Black carbon radiative effects highly sensitive to emitted particle size when resolving mixing-state diversity

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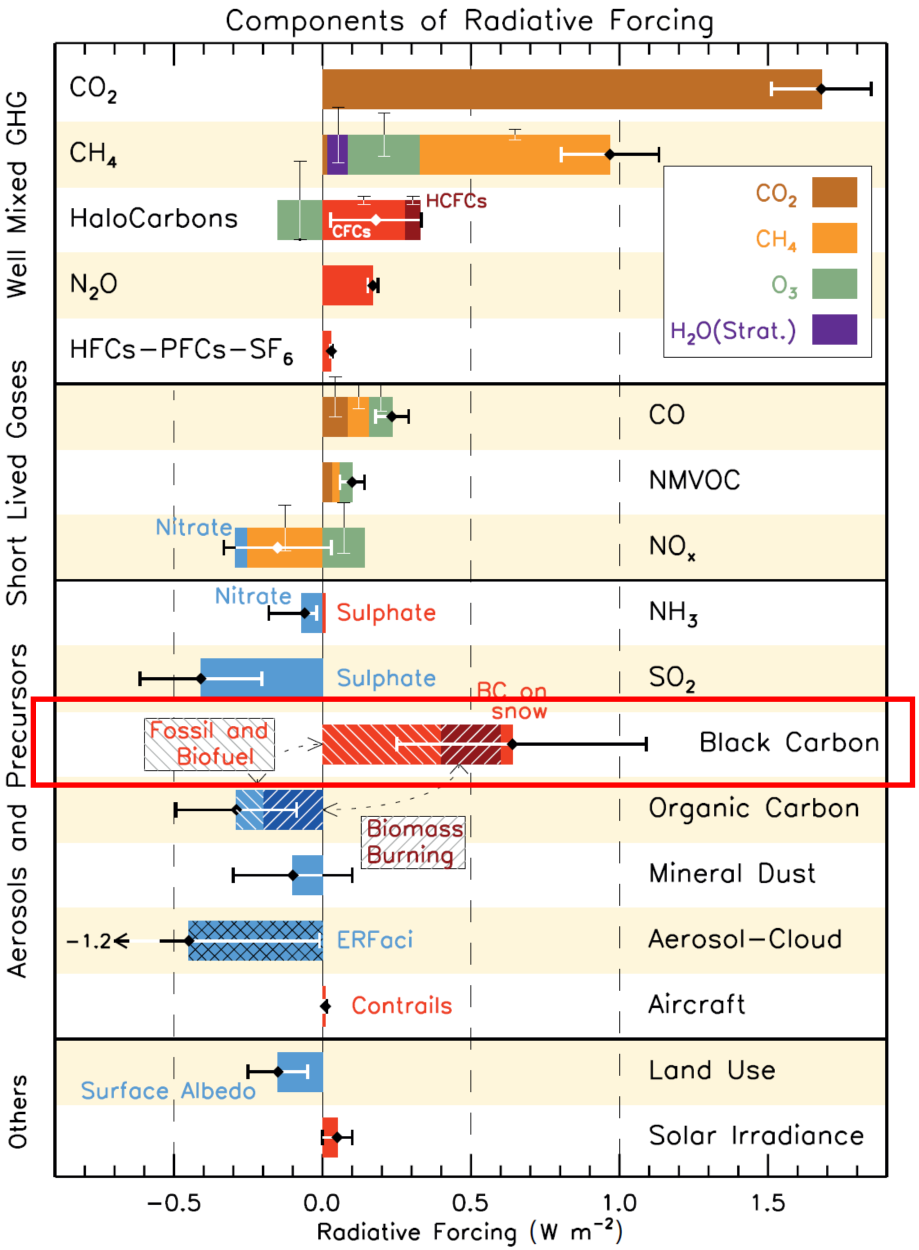
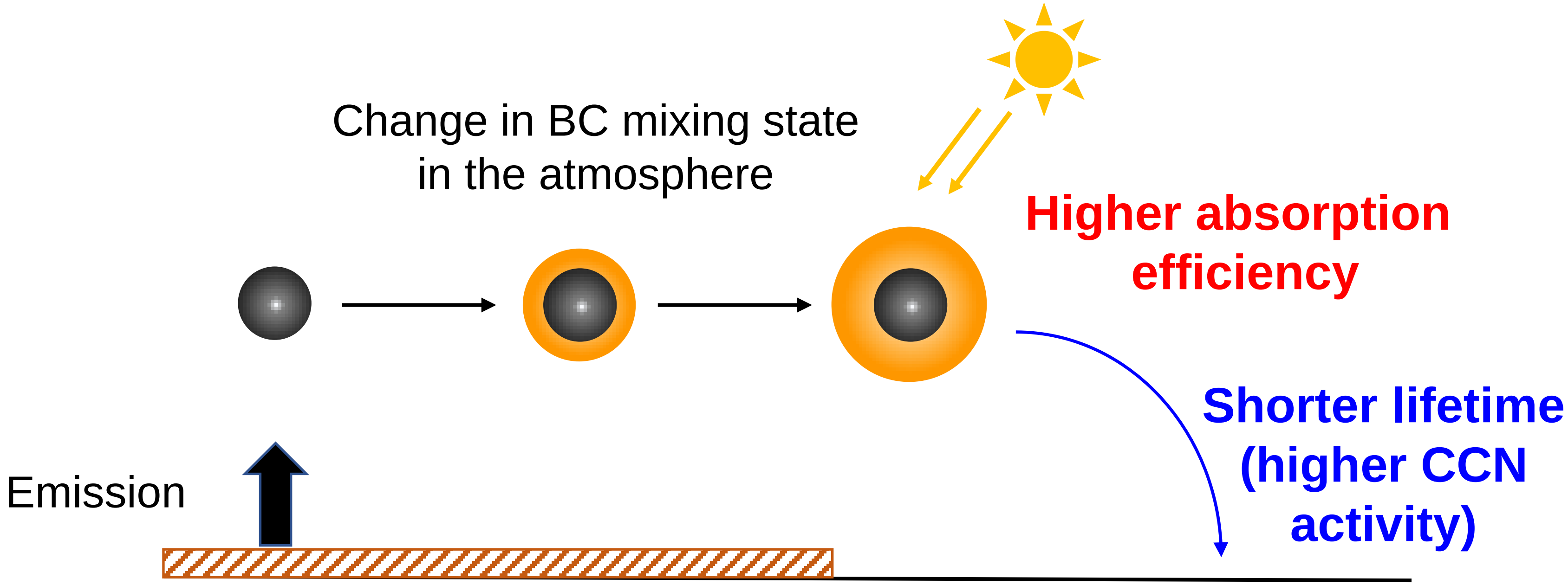
Importance of black carbon (BC)



[IPCC AR5]

- BC positive radiative forcing has a large uncertainty.
- Reducing this uncertainty is important to understand the total climate impact of anthropogenic aerosol. (e.g. the balance of heating/cooing by aerosols)

Importance of microphysical properties of BC

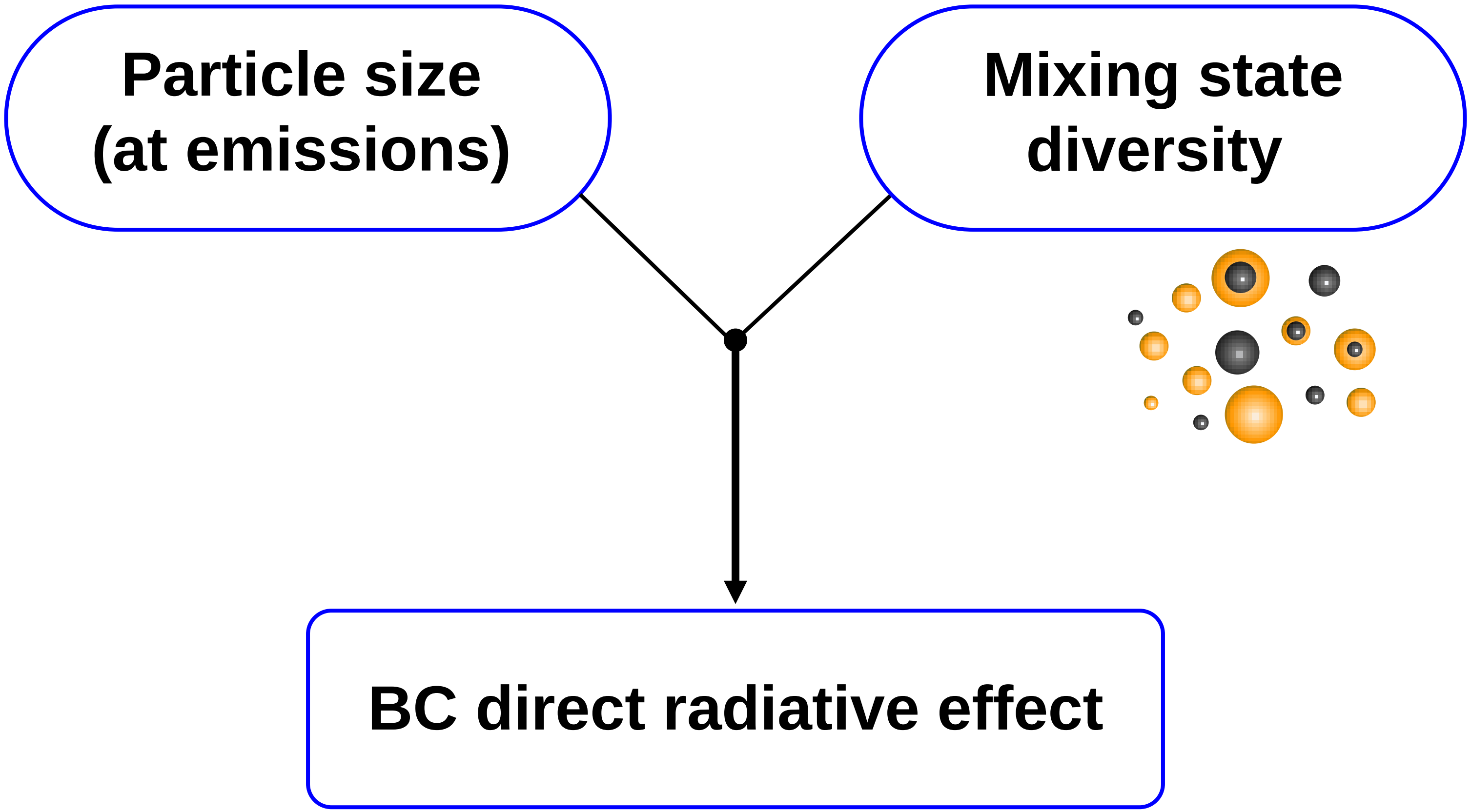

Change in BC mixing state in the atmosphere

Higher absorption efficiency

Shorter lifetime (higher CCN activity)

Emission

Particle size and mixing state strongly influence the solar radiation absorption efficiency and lifetime of BC, but… they have large uncertainties.



Particle size (at emissions)

Mixing state diversity

BC direct radiative effect

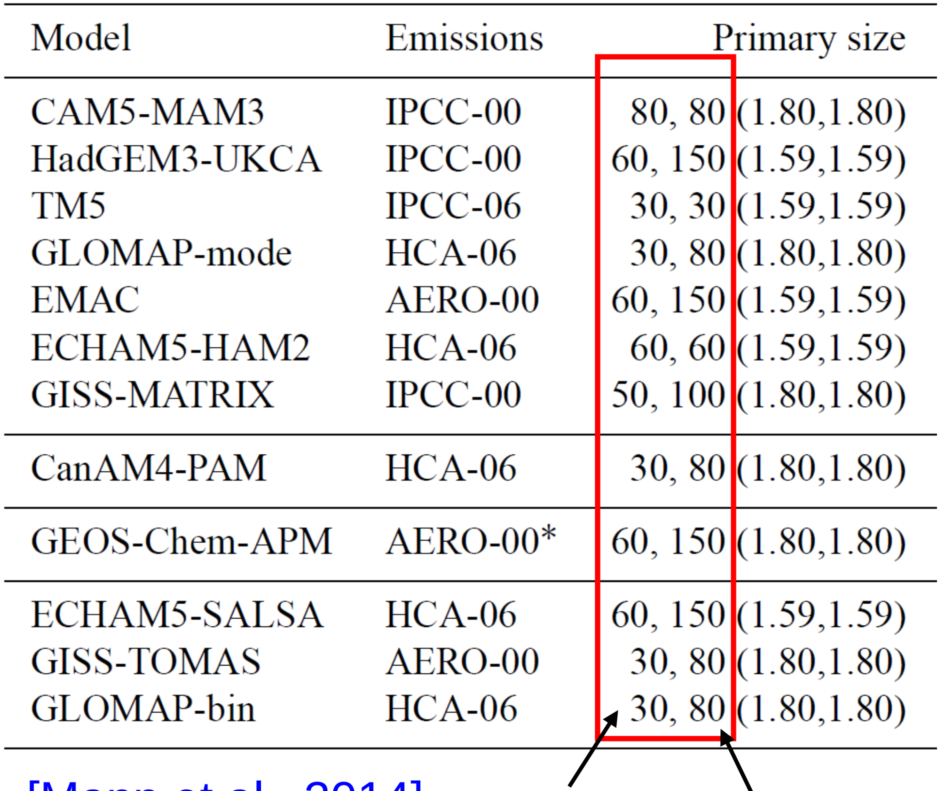
We show properly representing emission particle sizes and their interactions with BC mixing state diversity is essential when evaluating BC direct radiative effect.

Aerosol size distributions at emissions

Uncertainty ranges of emission sizes
(median diameters)

Fossil fuel (FF): 30-80 nm
Biofuel (BF): 50-200 nm
Biomass burning (BB): 50-200 nm

[Lee et al., 2013; Carslaw et al., 2013]



Model	Emissions	Primary size
CAM5-MAM3	IPCC-00	80, 80 (1.80,1.80)
HadGEM3-UKCA	IPCC-00	60, 150 (1.59,1.59)
TM5	IPCC-06	30, 30 (1.59,1.59)
GLOMAP-mode	HCA-06	30, 80 (1.80,1.80)
EMAC	AERO-00	60, 150 (1.59,1.59)
ECHAM5-HAM2	HCA-06	60, 60 (1.59,1.59)
GISS-MATRIX	IPCC-00	50, 100 (1.80,1.80)
CanAM4-PAM	HCA-06	30, 80 (1.80,1.80)
GEOS-Chem-APM	AERO-00*	60, 150 (1.80,1.80)
ECHAM5-SALSA	HCA-06	60, 150 (1.59,1.59)
GISS-TOMAS	AERO-00	30, 80 (1.80,1.80)
GLOMAP-bin	HCA-06	30, 80 (1.80,1.80)

[Mann et al., 2014]

FF

BF/BB

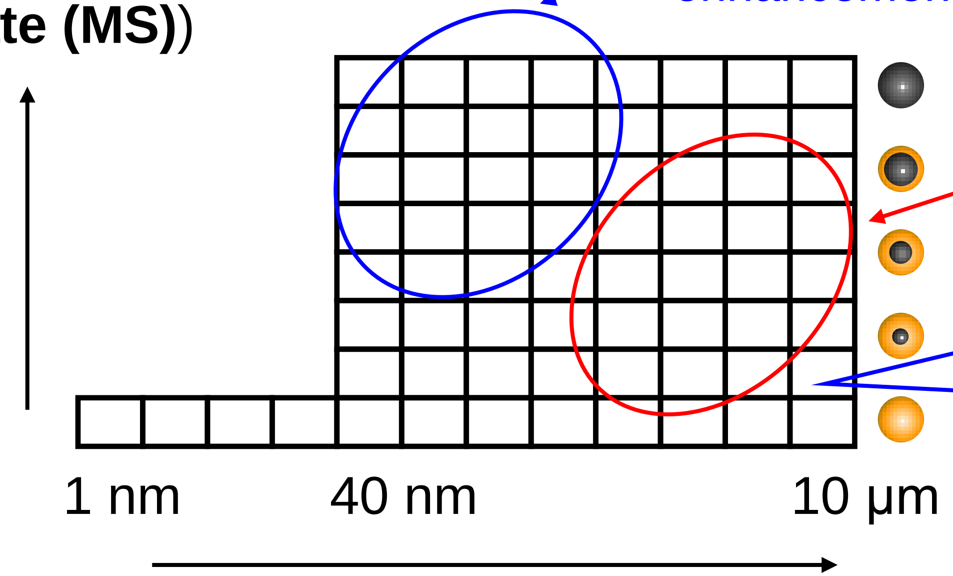
- The importance of emission size distributions to **aerosol number concentrations** and **aerosol-cloud interactions** is known.
  [e.g., Reddington et al., 2011; Spracklen et al., 2011]
- The impact on **BC radiative effect** is not understood well.

A 2-D sectional global aerosol model

ATRAS

[Matsui et al., ACP, 2014]

(Aerosol Two-dimensional bin model for foRmation and Aging Simulation)



BC mass fraction
(**mixing state (MS)**)

Lower absorption
enhancement & CCN activity

Higher
absorption
enhancement
& CCN activity

SO4, NH4, NO3
Dst, Sslt, OA
(POA, SOA), BC,
Number

1 nm 40 nm 10 µm

Particle dry diameter

12 size and 8 BC mixing state bins

BC size and mixing state → BC absorption → BC radiative effect → CCN activity of BC → BC lifetime/burden

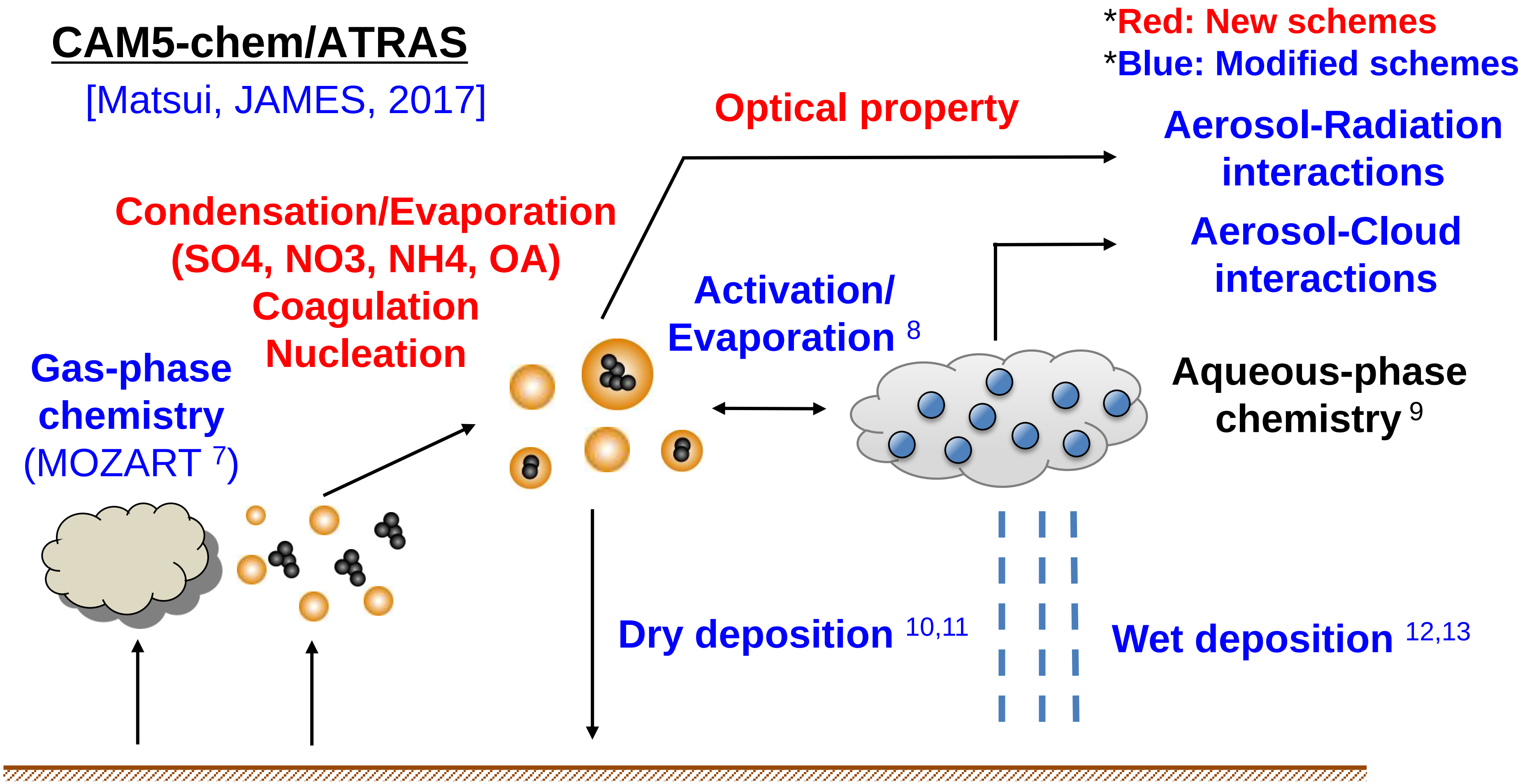
A 2-D sectional global aerosol model

CAM5-chem/ATRAS

[Matsui, JAMES, 2017]

*Red: New schemes

*Blue: Modified schemes



Optical property

Aerosol-Radiation interactions

Aerosol-Cloud interactions

Condensation/Evaporation (SO4, NO3, NH4, OA)

Coagulation

Nucleation

Activation/ Evaporation [8]

Gas-phase chemistry (MOZART [7])

Aqueous-phase chemistry [9]

Dry deposition [10,11]

Wet deposition [12,13]

Emission

(Anthropogenic [1],

Biogenic [1],

Biomass burning [1],

Dust [2,3,4], Sea salt [5,6])

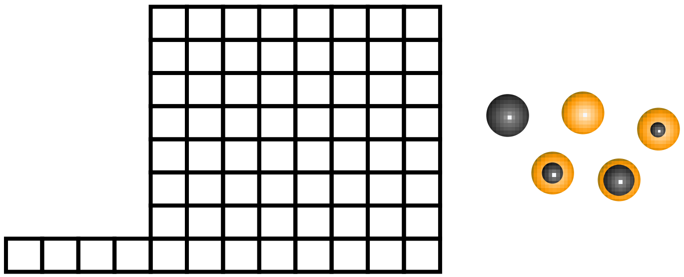
References:
1: Lamarque et al., 2010, 2: Zender et al., 2003, 3: Albani et al., 2014,
4: Kok, 2011, 5: Martensson et al., 2003, 6: Monahan et al., 1986,
7: Emmons et al., 2010, 8: Abdul-Razzak and Ghan, 2000,
9: Tie et al., 2001, 10: Wesely, 1989, 11: Zender et al., 2003,
12: Rasch et al., 2000, 13: Liu et al., 2012

Global aerosol model simulations

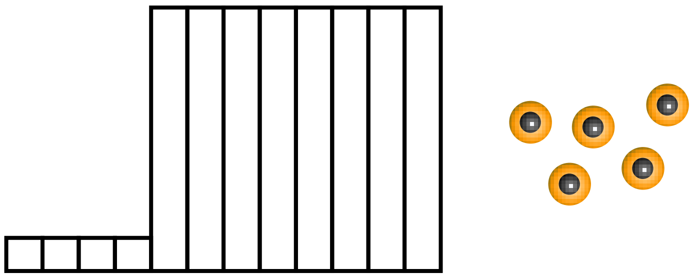
Present-day climate, 6-y online simulations (2.5 x 1.9 deg, 30 layers)

Simulation	Median diameter		Sigma	Comment
	FF	BF/BB		
Base	70	100	1.8	Matsui[3] and Matsui and Mahowald[4]
Large	80	200	1.8	Lee et al.[5] and Carslaw et al.[6]
Small	30	50	1.8	Lee et al.[5] and Carslaw et al.[6]
Sens 1	80	80	1.8	CAM5-MAM3 [7]
Sens 2	60	150	1.59	HadGEM3-UKCA [8], EMAC [9], and ECHAM5-SALSA [10]
Sens 3	30	30	1.59	TM5 [11]
Sens 4	30	80	1.8	GLOMAP [12,13], CanAM4-PAM [14], and GISS-TOMAS [15]
Sens 5	60	60	1.59	ECHAM5-HAM2 [16]
Sens 6	50	100	1.8	GISS-MATRIX [17]
Sens 7	60	150	1.8	GEOS-Chem-APM [18]

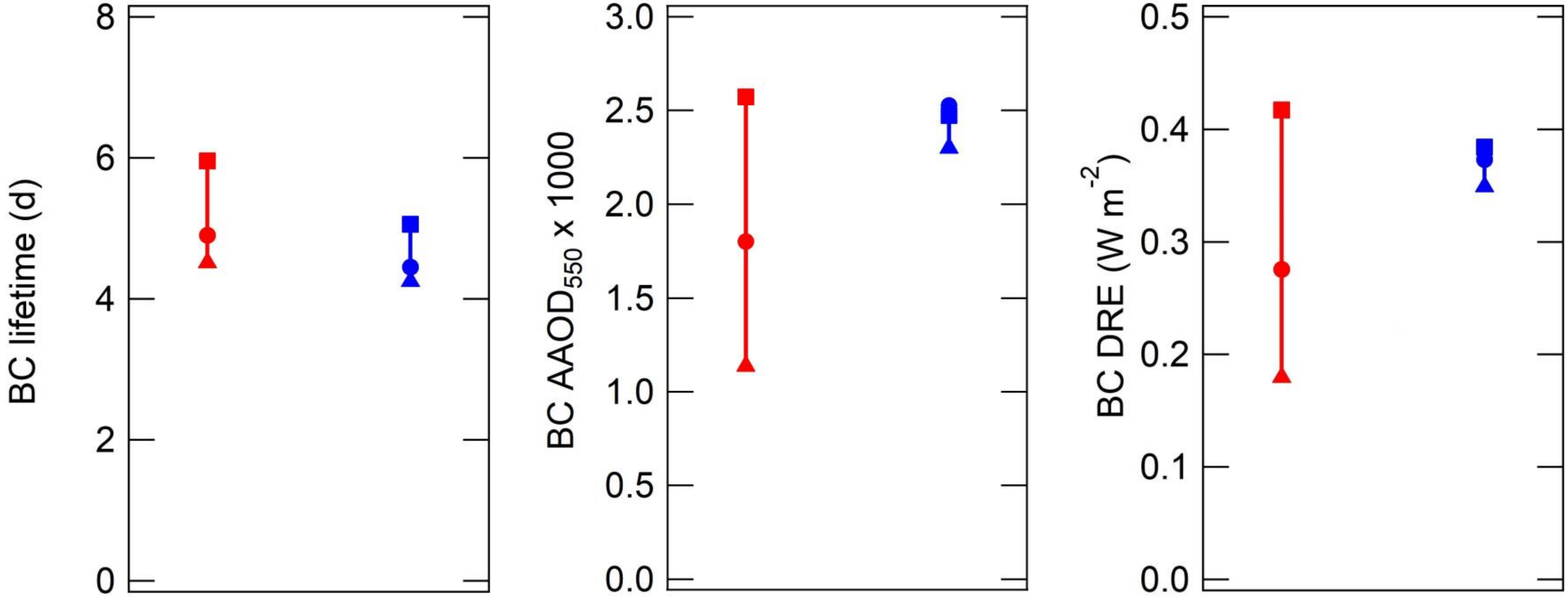
Multiple MS (12 size x 8 MS)



Single MS (12 size x 1 MS)



Sensitivity of BC properties to emission particle sizes



(global mean)

Multiple MS (12 size x 8 MS)

Single MS (12 size x 1 MS)

■ Small, ● Base, ▲ Large

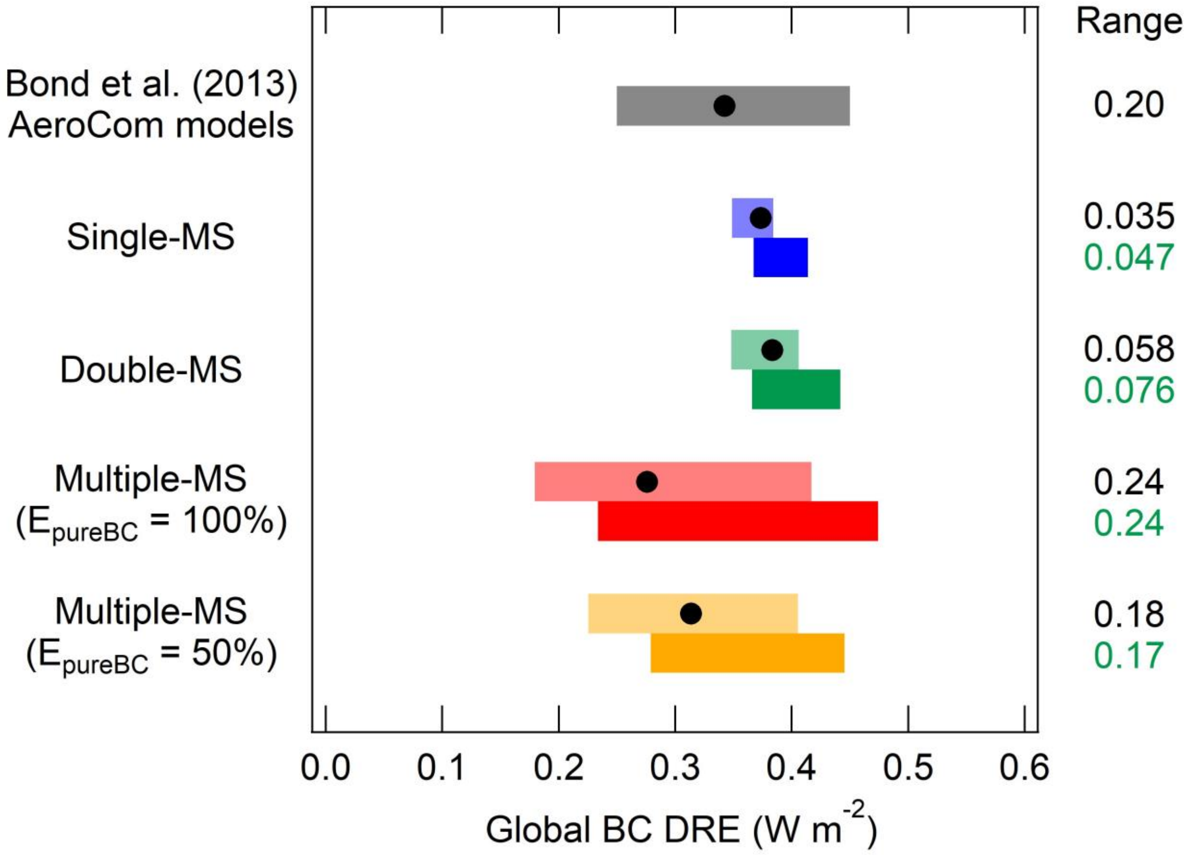
Small / Large ratio

	Multiple-MS	Single-MS
BC lifetime	1.3	1.2
BC AAOD$_{550}$	**2.3**	1.1
BC DRE	**2.3**	1.1

* AAOD$_{550}$: absorption aerosol optical depth at 550 nm

* DRE: direct radiative effect

Sensitivity of BC direct radiative effect



Small, Base, Large
AeroCom size
(from 7 sensitivity simulations)

- BC direct radiative effect (DRE) is 5-7 times more sensitive to changes in aerosol emission size distributions when the diversity in BC mixing state is resolved.
- The range of BC DRE is slightly larger than the multi-model variability in BC DRE estimated by global aerosol models.

Causes of high sensitivity by mixing state diversity

BC AAOD

$$= \underbrace{\text{BC burden}}_{\textbf{BC lifetime}} \times \underbrace{\frac{\text{BC AAOD (BC core)}}{\text{BC burden}}}_{\textbf{Absorption of BC core}} \times \underbrace{\frac{\text{BC AAOD (BC core + coating)}}{\text{BC AAOD (BC core)}}}_{\textbf{Absorption enhancement (by coating species)}}$$

Small / Large ratio

	BC lifetime	Absorption of BC core	Absorption enhancement
Multiple MS	**1.3**	**1.4**	**1.2**
Single MS	**1.2**	**0.95**	**0.96**

Causes of high sensitivity by mixing state diversity

$$\text{BC AAOD} = \text{BC burden} \times \frac{\text{BC AAOD (BC core)}}{\text{BC burden}} \times \frac{\text{BC AAOD (BC core + coating)}}{\text{BC AAOD (BC core)}}$$

BC lifetime

Absorption of BC core

Absorption enhancement (by coating species)

Small / Large ratio

	BC lifetime		Absorption of BC core		Absorption enhancement		AAOD
Multiple MS	**1.3**	**x**	**1.4**	**x**	**1.2**	**=**	**2.2**
Single MS	**1.2**	**x**	**0.95**	**x**	**0.96**	**=**	**1.1**

Causes of high sensitivity by mixing state diversity

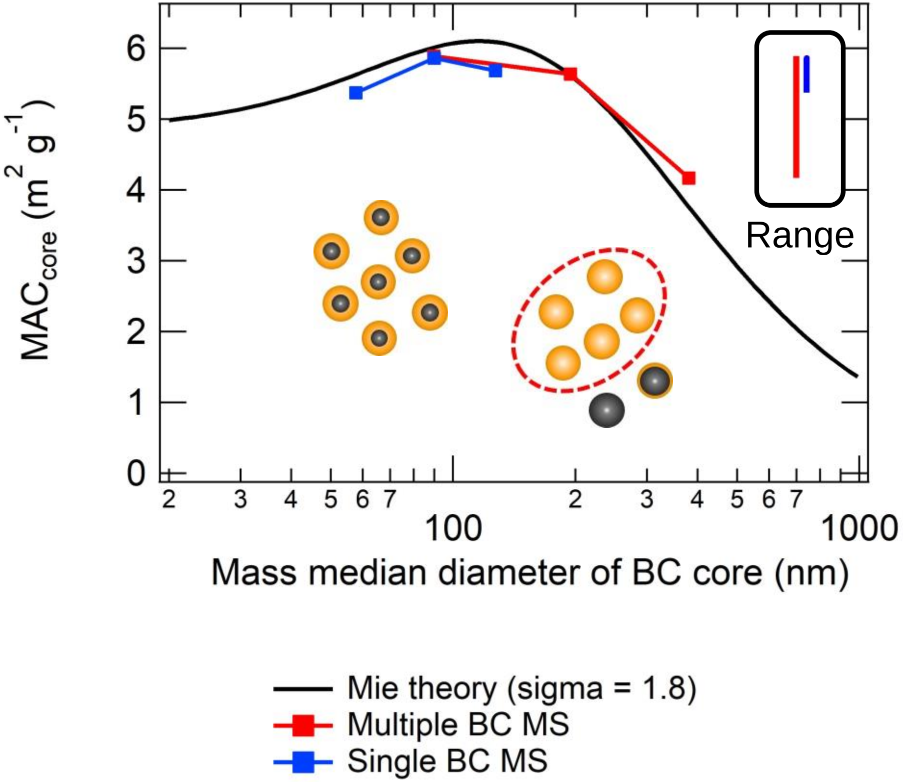
BC AAOD

$$= \text{BC burden} \times \frac{\textbf{BC AAOD (BC core)}}{\textbf{BC burden}} \times \frac{\text{BC AAOD (BC core + coating)}}{\text{BC AAOD (BC core)}}$$

Wavelength: 550 nm

Observations of BC core size (~ 200 nm)

[e.g., Schwarz et al., 2010;
Kondo et al., 2011; Moteki et al., 2012]



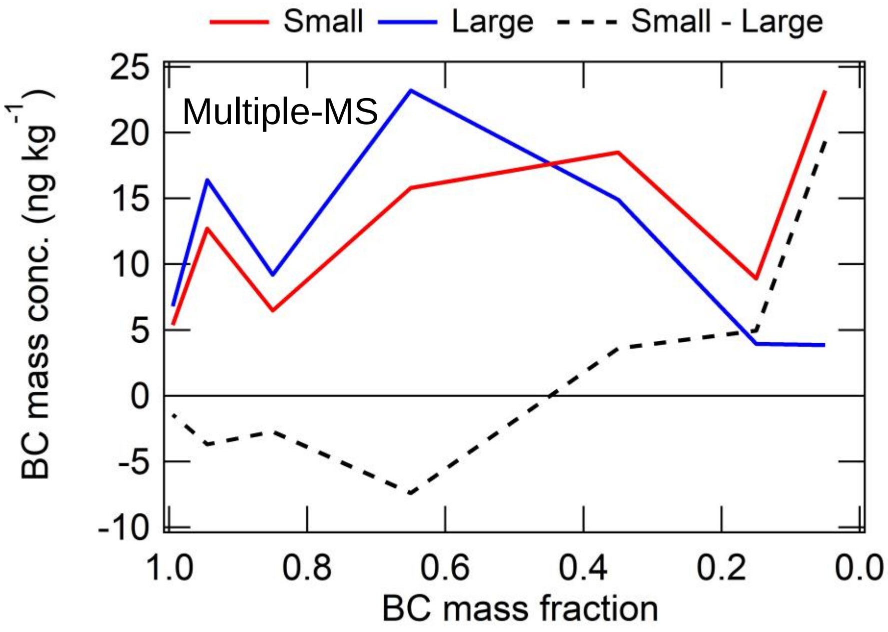
MAC_{core} =
BC absorption coefficient / BC mass

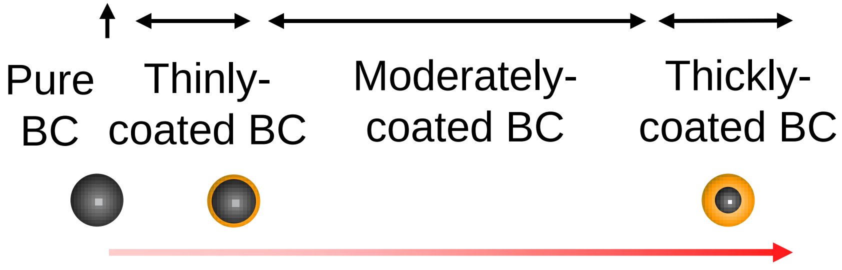
BC core size and the range of MAC_{core} are larger when BC-free particle are treated properly.

Causes of high sensitivity by mixing state diversity

BC AAOD

$$= \text{BC burden} \times \frac{\text{BC AAOD (BC core)}}{\text{BC burden}} \times \frac{\textbf{BC AAOD (BC core + coating)}}{\textbf{BC AAOD (BC core)}}$$





Pure BC

Thinly-coated BC

Moderately-coated BC

Thickly-coated BC

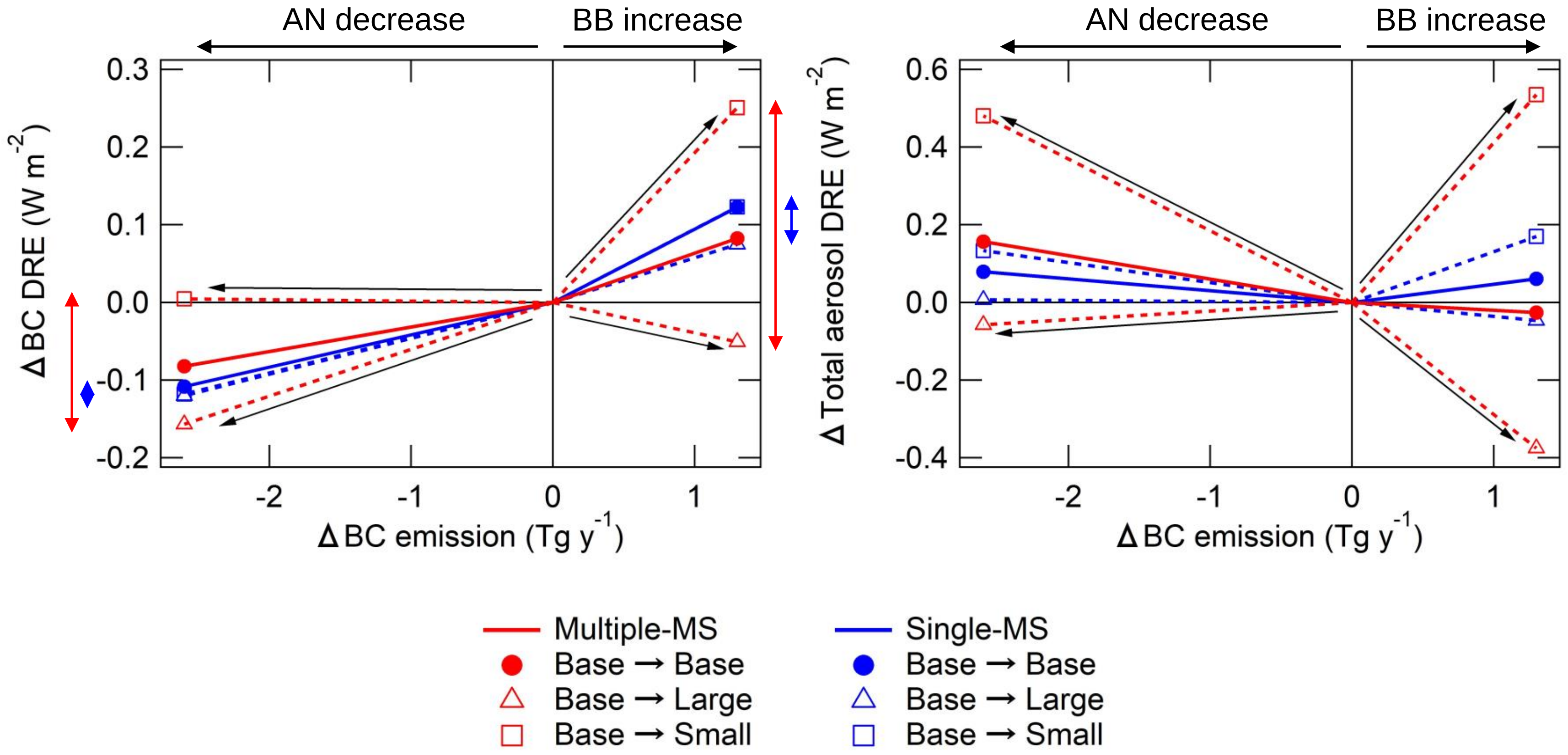
Lower absorption enhancement

Higher absorption enhancement

Small-size simulation (Multiple-MS) has 5 times more thickly-coated BC than Large-size simulation.

→ Higher absorption enhancement in Small-size simulation

Sensitivity to emission changes in the future



The present-day to future changes of BC and total aerosol direct radiative effect are 6-13 and 4 times more sensitive to changes in emission size distributions, respectively, by resolving mixing state diversity with a potential to change the sign of their DREs (warming or cooling effect) for both anthropogenic (AN) and biomass burning (BB) sources.

Summary

- The sensitivity (range) of present-day BC direct radiative effect, due to current uncertainties in emission size distributions, is amplified 7 times (0.18-0.42 W m^{-2}) when the diversity in BC mixing state is sufficiently resolved.

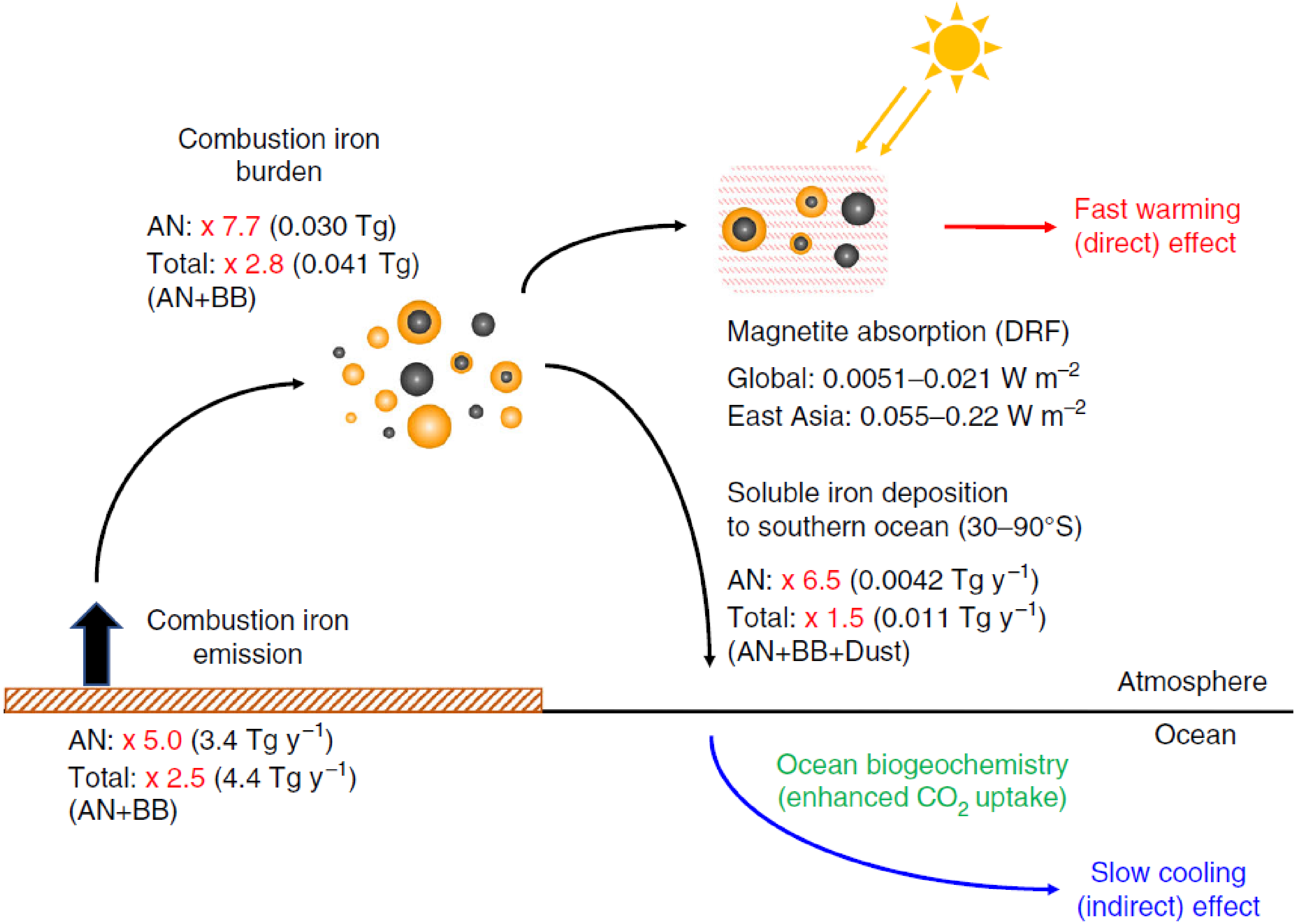
- This particle size and mixing state effect should be considered as an uncertainty in BC direct radiative effect.

- BC and total aerosol direct radiative effects and their future changes cannot be calculated accurately without 1) reducing large uncertainties in aerosol size distributions in emissions and 2) considering BC mixing state diversity.

Matsui, H., D. S. Hamilton & N. M. Mahowald (2018),
Black carbon radiative effects highly sensitive to emitted particle size when resolving mixing-state diversity, *Nature Commun*., 9, 3446.

E-mail: matsui@nagoya-u.jp

Matsui, H., N. M. Mahowald, N. Moteki et al. (2018), Anthropogenic combustion iron as a complex climate forcer, *Nature Commun*., 9, 1593.



Combustion iron
burden

AN: x 7.7 (0.030 Tg)
Total: x 2.8 (0.041 Tg)
(AN+BB)

Fast warming
(direct) effect

Magnetite absorption (DRF)

Global: 0.0051–0.021 W m^{-2}
East Asia: 0.055–0.22 W m^{-2}

Soluble iron deposition
to southern ocean (30–90°S)

AN: x 6.5 (0.0042 Tg y^{-1})
Total: x 1.5 (0.011 Tg y^{-1})
(AN+BB+Dust)

Combustion iron
emission

AN: x 5.0 (3.4 Tg y^{-1})
Total: x 2.5 (4.4 Tg y^{-1})
(AN+BB)

Atmosphere

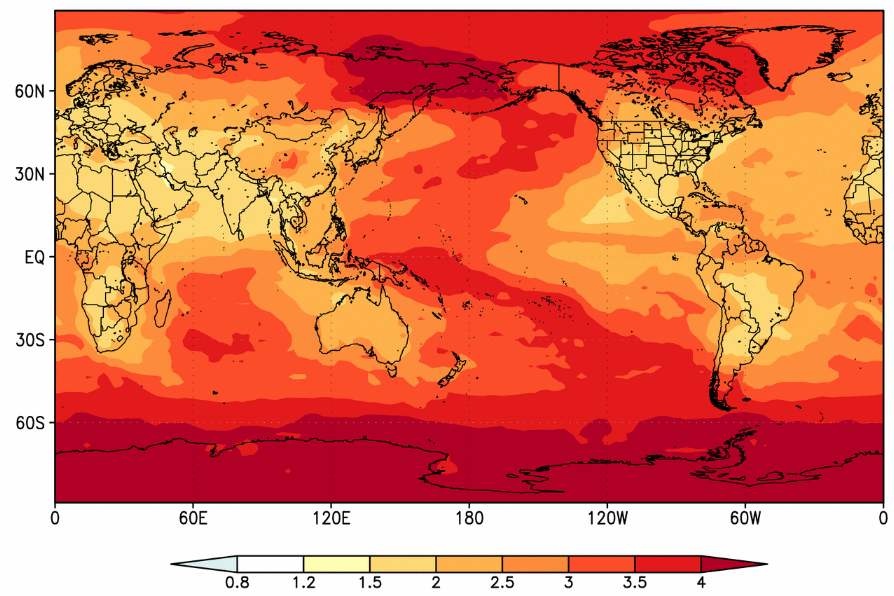
Ocean

Ocean biogeochemistry
(enhanced CO$_2$ uptake)

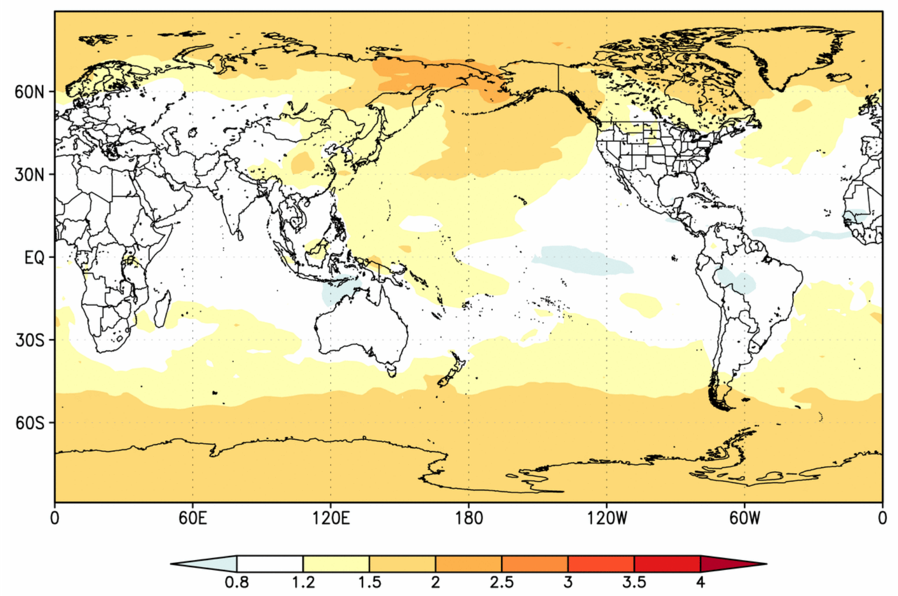
Slow cooling
(indirect) effect

- AN magnetite DRF can be comparable to BrC DRF.
- AN iron may be an important source of soluble iron deposition to oceans.

The values shown by red are the Small/Large ratios.



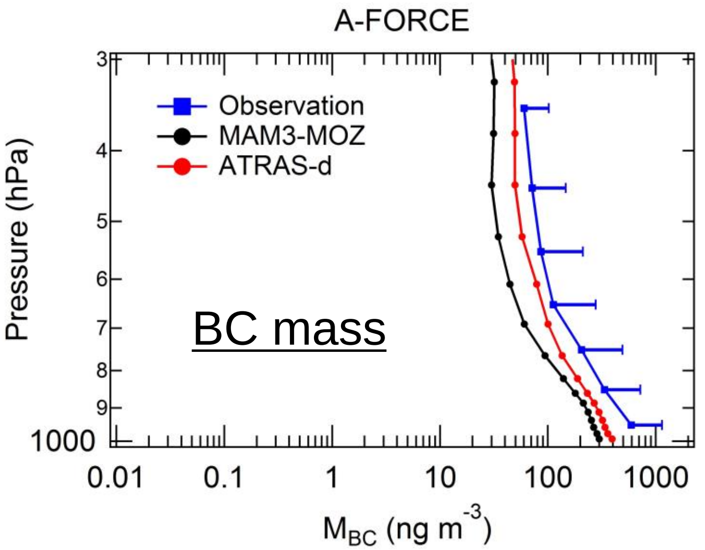
BC DRE ratio (Multiple-MS, Small / Large)

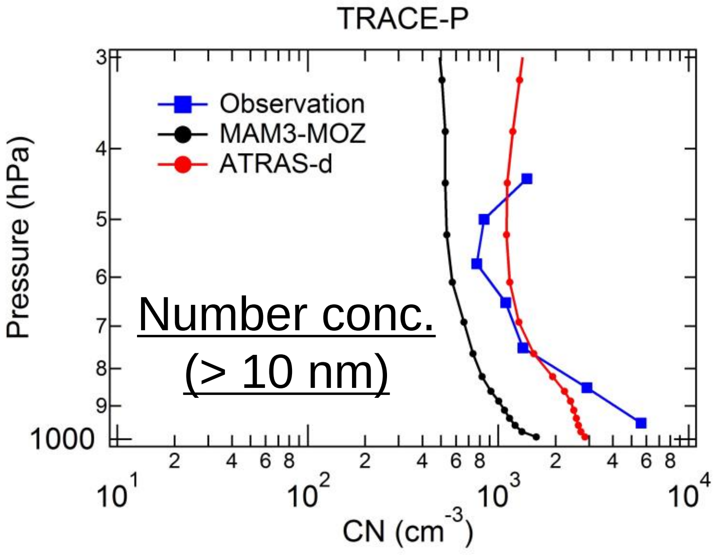


BC DRE ratio (Single-MS, Small / Large)

Comparison with measurements

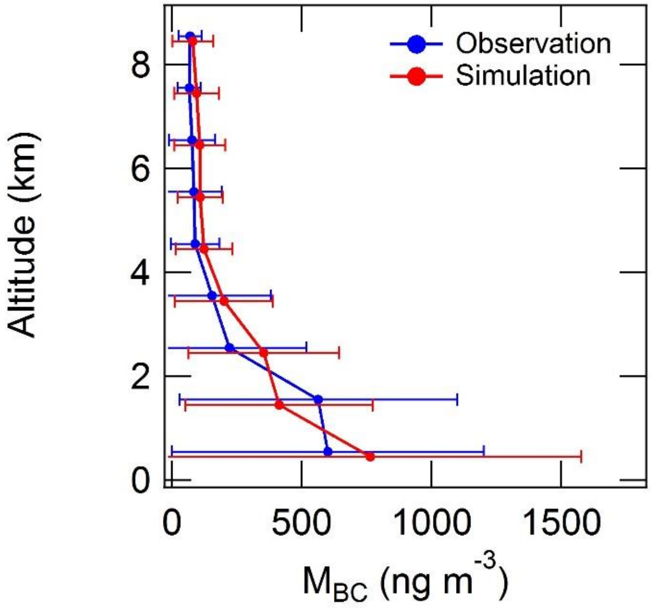
Data	Species	Average of all sites		
		Obs.	MAM3	ATRAS
AMS	SO4	3.8	**4.1**	3.3
(34 sites)	NH4	1.6	0.76	**1.0**
	NO3	1.3	---	**1.3**
	OA	4.9	**4.7**	5.9
IMPROVE	SO4	1.4	2.4	**2.2**
(193 sites)	OA	1.8	4.3	**3.9**
	BC	0.48	0.24	**0.26**
EMEP	SO4	2.2	**2.1**	2.4
(151 sites)	OA	5.0	3.5	**4.7**
	BC	0.56	0.41	**0.43**
				(μg m^{-3})
MODIS	AOD	0.16	0.096	**0.14**
AERONET	AOD	0.25	0.11	**0.17**
(603 sites)	SSA	0.92	0.90	0.94

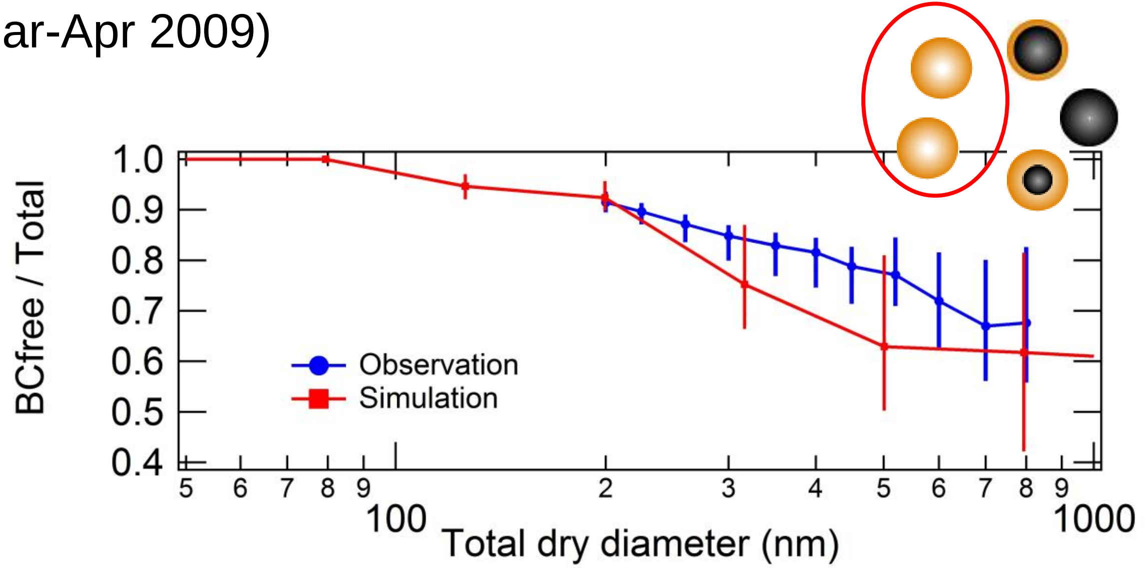


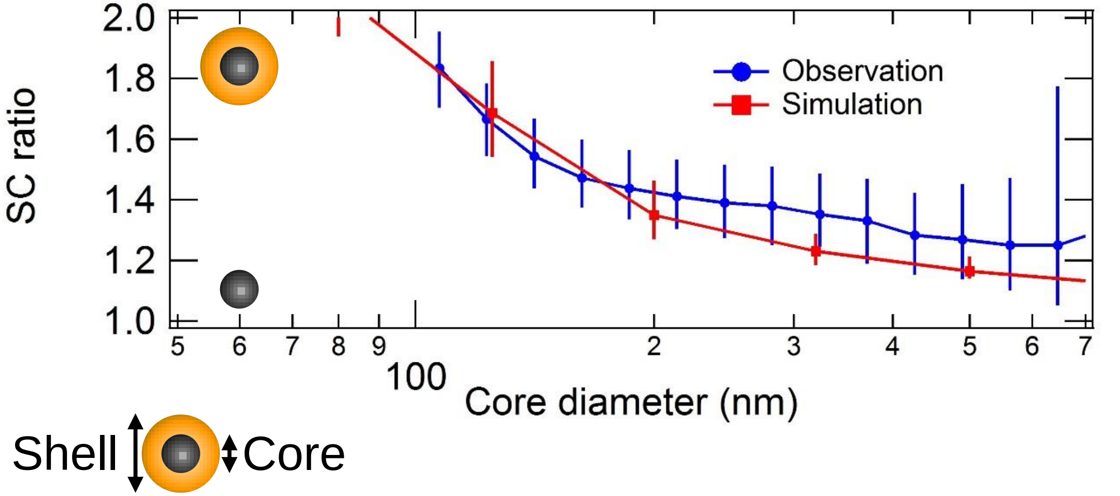


Validation of ATRAS (BC mixing state)

A-FORCE aircraft campaign (21 flights)
(Yellow Sea, Western Japan, Mar-Apr 2009)



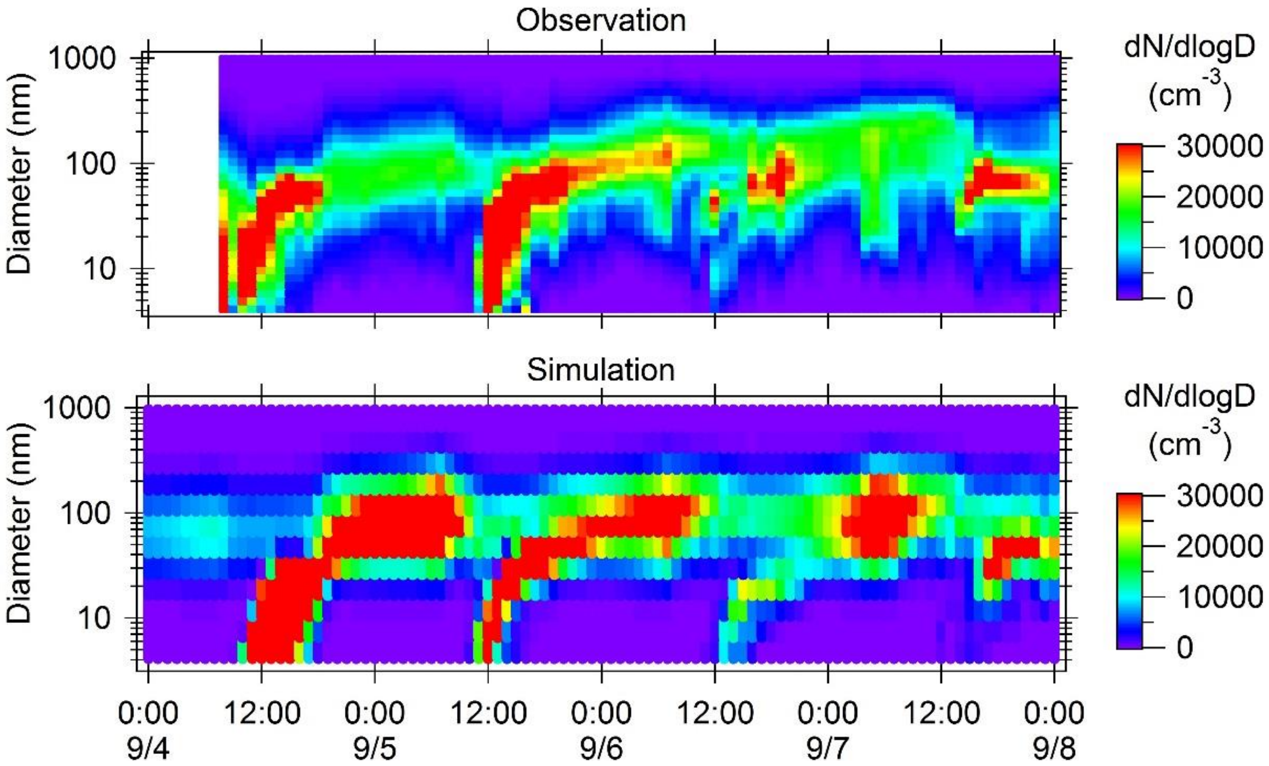




[Matsui et al., JGR, 2013b]

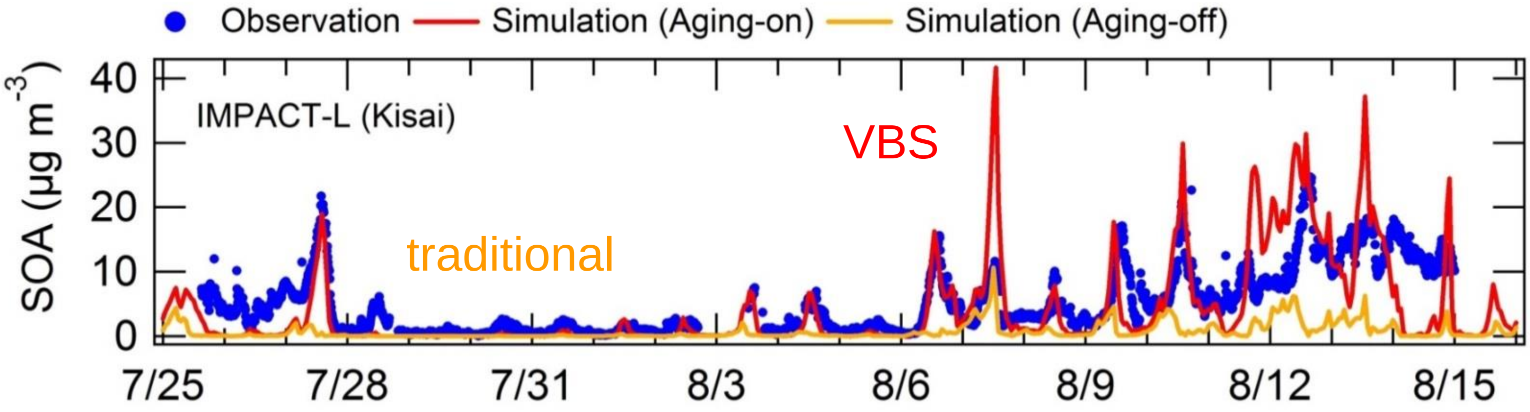
Validation of ATRAS (NPF, OA formation)

CARE-Beijing 2006 campaign
(Beijing, Aug-Sep 2006)



[Matsui et al., JGR, 2011]

IMPACT campaign
(Tokyo, Jul-Aug 2004)



[Matsui et al., ACP, 2014a]