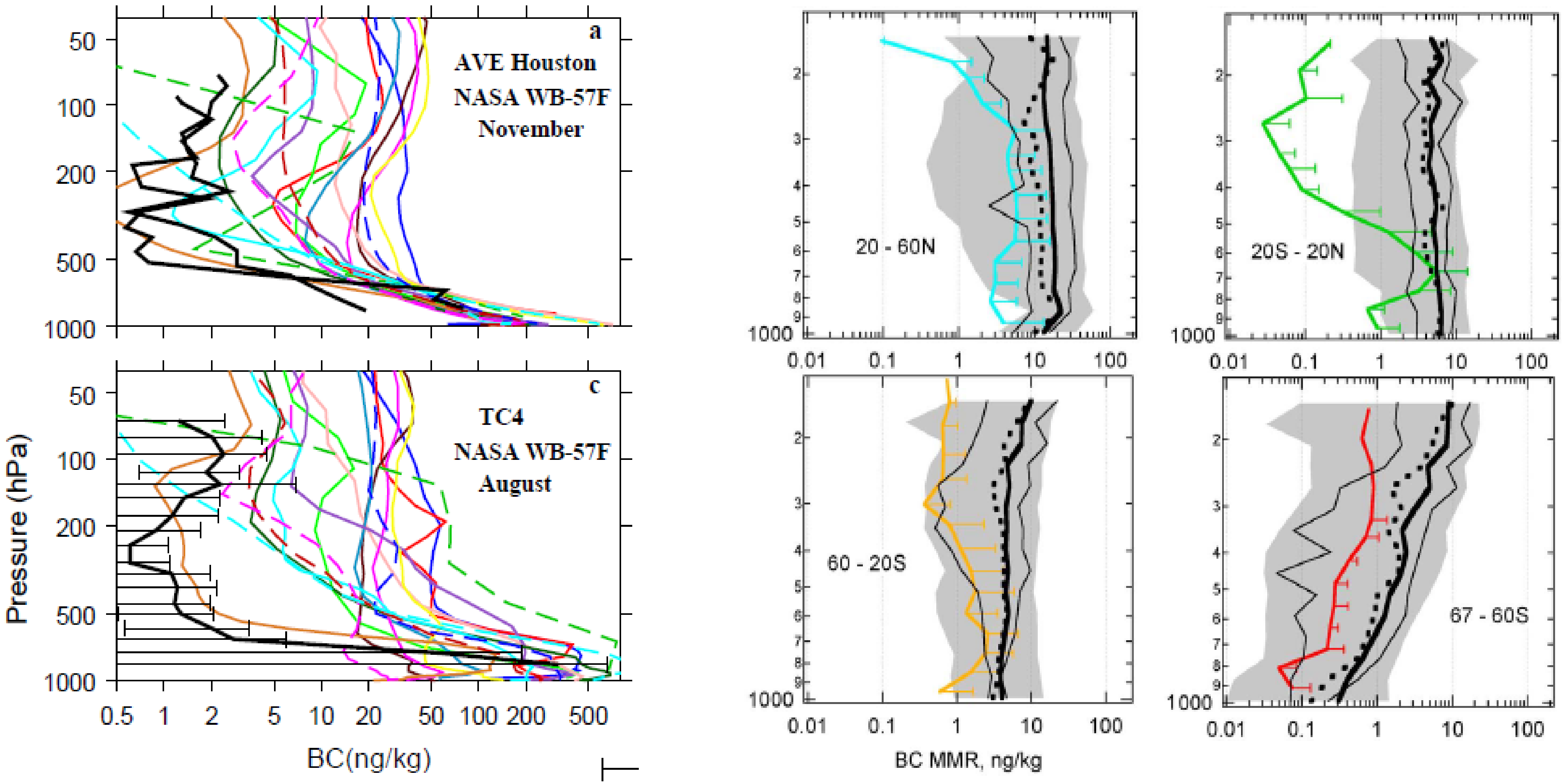
(1) Global models *underestimate* absorption AOD



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]

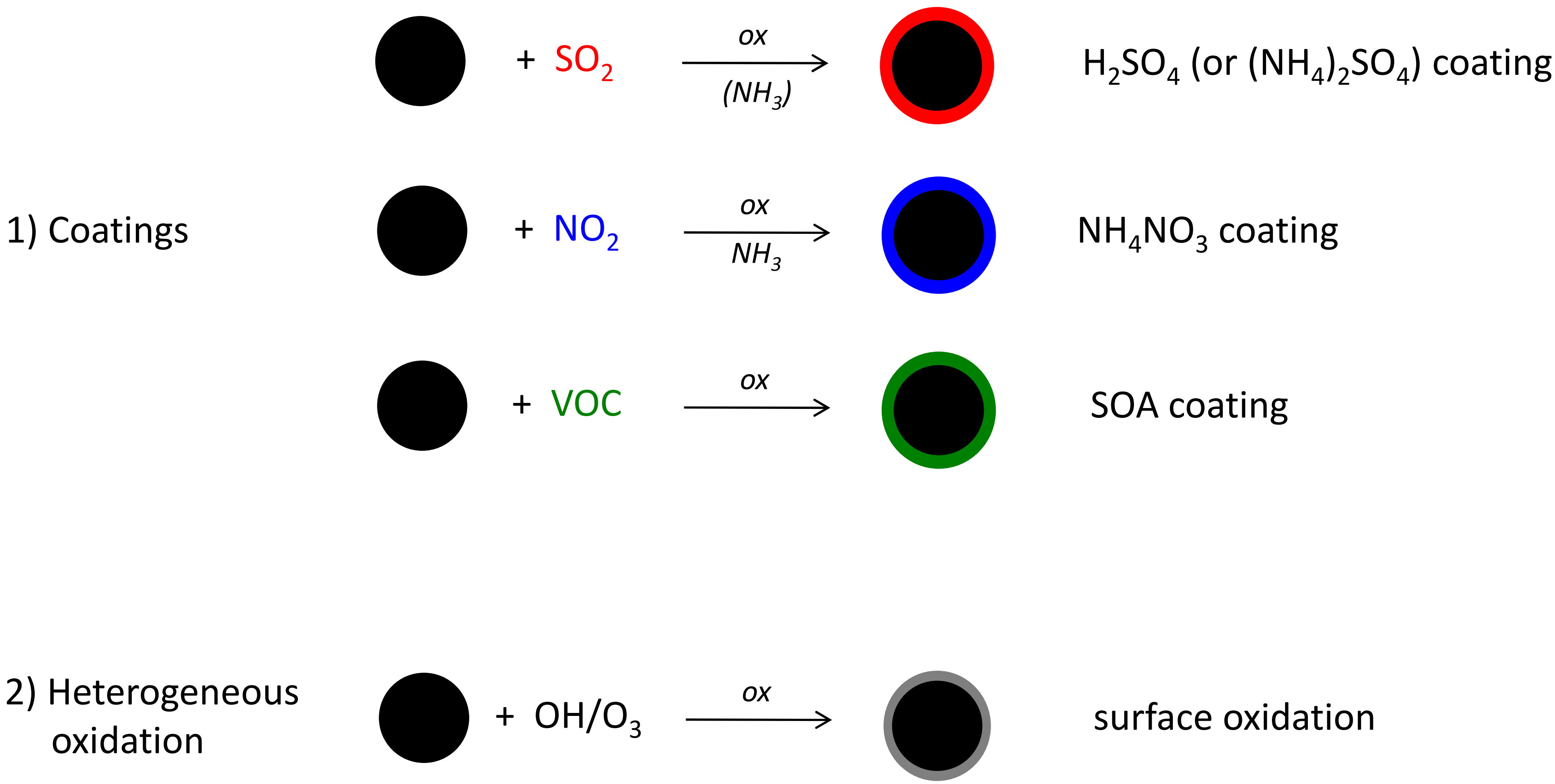
[Schwarz et al., 2010]

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.

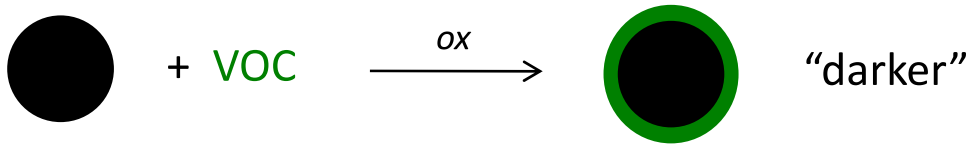
Key BC aging reactions



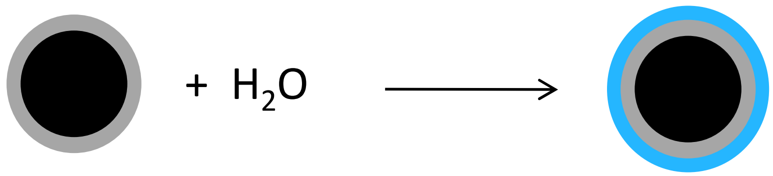
Effects of aging on BC properties

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

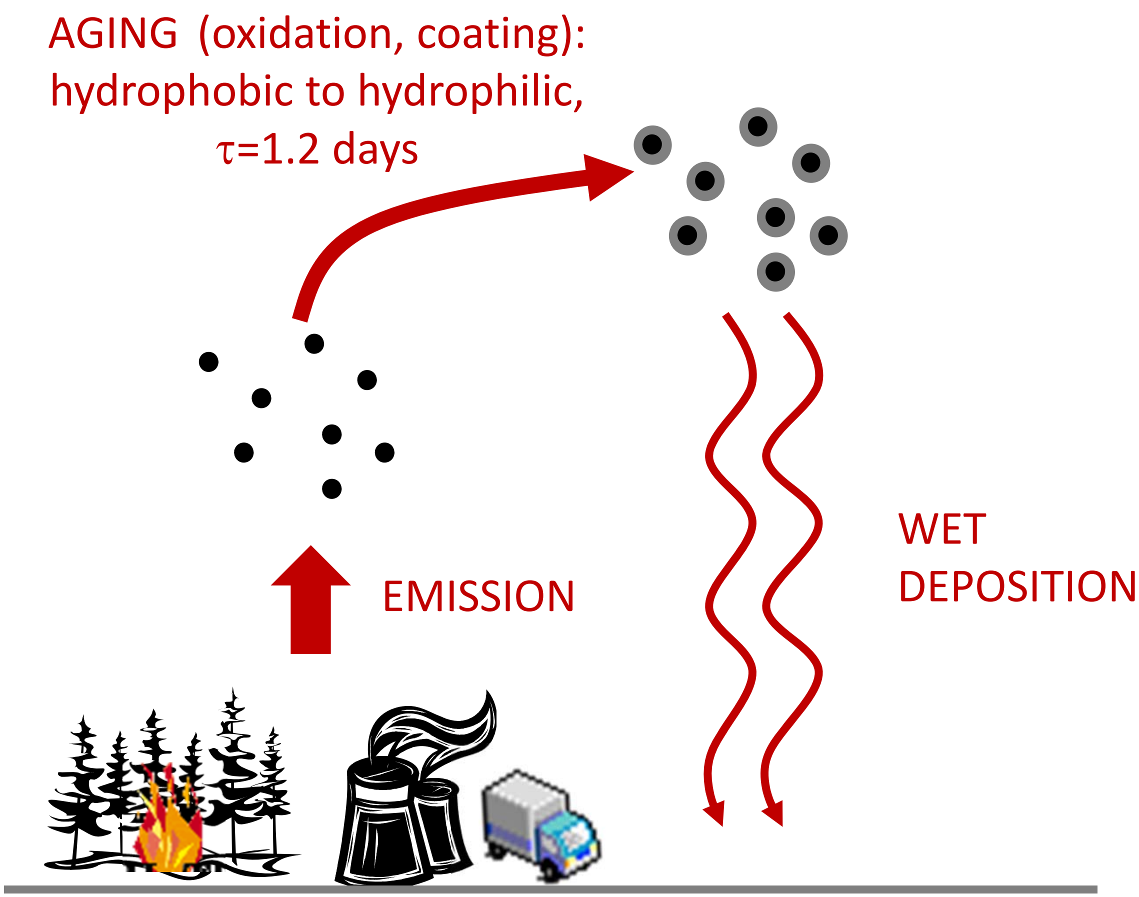


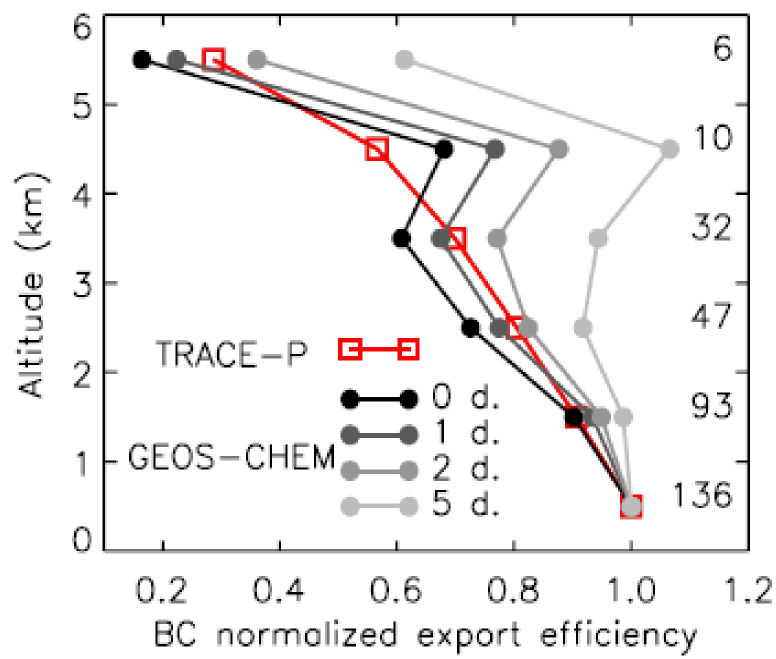
Higher hygroscopicity can lead to …

- 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

Koch et al. 2009

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)

N = no aging

A = aging at a fixed lifetime

I = aging with coagulation and condensation

= aging affects optical properties

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.

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?

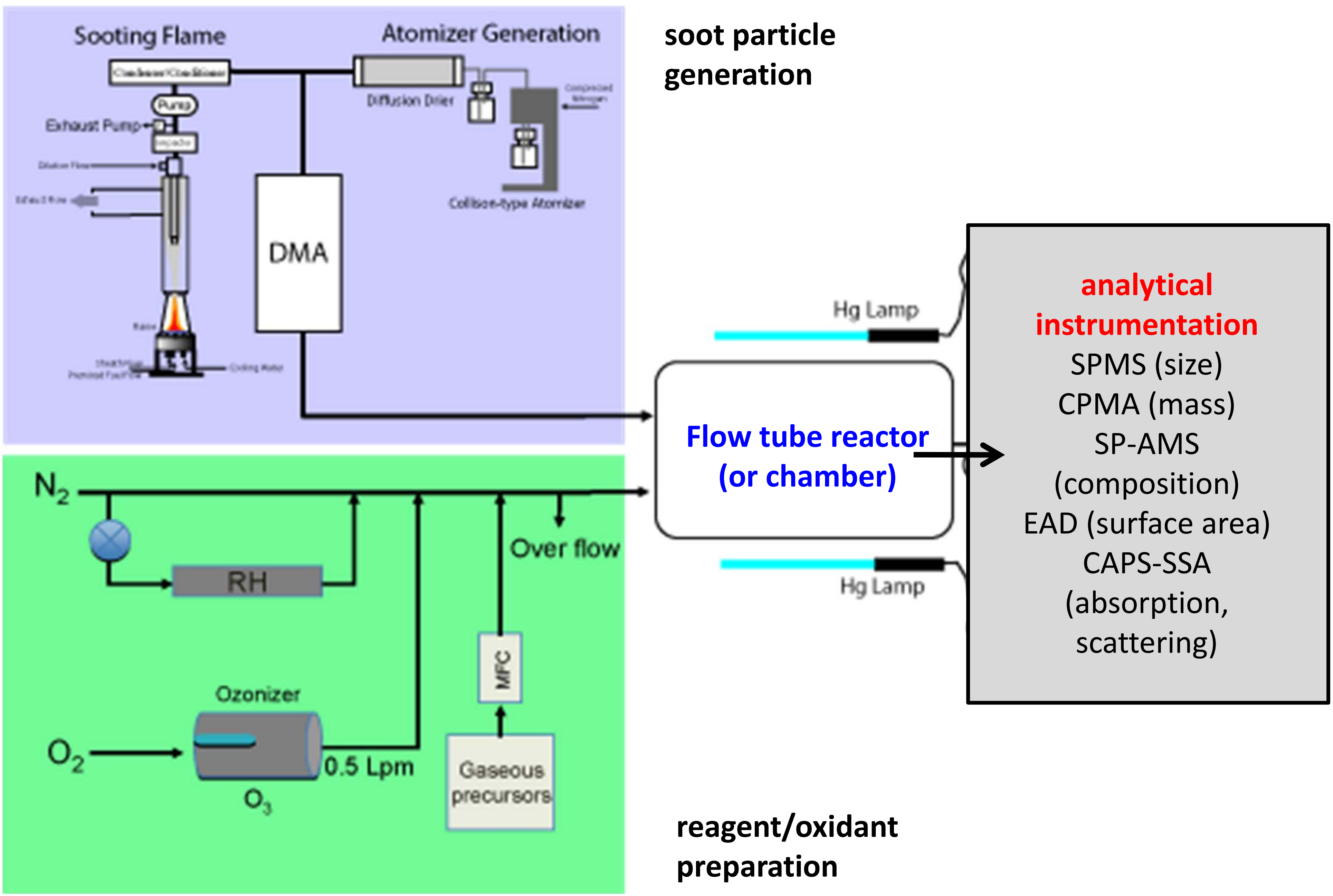
Approach: Laboratory + Modeling

Laboratory studies of
BC aging reactions:
Chemical, optical properties

Development of
global modeling framework
for representation of aging,
climate effects

Global modeling using
aging parameters from laboratory
studies for improved
calculation of DRF

Next steps



Sooting Flame
Atomizer Generation
Pump
Exhaust Pump
Diffusion Drier
Collison-type Atomizer
DMA
soot particle generation
Hg Lamp
Flow tube reactor (or chamber)
Hg Lamp
analytical instrumentation
SPMS (size)
CPMA (mass)
SP-AMS (composition)
EAD (surface area)
CAPS-SSA (absorption, scattering)
N2
RH
Over flow
MFC
Ozonizer
O2
0.5 Lpm
O3
Gaseous precursors
reagent/oxidant preparation

Experimental matrix

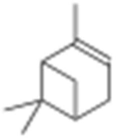
Based upon "BC^2" intercomparison study, 318 runs in 1 month *[Cross et al. 2010]*

BC source
- fractal soot from McKenna burner (denuded at 300°C)
- also atomized black carbon spheres

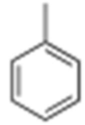
Particle size
- monodisperse, 30-300 nm

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

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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