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

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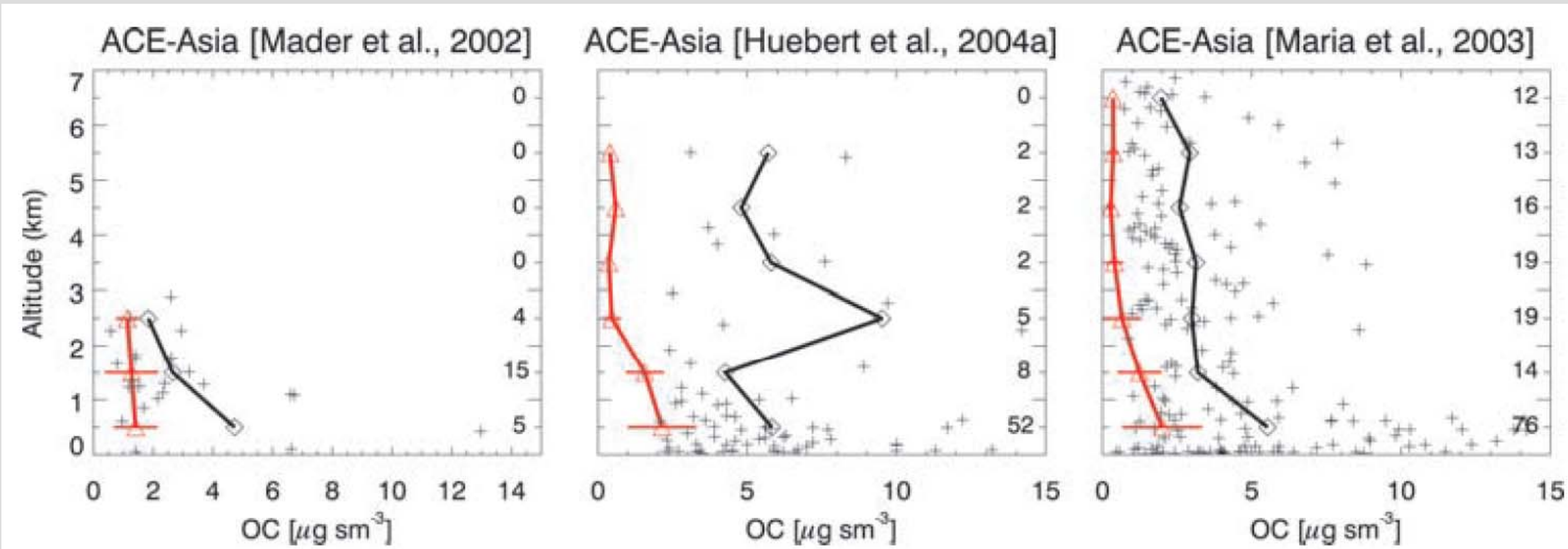
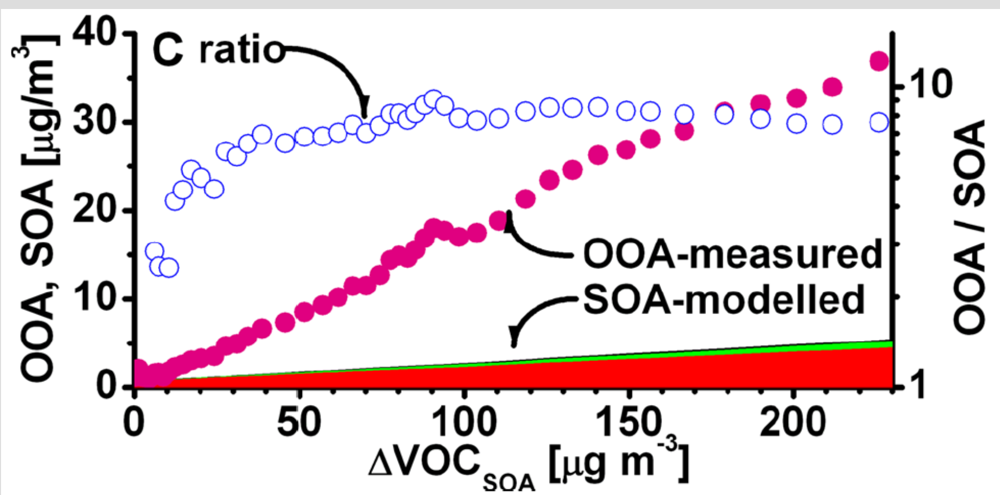
Organic aerosol (circa 2005)

Discrepancies in mass loadings between modeled, ambient OA:



Heald et al. 2005

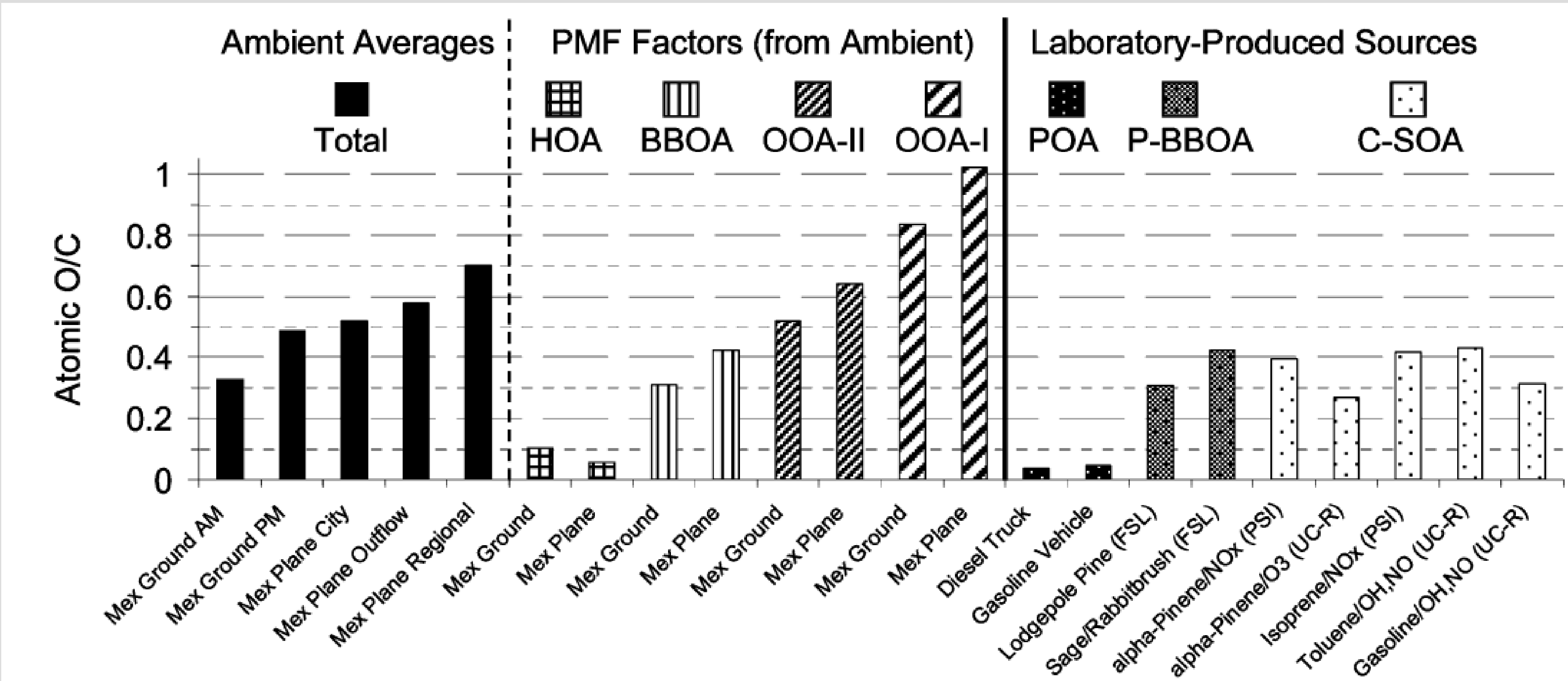


Volkamer et al. 2006

also: de Gouw et al. 2005, Kleinman et al. 2008, etc...

Organic aerosol, cont.

Discrepancies in chemical composition (degree of oxidation) between lab, ambient OA:

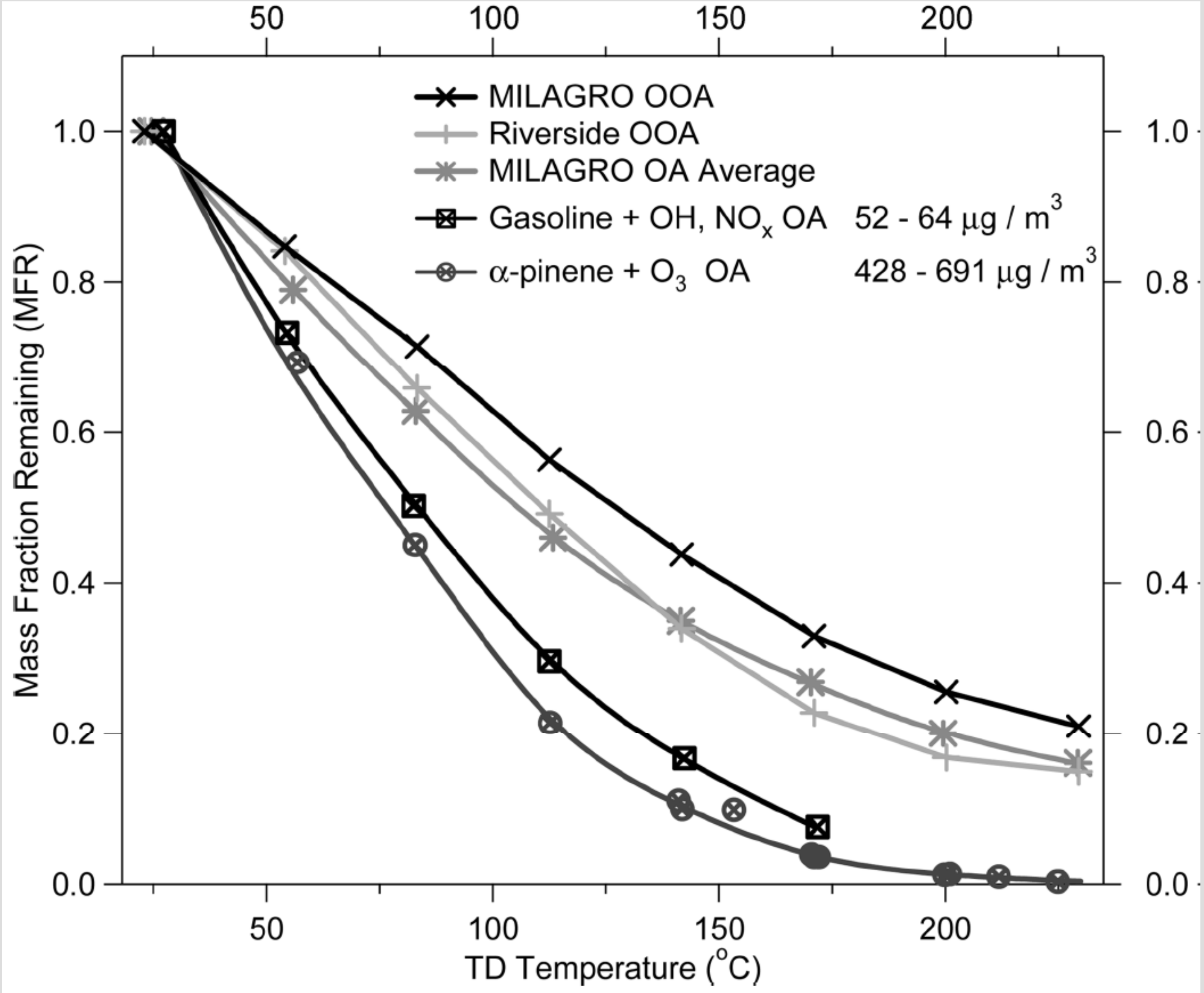


Aiken et al. 2008

also: Bahreini et al. 2005, Heald et al. 2010, Kroll et al. 2011, etc…

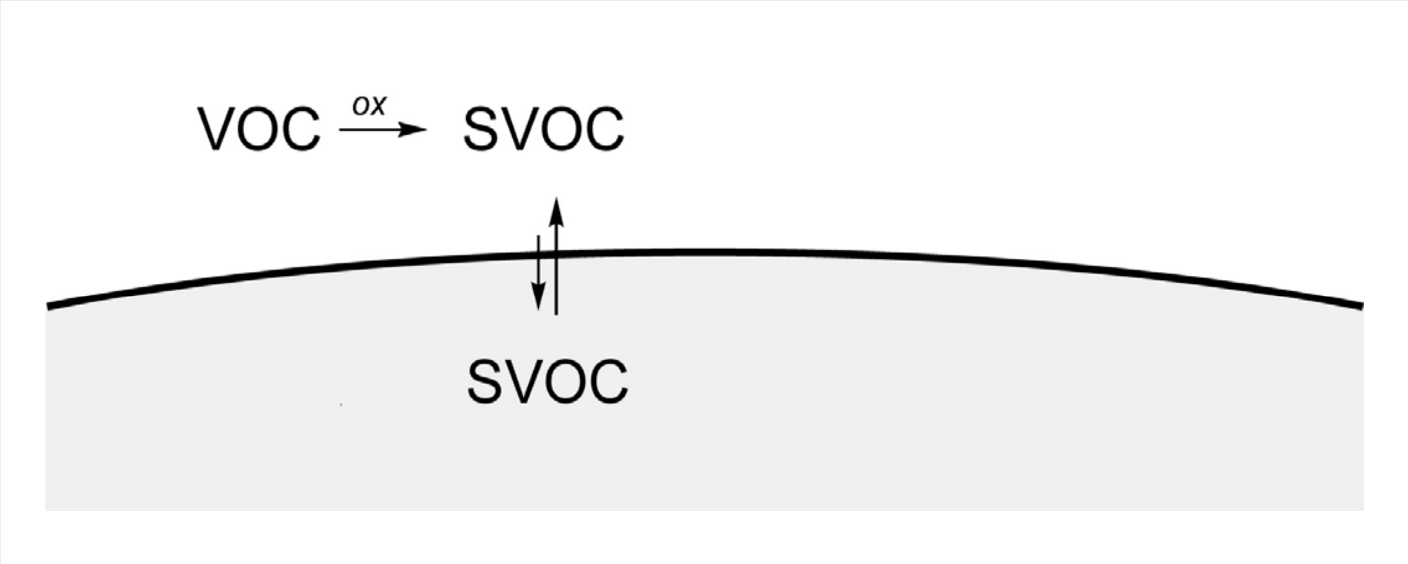
Organic aerosol, cont.

Discrepancies in key properties (volatility) between lab, ambient OA:



Huffman et al. 2009

“Aging” of organic aerosol



Robinson et al. 2007
Jimenez et al. 2009

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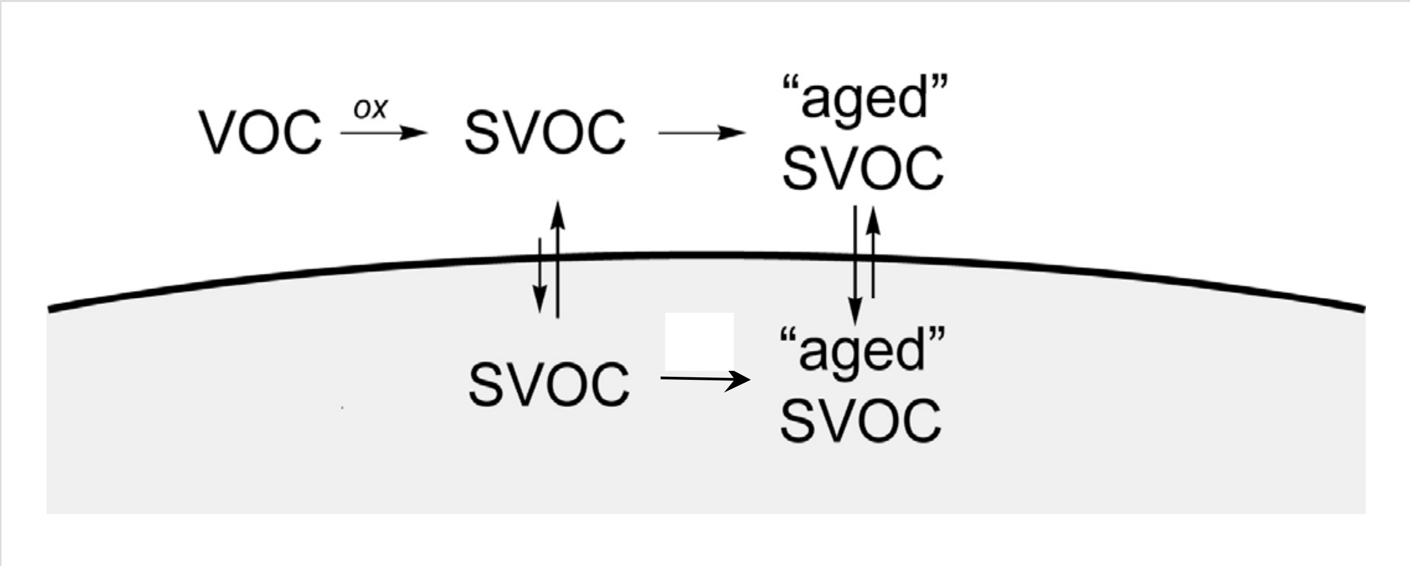

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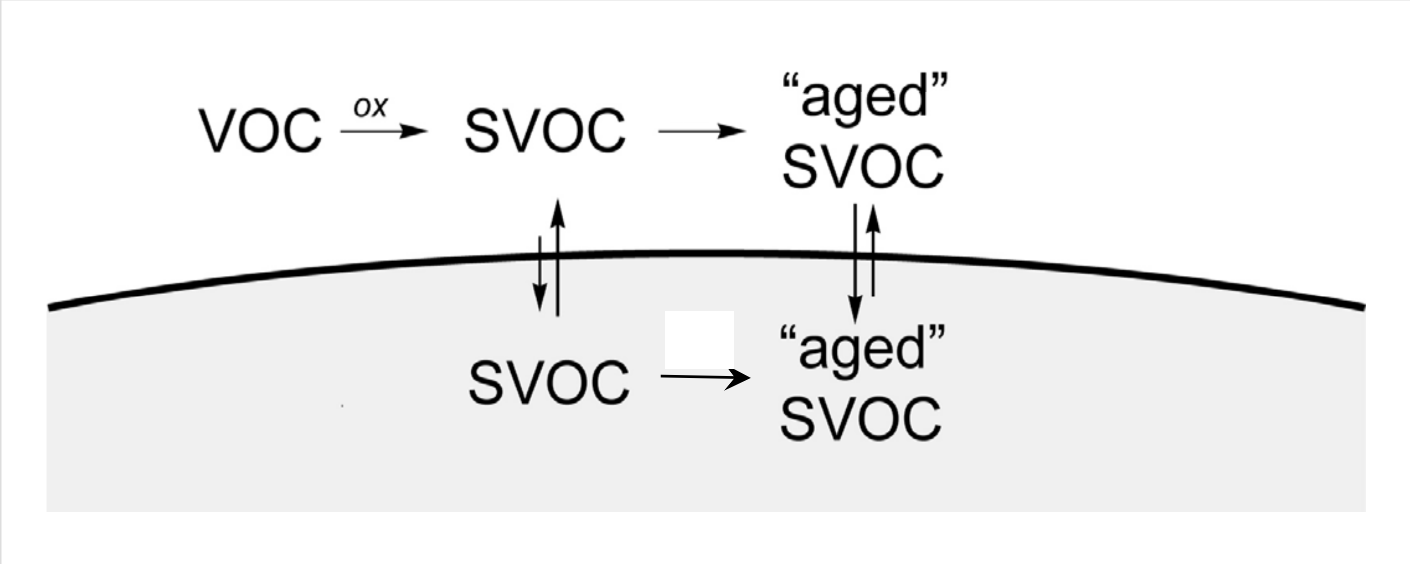
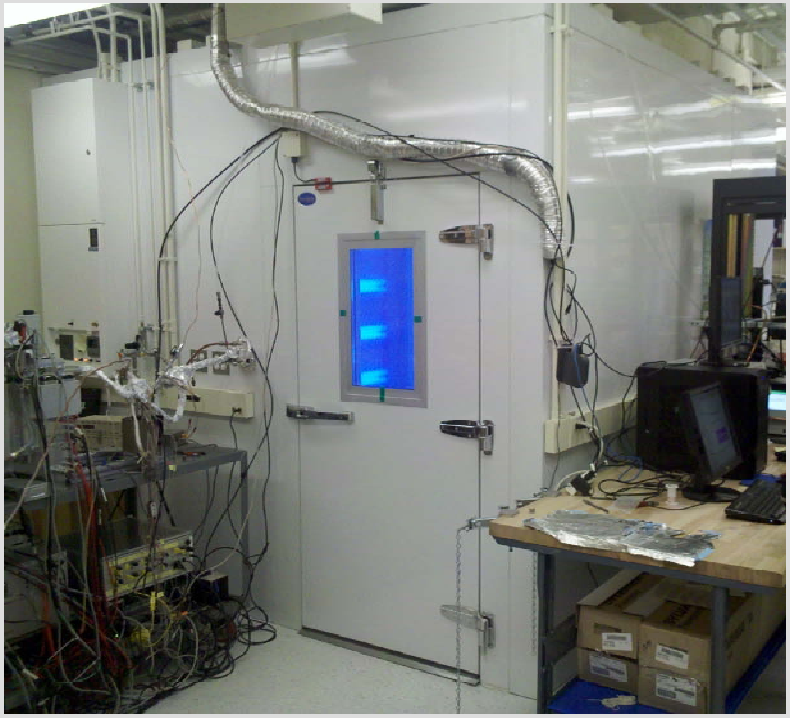


Robinson et al. 2007
Jimenez et al. 2009

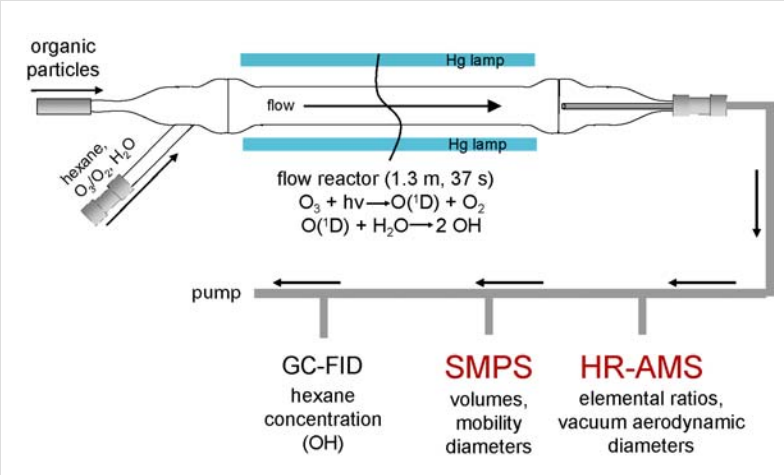
Aging of OA can describe a large number of processes:

- Oxidation reactions (gas- or condensed-phase)
- Nonoxidative chemical transformations
- Evaporation/condensation

Laboratory experiments of OA aging

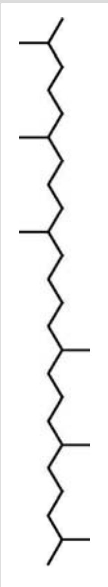


1) environmental chamber

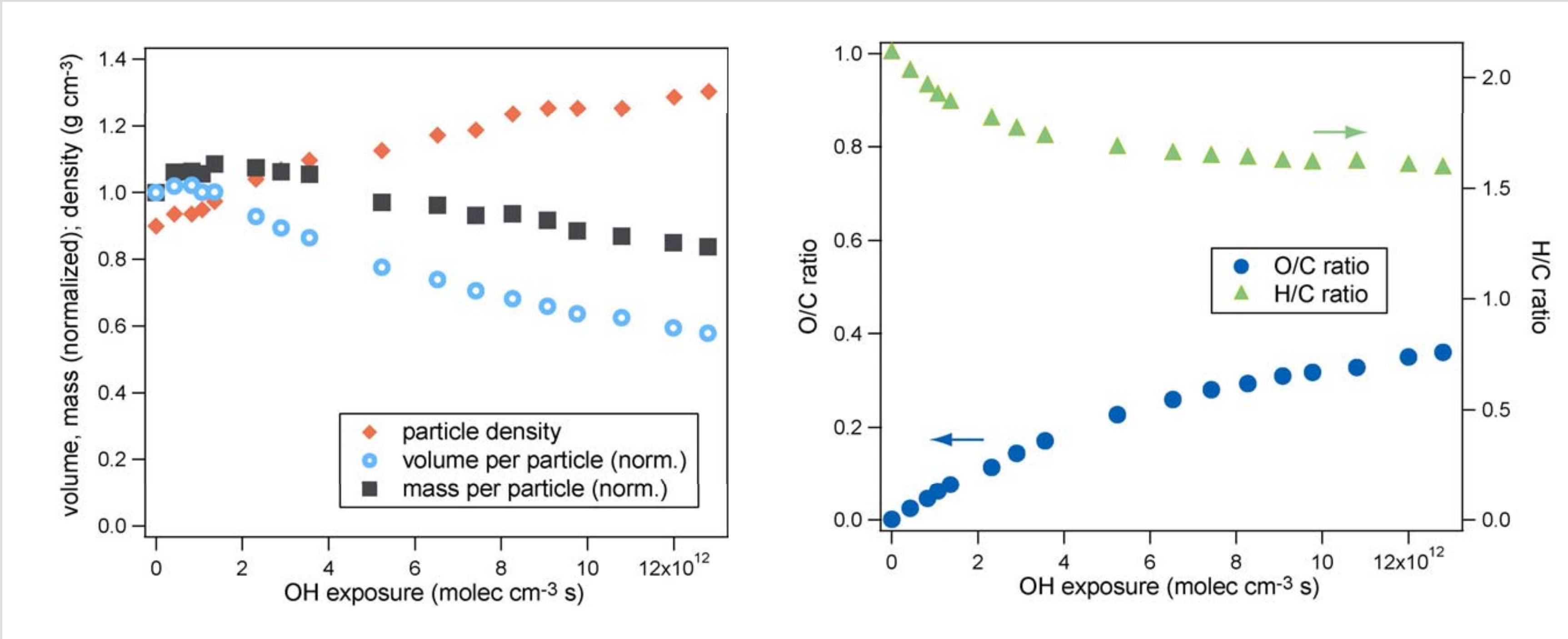


2) flow tube reactor

Oxidative aging of OA: Lab studies

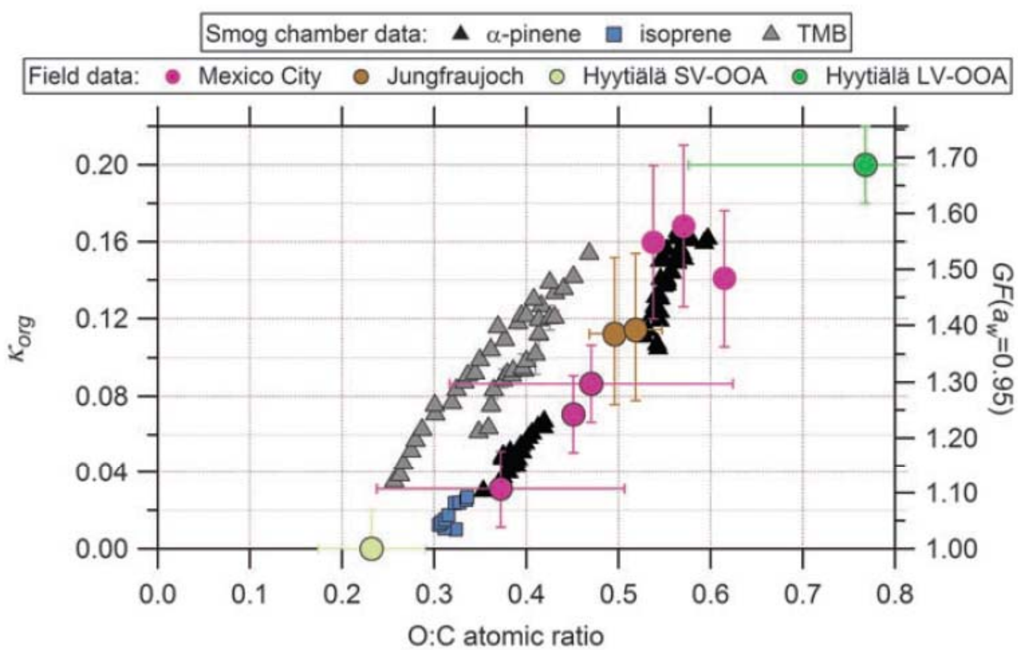


squalane
$C_{30}H_{62}$
+ OH



Oxidative aging: Effects on OA properties

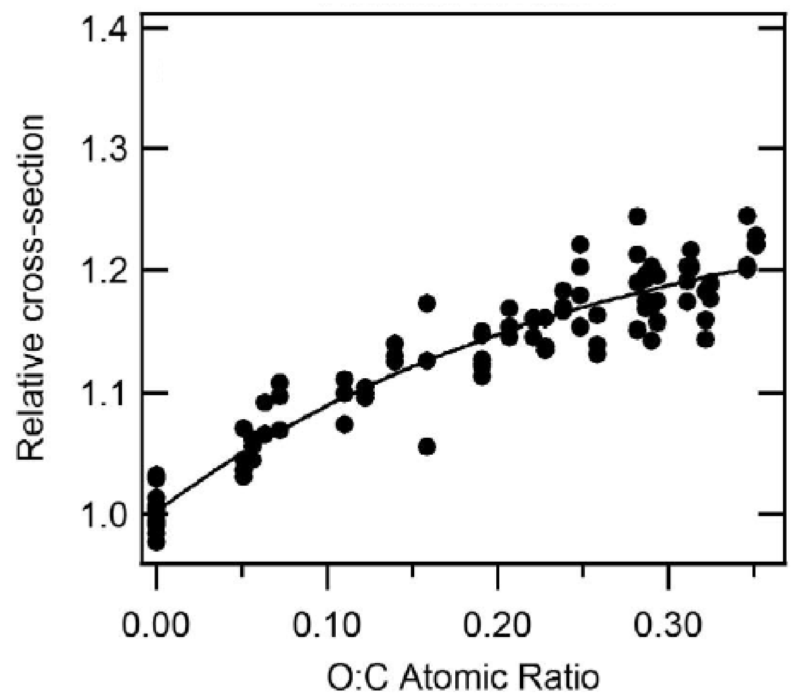
Water uptake



Jimenez et al. 2009

also: Chang et al. 2010,
Massoli et al. 2010,
Cappa et al. 2011, etc.

Optical properties



Cappa et al. 2011

This project: Aging of black carbon aerosol

Hypothesis: *Similar to the emerging view of organic aerosol, black carbon aerosol is a chemically dynamic system, subject to dramatic changes in physicochemical properties, and therefore climate forcing effects, as a result of atmospheric aging.*

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Major questions:

- what are the most important atmospheric aging transformations of BC?
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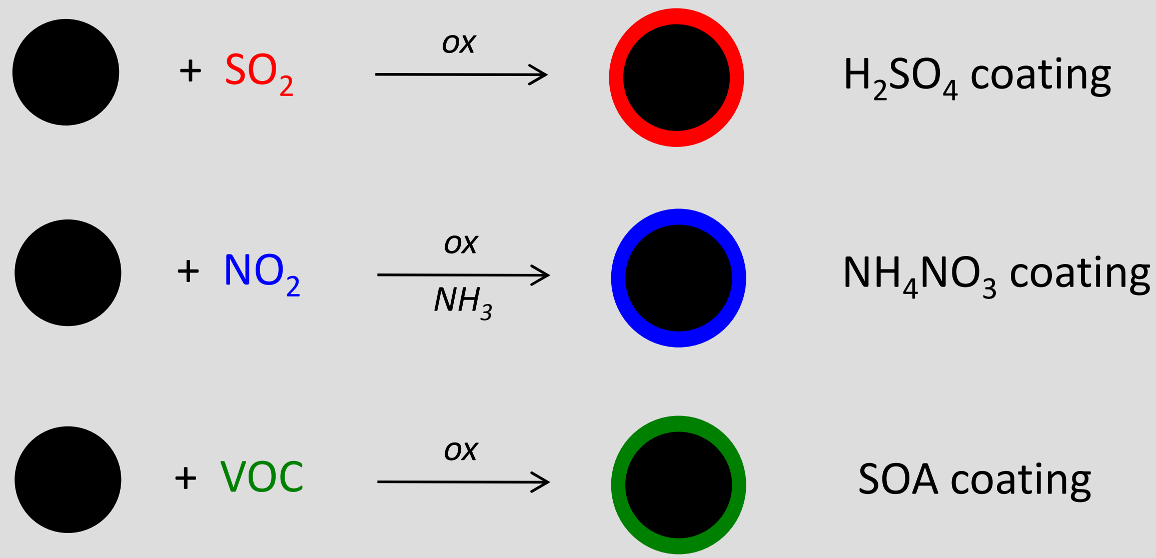
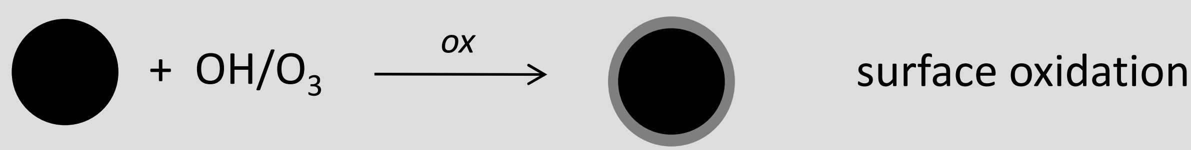
Key BC aging reactions

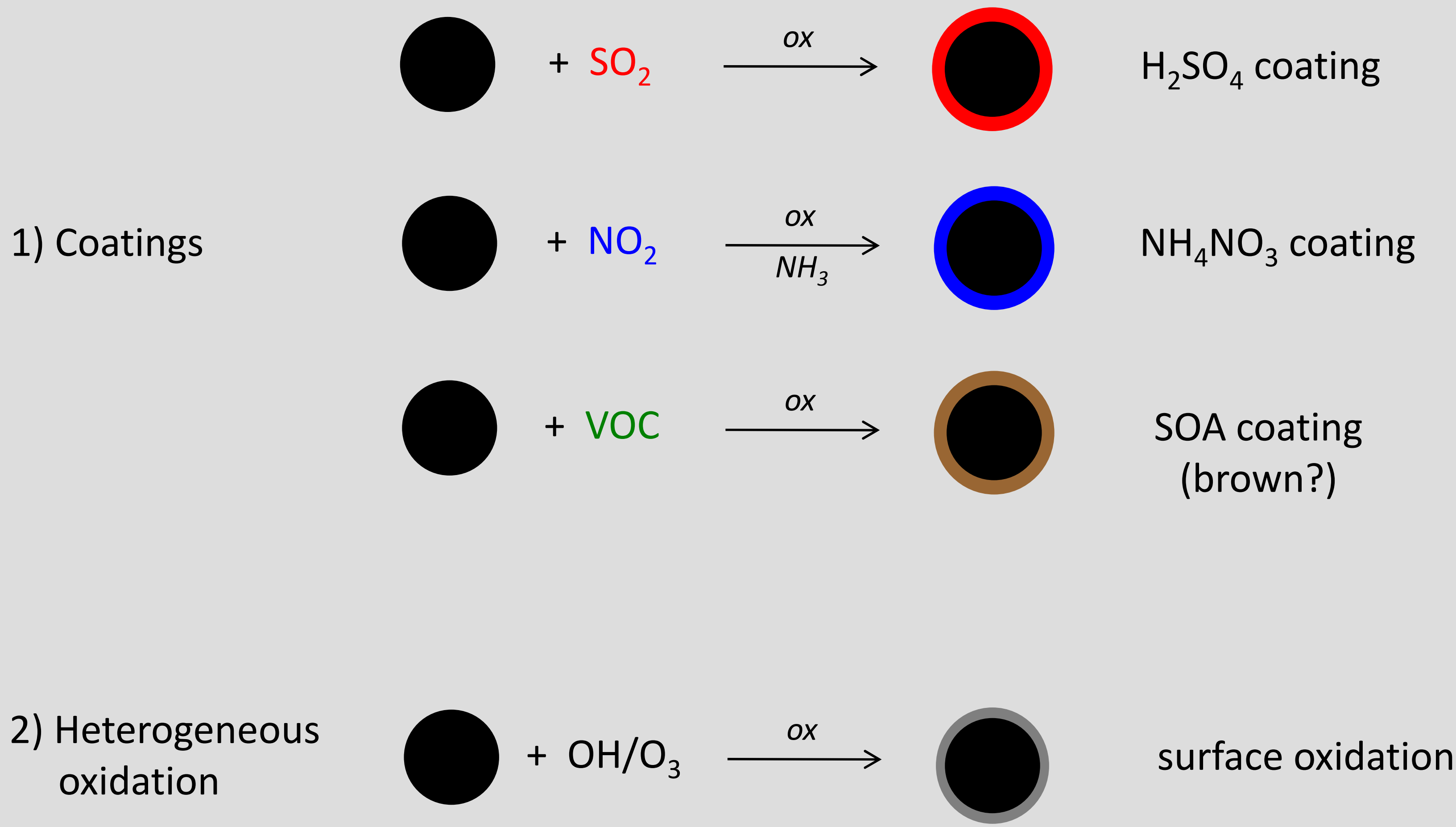
1) Coatings



2) Heterogeneous oxidation



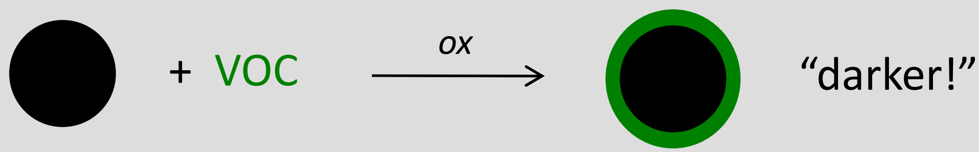
Key BC aging reactions



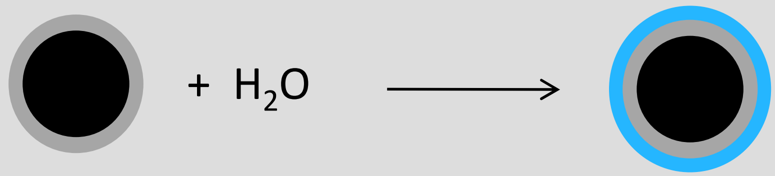
Effects of aging on BC properties

1) Enhancement of light absorption by coatings

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



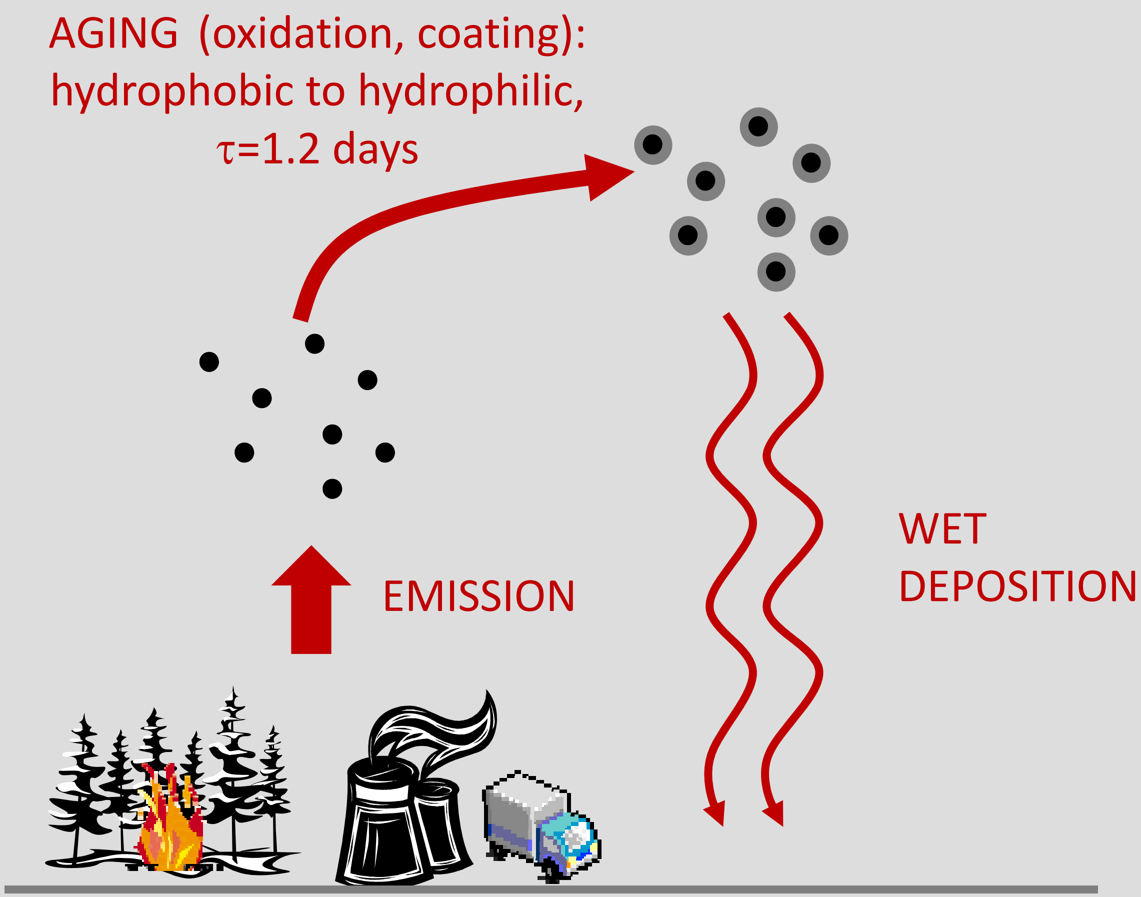
2) Increased water-uptake ability by coated or oxidized BC

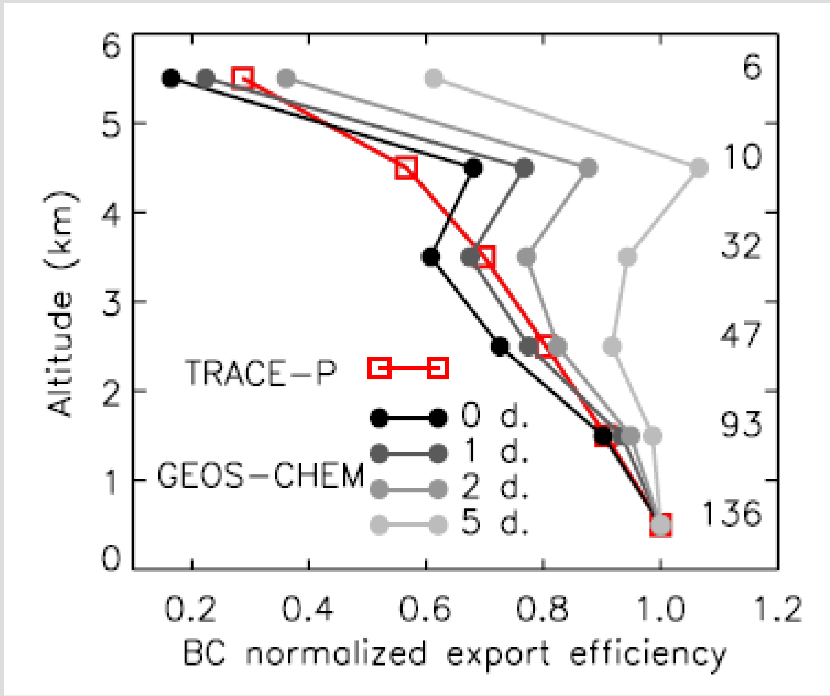


This additional hygroscopicity can potentially lead to …

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

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 to be used here

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

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Major questions:

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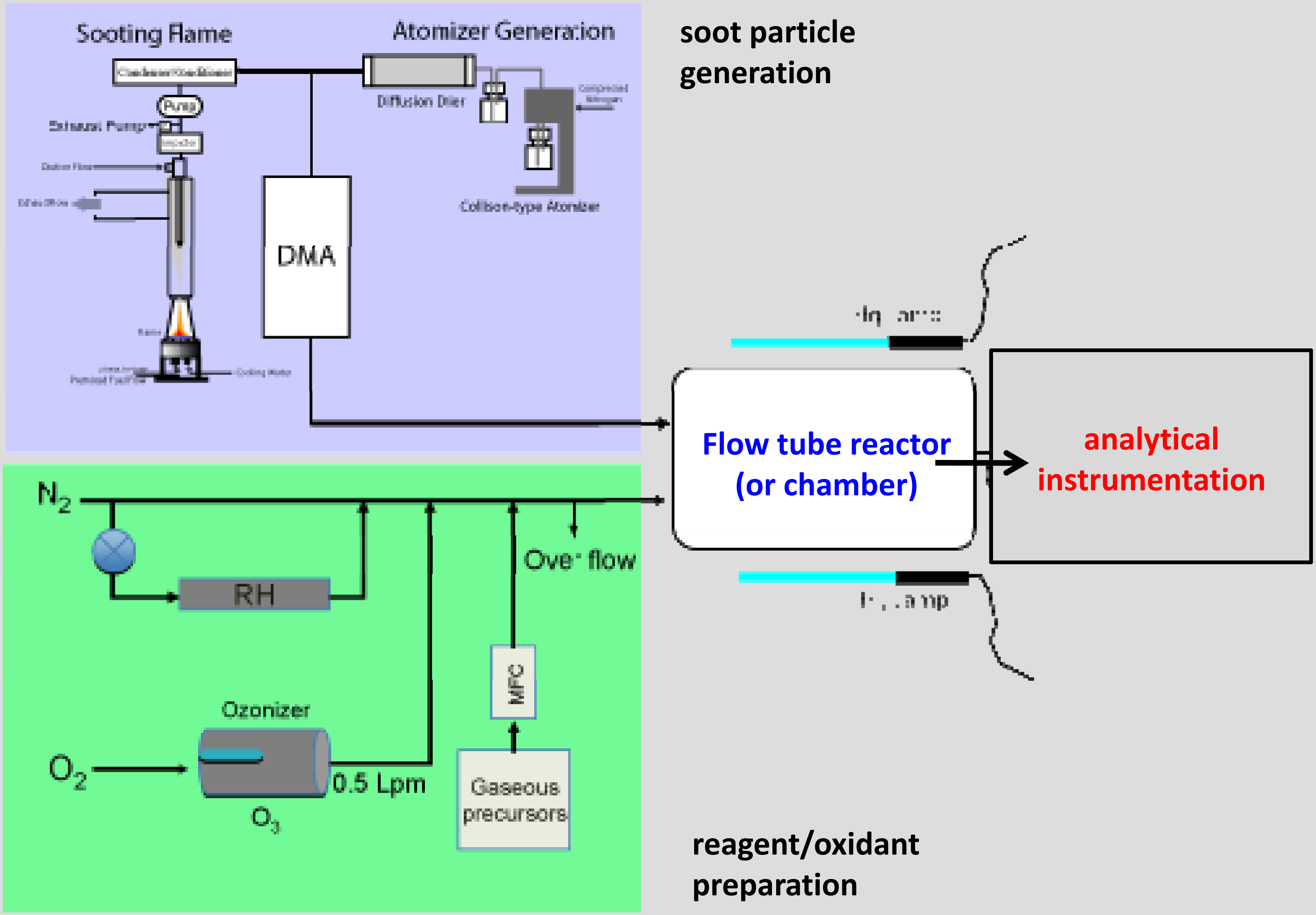
Approach:

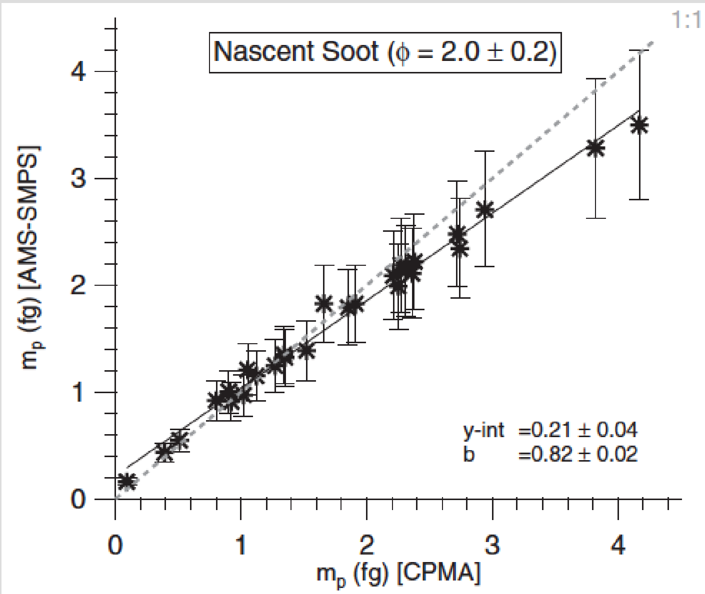
- detailed **laboratory studies** of individual aging reactions
- **parameterization of optical properties** as a function of size, RH, mixing state
- **incorporation into a global model** framework for calculation of direct radiative forcing

Laboratory setup

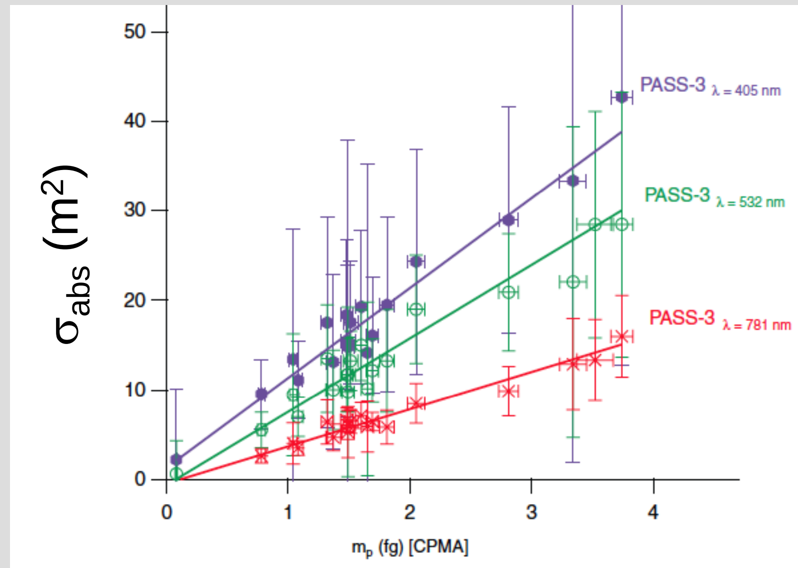


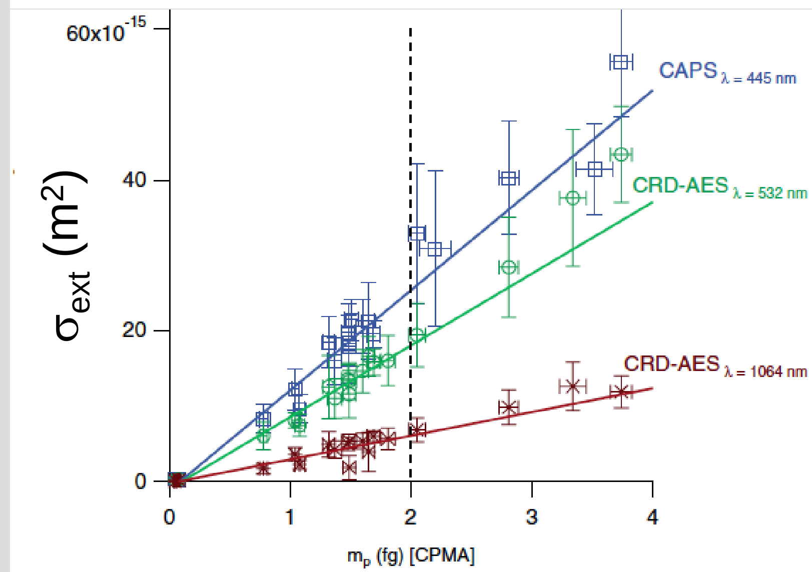
Key measurements

Particle mass, density



Multi-wavelength absorption, extinction, scattering (MAC, SSA, etc.)





Particle chemical composition

Particle hygroscopicity (κ)
CCN activity (κ)

Cross et al. 2010

Experimental matrix

Based upon “BC^2” intercomparison study, 318 runs *[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

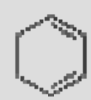
- controlled after reactor, but before instruments (multiplex of 0%, 30%, 60%, 90%)

Aging type

- Heterogeneous oxidation (OH, O_3)
- coated with sulfuric acid (SO_2)
- coated with SOA (fresh, aged)
- SOA + H_2SO_4

α-pinene SOA

toluene SOA



+ NO_x

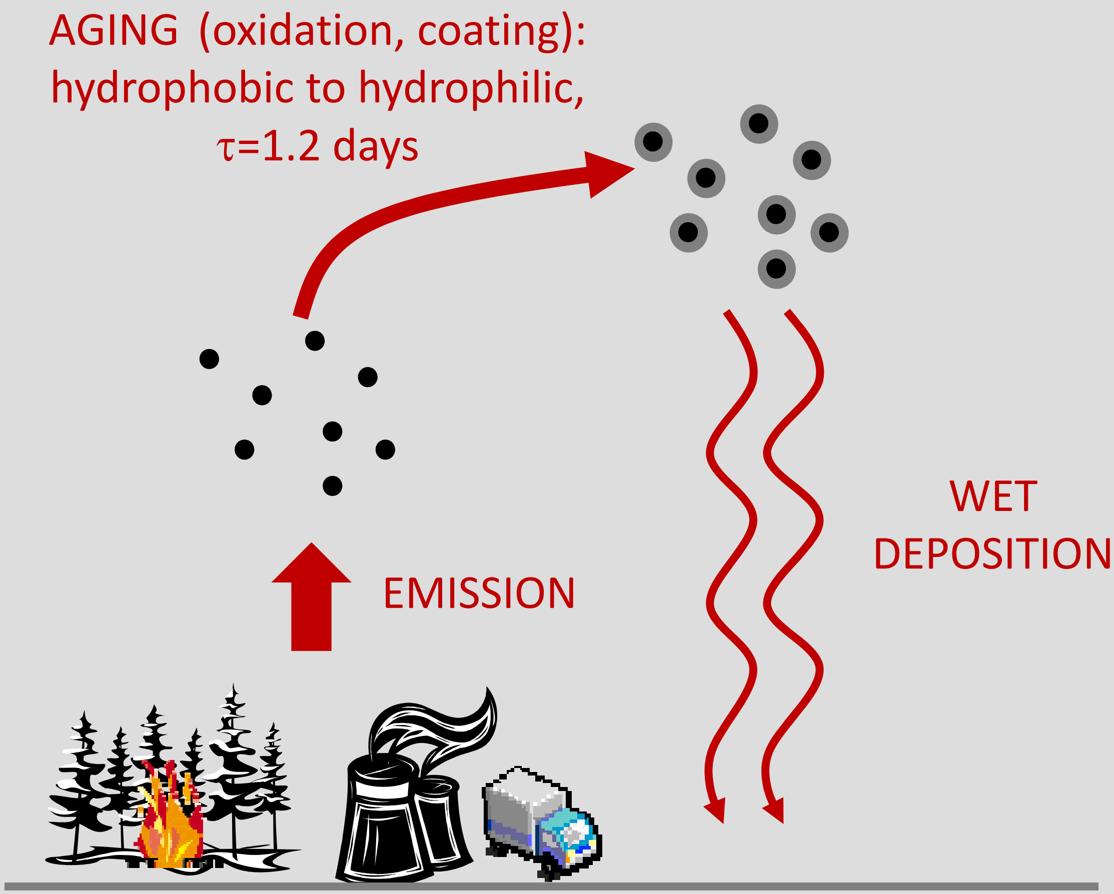
“brown SOA”

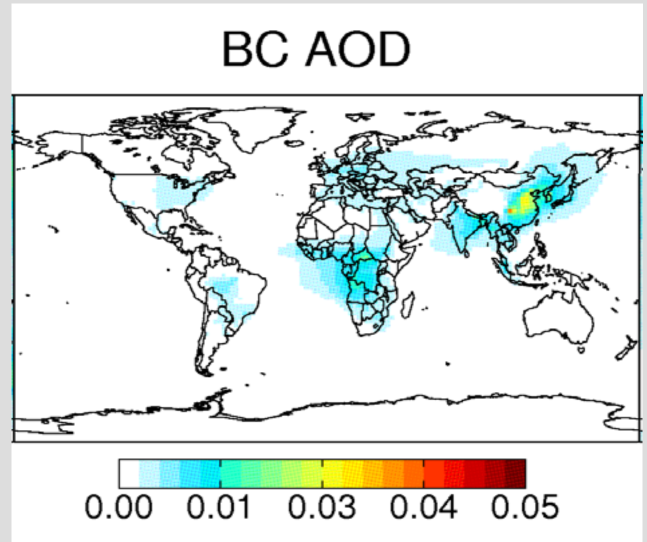
Parameterization

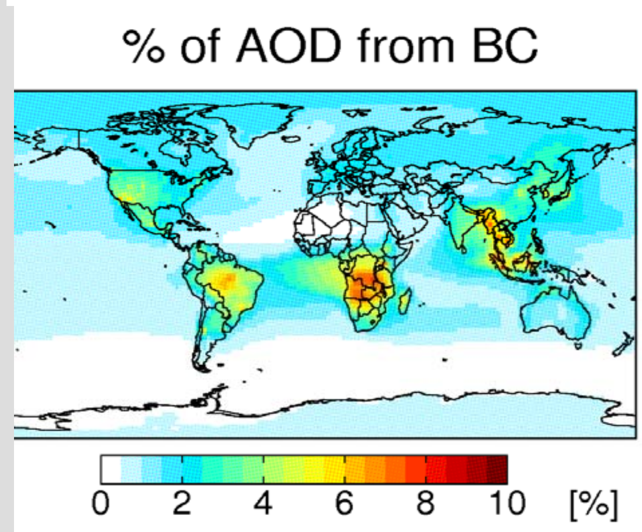
- Calculation of radiative forcing in models requires knowledge of key optical parameters as a function of particle properties
- This will be done by construction of a “lookup table” (or interpolated function) based on experimental results

particle size		mass absorption efficiency
relative humidity		single scattering albedo
wavelength	$\longrightarrow$	phase functions
mixing state (BC:org:SO_4)		asymmetry parameters

BC in GEOS-Chem (current treatment)







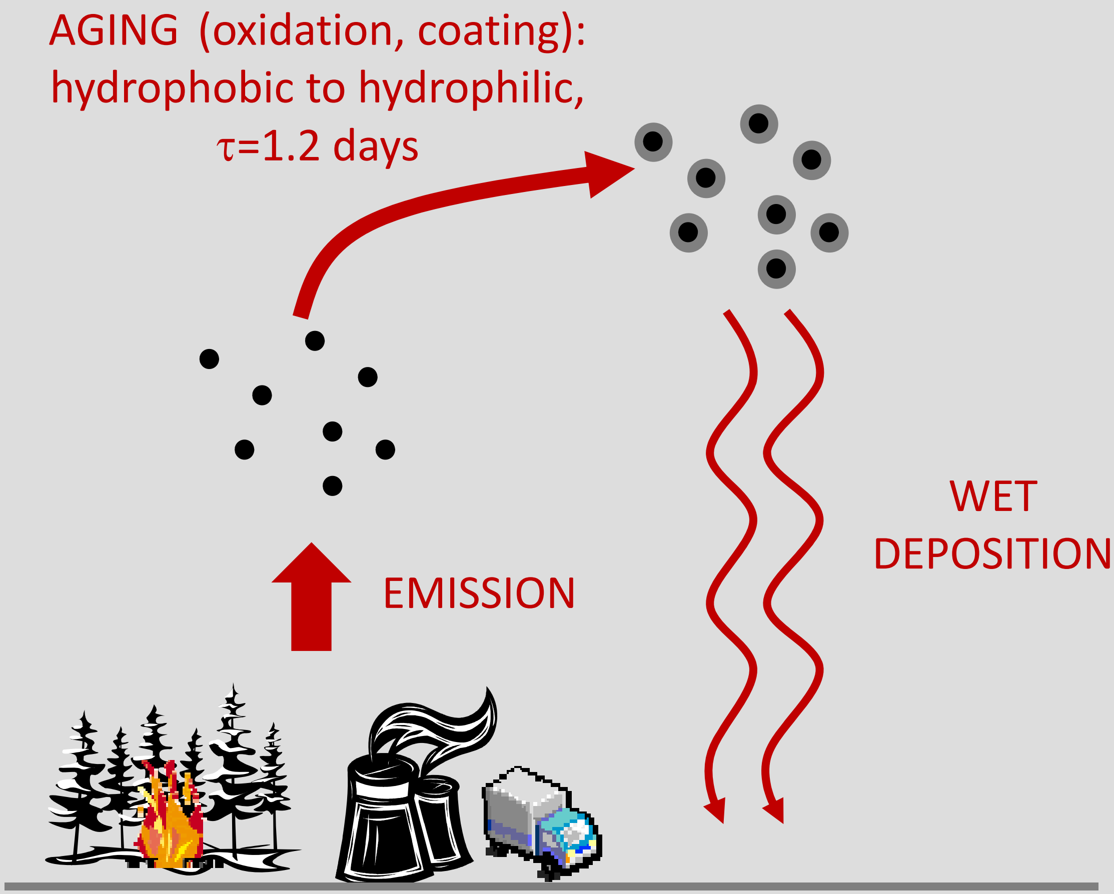
$$\tau = \frac{3}{4}\frac{Q_{ext}M}{r_{eff}\rho} = \alpha M$$

Currently aerosol is treated as (optically) externally mixed, with hydrophilic and hydrophobic BC having the same optical properties (with the exception of water uptake).

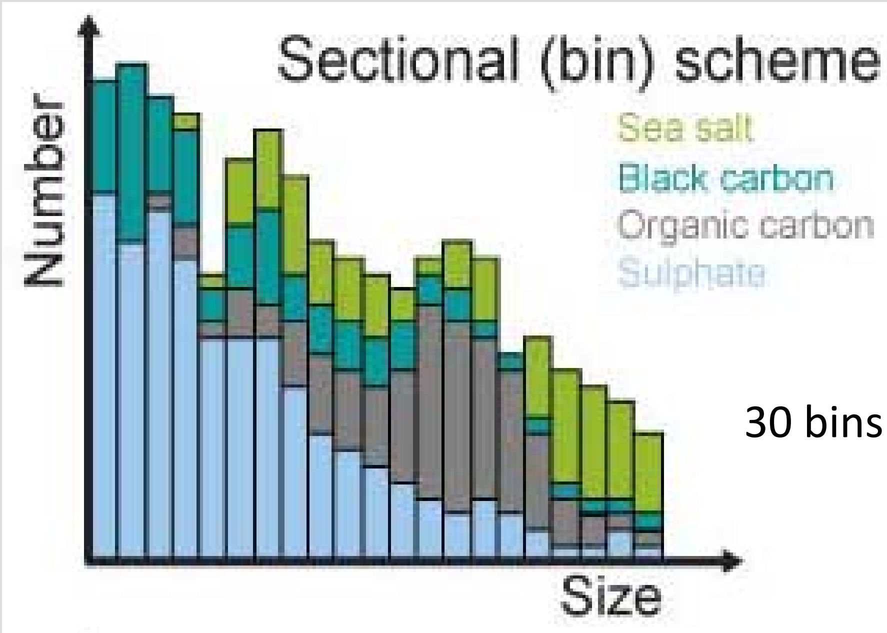
[Tegen and Lacis, 1996]

Modeling BC aging in GEOS-Chem

MASS-ONLY SCHEME



SECTIONAL SCHEME (TOMAS)



30 internally mixed bins from 0.01-10 μm (dry)

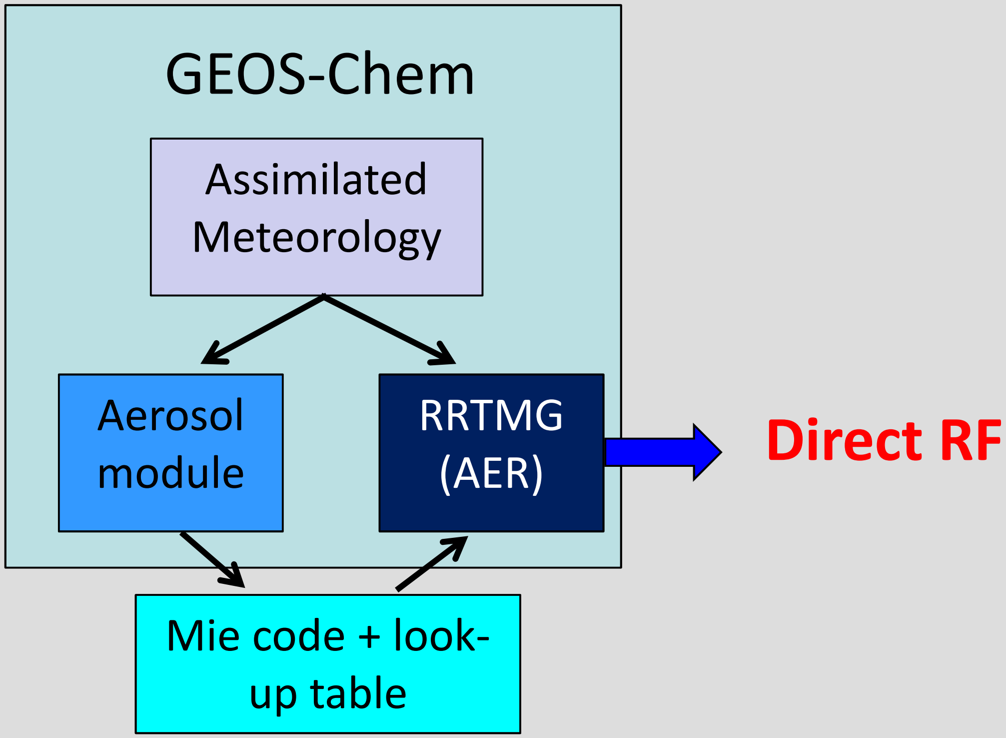
[Trivitayanurak et al., 2008]

Here: allow for physical aging through either (1) 1-day timescale (2) assume internal mixing (for all or within bins) or (3) implement a kappa-based transition to soluble aerosol

Calculation of direct radiative forcing by BC

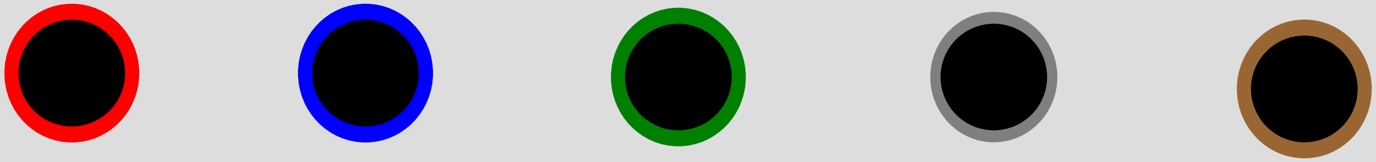
(1) AER's *Rapid Radiative Transfer Model (RRTMG)* will be integrated into GEOS-Chem (online) this Fall for application here. (RRTMG utiliizes the correlated-k approach to calculate fluxes and heating rates efficiently and accurately. Has been applied in many GCMs, well-documented comparison with line-by-line code.)

(2) Optical properties (mass extinction efficiency, SSA, asymmetry parameters and phase function) will be applied from lookup table in shortwave to GEOS-Chem simulated BC to estimate DRF.

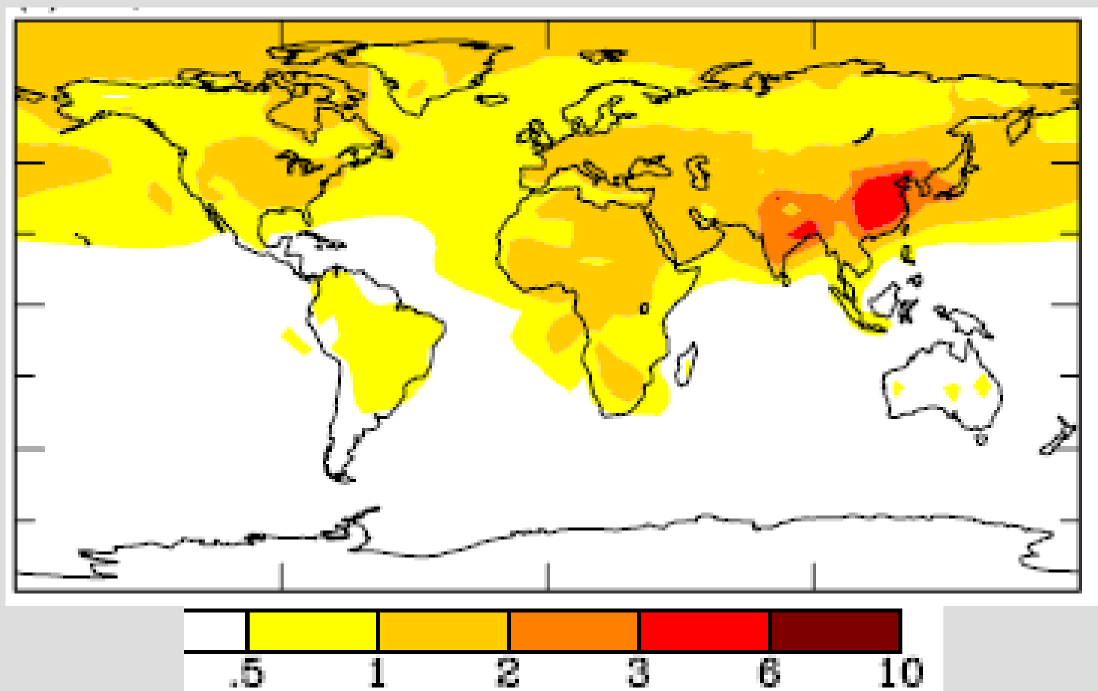


Investigating the sensitivity of DRF to BC aging

Laboratory measured optics



Simulated direct radiative forcing



The modeling, based upon detailed laboratory studies of BC aging, will allow for the estimation of how chemical transformations of BC particles impact direct radiative forcing (and export efficiency, deposition to the Arctic, other ancillary effects of aging timescales).