Definitional Issues

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2 May 2005

IPCC Aerosol Meeting

Geneva, Switzerland

Or, in other words…

whaaat? we don't even know what it is and you want us to add up emissions?

I mean, it *is* rather uncertain and all…

Relax.

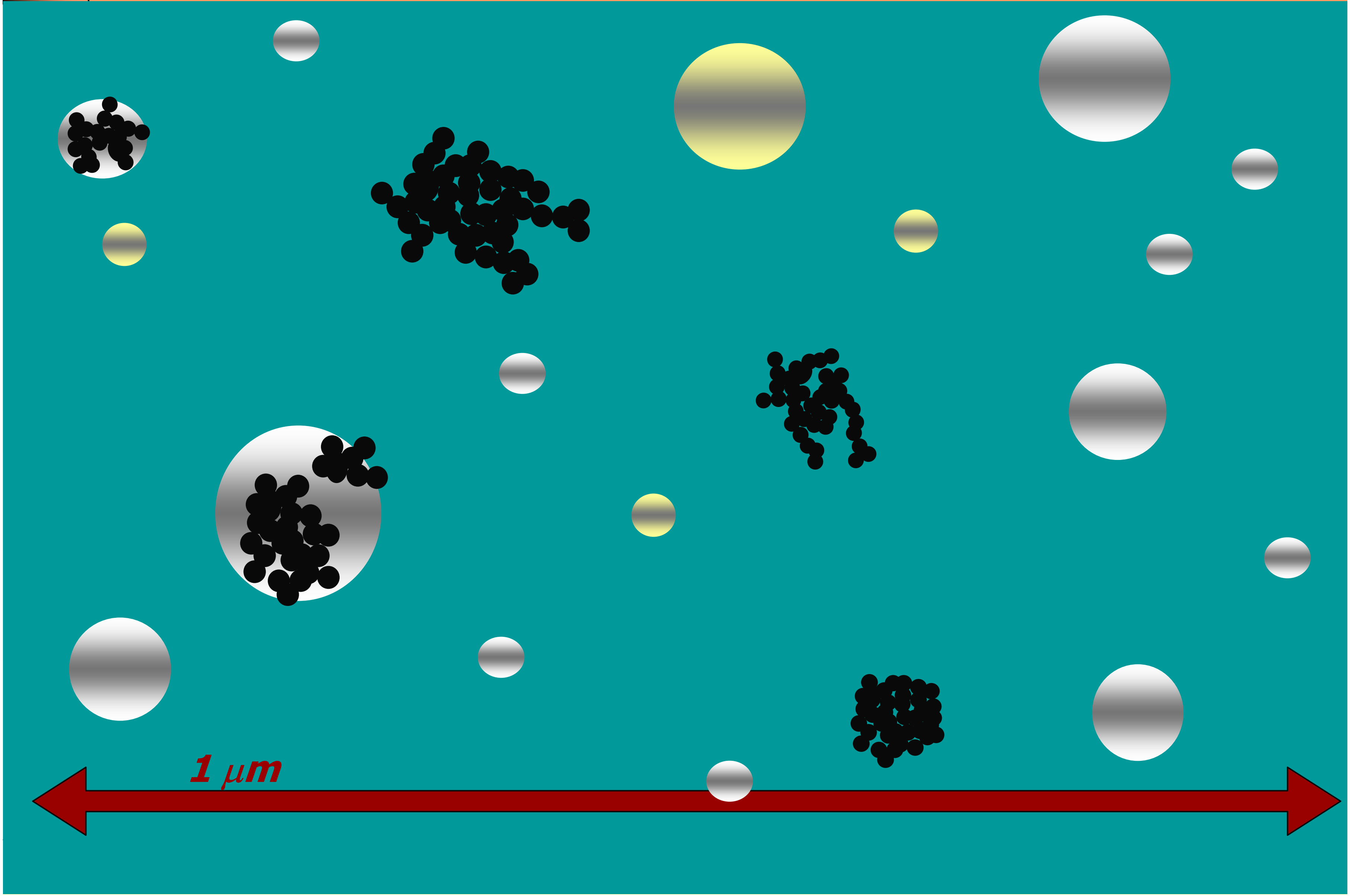
Questions I think we are asking

- Climatic effects of carbonaceous particles depend on composition.
- There are thousands of carbon compounds.
  (obviously, we will not model each individually)
- What divisions between carbon types *must* we draw to represent effects on climate?
- Can we measure those divisions in practice?

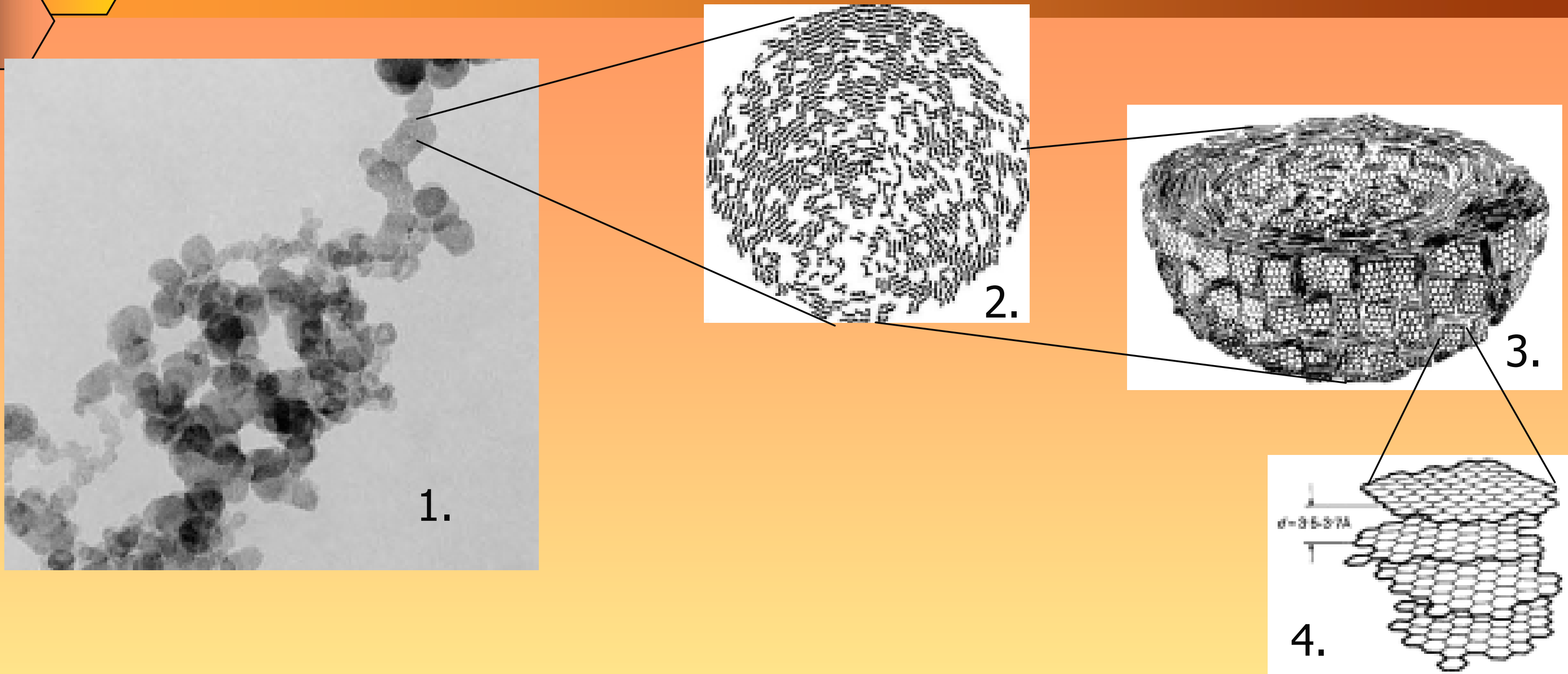
What I'll cover here

1. carbon particles… hmm.
   a. could you identify a black carbon particle
      …if you had nanovision?
   b. Houston, we have a (measurement) problem
   c. what can we do with all these names?
2. modeled climate forcing
   a. overview: forcing calculations
   b. the pieces: direct forcing
   c. some comments: indirect forcing
3. summary: role of definitions

If you had microvision…



Nanovision (and a very small scalpel)



1. *Micrograph of diesel soot:* Stanmore, Brilhac, and Gilot, *Carbon.* **2001**, *39* , 2247-2268.
2. *Structure of spherule extracted from HRTEM image*: Palotás *et al.*, *Energy and Fuels* **1996**, *10* , 254-259.

3, 4. *Structure of spherule and layers inferred from electron microscopy and X-ray diffraction:* Heidenreich, Hess and Ban, *J. Appl. Crystallography* **1968**, 1, 1-19

Structure has been known for many years.
Heckman, F.A. Microstructure of carbon black.
Rubber Chem. Technol. **1964**, *37* , 1245-1298.

Nanovision (and a very good chemistry set)

thousands of compounds
much carbon and hydrogen
sometimes oxygen &
a little nitrogen

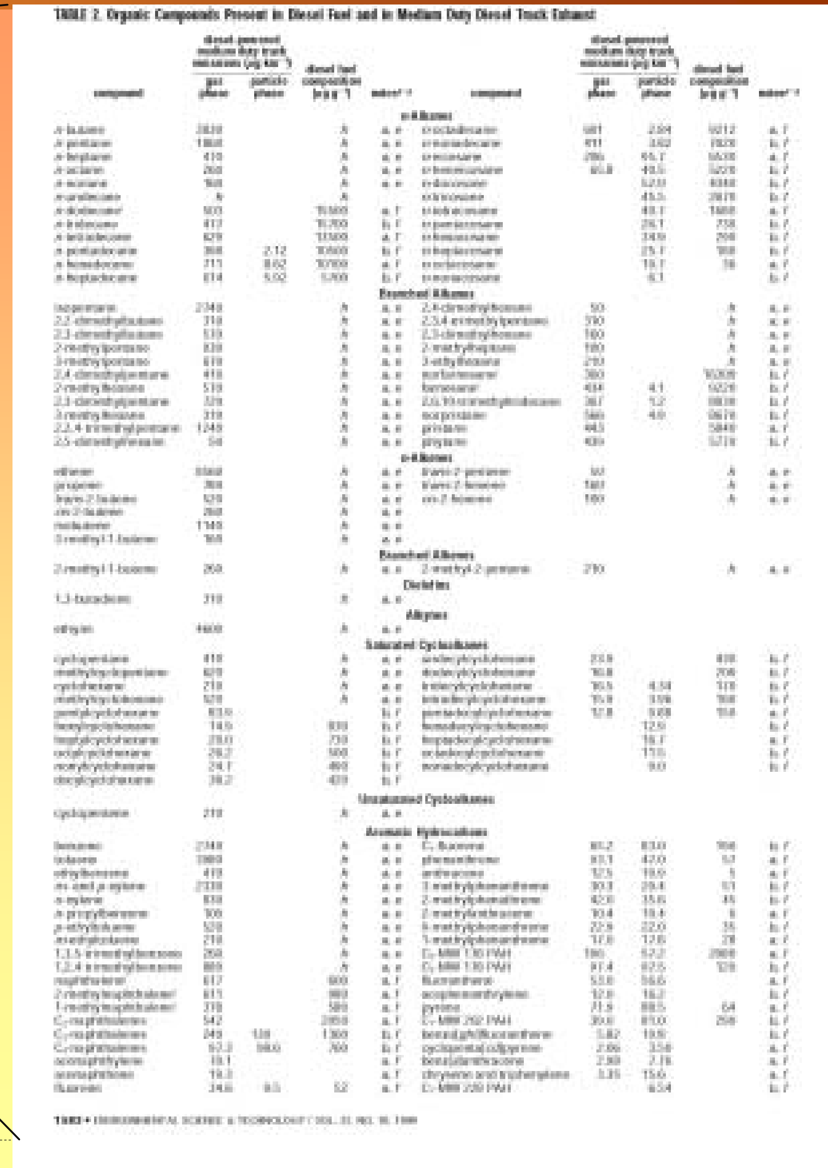


TABLE 2. Organic Compounds Present in Diesel Fuel and in Medium Duty Diesel Truck Exhaust

n-Alkanes

Branched Alkanes

n-Alkenes

Branched Alkenes

Diolefins

Alkynes

Saturated Cycloalkanes

Unsaturated Cycloalkanes

Aromatic Hydrocarbons

Schauer, J.J.; Kleeman, M.J.; Cass, G.R.; Simoneit, B.R.T., *Environ. Sci. Tech.* **1999**, *33* , 1578-1587.

Information from combustion research

- "soot" appears to form in a very narrow region (now you don't see it, now you do)
- implies there is a sharp difference between "special" BC and other
- caveat: most formation studies look only at simplified situations; is this true for real combustion?

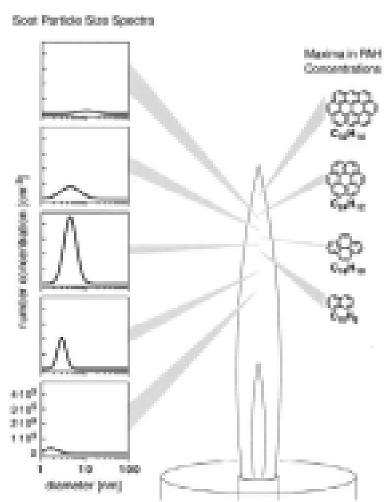
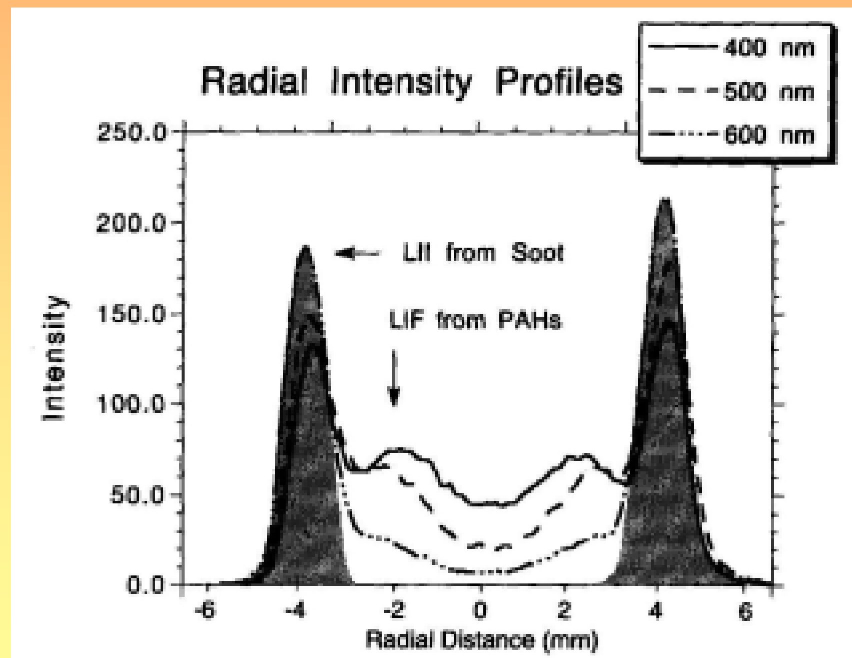


Fig. 2. A representation of the methane flame with PAH-maxima and soot particle size distributions, assigned to the respective height in the flame.

Siegmann, K.; Sattler, K.; Siegmann, H.C. *J. Electron Spectrosc. Rel. Phenom.* **2002**, *126*, 191-202.



Van der Wal, R.L.; Jensen, K.A.; Choi, M.Y. *Comb. Flame.* **1997**, *109*, 399-414.

Measurement goals (I)

We can't measure climate forcing by individual constituents.

- Aerosol concentrations are patchy

  Individual measurements don't represent the globe.

- Satellites can measure globally

  but they can't distinguish components, nor tell the difference between anthropogenic and natural.

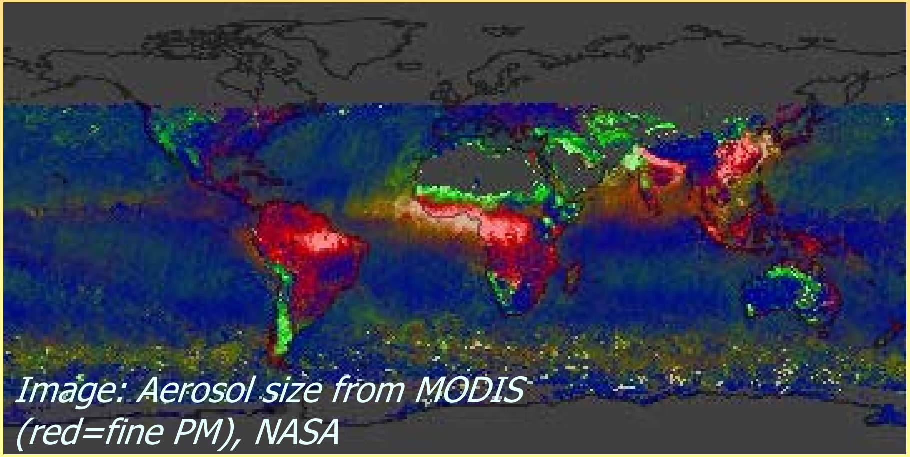


Image: Aerosol size from MODIS (red=fine PM), NASA

Measurement goals (II)

Thus, we have to model the forcing. Measurements can:

- Provide model inputs
- Corroborate model results

For this purpose, the measurements must:

- Distinguish between important groups of carbon compounds
- Measure a *conserved* property

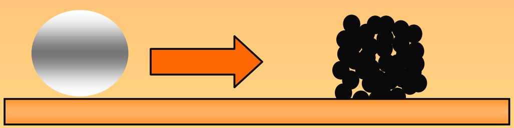
measurement tools

- combustion analysis → total carbon — OK *
- light absorption → dark carbon — next
- thermal-optical analysis
  → some carbon division (light/dark?) — next
- gas chromatography/mass spectrometry
  → individual species — identifies 10-20% of carbon
- Fourier Transform Infrared
  → functional groups — useful but expensive
- soluble fraction — real-time ability is new
- single-particle analysis — useful but expensive

* *not immune to measurement artifacts, though*

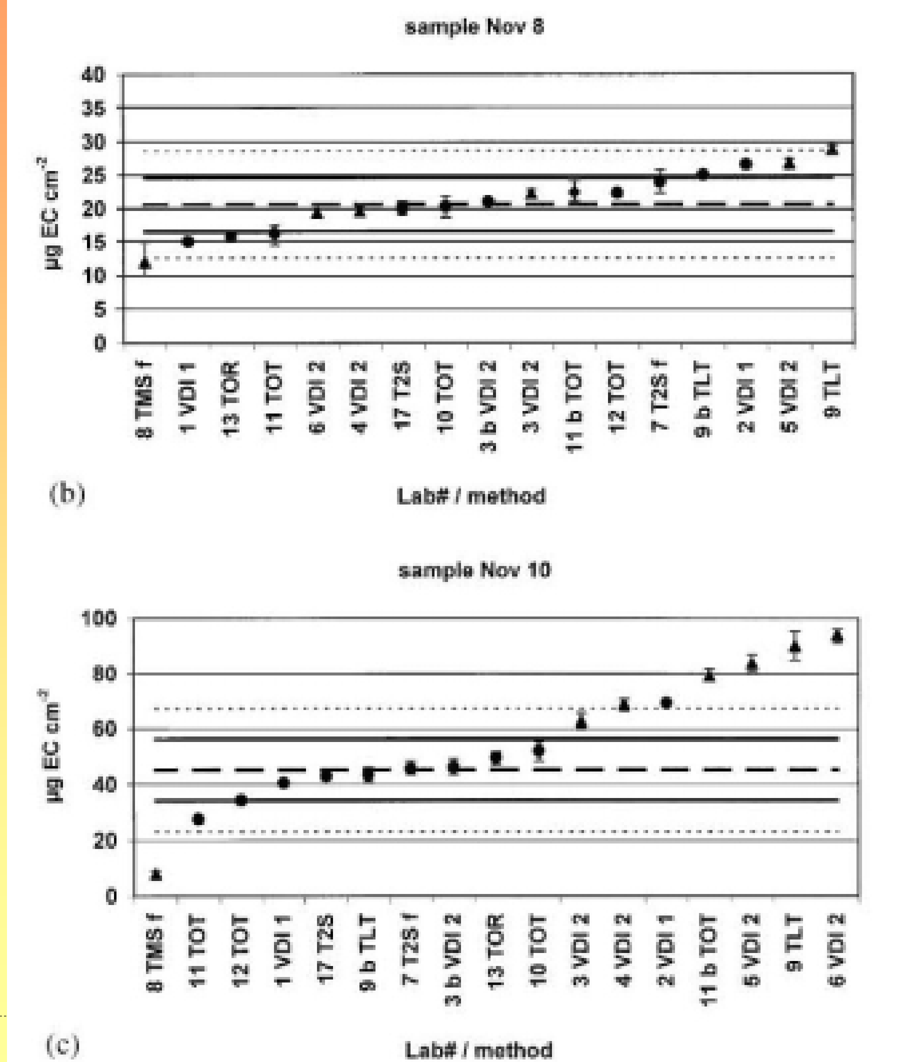
measurements: thermal

- Principle: Heat, measure carbon released at different temperatures
- Complication 1: Charring during analysis



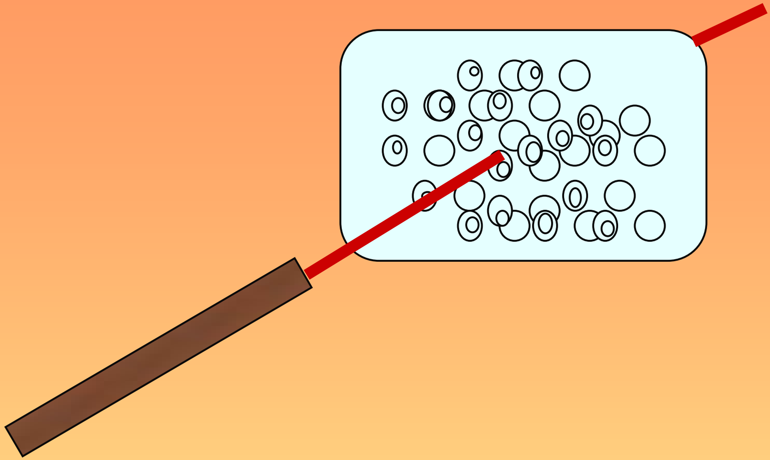
- Complication 2: Oxidation rates can vary in different mixtures
- Different protocols (rates/ magnitudes of heating) yield different results

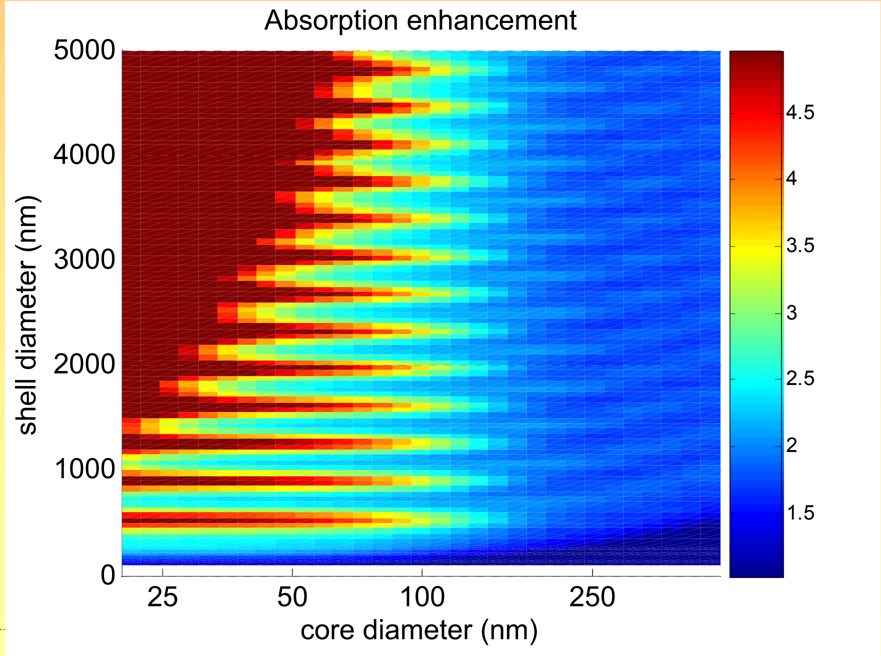
Interlab comparison of same filter



measurements: optical

- Principle: Collect particles on filter; monitor transmittance
- Complication 1: Amplification by filter (x2-4)
- Complication 2: Absorption per mass changes with particle form
- Complication 3: Non-carbon absorbers (dust?)
- Drawback: Doesn't measure negligibly-absorbing particles
- Different protocols (filters, wavelengths) and locations yield different results





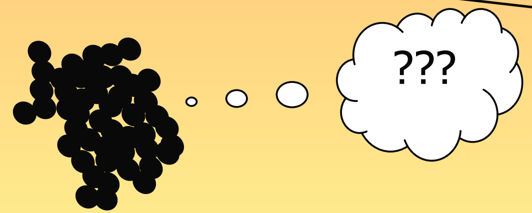
All those names!

By the way, there's little enough information that we use *any* of these to inform models.

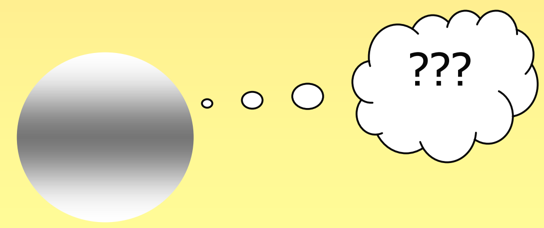
graphitic carbon
"substance that almost looks like graphite"
"result of Raman spectroscopy"

elemental carbon
"substance we talk about for air quality"
"result of thermal measurement"

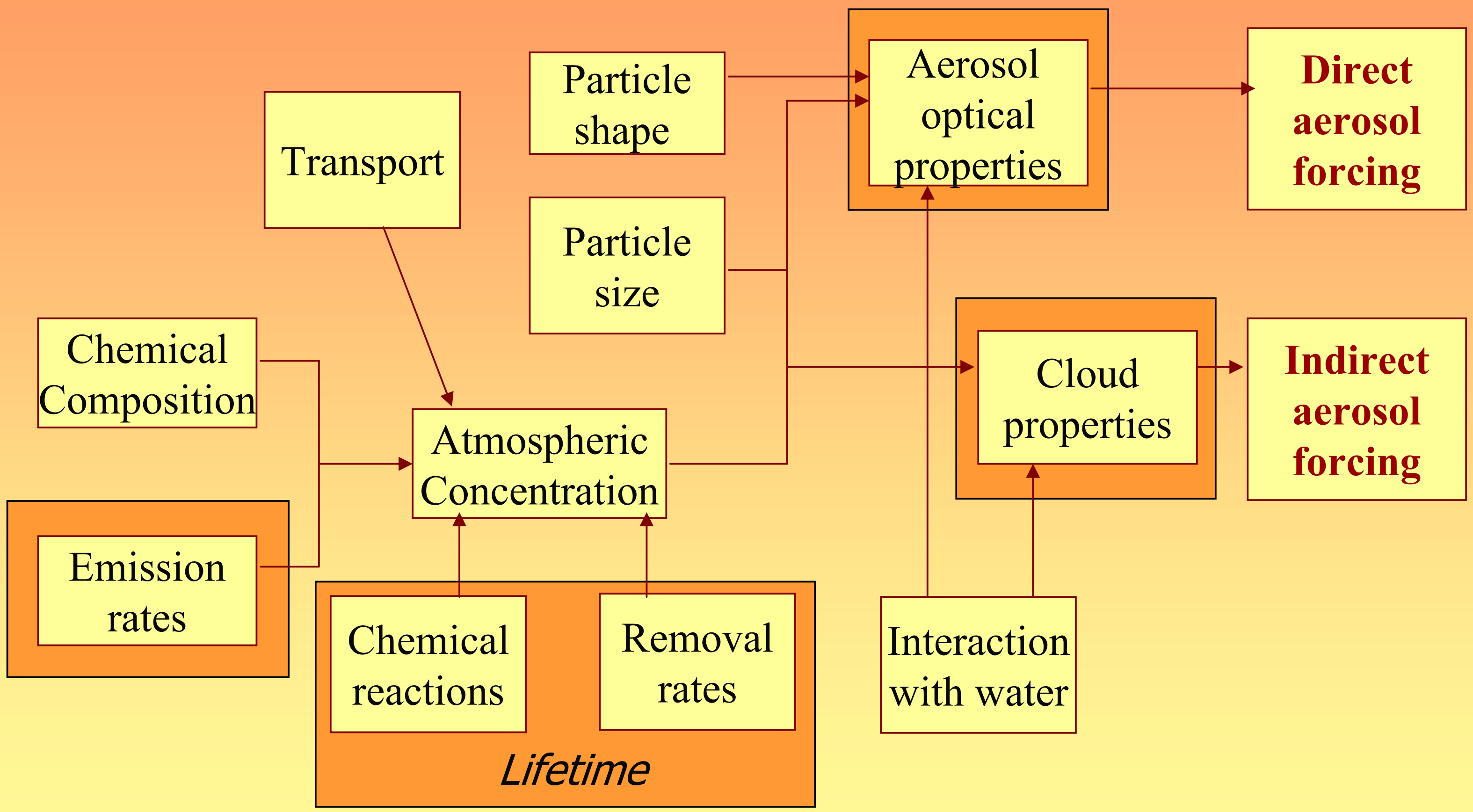
black carbon
"substance used in climate models"
"result of optical measurement"



organic carbon
"everything else"



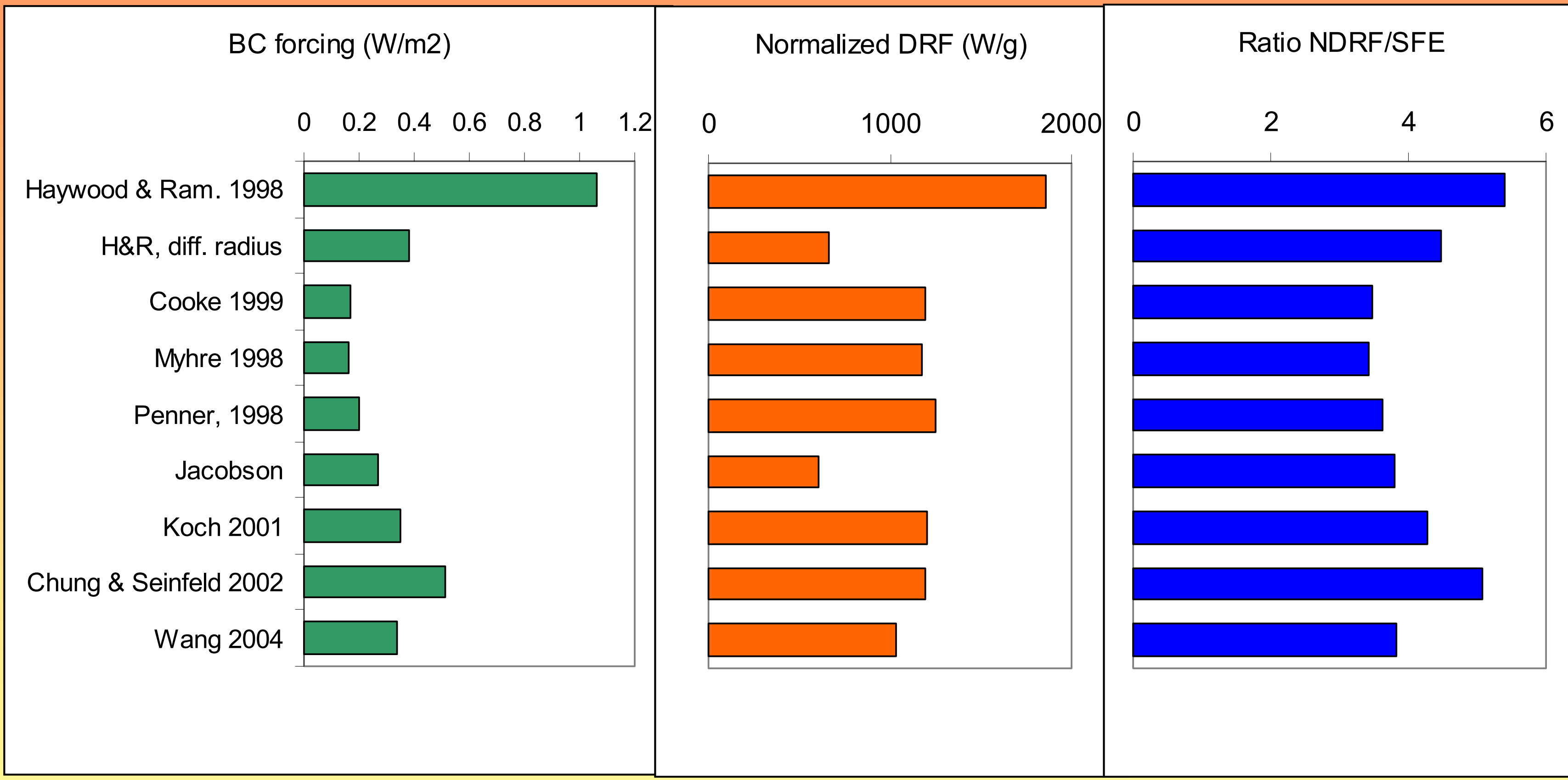
Modeling procedure



Aerosol lifetime estimates

- Few ways to measure; must be modeled
  - Depends on meteorology (esp. rainfall)
  - Estimates are about ~1 week
- Model <u>corroborated</u> by comparison with measurements
- Hard to make that comparison unless quantity measured is invariant!

Model comparisons: black carbon



coef. var: 73% (unfair)

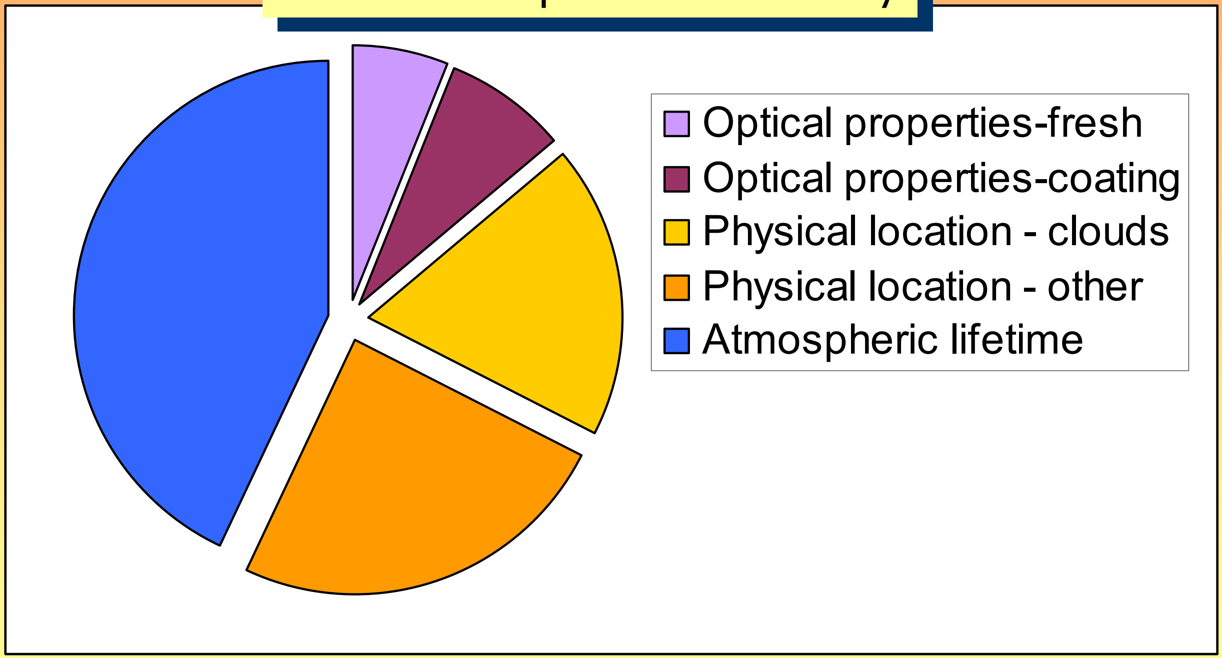
accounting for emission & lifetime differences coef. var: 32%

accounting for optical property differences coef. var: 17%

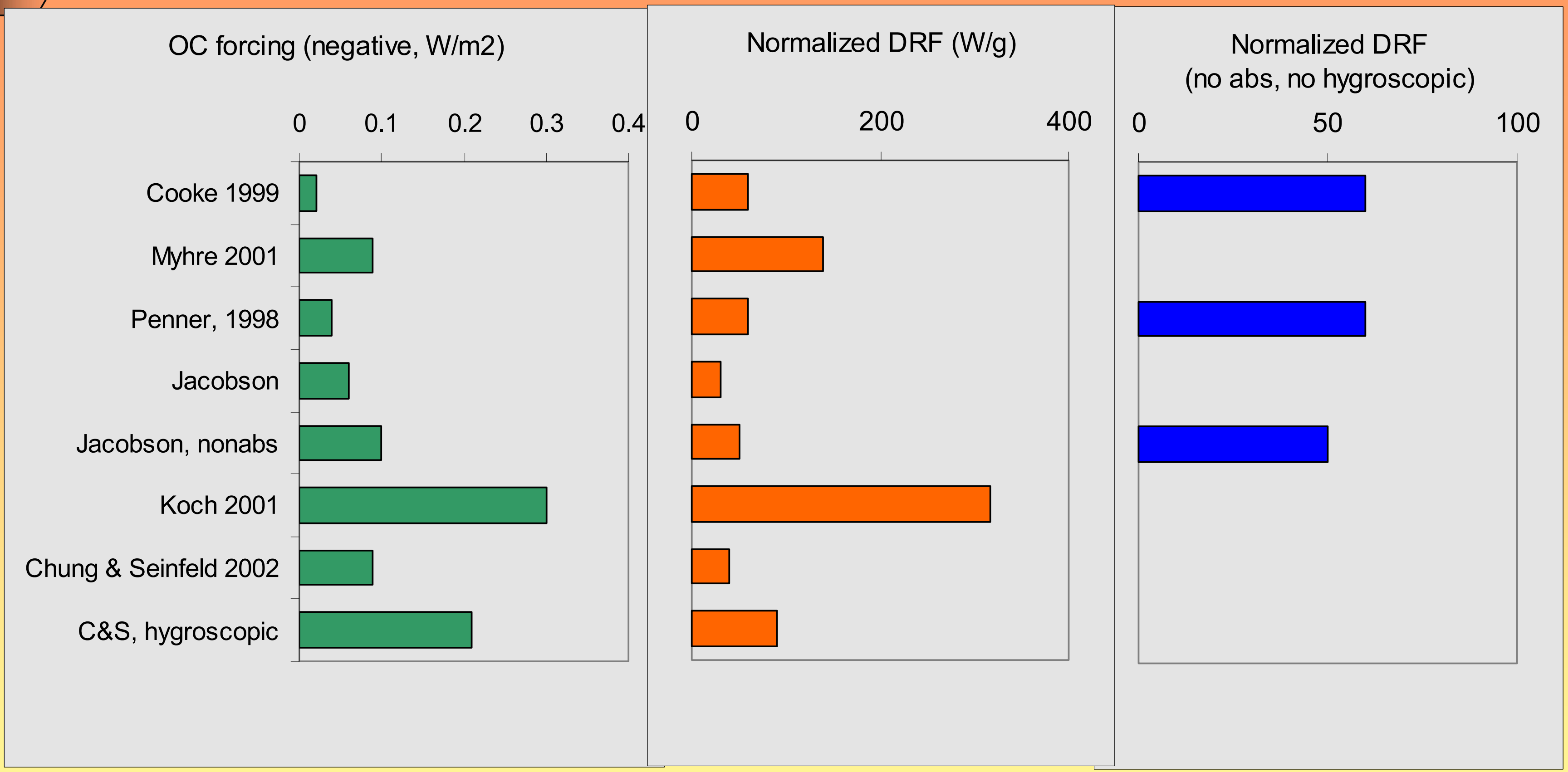
Variation in model results

Modeled integrated forcing by BC during its lifetime:
860 MJ/gram emitted (350-2000)

Sources of squared uncertainty



Model comparisons: organic carbon



coef. var: 83% (unfair)

accounting for emission & lifetime differences coef. var: 96%

accounting for some property differences coef. var: 10%

Indirect forcing: some questions

- Which aerosols dominate number concentration in critical regions?
  - Primary? (BC, dust, some organic)
  - Precursors of nucleation?
- Which aerosols/precursors affect cloud droplet number and size?
  - Solubility, or other better metric?

How defined classes of carbon affect forcing estimates

Uncertainties remain in:

- Emission estimates
  (what's the conserved quantity?)
- Model corroboration
  (what's the lifetime?)
- Species representation
  (what are the properties?)