Global Aerosol Microphysics Models: Tools for Assessing Uncertainties in the Indirect Effect

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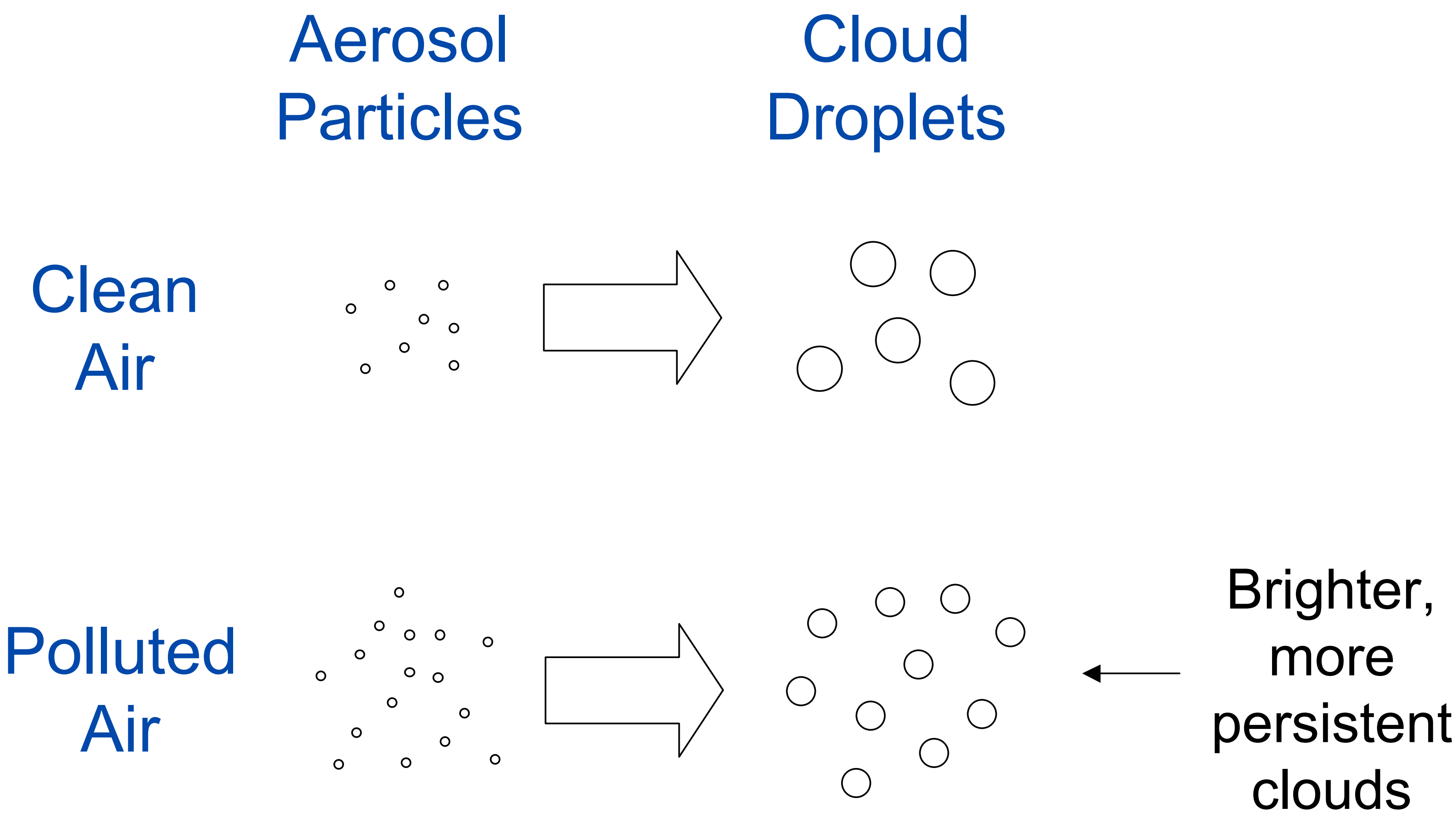
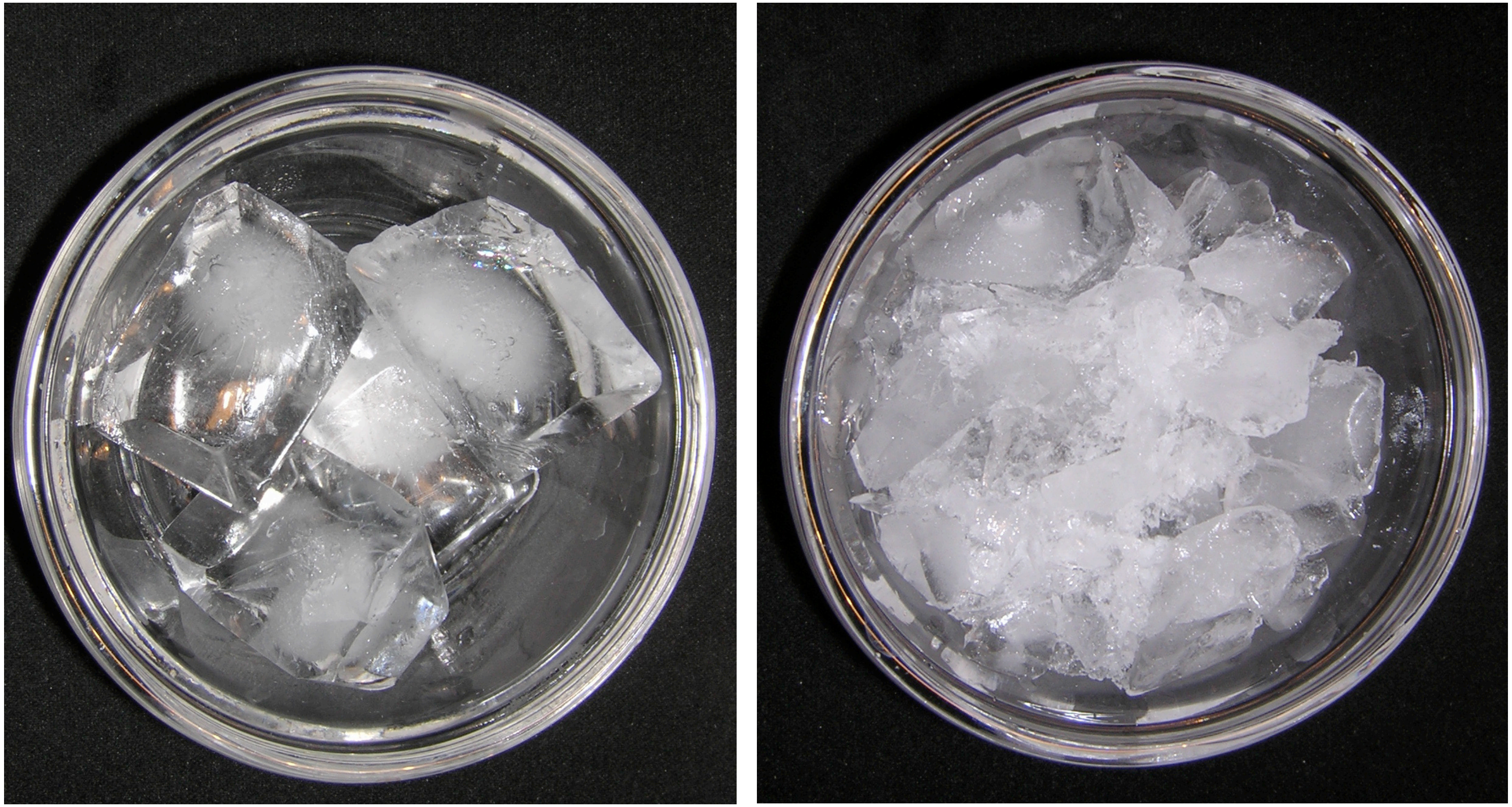
Themes and Overview

- ♣Goal: Global CCN prediction
- ♣Methodological
  - Mechanistic models
  - Quantify importance of processes
- ♣Sea-salt and mineral dust
- ♣Carbonaceous
  - Size/number vs chemical solubility
  - POA vs SOA

Indirect Effect on Climate



More Surface Area = More Scattering



Photos by Amy Sage at CMU

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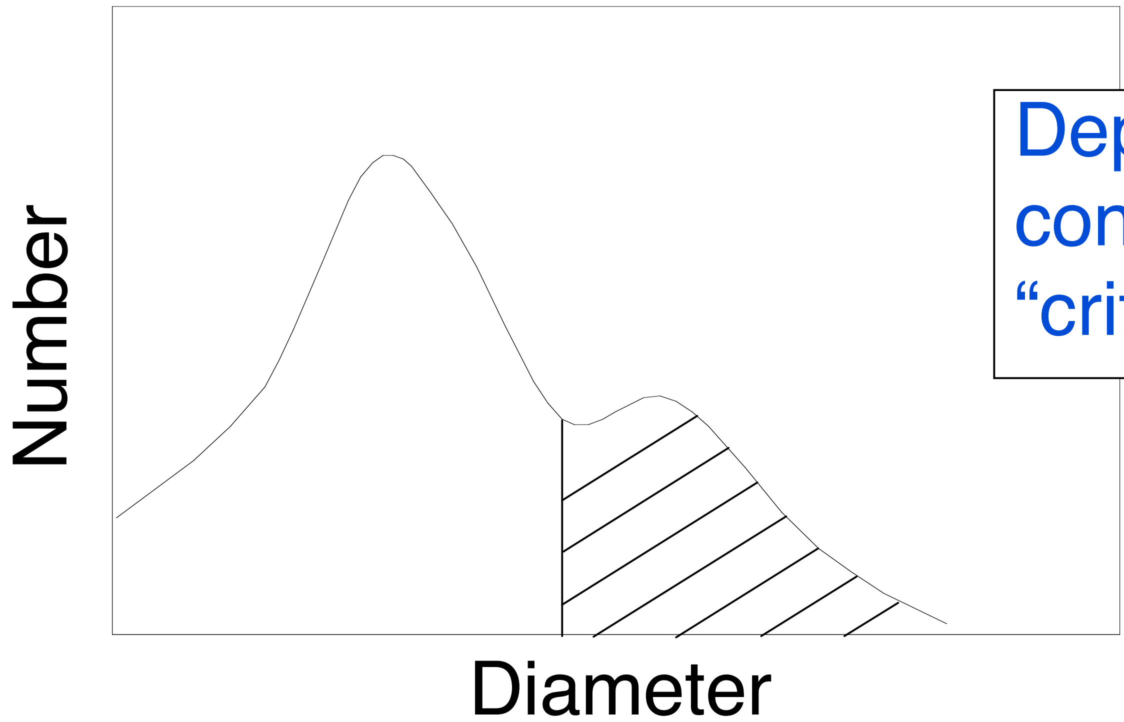
Cloud Condensation Nuclei (CCN)

- "Activation" = formation of cloud droplet from aerosol particle
- involves a competition between solute and surface tension effects (Kohler)

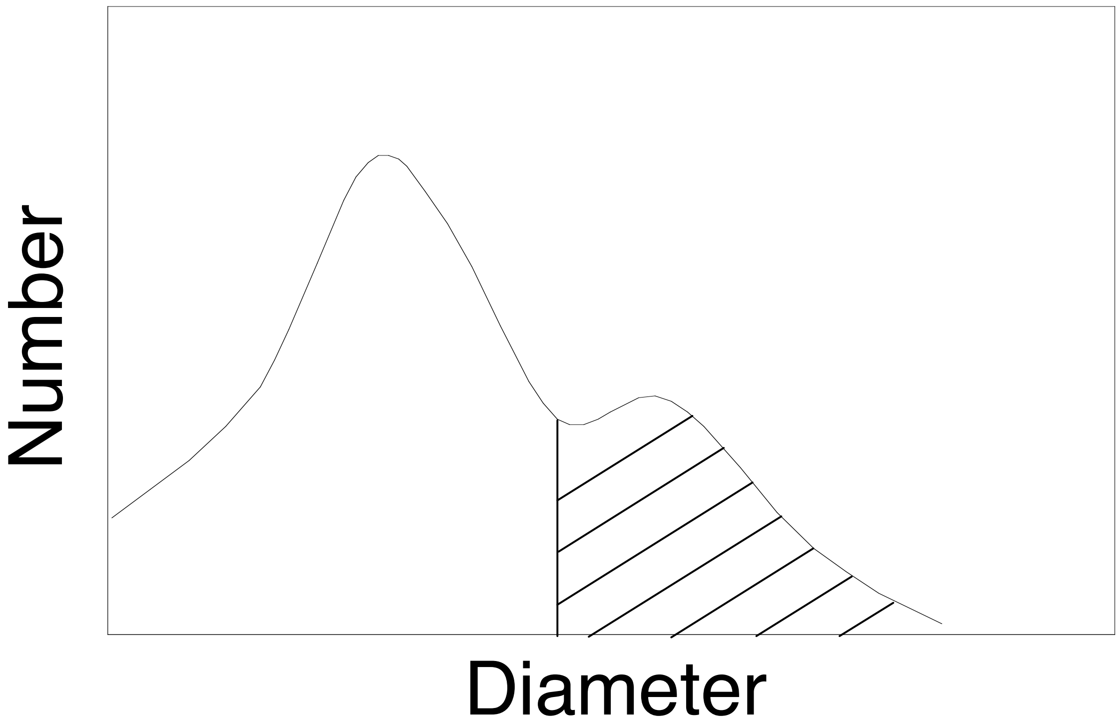


Depends on number concentration above "critical diameter"

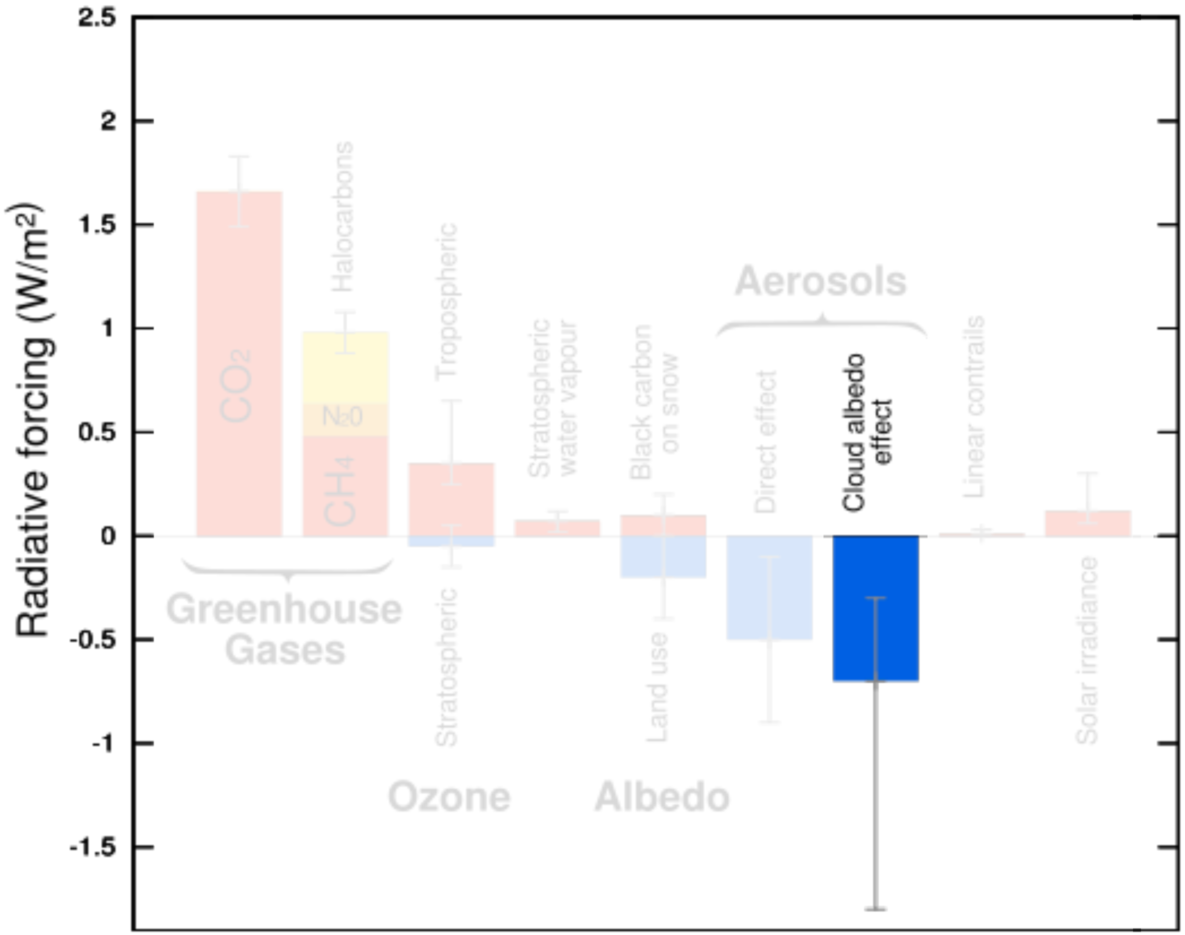
Cloud Condensation Nuclei (CCN)

Critical diameter depends on:

- Composition
- Mixing state
- Ambient supersaturation (S = 0.2% as a metric)



Forcings Comparison



Intergovernmental Panel on Climate Change (IPCC): 4th assessment report

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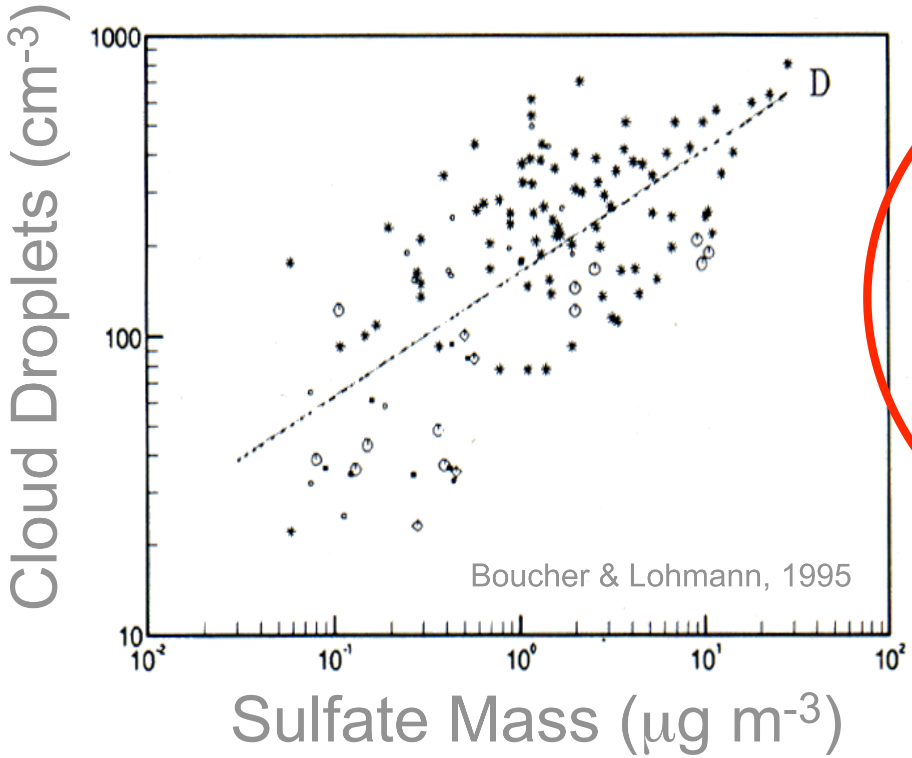
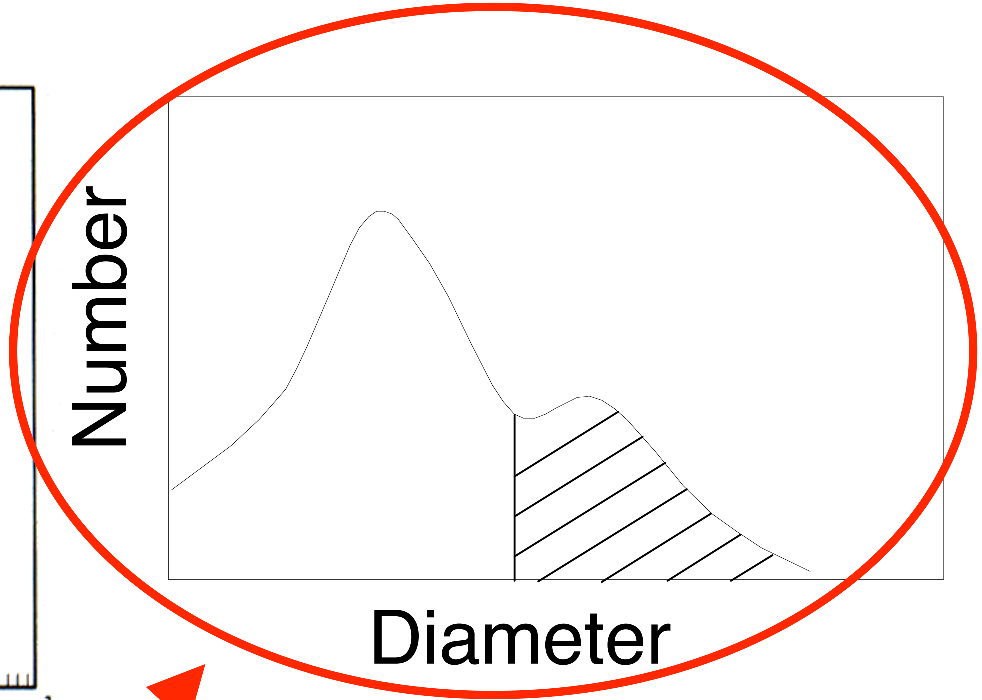
Two Model Types





Empiri

- ♣ Bulk
- ♣ Emp

We have built this for a global climate model

Mechanistic:

- ♣ Microphysical model
- ♣ Modified Köhler theory

Limitations of Empirical Approach

I: *Martin et al*. [1994]:

-0.68 W/m^2

II: *Martin et al.* with background CCN:

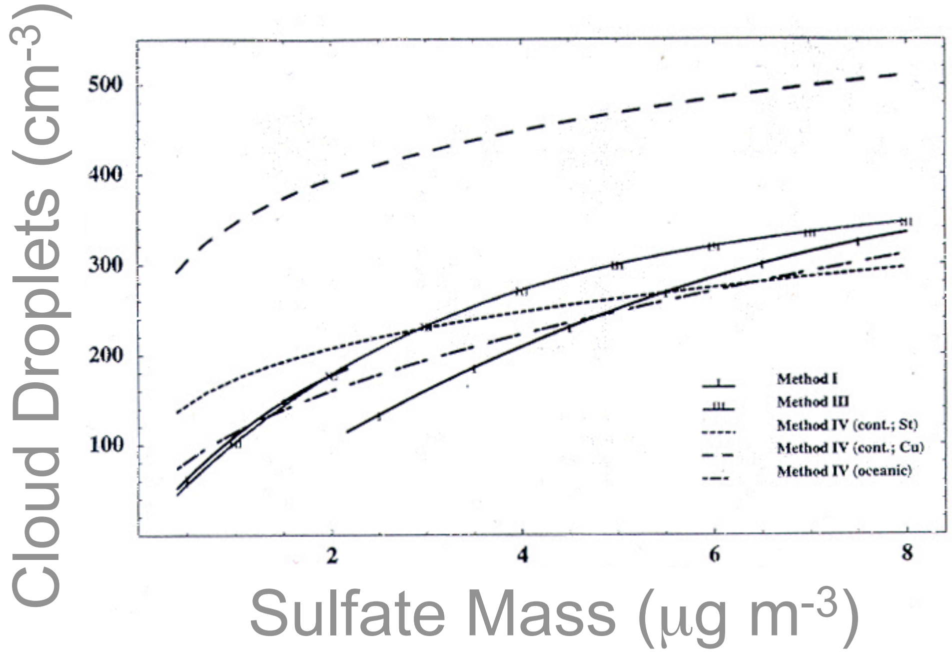
-0.40 W/m^2

III: *Jones et al.* [1994]:

-0.80 W/m^2

IV: *Boucher and Lohmann* [1995]:

-1.78 W/m^2



"It is argued that a less empirical and more physically based approach is required…"

Kiehl et al. [2000]

Microphysical Models

- Offer *potential* to narrow uncertainty in indirect forcing...

Size-Resolved Models

- ♣ Offer _<u>potential</u>_ to narrow uncertainty in indirect forcing...
- ♣ …subject to important model inputs:
  - nucleation mechanism / parameterization
  - carbonaceous aerosol solubility
  - mixing state
  - emissions
    - mass: sea-salt, mineral dust etc.
    - <u>number</u> of primary emissions
  - scavenging lifetime
  - pre-industrial aerosols

Size-Resolved Models

- Offer *potential* to narrow uncertainty in indirect forcing...

It will be some time before microphysics models can predict

•CCN activity

•indirect radiative forcing

with a tolerable degree of uncertainty

- pre-industrial aerosols

“Controlling Factors”

"Controlling Factors"

- What the world does <u>not</u> need:

- What the world <u>does</u> need:

"Controlling Factors"

- What the world does <u>not</u> need:
  - Another radiative forcing estimate
  - (unless it's outside the IPCC range)
- What the world <u>does</u> need:

“Controlling Factors”

- ♣ What the world does not need:
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  - (unless it’s outside the IPCC range)
- ♣ What the world does need:
  - To know what physical processes and model inputs account for uncertainty range
  - Test significance of new processes

“Controlling Factors”

- ♣ What the world does <u>not</u> need:
  - Another radiative forcing estimate
  - (unless it’s outside the IPCC range)
- ♣ What the world <u>does</u> need:
  - To know what physical processes and model inputs account for uncertainty range
  - Test significance of new processes
- ♣ Models can do these things

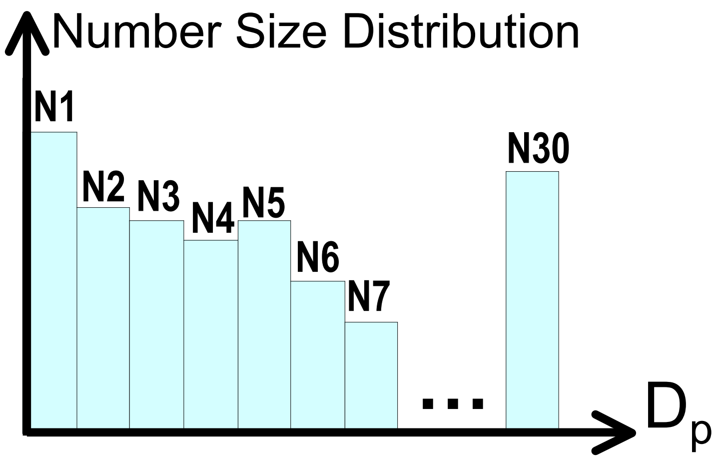
  ⇒ tool for defining research priorities

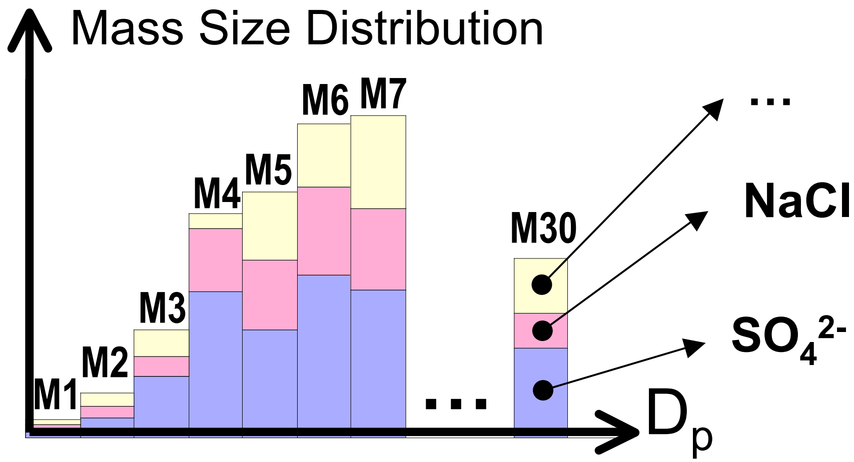
TOMAS Model

- Meteorology (host model):
  - GISS GCM II-prime (predicted meteorology)
  - GEOS-CHEM (assimilated meteorology)
- Microphysics: TwO-Moment Aerosol Sectional (TOMAS)
- Species: Sulfate, sea-salt, EC, OC, dust
- Processes
  - Emissions (size-resolved)
  - Chemistry: sulfur chemistry and SOA formation
  - Microphysics: condensation, coagulation, nucleation
  - Cloud processing: oxidation of SO_2
  - Size-resolved dry / wet deposition

TOMAS

- TwO-Moment Aerosol Sectional algorithm
  - Moments = aerosol <u>number</u> and <u>mass</u>
  - 30 sections from D_p ~10 nm to ~10 µm
  - conserves mass (during coagulation) and number (during condensation)
  - Single-moment sectional: generally do not conserve number during condensation





[*Adams and Seinfeld*, 2002]

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Aerosol Microphysics

General Dynamic Equation

$$N_k = \int_{m_k}^{m_{k+1}} n_k(m)dm$$

$$M_k = \int_{m_k}^{m_{k+1}} mn_k(m)dm$$

- ~30,000 grid cells
- >= 1 year
- Adaptive time steps

Coagulation:

$$\frac{dN_k}{dt} = \frac{1}{2}K_{k-1,k-1}N_{k-1}^2 + \psi_{k-1}\sum_{i=1}^{k-2}K_{k-1,i}M_i$$
$$+\frac{\psi_k - f_k}{2x_k}\xi\sum_{i=1}^{k-1}K_{k,i}M_i\overline{m}_i - K_{k,k}N_k^2 - N_k\sum_{i=k+1}^{I}K_{k,i}N_i$$
$$-\frac{\psi_{k-1} - f_{k-1}}{2x_{k-1}}\xi\sum_{i=1}^{k-2}K_{k-1,i}M_i\overline{m}_i - \psi_k\sum_{i=1}^{k-1}K_{k,i}M_i$$

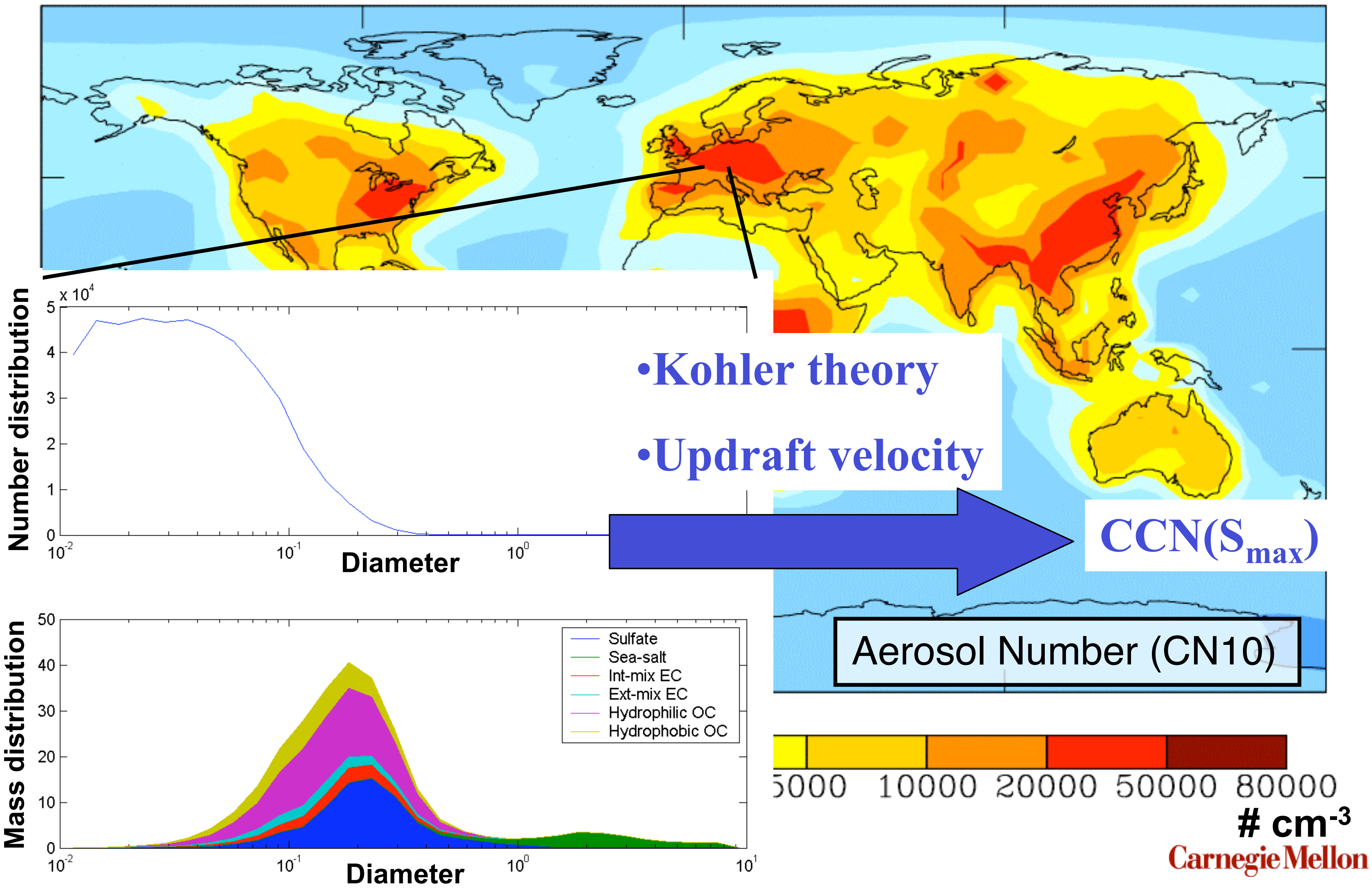
Condensation:

$$M_k(t+\Delta t) = \int_{m_{k,f}}^{m_{k+1,f}} \left[m^{2/3} + \frac{2}{3}\tau\right]^{3/2} n_k(m)dm$$

$$m_{k,f} = \left[m_{k,i}^{2/3} - \frac{2}{3}\tau\right]^{3/2}$$

$$\tau = \int_t^{t+\Delta t} C\Delta S(t)dt$$

TOMAS Sample Predictions



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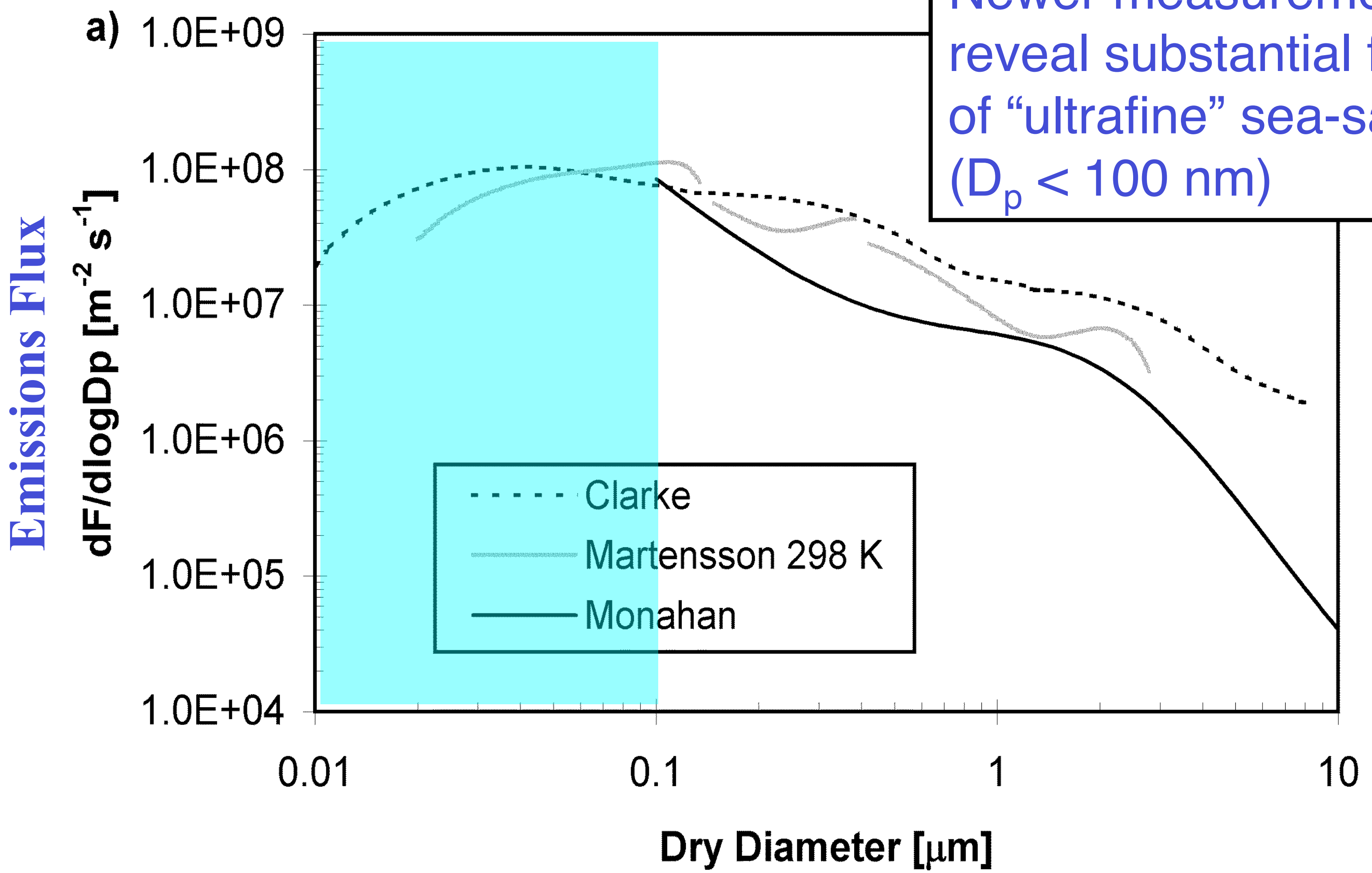
Sea-salt and CCN

Sea-salt Emissions Parameterizations

Newer measurements reveal substantial flux of “ultrafine” sea-salt (D_p < 100 nm)



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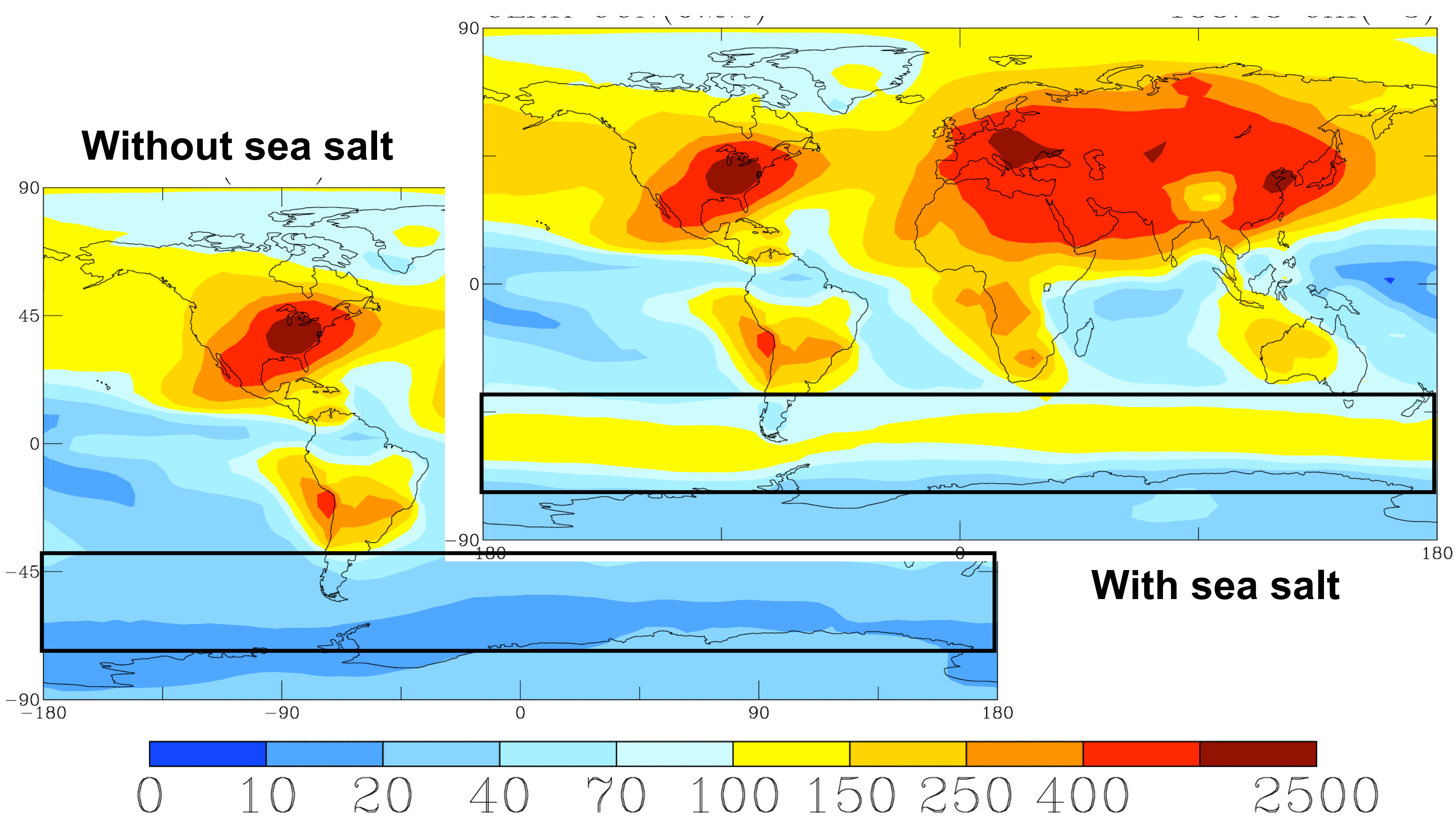
Sea-salt: Questions

- ♣ How much does sea-salt contribute to the natural background CCN concentrations?
- ♣ Do ultrafine sea-salt emissions enhance CCN concetrations?

CCN(0.2%) concentrations (cm^{-3})



Pierce, J. R. and Adams, P. J.: Global evaluation of CCN formation by direct emission of sea salt and growth of ultrafine sea salt, *Journal of Geophysical Res*earch, 111, 2006.

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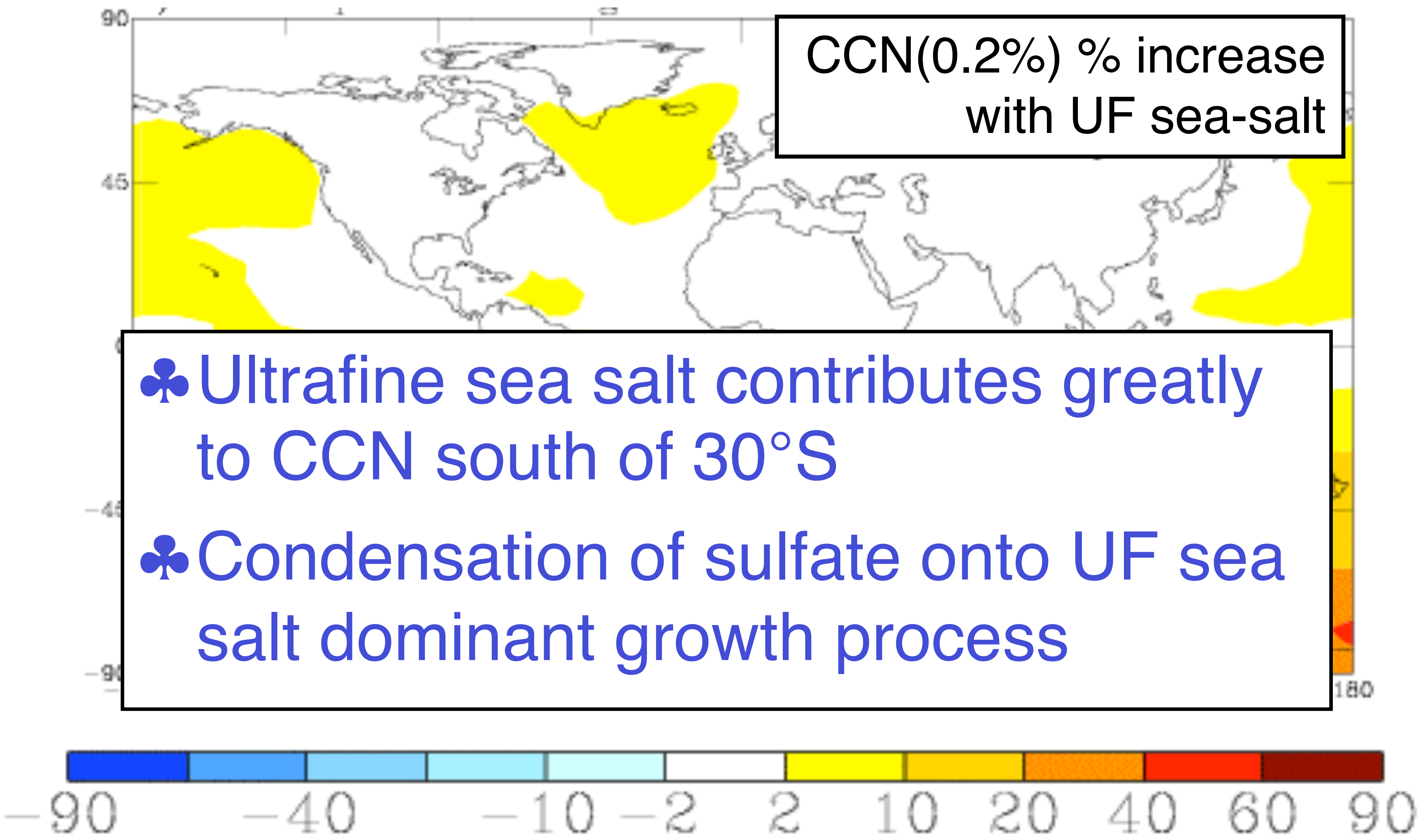
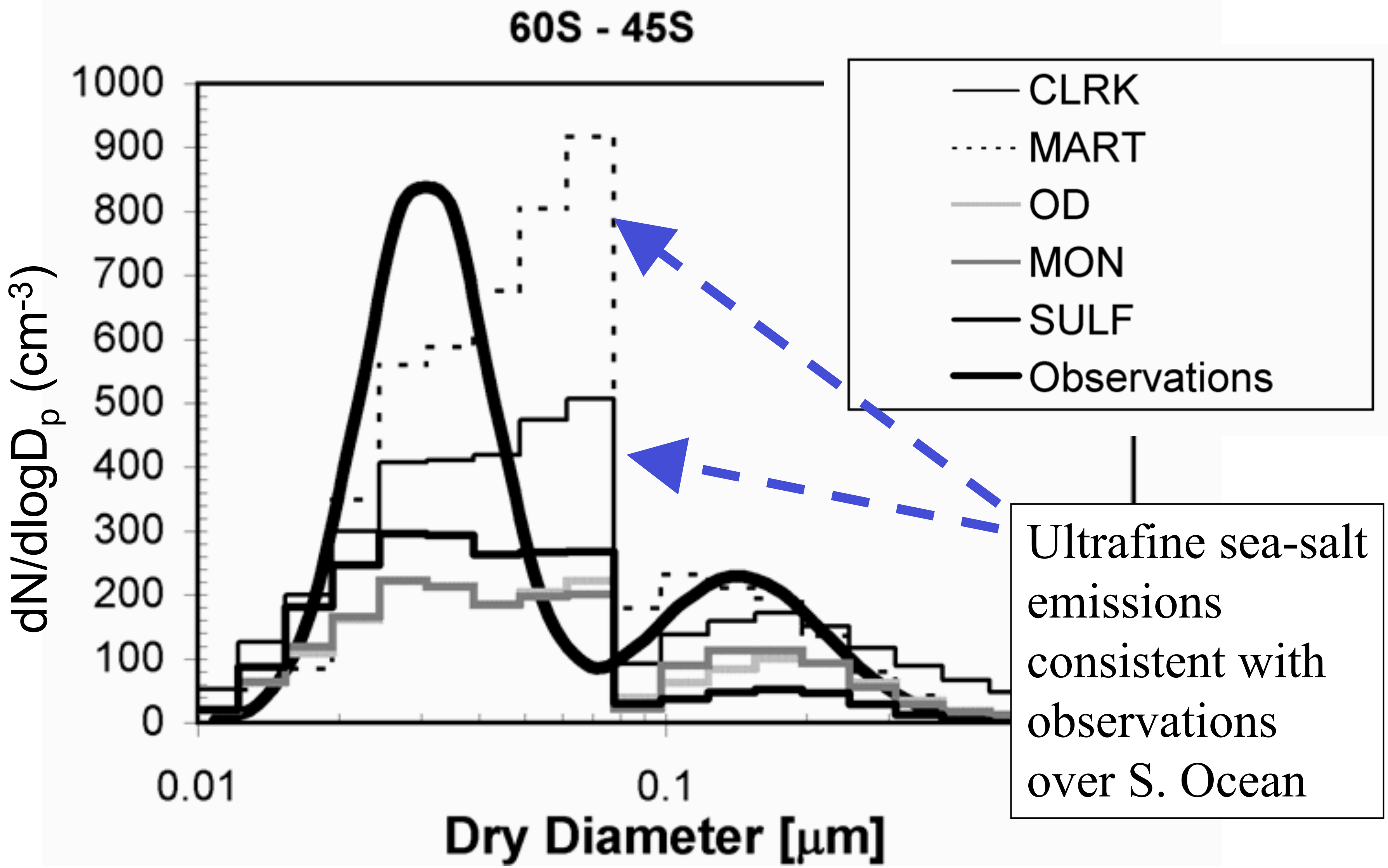
CCN from Ultrafine Sea-salt



- ♣ Ultrafine sea salt contributes greatly to CCN south of 30°S
- ♣ Condensation of sulfate onto UF sea salt dominant growth process

Observed Number Distributions



Observations from *Heintzenberg et al.* [2000]

Mineral Dust and CCN

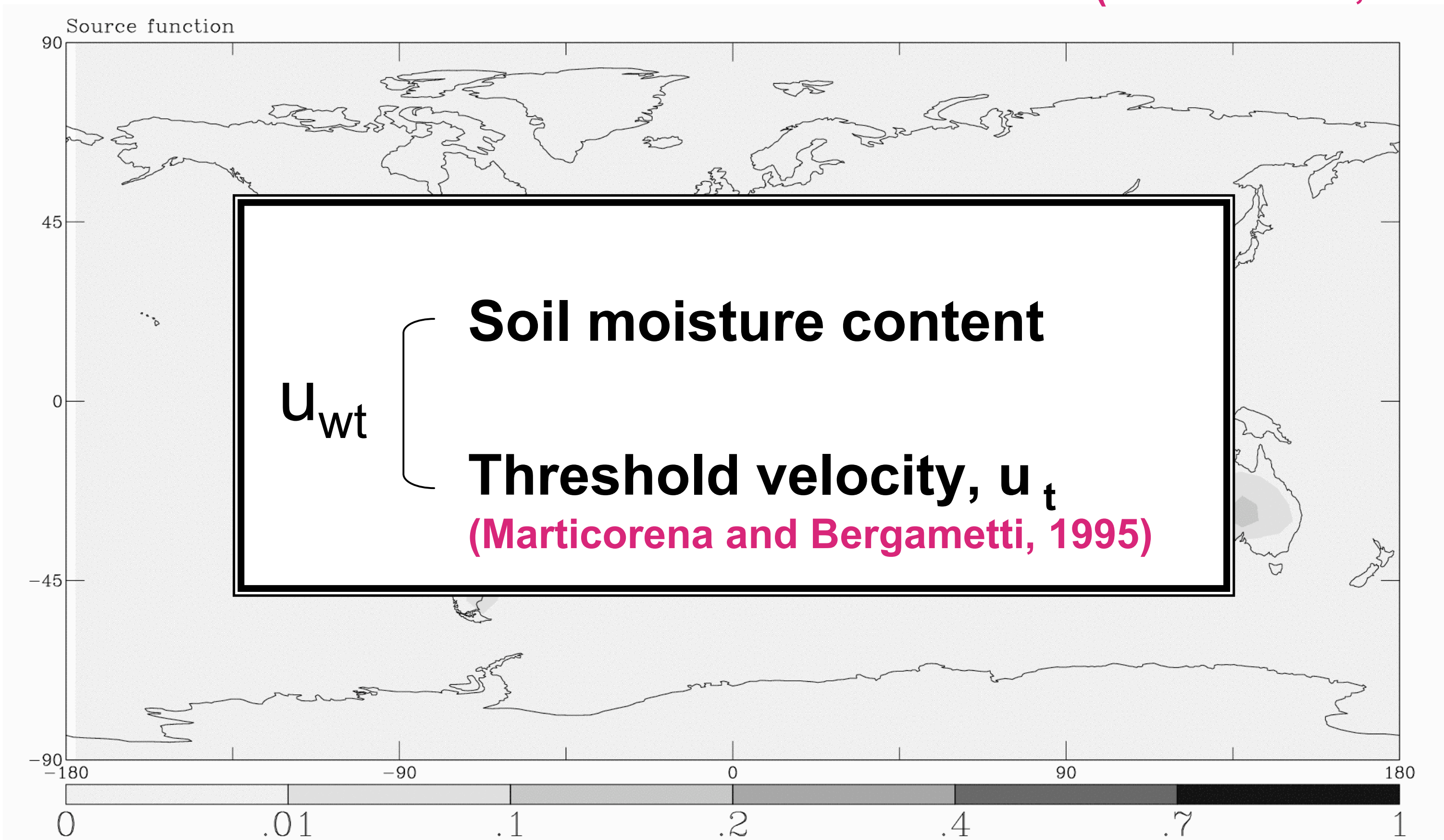
Dust Model Description

$$F_p = \left\{ \begin{array}{c} C S s_p u^2_{10} (u_{10} - u_{wt}) : u_{10} > u_{wt} \\ 0 : u_{10} \le u_{wt} \end{array} \right\}$$

S: Source function

s_p: Soil fraction

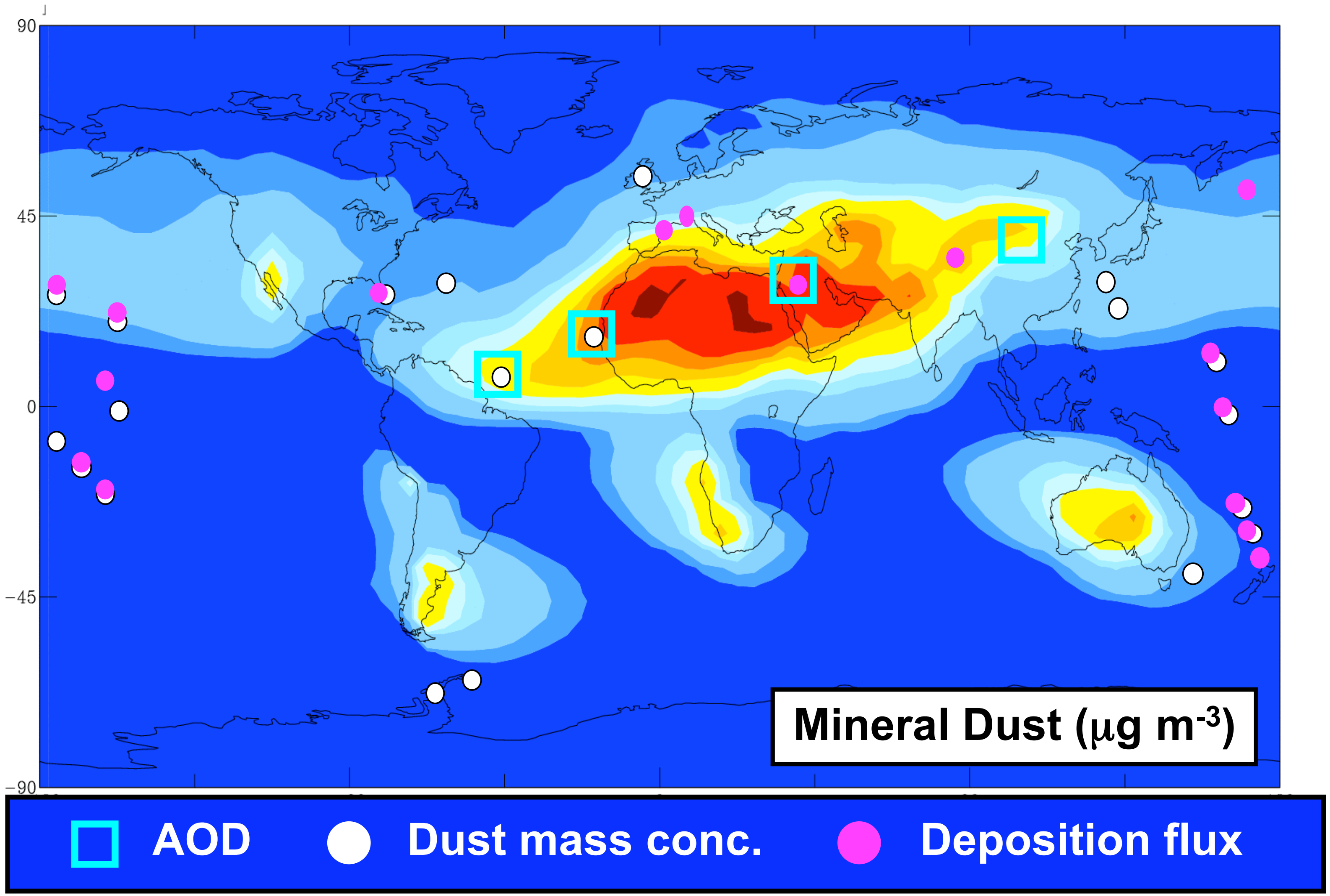
(Ginoux et al., 2004)

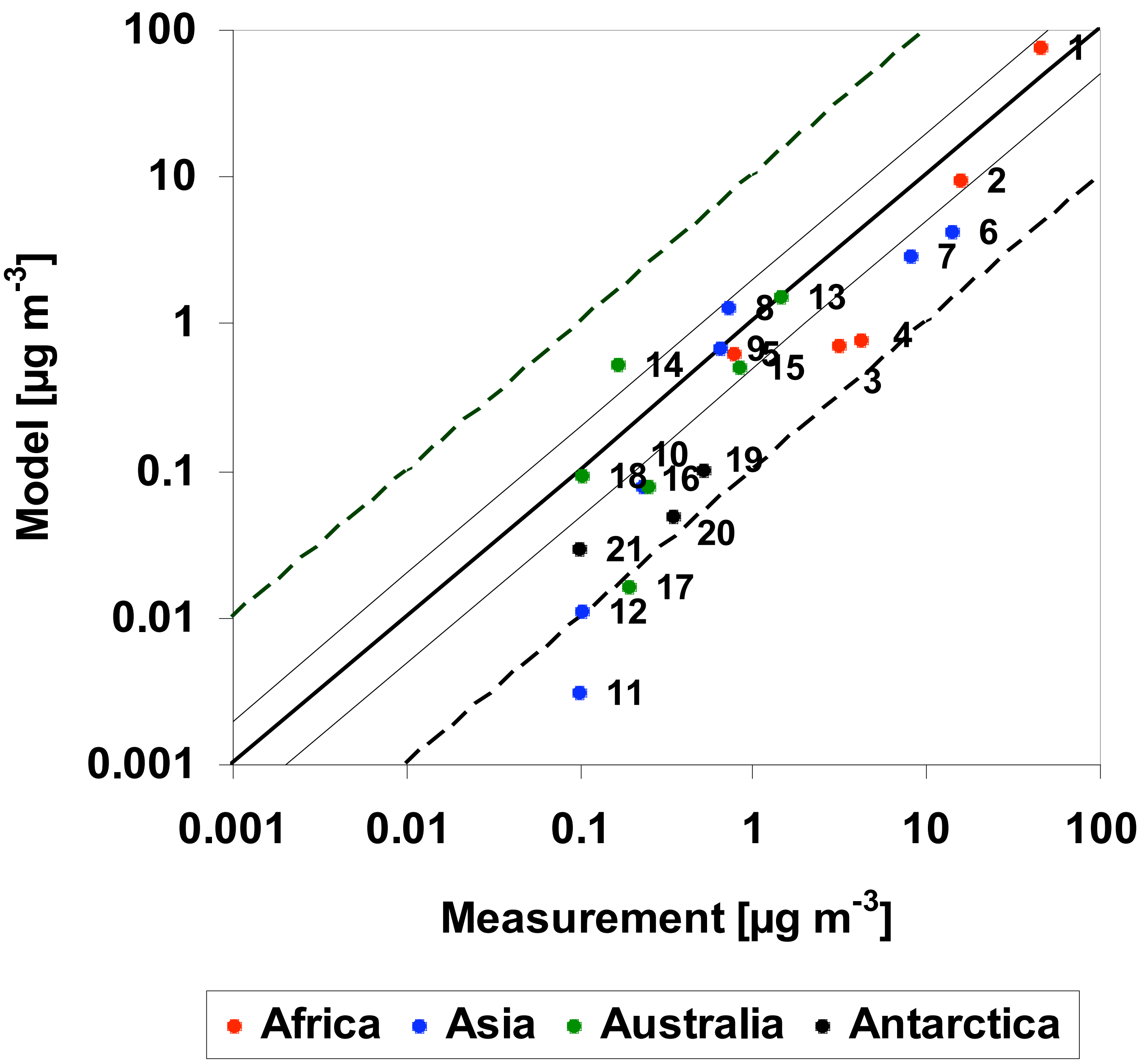


u_{wt} { **Soil moisture content**, **Threshold velocity, u_t** }

(Marticorena and Bergametti, 1995)

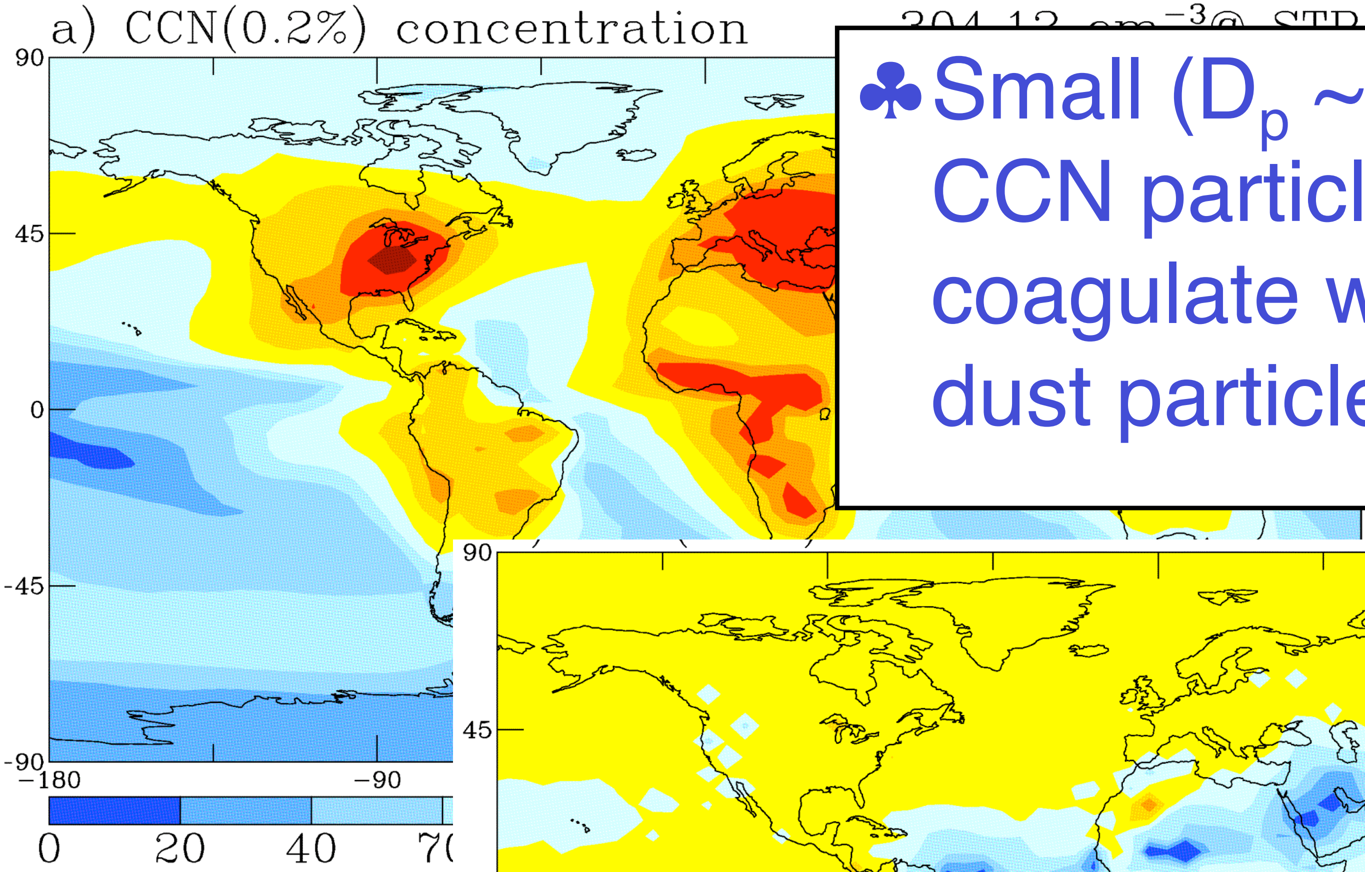
Dust Mass Concentrations

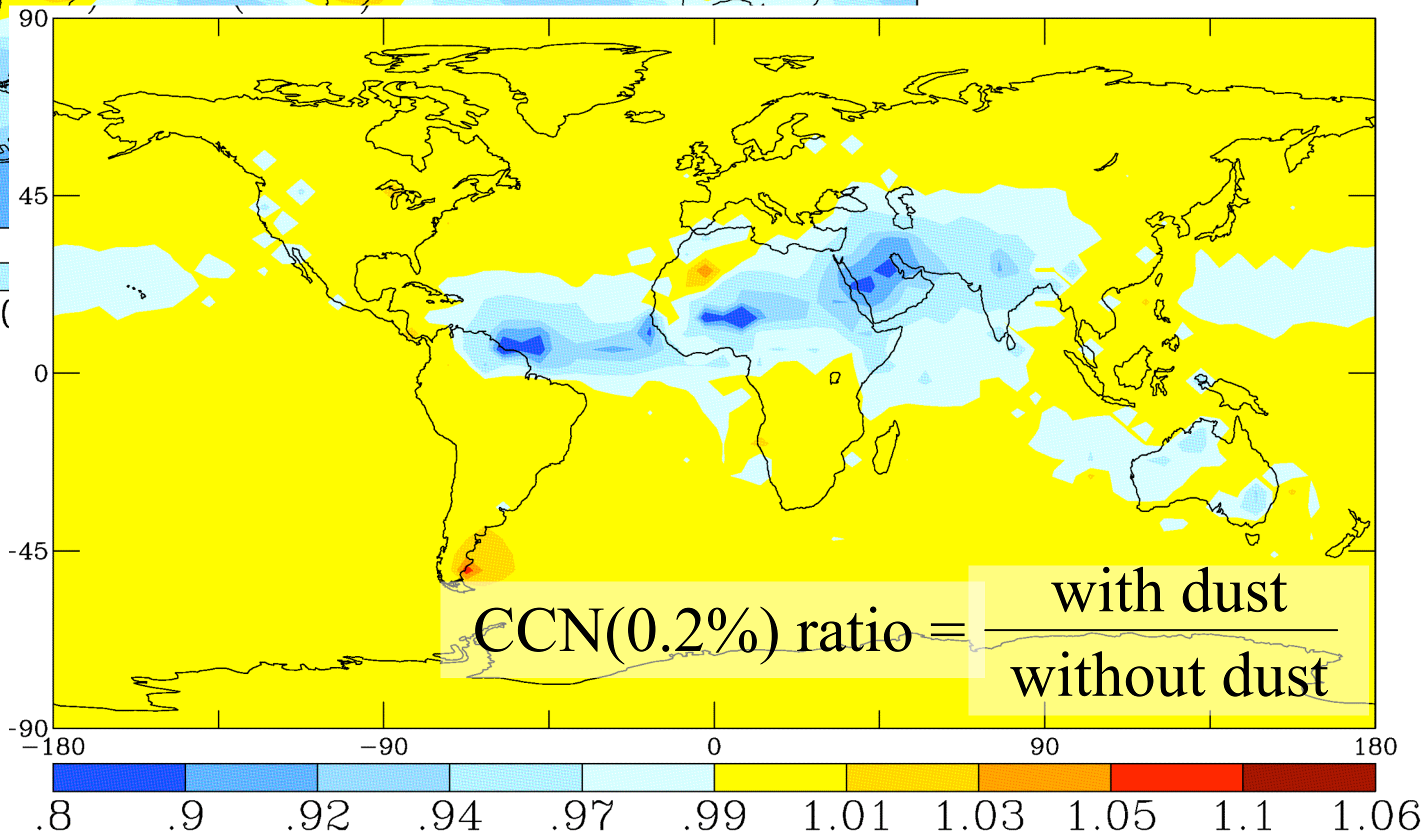




100
10
1
0.1
0.01
0.001
0.001
0.01
0.1
1
10
100
Model [µg m-3]
Measurement [µg m-3]
1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
Africa
Asia
Australia
Antarctica

♣ Small (D_p ~ 100 nm) CCN particles coagulate with larger dust particles





Carbonaceous Aerosol and CCN

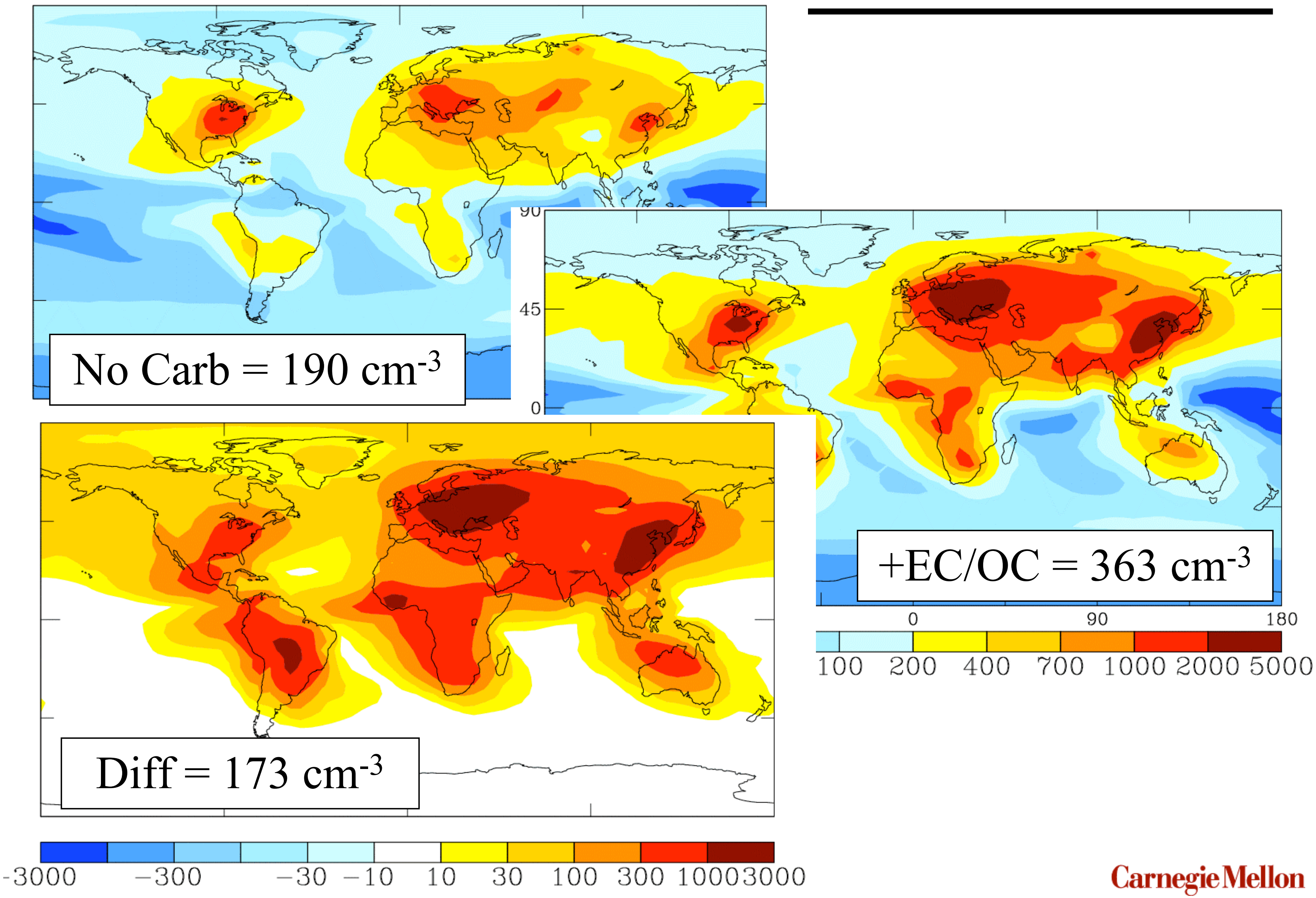
Carbonaceous Simulation

- Species: Sulfate, sea-salt, EC, OM
  - EC and OM are both divided into hydrophilic/hydrophobic fractions
- Emissions: Bond et al., 2004
  - EC: 8 Tg yr^{-1} (80% hydrophobic initially)
  - OC: 33.9 Tg yr^{-1} (50% hydrophobic initially)
- Aging of hydrophobic to hydrophilic with
  - $\tau = 1.5$ days
- Primary emissions: assumed lognormal
  - D_{pg} (mass) = 100 nm and $\sigma = 2$
  - D_{pg} (number) = 30 nm
- SOA: later in this talk

CCN Activity of EC/OC

- ♣ Modified Köhler theory
  - Slightly soluble species
  - Insoluble species
  - Surfactant effects <u>not</u> included
- ♣ Base case: assume two aerosol populations
  - Population 1: hydrophobic EC
  - Population 2: internal mixture of other species
    - Sulfate and sea-salt
    - Hydrophilic EC (insoluble core)
    - Hydrophobic OC (slightly soluble, 0.009 g /100 cm^3 H_2O)
    - Hydrophilic OC (D_{crit} = 45 nm and 140 nm at S = 1% and 0.2%)

CCN(0.2%) Impacts



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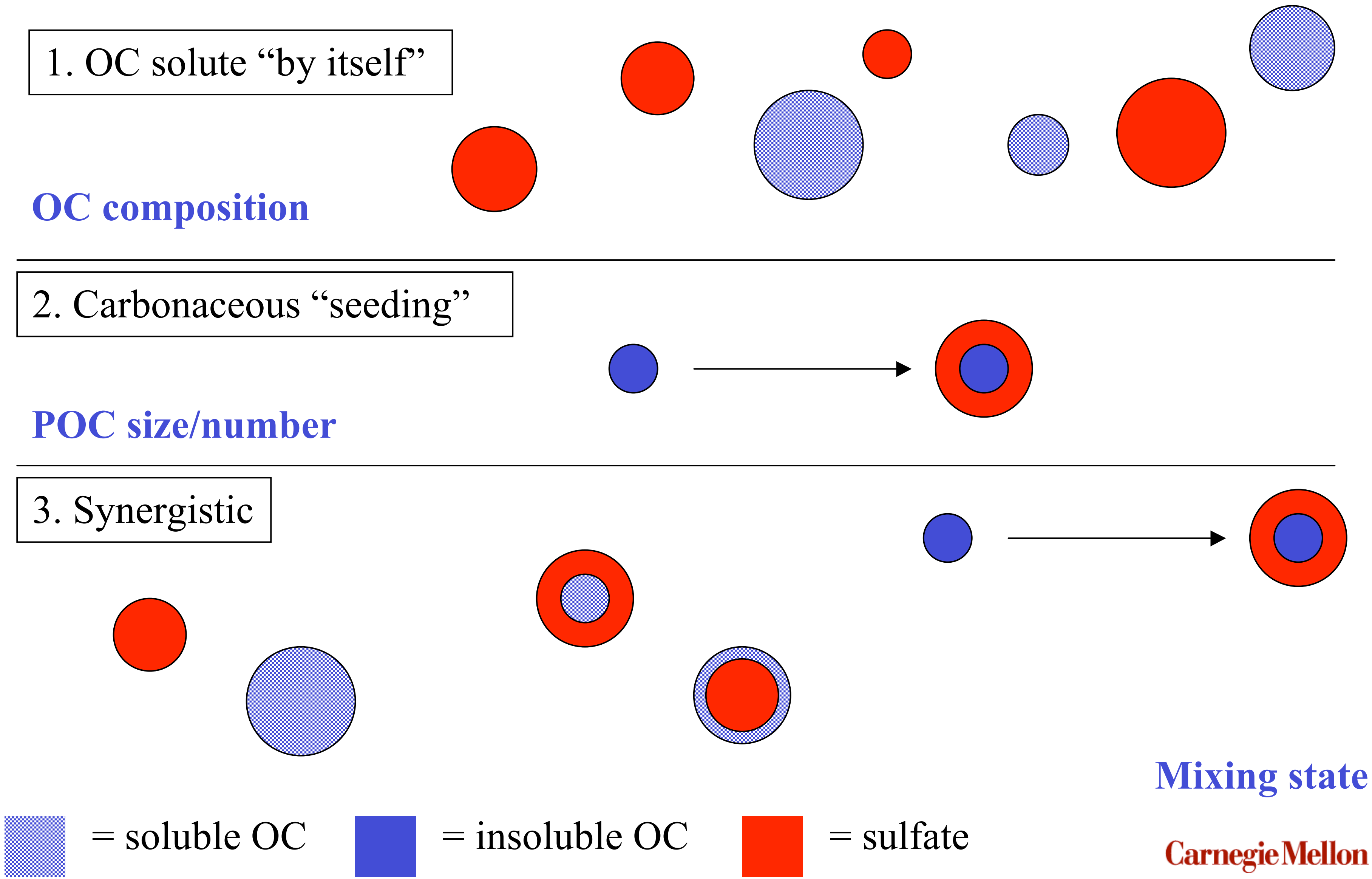
Conceptual Models



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Sensitivity to Mixing / Solubility

Repeat simulations of CCN(0.2%) for alternative activation scenarios:

1) Insoluble ("Carbonaceous seeding"):
   - EC/OC are insoluble cores
   - Internally mixed with sulfate/sea-salt
2) Externally-mixed* ("OC solute by itself"):
   - External mixtures: sulfate, sea-salt, EC/OC
   - Solubility/activation same as base case

*CCN activation/wet deposition calculated with external mixing but microphysics still internal mixing

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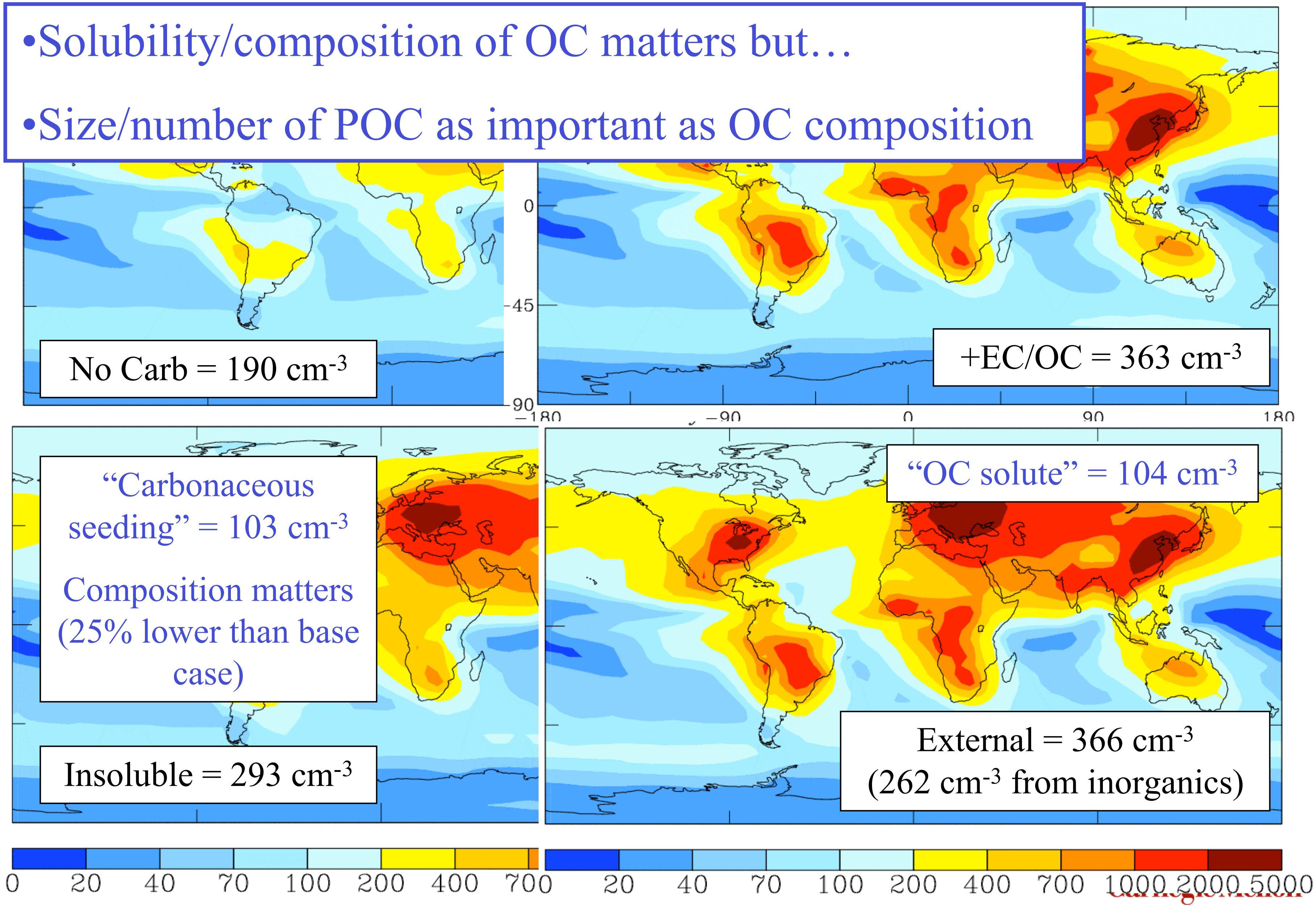
CCN(0.2%) Impacts

- Solubility/composition of OC matters but…
- Size/number of POC as important as OC composition



What About SOA?

- SOA = secondary organic aerosol (chemically produced from gaseous VOCs)
- Sources and chemistry of SOA highly uncertain
- Most global/regional models suggest that SOA is <30% of total OA
- Recent work suggests much POA evaporates in atmosphere
- AMS measurements suggest that SOA may be ~90% of total OA

POA vs. SOA

- **POA and SOA have different effects on aerosol size distribution**

⇒We expect different effects on CCN concentration

SOA	POA
♣ Adds only mass ♣ No new particles ♣ Mass condensing onto existing particles ◊ growth ♣ Impact on CCN depends on what size SOA condenses	♣ Adds mass and number (both UF and CCN) ♣ UF provide condensational sink ◊ growth to CCN ♣ Adding number leads to coagulation ◊ reduces number ◊ also growing mass to larger sizes

Simulations

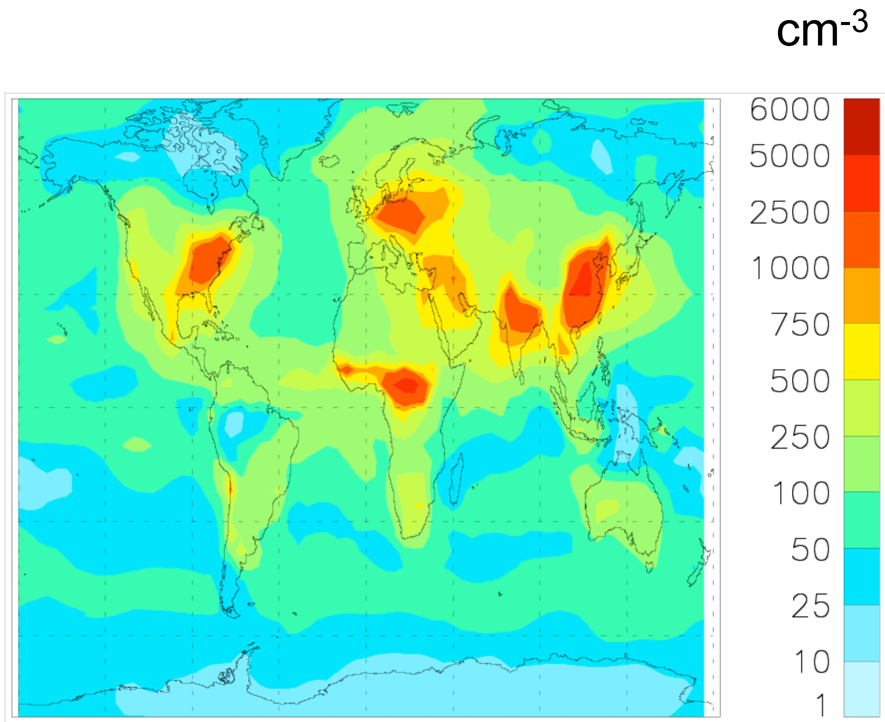
- ♣ GEOS-CHEM global model
  - SOA condenses according to absorptive partitioning theory
  - Two months "spin up" (Nov and Dec)
  - All results are Jan average
- ♣ POA Case: POA only; no SOA
  - POC = 26 Tg/yr
  - Emissions from Bond [2004], Cooke [1999], Park [1998] and GFED2
- ♣ Base Case
  - POA plus 8.6 Tg/yr of SOA (10% of monoterpene emissions)
- ♣ Sensitivity Cases
  - Total OC = 26 Tg/yr
  - Vary %SOA: 0%, 10%, 50%, 90%, 100%

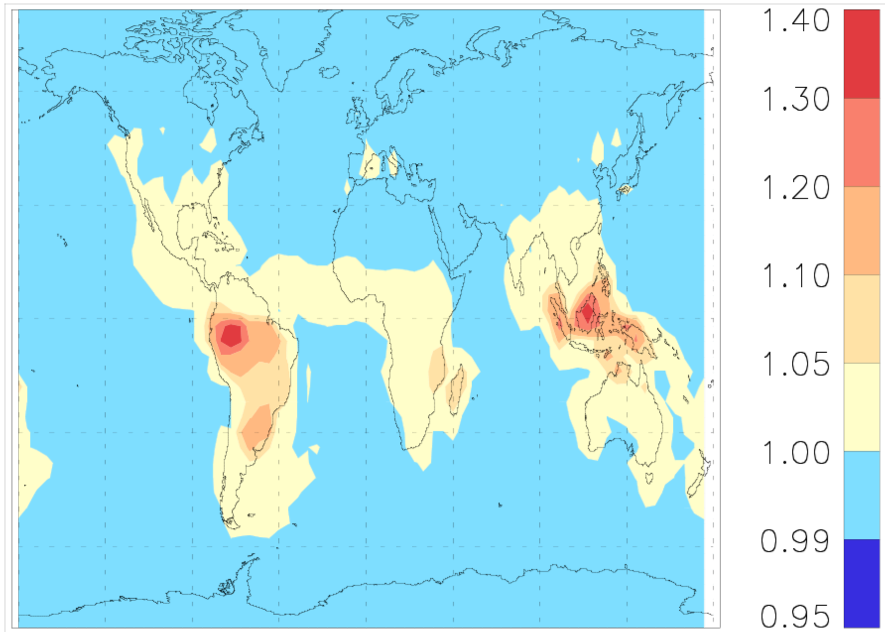
Effect of Biogenic SOA

- ♣ Surface CCN(0.2%) without SOA

- ♣ CCN(0.2%) ratios

  $$= \frac{\text{CCN w/ biogenic SOA}}{\text{CCN w/o SOA}}$$



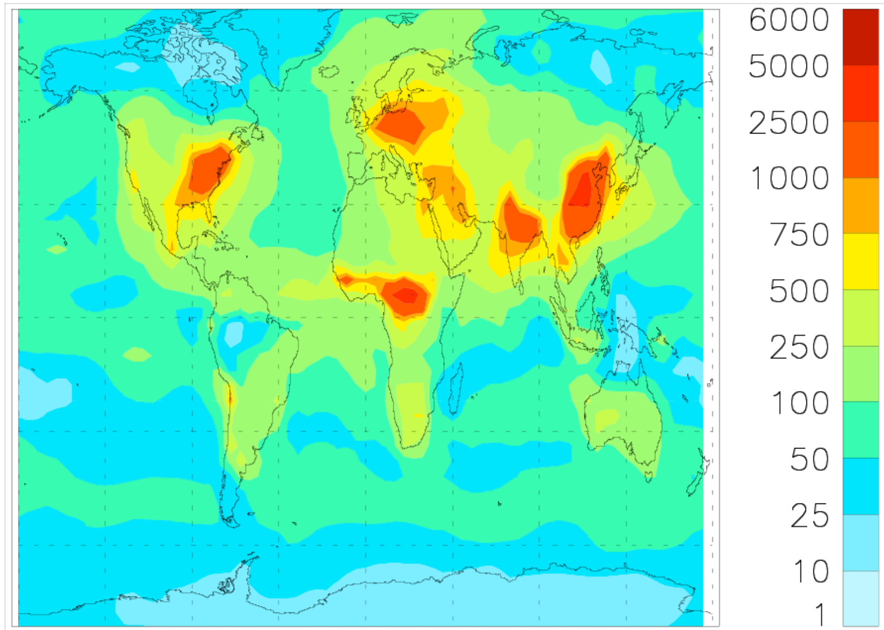


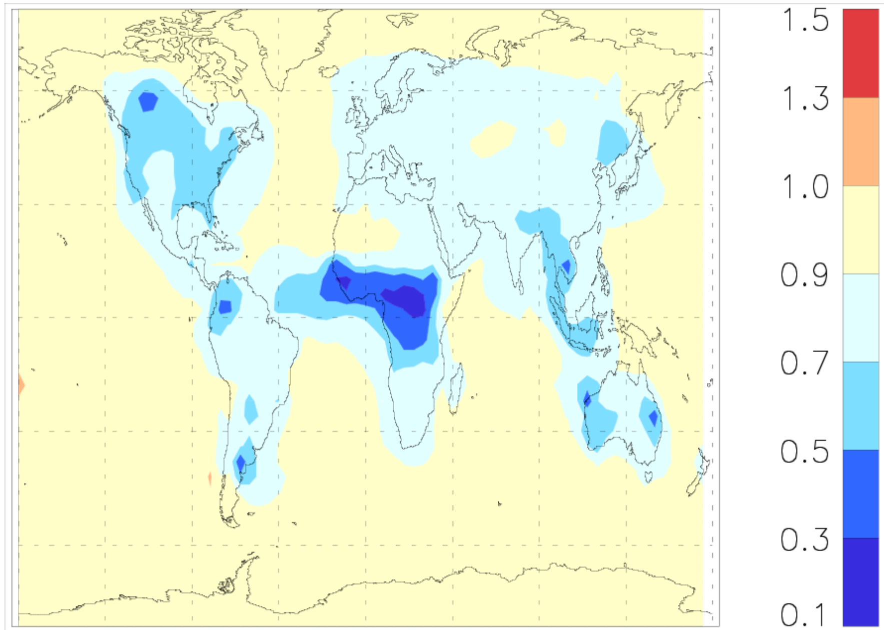
POA vs. SOA effect on CCN

- ♣ Surface CCN(0.2%) 100%POA case
- ♣ CCN(0.2%) ratios

$$= \frac{\text{CCN 100\%SOA}}{\text{CCN 100\%POA}}$$

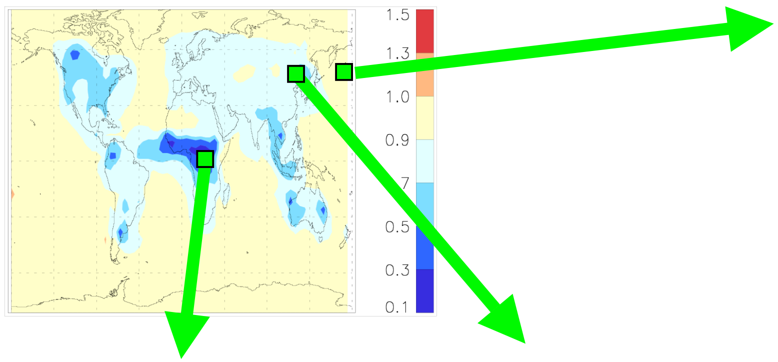
cm^{-3}

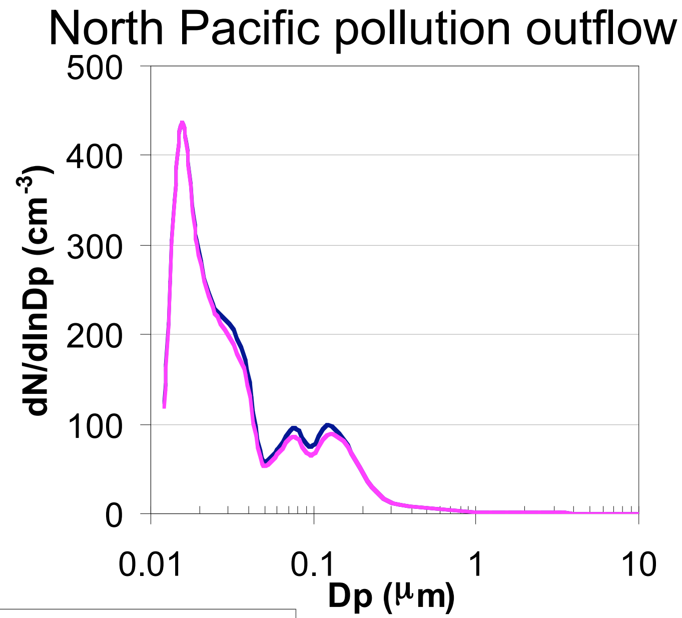


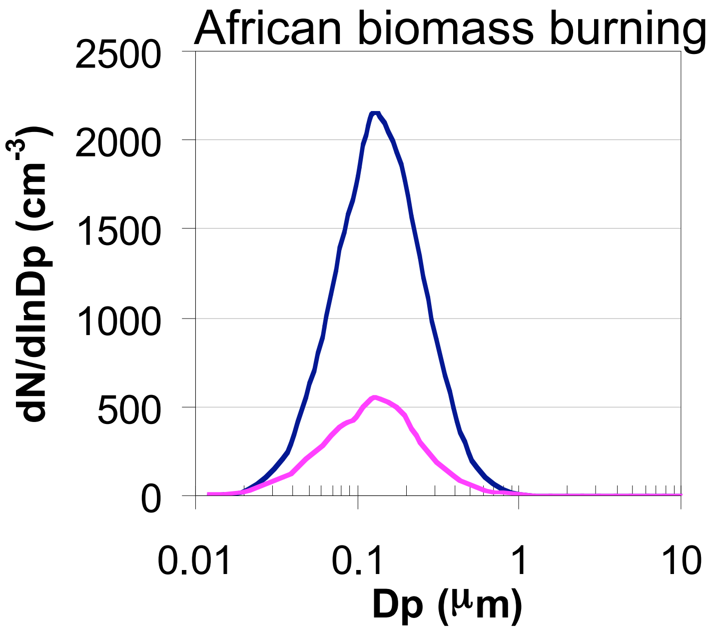


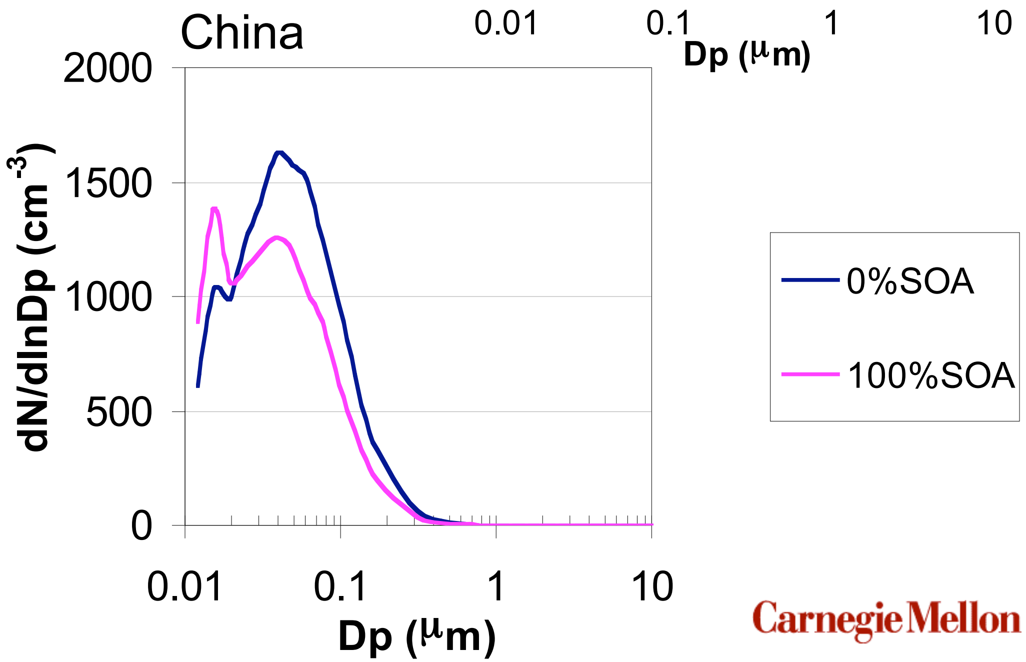
POA vs. SOA effect on CCN

- ♣Number size distribution









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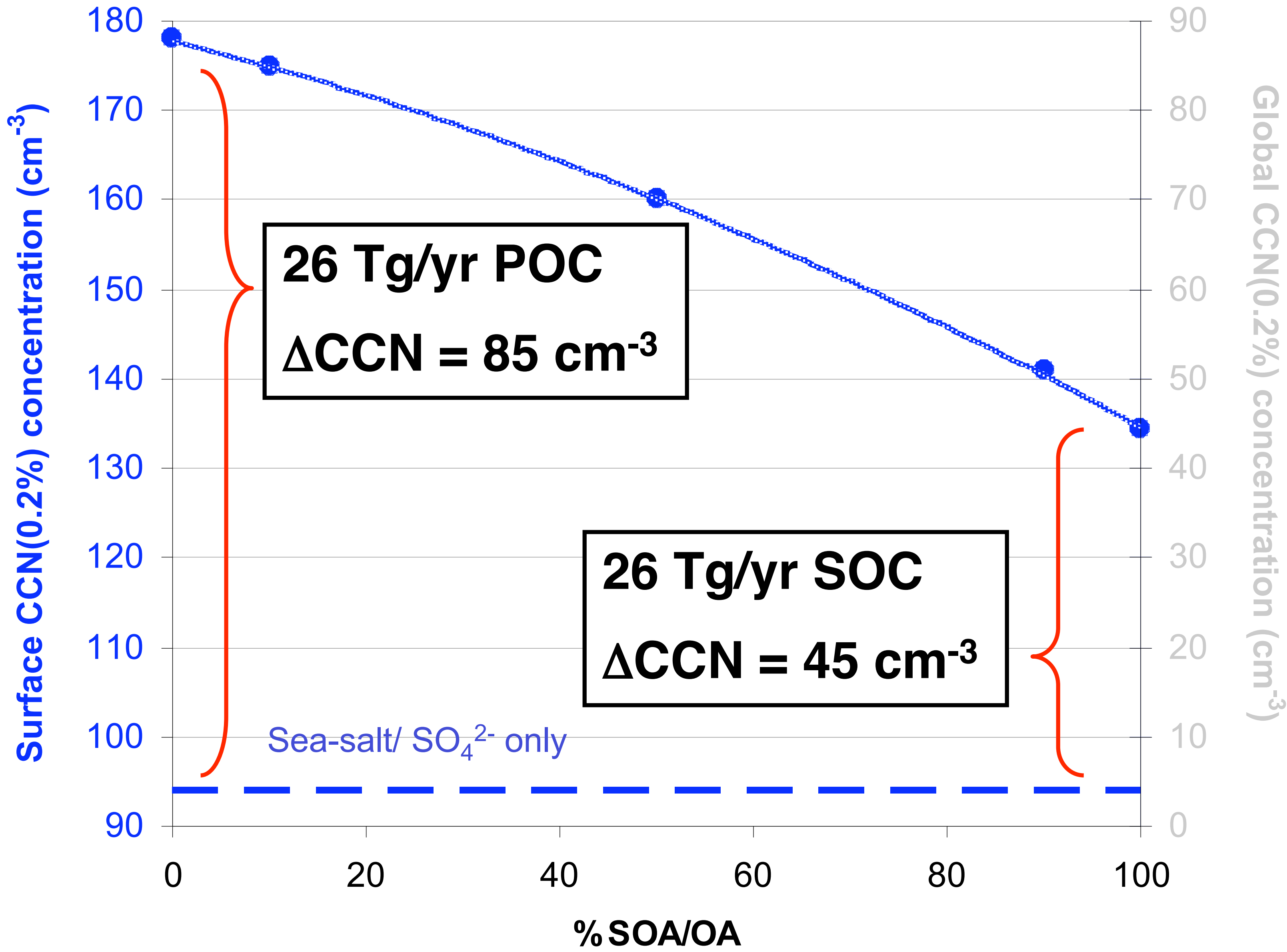
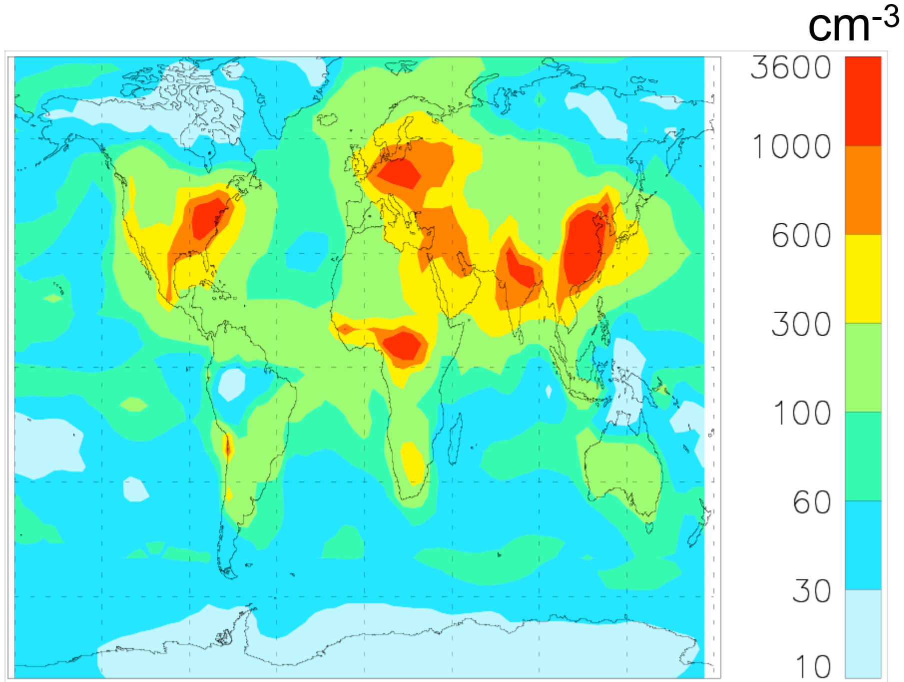
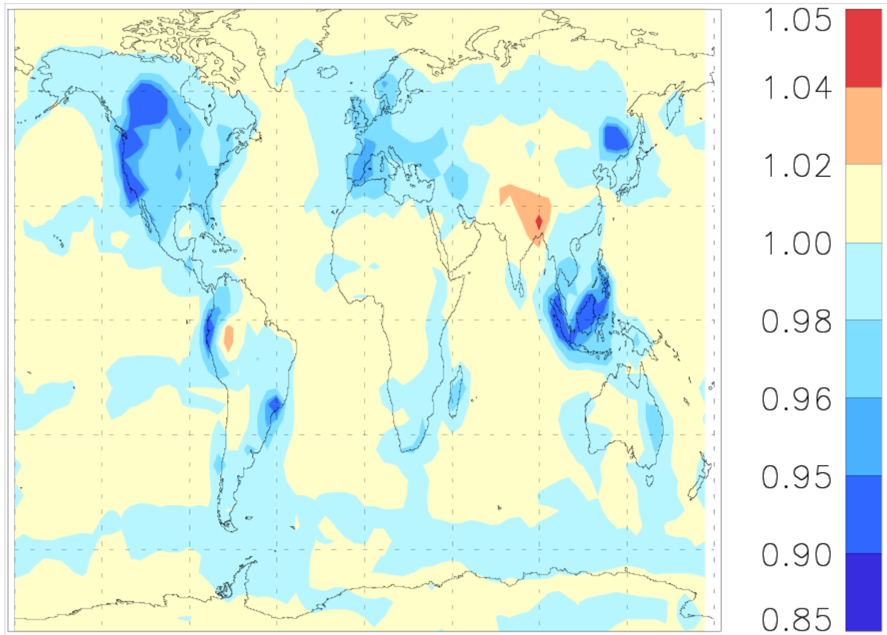
POA vs. SOA effect on CCN



Absorptive Partitioning: Organic vs. Aqueous

- ♣ Surface CCN(0.2%) for 50%SOA case
- ♣ CCN(0.2%) ratios = $\frac{\text{cond to aqueous}}{\text{cond to organic}}$





Conclusions

- ♣ Microphysical models
  - offer *potential* to reduce uncertainty in the future
  - provide insight into physical processes controlling uncertainties now
- ♣ Ultrafine sea-salt important for natural CCN in S. Ocean
- ♣ Dust reduces CCN(0.2%) via coagulational scavenging
- ♣ Microphysics (size, number) of carbonaceous as important as chemistry
- ♣ Primary emissions generally create more CCN than secondary

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