toward resolution on the optics of light-absorbing carbon

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16 December 2004

American Geophysical Union

San Francisco, California

carbon neutral travel

don't just travel. travel neutral.

www.co2.org

funding: U.S. EPA, NASA-ACMAP, NASA-IDS,
NASA-EOS, NOAA-ACIP

material presented here

"Light absorption by carbonaceous particles: An investigative review"

T. C. Bond and R. W. Bergstrom, *perpetually in prep*

(~250 references)

Approach: Review (not just tabulate) all previous literature

"Classifying climate-relevant properties of primary carbonaceous particles"

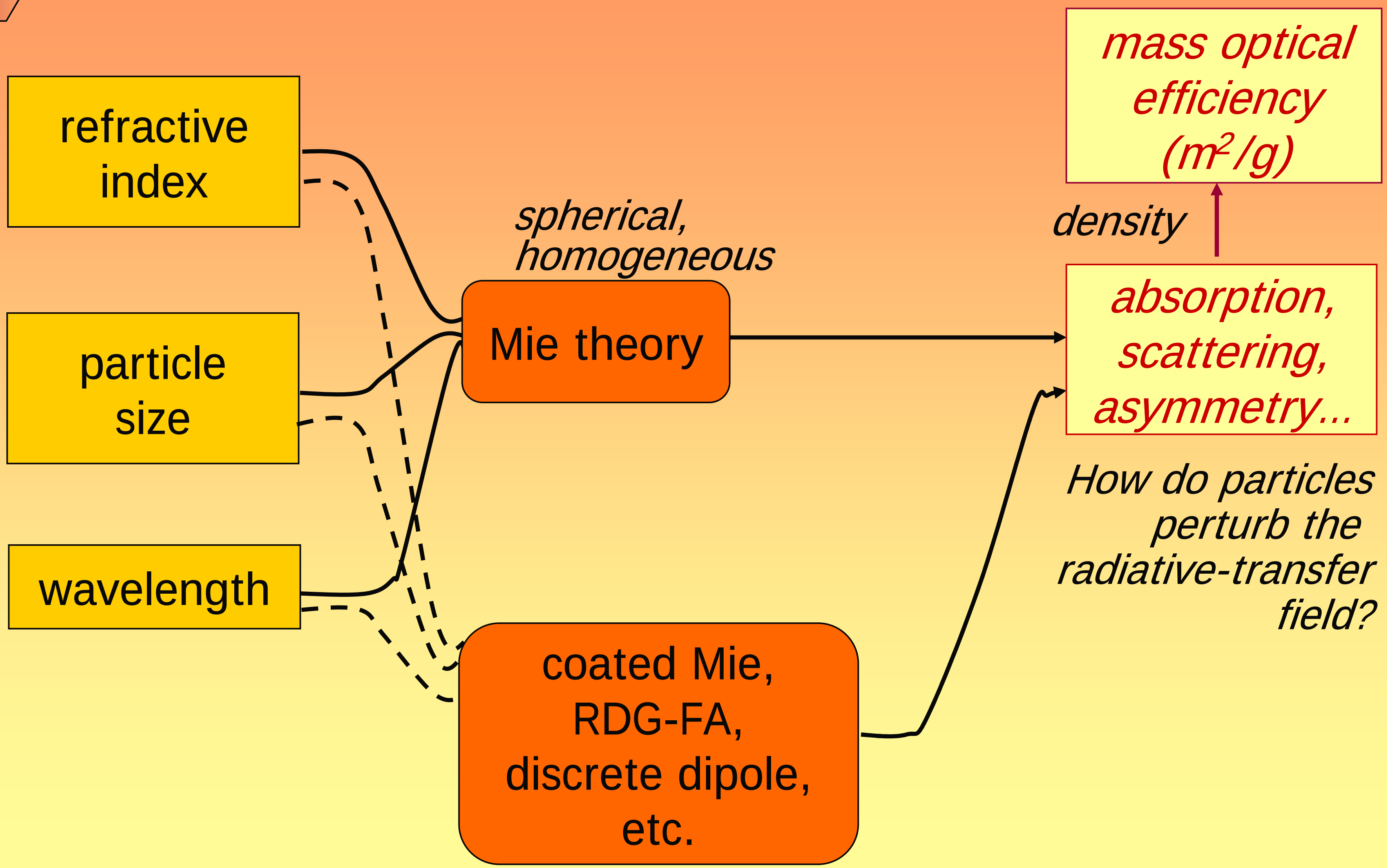
H. Sun and T. C. Bond, *in prep*; poster Friday morning

"Can reducing soot emissions save us from global warming?"

T. C. Bond and H. Sun, *submitted*

scattering, backscattering...
fractal geometry, particle restructuring...
wavelengths other than 550 nm...
organic carbon, water uptake...

modeling optical properties



refractive index

particle size

wavelength

spherical, homogeneous

Mie theory

coated Mie, RDG-FA, discrete dipole, etc.

absorption, scattering, asymmetry…

density

mass optical efficiency (m^2/g)

How do particles perturb the radiative-transfer field?

why do we need that refractive index?
...to represent changes in morphology

background

- large variability reported
  - in refractive index, absorption cross-section, and direct climate forcing
  - the buzz: “very uncertain”
- goal of our work
  - long-term: achieve *predictive capability* for the climatic impacts of carbonaceous aerosols
  - this presentation: obtain *most defensible estimates* of optical properties for uncoated (“naked”) soot

Secret fear:

What if this variability is unpredictable and we are stuck with a large uncertainty in radiative forcing?

take-home messages

These pertain to the material
called “black carbon” in climate models

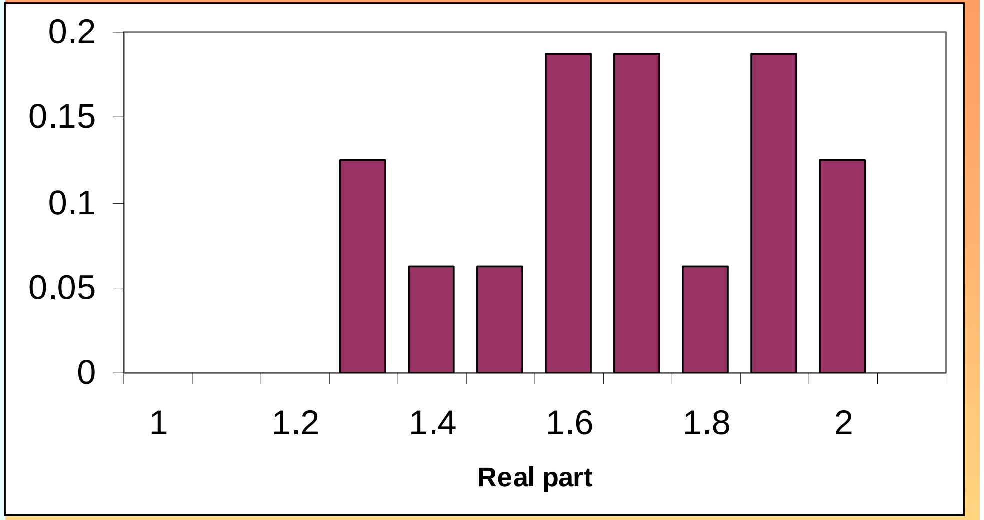
Default (OPAC) ***refractive index*** *should be changed. It represents an aerosol that is not dark enough.*

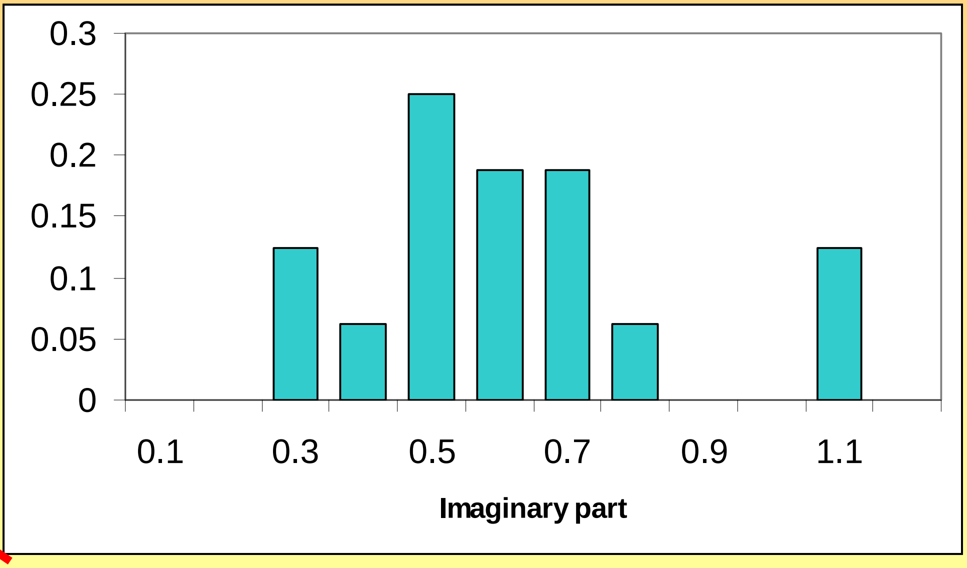
The central estimate of ***absorption cross-section*** *for uncoated carbon is 7.5* m^2/g.

Most variations in ***modeled direct forcing*** *can be explained by differences in particle optics and lifetime.*

refractive index: measurements

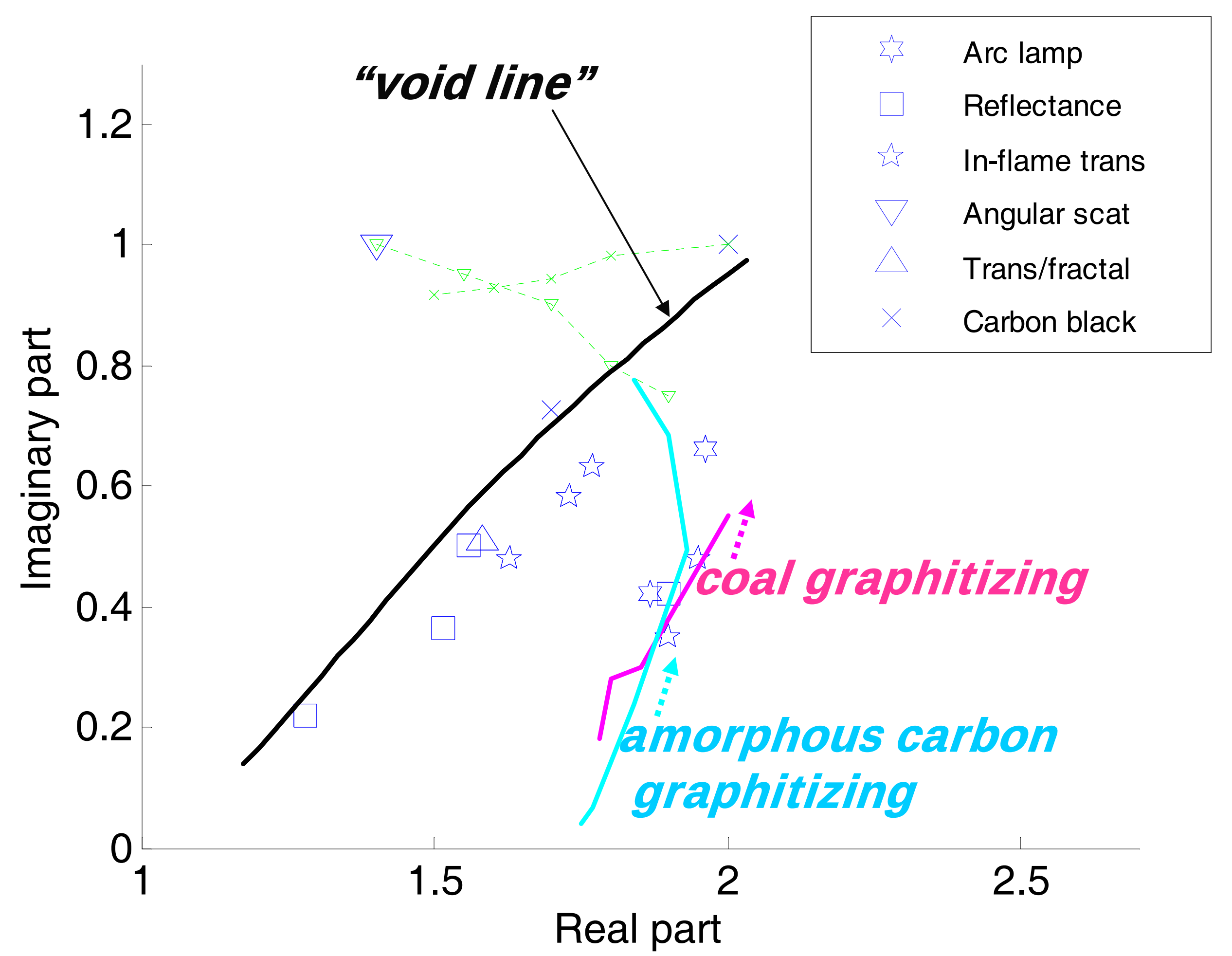
Reference	*Value*	TW	MR	H	FMK
Coal					
McCartney et al, 1965	(1.7-2.0)-(0.25-0.5)i	x			
Flame-generated carbon					
Batten, 1985	(1.20-1.35)-(0.1-0.22)i			x	
Dalzell and Sarofim, 1969	1.56-0.47i			x	
Janzen, 1979	2.00-1.00i				x
Pluchino et al, 1980	(1.7-1.8)-(0.6-0.8)i			x	
Senftleben and Benedict, 1917	1.9-0.65i	x			
Graphite					
Greenaway et al, 1969	2.6-1.25i	x			
"	1.5-0.005i	x			
Derived values					
McCartney and Ergun, 1961	2.02-0.56i		x		
Dalzell and Sarofim, 1969	1.84-0.46i		x		
Janzen, 1979	1.25-0.25i			x	x
Janzen, 1979	1.50-0.5i			x	x
Secondary references					
Ackermann and Toon, 1981	1.94-0.66i			x	
Bergstrom, 1972	1.95-0.66i				x
Bergstrom, 1973	2.0-0.66i			x	
Twitty and Weinman, 1971	1.80-0.8i				x
Hess and Herd, 1993	2.00-1.00i				x
Hanel, 1987	1.9-1.0i			x	
Jaenicke, 1988	1.75-0.44i			x	
Kattawar and Heard, 1976	1.95-0.66i			x	
Ouimette and Flagan, 1982	1.56-0.47i			x	
Roessler & Faxvog, 1980	1.96-0.66i			x	
Roessler & Faxvog, 1980	2.0-0.66i			x	
Shettle and Fenn, 1972	1.76-0.45i			x	
"various textbooks"	2.0-1.0i			x	





reviews:TW=Twitty & Weinman, 1971; MR=Medalia & Richards, 1981; H=Horvath, 1993; FMK=Fuller et al, 1999

refractive index: explanations



- lots of variation!
- some can be explained by material voids
- some meas. did not provide unique constraints
- some particles are partially graphitized

OPAC refractive index

origin:

Twitty & Weinman, 1971

Shettle & Fenn
WMO
d'Almeida, 1991

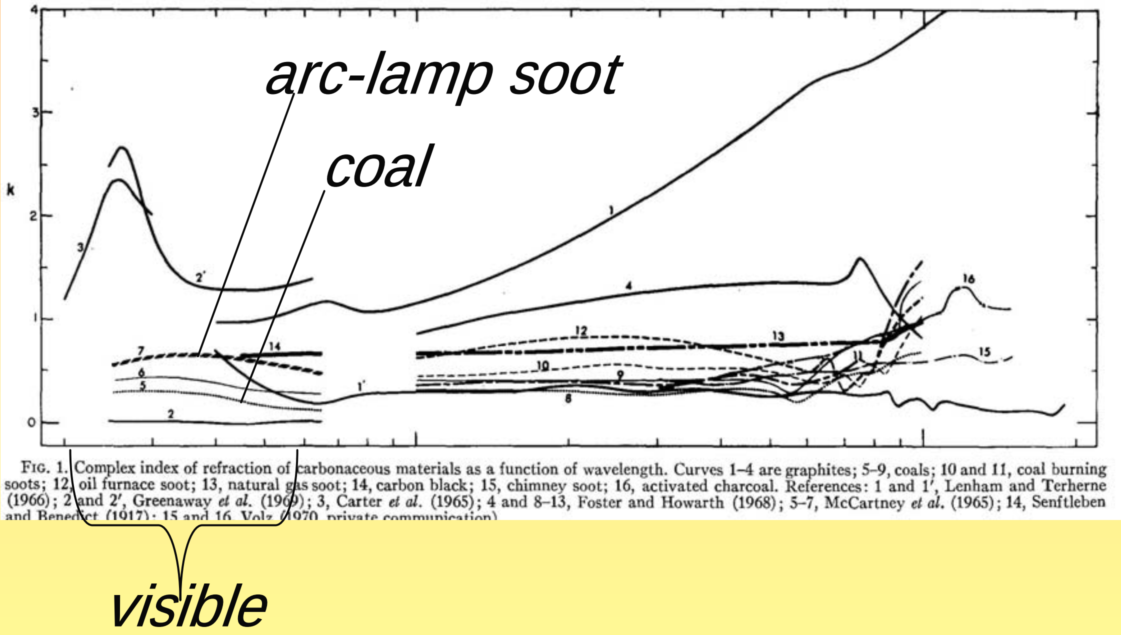
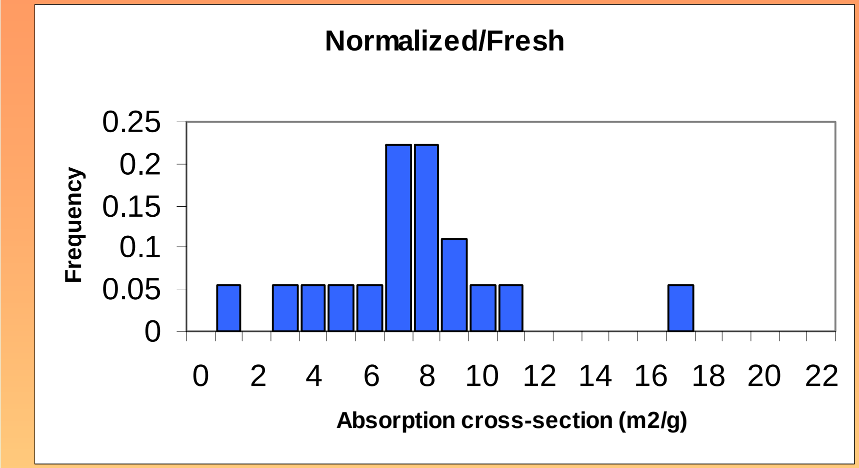


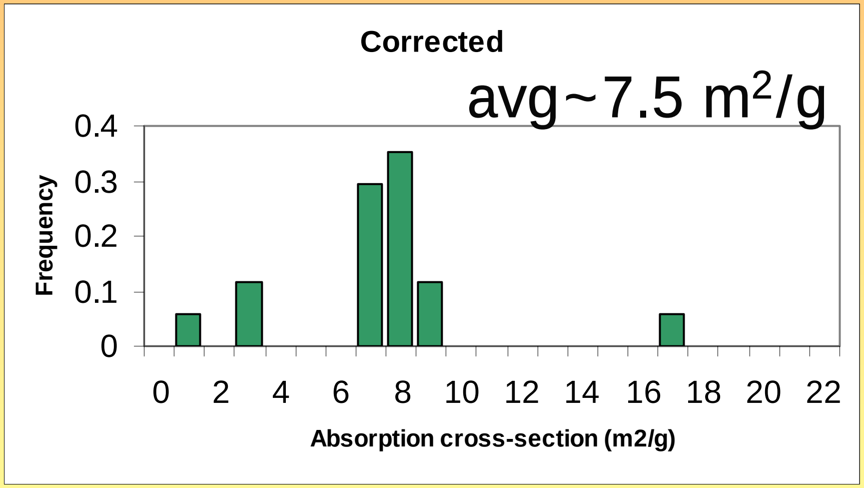
FIG. 1. Complex index of refraction of carbonaceous materials as a function of wavelength. Curves 1–4 are graphites; 5–9, coals; 10 and 11, coal burning soots; 12, oil furnace soot; 13, natural gas soot; 14, carbon black; 15, chimney soot; 16, activated charcoal. References: 1 and 1′, Lenham and Terherne (1966); 2 and 2′, Greenaway *et al.* (1969); 3, Carter *et al.* (1965); 4 and 8–13, Foster and Howarth (1968); 5–7, McCartney *et al.* (1965); 14, Senftleben and Benedict (1917); 15 and 16, Volz (1970, private communication).

Default (OPAC) ***refractive index*** *should be changed.*
It represents an aerosol that is not dark enough.

absorption cross-section: measured

Reference	*Cited by*	*Value*	*Wavelength*	*Adjusted to 550 nm **
MEASUREMENTS				
Diesel engine soot				6.8
Truex and Anderson, 1979	H, L	17	514	17
Japar and Szkarlat, 1981		8.9	514.5	8.3
Japar et al., 1984		8.5	514.5	8.0
Roessler and Faxvog, 1980	L	8.3	514.5	7.8
Schnaiter et al., 2003		6.6+/-0.4	550	6.6
Scherrer et al., 1980		9.0	550	7.4
Szkarlat and Japar, 1981	H, L	8.28	514	7.7
Szkarlat and Japar, 1983	H	10.9	500	8.1
Other combustion aerosol				
Bruce et al, 1991		4.55	488	4.0
Colbeck et al, 1987		7.5	632	8.7
"		7.5+/-0.6	632	8.7
Japar and Killinger, 1979		1.5		1.5
Lee, 1980		8.0	550	6.5
Mullins and Williams, 1984		4.1+/-0.1	450	3.2
Mulholland and Choi, 1998		5.9±0.1	633	6.8
"		6.9±0.1	633	7.9
Carbon black				
Donoian and Medalia, 1967		9.68	550	7.5
Other sources				
Schnaiter et al., 2003		2.9 +/- 0.5	550*	2.9 +/- 0.5
Ambient - fine				
Adams et al., 1990		10	514	9.3
Edwards et al., 1983	L	7-12	550	**need to look**
Groblicki et al., 1981	L	11.8	550	
Gundel et al., 1984	L	25.4		
Japar et al.,1981		8.0+/-0.4	550	
Japar et al., 1984		9.1	500	8.3
Japar et al., 1986		11.0, 12.1		check
Japar et al., 1986	L	9.8-12		check
Liousse et al., 1993		5	broadband	
"	L	20		
Wolff et al, 1980	L	12.7	550	10.6
Ambient - coarse				
Wolff et al, 1980	L	3.8	550	
CALCULATIONS				
Chylek et al, 1981	H	5	514	
Clarke and Noone, 1985	H	6.6-8		**check**
Jennings and Pinnick, 1980	L	3.5-8.6		
Nelson, 1989	L	8-10	550	
Rosen and Hansen, 1984	L	8.3-18.1	500	
SECONDARY REFERENCES				
Clarke, 1989	H	9.68		
Heintzenberg, 1982	L	9.68		
Clarke et al, 1987	L	9.68		
Japar et al, 1984		9.8		
Patterson et al., 1986	L	6.75		
Roessler, 1984	H	5		
Waggoner et al., 1981	L	5-11		


Normalized/Fresh


Corrected

avg~7.5 m^2/g

absorption cross-section

origin of 10 m²/g:

Donoian and Medalia, 1967

in water

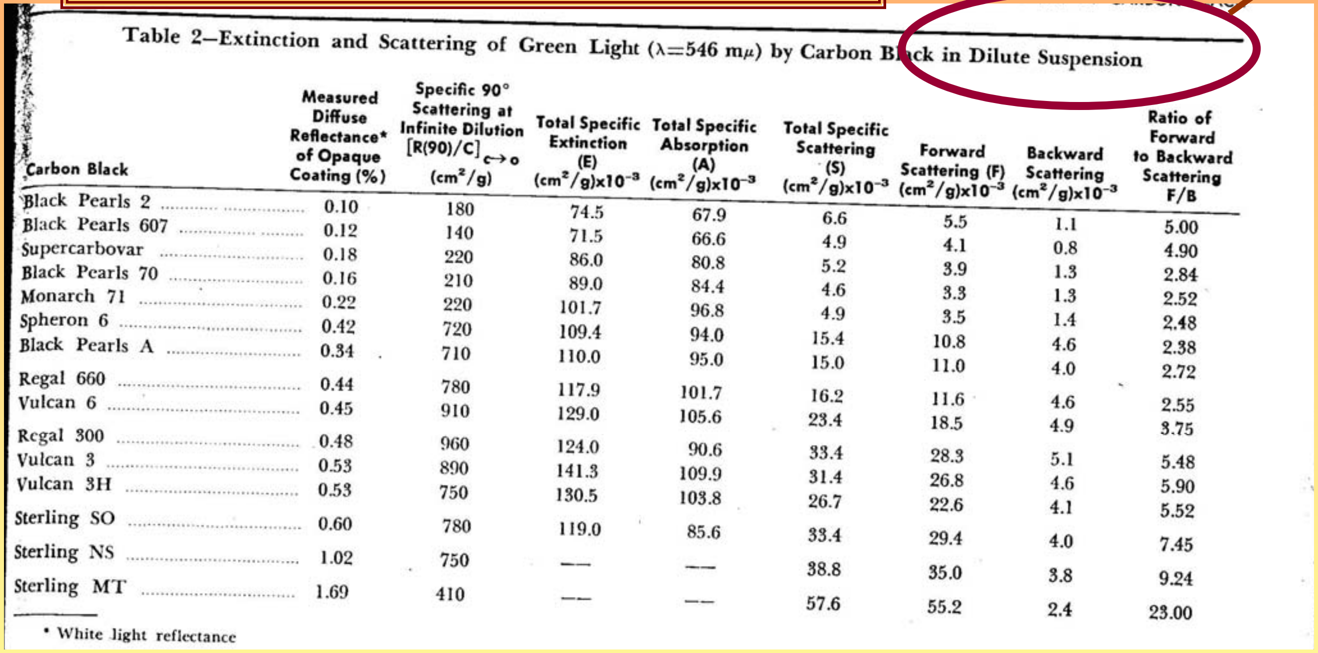


Table 2—Extinction and Scattering of Green Light (λ=546 mμ) by Carbon Black in Dilute Suspension

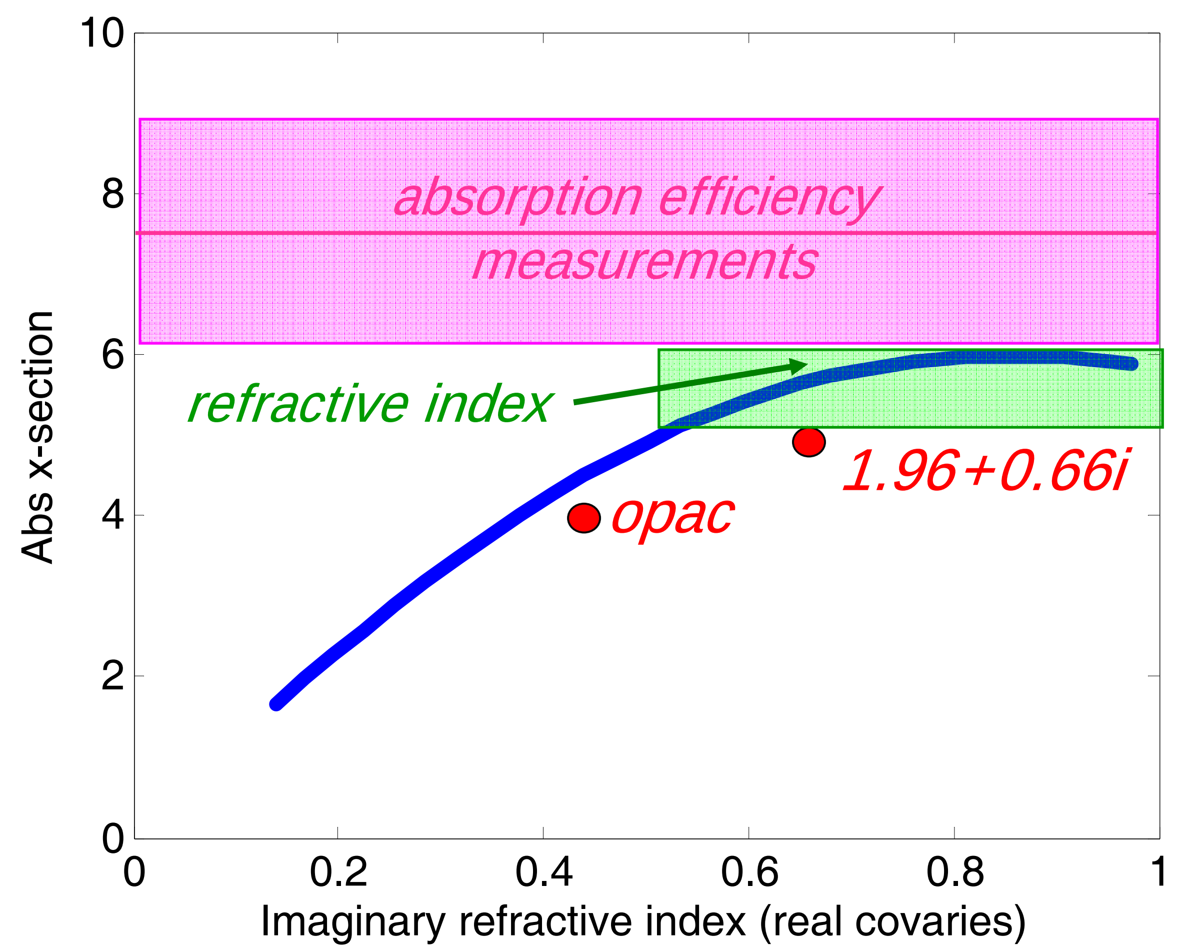
Carbon Black	Measured Diffuse Reflectance* of Opaque Coating (%)	Specific 90° Scattering at Infinite Dilution $[R(90)/C]_{c\to o}$ (cm²/g)	Total Specific Extinction (E) (cm²/g)x10⁻³	Total Specific Absorption (A) (cm²/g)x10⁻³	Total Specific Scattering (S) (cm²/g)x10⁻³	Forward Scattering (F) (cm²/g)x10⁻³	Backward Scattering (cm²/g)x10⁻³	Ratio of Forward to Backward Scattering F/B
Black Pearls 2	0.10	180	74.5	67.9	6.6	5.5	1.1	5.00
Black Pearls 607	0.12	140	71.5	66.6	4.9	4.1	0.8	4.90
Supercarbovar	0.18	220	86.0	80.8	5.2	3.9	1.3	2.84
Black Pearls 70	0.16	210	89.0	84.4	4.6	3.3	1.3	2.52
Monarch 71	0.22	220	101.7	96.8	4.9	3.5	1.4	2.48
Spheron 6	0.42	720	109.4	94.0	15.4	10.8	4.6	2.38
Black Pearls A	0.34	710	110.0	95.0	15.0	11.0	4.0	2.72
Regal 660	0.44	780	117.9	101.7	16.2	11.6	4.6	2.55
Vulcan 6	0.45	910	129.0	105.6	23.4	18.5	4.9	3.75
Regal 300	0.48	960	124.0	90.6	33.4	28.3	5.1	5.48
Vulcan 3	0.53	890	141.3	109.9	31.4	26.8	4.6	5.90
Vulcan 3H	0.53	750	130.5	103.8	26.7	22.6	4.1	5.52
Sterling SO	0.60	780	119.0	85.6	33.4	29.4	4.0	7.45
Sterling NS	1.02	750	—	—	38.8	35.0	3.8	9.24
Sterling MT	1.69	410	—	—	57.6	55.2	2.4	23.00

* White light reflectance

10 m²/g
for fresh
“soot”?

The central estimate of ***absorption cross-section*** *for uncoated carbon is 7.5 m²/g.*

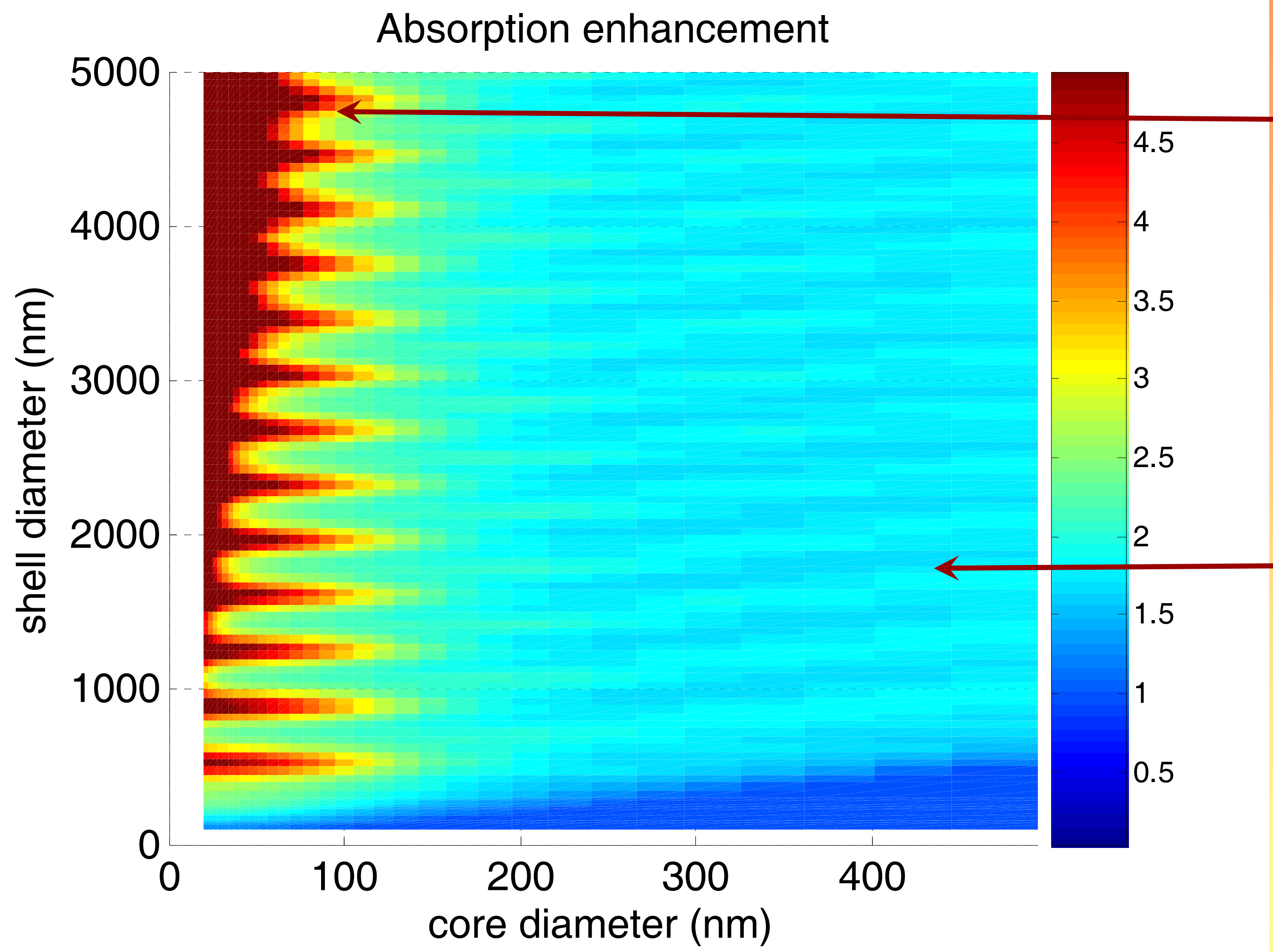
closure: refractive index & absorption



- refractive index for *graphitized carbon*: somewhere on “void line”
- consistent values: 1.7+0.7i, 2+1i
- lower imaginary r.i. possible; *depends on formation*
- cross-section higher than predictions ...agglomeration?

assume: small particles (constant mass absorption efficiency), density=1.8 g/cm^3

morphology: coating (overview)

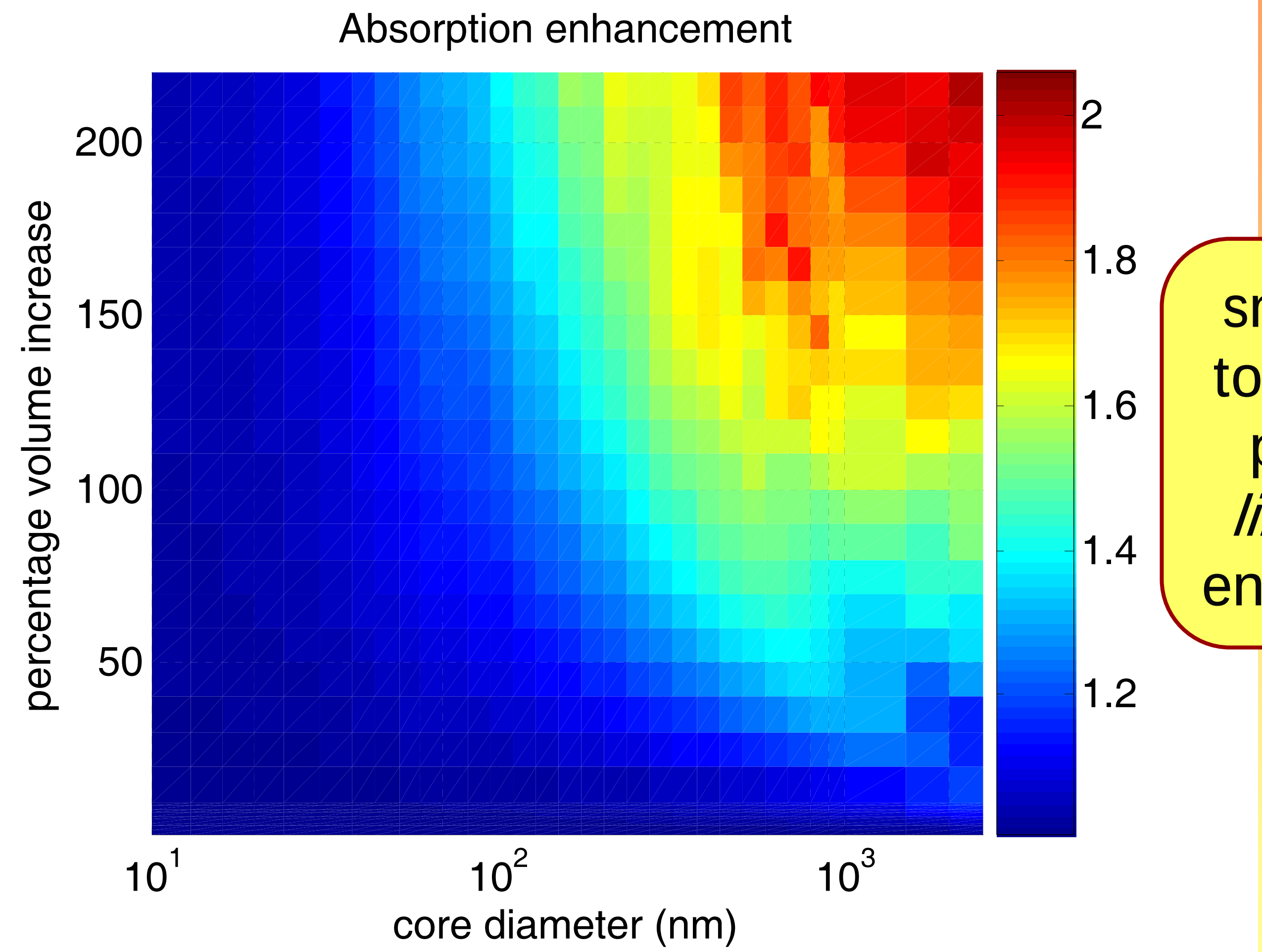


Can be very high, but very unlikely

Mostly constant ~1.8

Main goal of this investigation:
identify “mixing regimes” appropriate for use in models

morphology: coating (example)

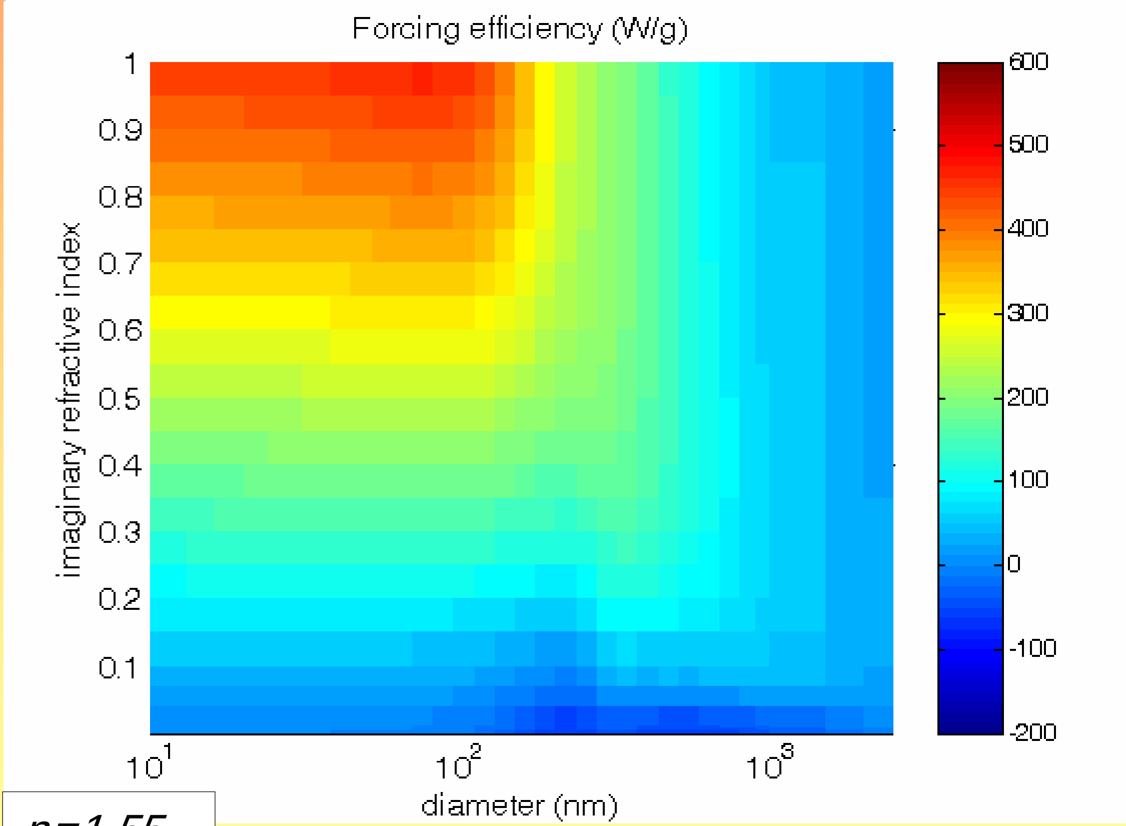


small vol. additions to combustion-sized particles result in *limited* absorption enhancement (<1.4)

“simple forcing efficiency”

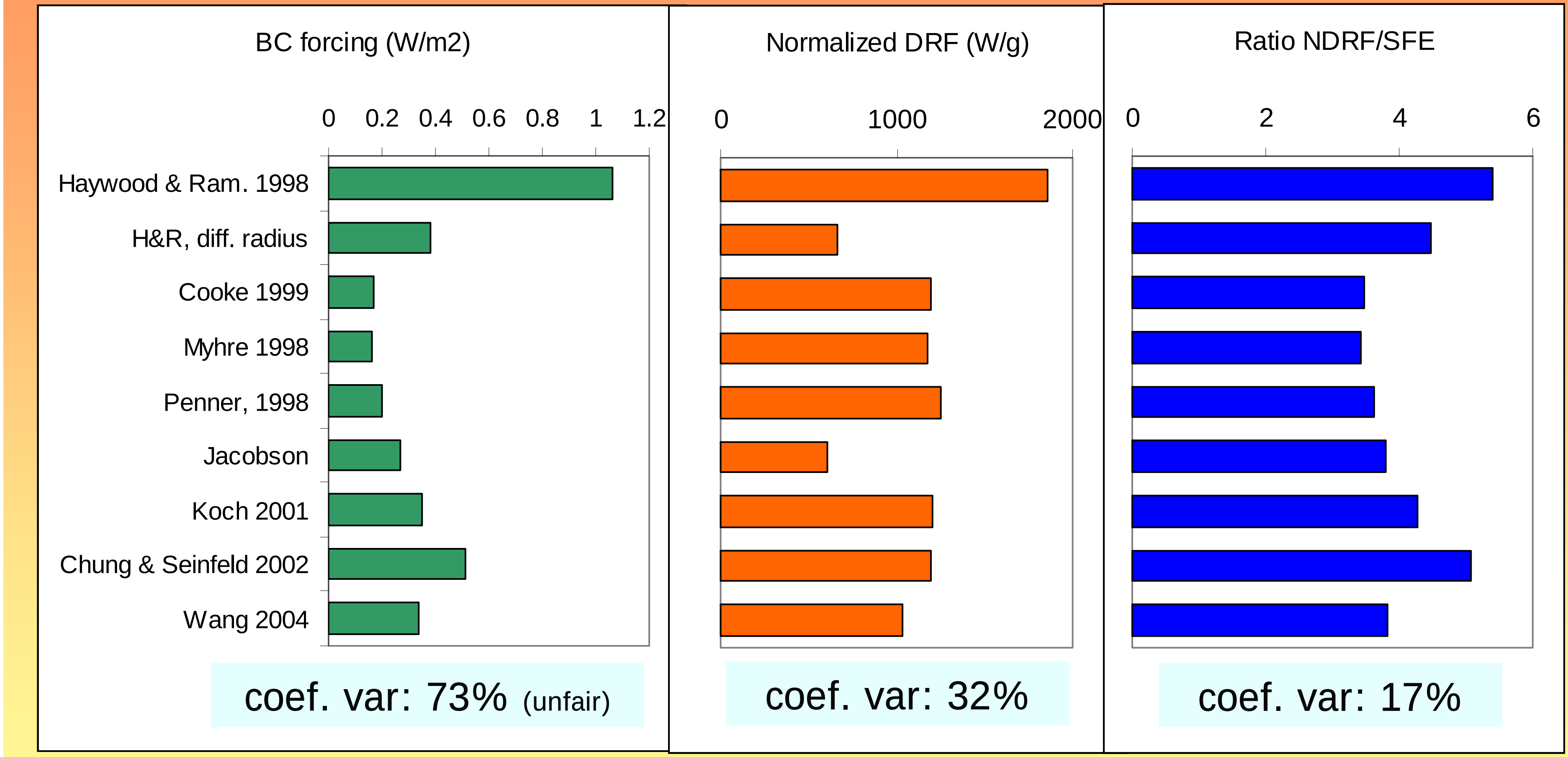
simplistically,
(Chylek&Wong 1995)

$$\frac{S_0}{4}\cdot\left(\tau_{atm}\right)^2\cdot\left(1-F_c\right)\cdot\left[2\cdot\left(1-a_s\right)^2\cdot\beta\cdot MSC-4\cdot a_s\cdot MAC\right]$$



n=1.55
uncoated

comparison of modeled forcing



coef. var: 73% (unfair)

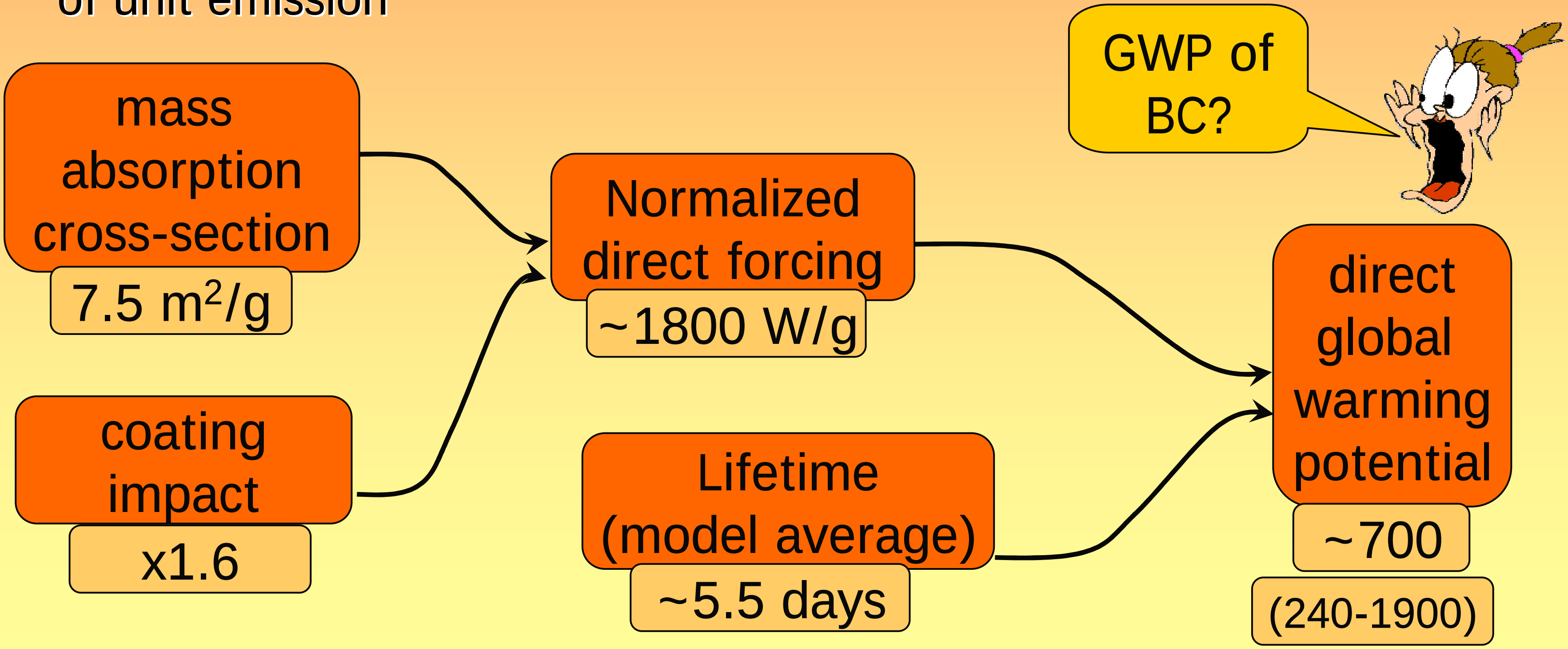
coef. var: 32%

coef. var: 17%

*Most variations in **modeled direct forcing** can be explained by differences in morphology and aerosol lifetime.*

“Consensus”?

- positive forcing is *nearly linear* in absorption coefficient
- simple forcing efficiency + lifetime explain most differences in predicted forcing
- only lifetime & optics are needed to estimate *direct radiative* impact of unit emission



mass absorption cross-section

7.5 m^2/g

coating impact

x1.6

Normalized direct forcing

~1800 W/g

Lifetime (model average)

~5.5 days

direct global warming potential

~700

(240-1900)

GWP of BC?

take-home messages

These pertain to the material
called “black carbon” in climate models

Default (OPAC) ***refractive index*** *should be changed. It represents an aerosol that is not dark enough.*

The central estimate of ***absorption cross-section*** *for uncoated carbon is 7.5* m^2/g.

Most variations in ***modeled direct forcing*** *can be explained by differences in particle optics and lifetime..*

We combine model outputs to produce a ***GWP for BC***.