CCN Formation: Microphysics and Chemistry

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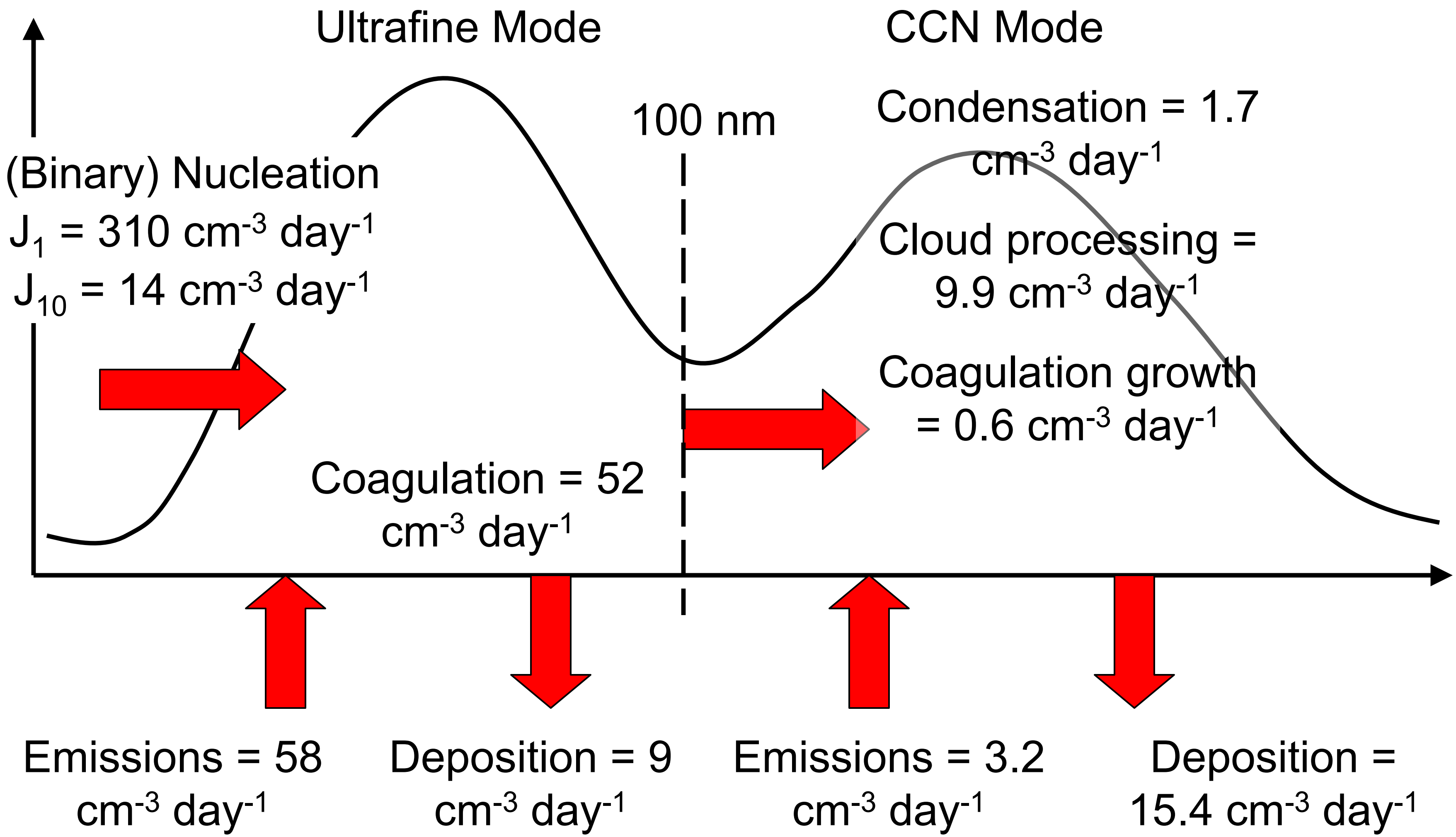
Two Themes

1) Interactions of *chemistry* and *microphysics* in the global CCN cycle

2) The *CCN mode* is tightly coupled to and largely derived from the *ultrafine mode*

CCN Budget and Ultrafines



GISS-GCM; global average; Sulfate, sea-salt, carbonaceous, dust

Science Questions

- How do CCN concentrations depend on:
- Nucleation rate and mechanism?
  - Fast (ternary) vs slow (binary) nucleation
- Primary particle emissions?
  - Important uncertainty for CCN
  - Do BC reductions slow global warming?
- OC composition / hygroscopicity?
- SOA formation rates?

Model Description

Aerosol species

- Sulfate
- Sea-salt
- EC: ext/int mixed
- OC: hydro-phobic/philic
- Mineral dust

Microphysics

- TOMAS algorithm
- Condensation/coagulation
- Nucleation (binary, ternary, empirical "activation", ion-induced)

Host models

- GISS (GCM)
- GEOS-CHEM (CTM)

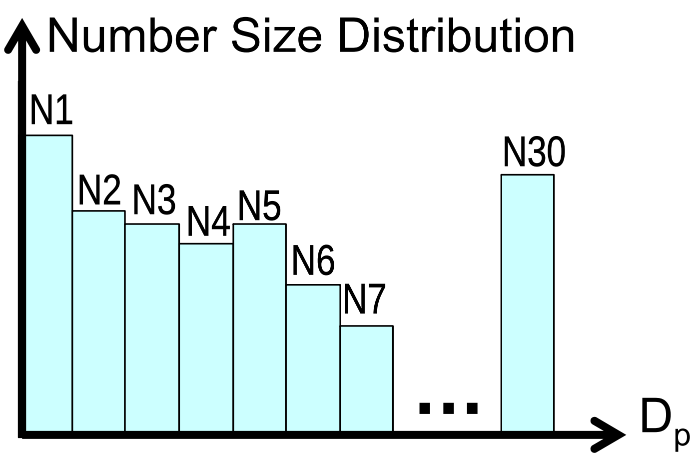
Size-resolved emissions w/ subgrid coagulation

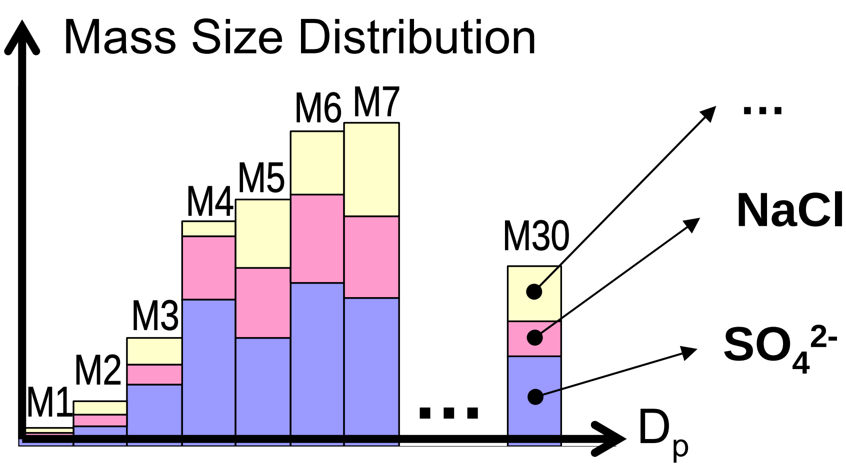
Chemistry

- DMS→ SO_2 → sulfate; SO_2 → $H_2SO_4(g)$
- Dial in SOA "mechanism"
- EC/OC "aging": 1.5 days
- Modified Kohler theory (hydrophilic OM: $\kappa = 0.12$)

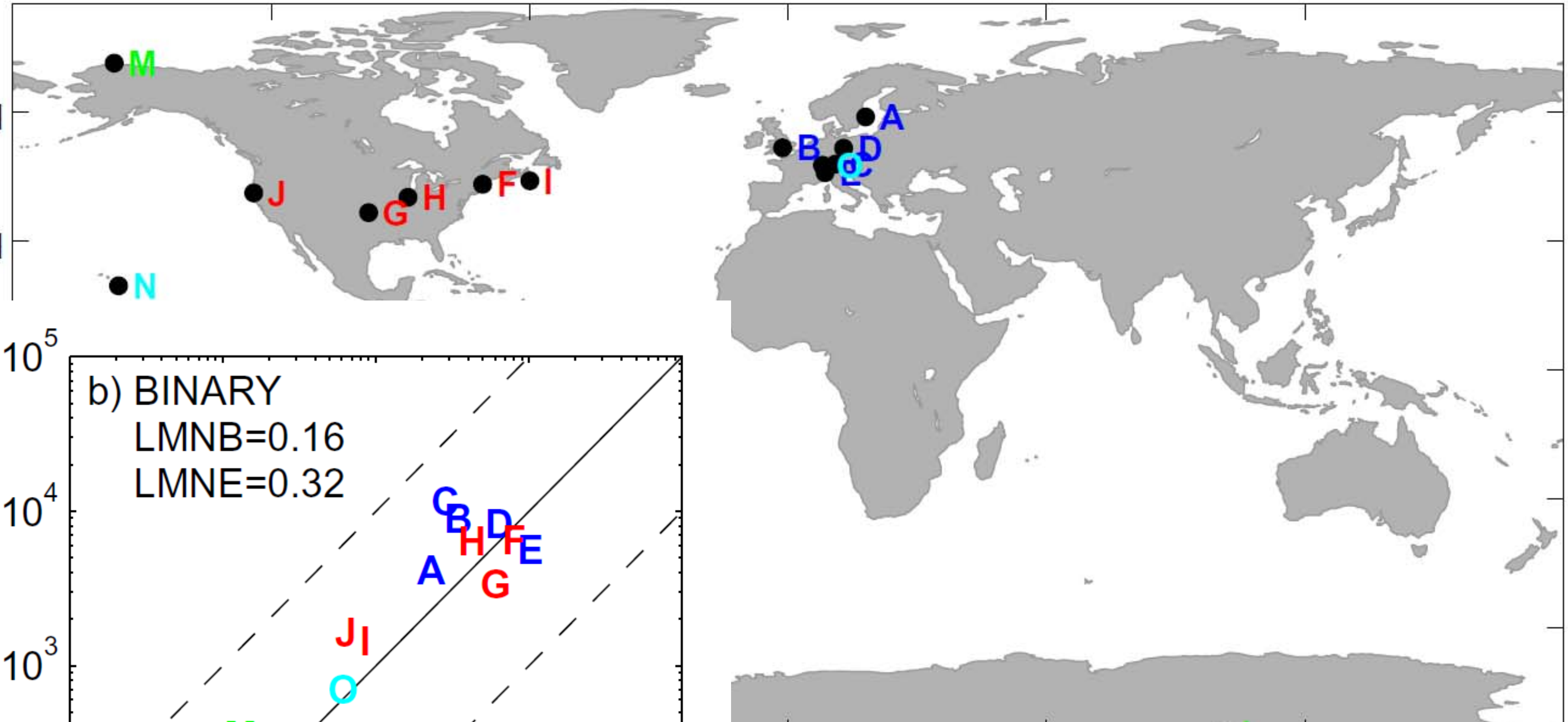
TOMAS Overview

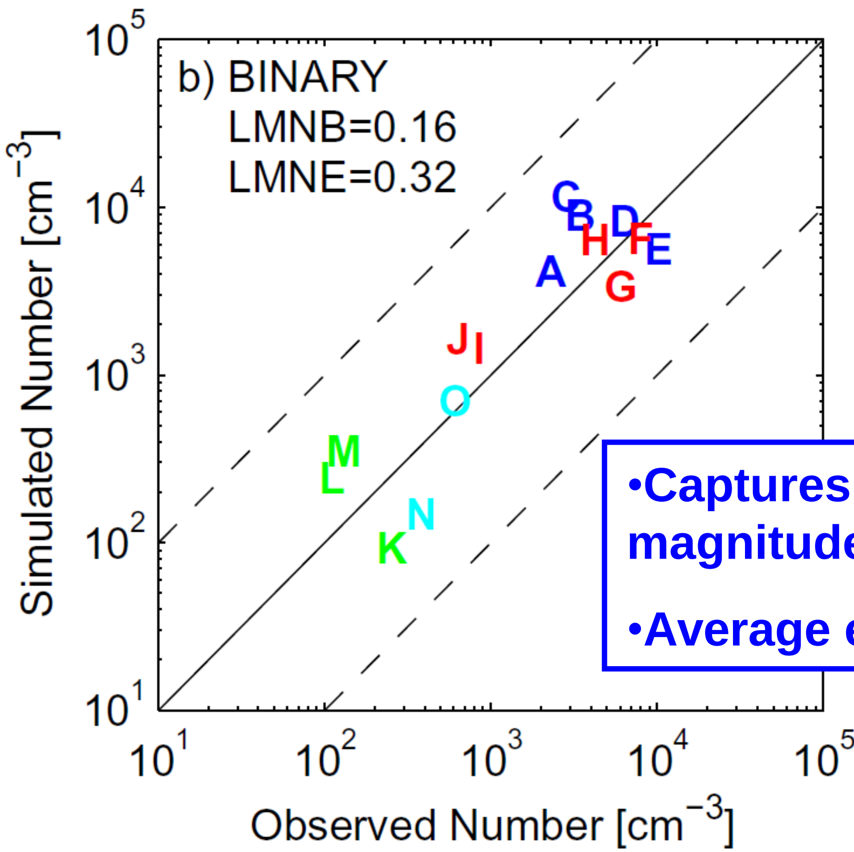
- TwO-Moment Aerosol Sectional algorithm
  - Size range usually 10 nm – 10 µm
  - 30 size sections (10 per decade of diameter)
  - Parameterized nucleation mode
  - Sometimes extended down to 1 nm (nucleation mode)
  - Moments = 1) aerosol number and 2) aerosol mass





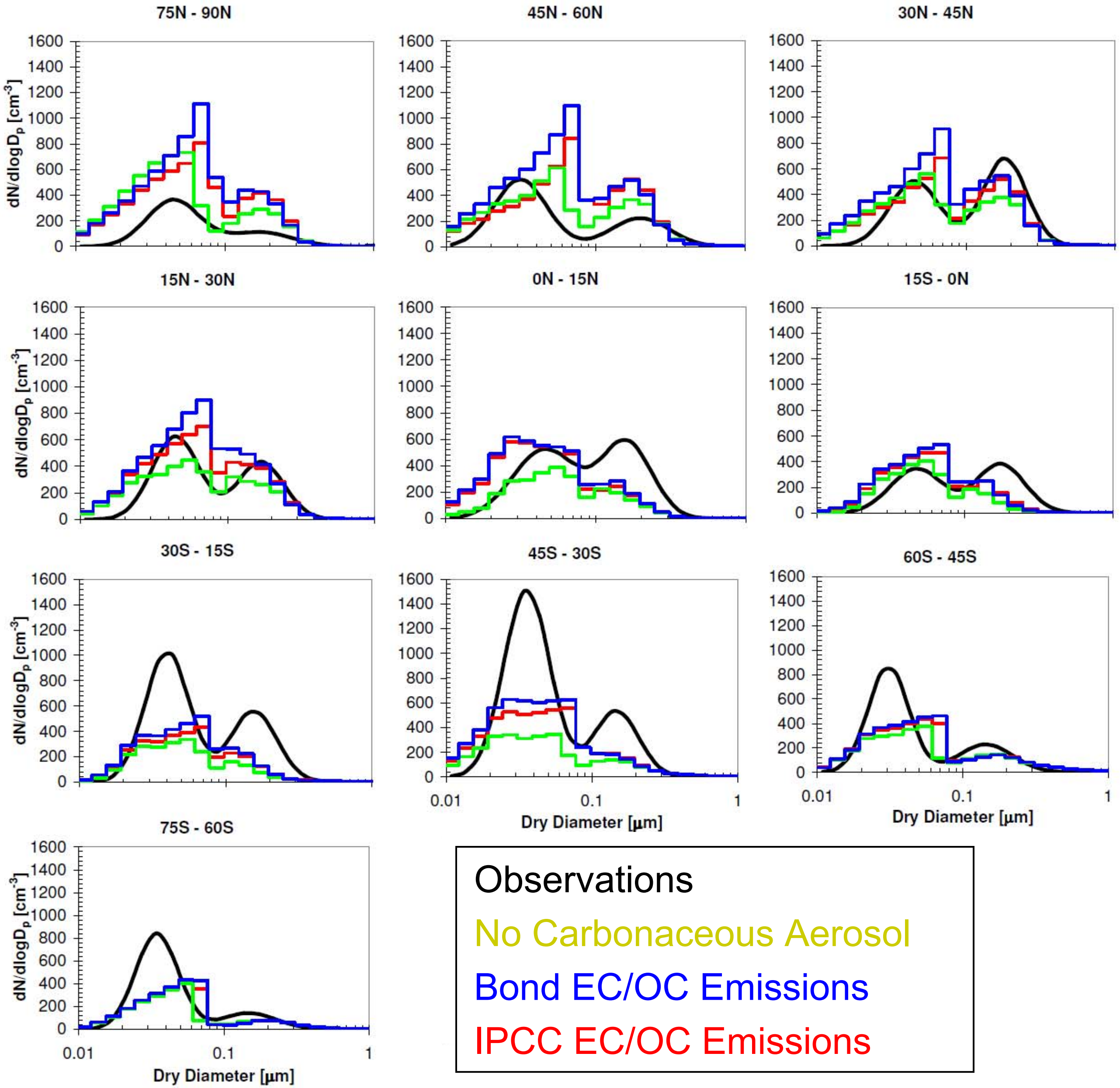
Model Evaluation: CN10





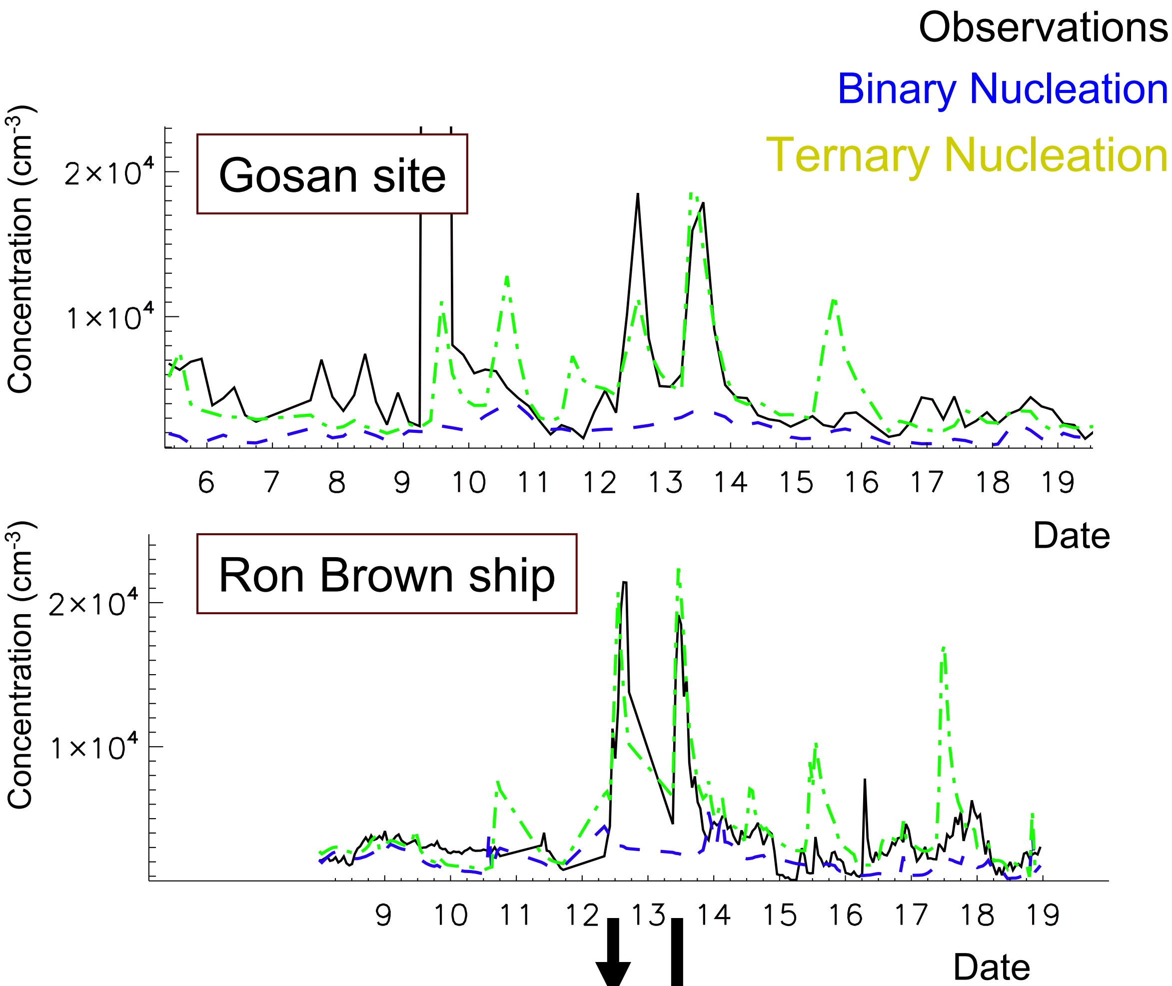
- **Captures variation in CN10 over two orders of magnitude**
- **Average error (LMNE) is a factor of 2**

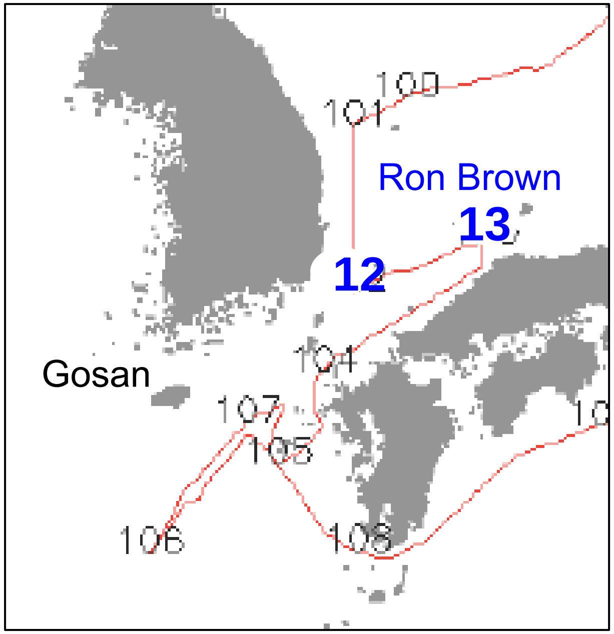
Model Evaluation: Marine Size Distributions



Civil and Environmental ENGINEERING

Regional Nucleation Events





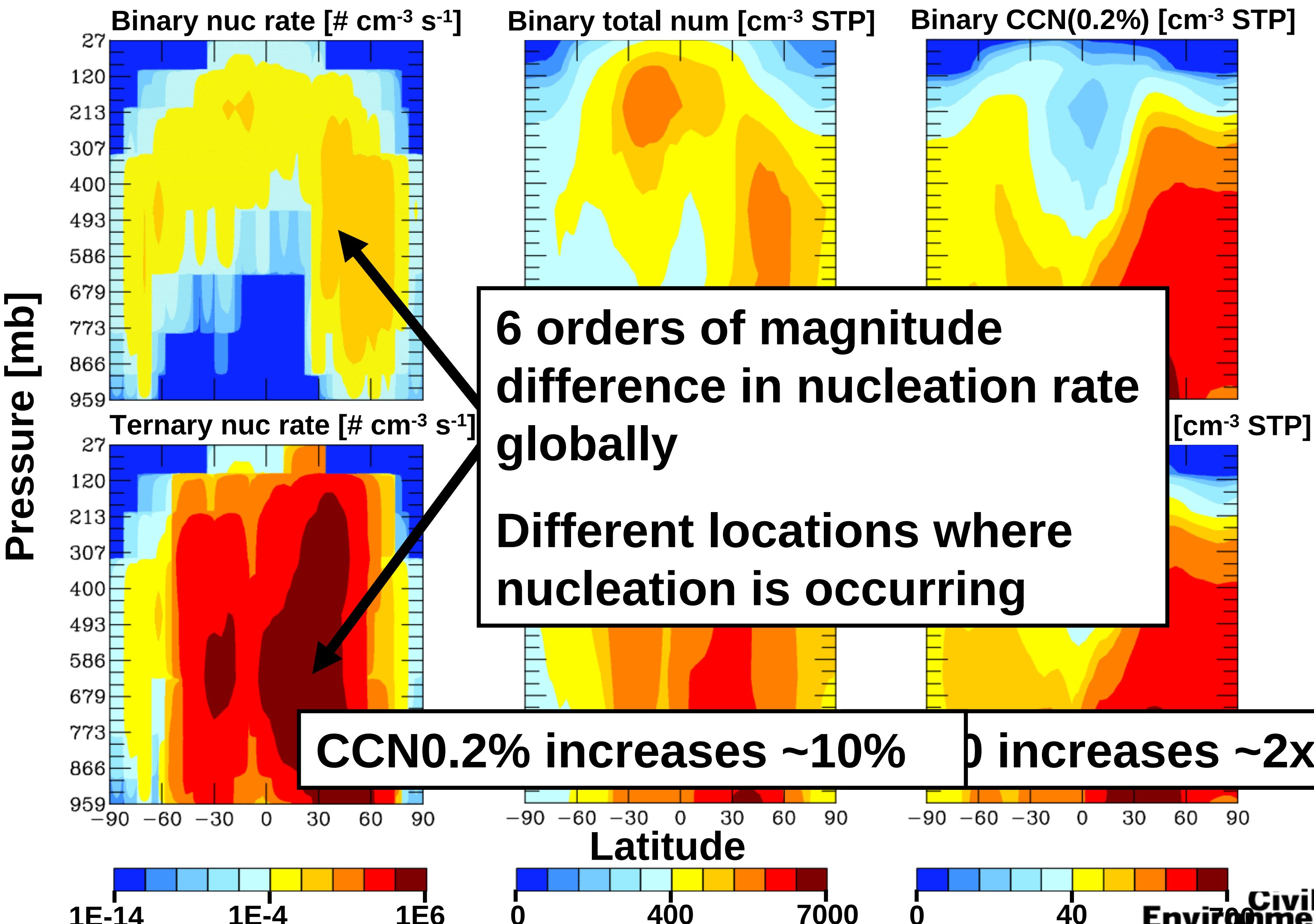
~400 km away

~600 km away

Nucleation and CCN

- Nucleation chemistry not understood
- Several proposed nucleation mechanisms
  - Binary (H_2SO_4-H_2O)
  - Ternary (H_2SO_4-NH_3-H_2O)
  - Ion-induced nucleation (also involves H_2SO_4)
- Nucleation rates vary by many orders of magnitude
- Scenario 1: Slow (binary) nucleation (Vehkamaki 2002)
- Scenario 2: Fast (ternary) nucleation (Napari 2002)

Nucleation and CCN



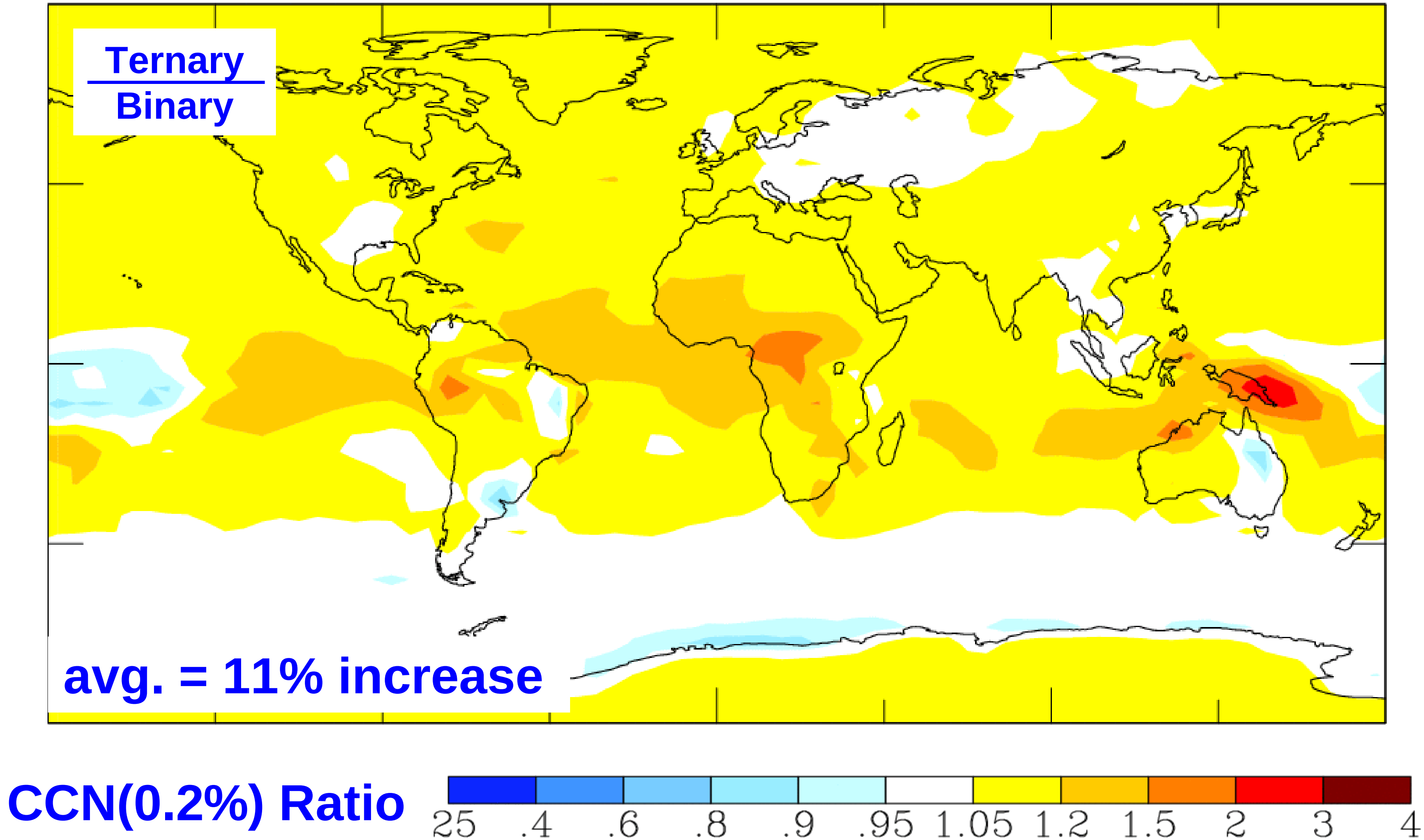
6 orders of magnitude difference in nucleation rate globally

Different locations where nucleation is occurring

CCN0.2% increases ~10%

) increases ~2x

Nucleation and CCN



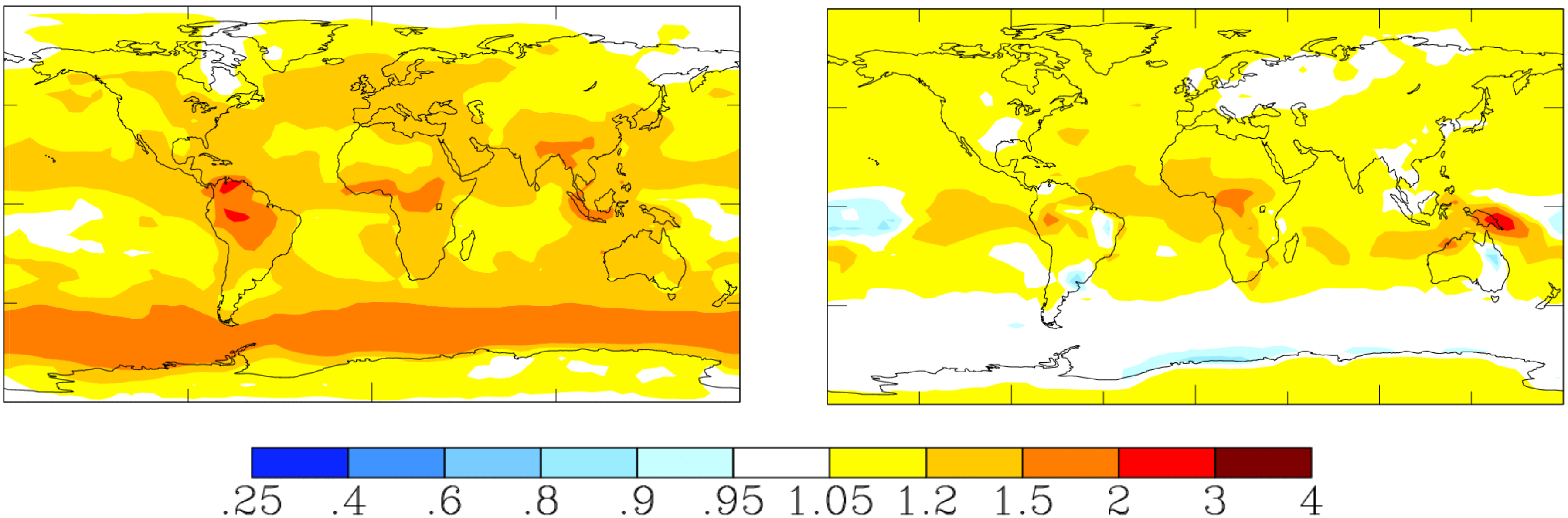
Primary Emissions

PE / PE/3

Sensitivity of CCN to emissions — **avg. = 27% increase**

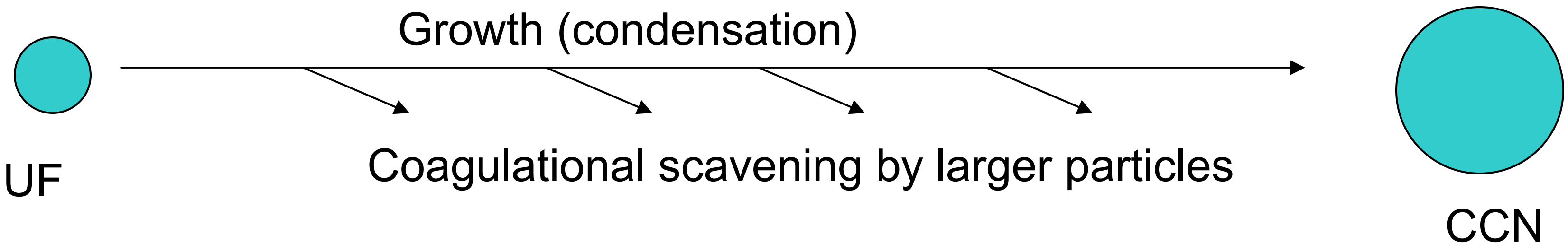
Ternary / Binary

Sensitivity of CCN to nucleation — **avg. = 11% increase**



- Primary emissions: EC/OC, plume sulfate, sea-salt
- Uncertainty from primary emssions greater
- Results subject to choice of “bounding values”
- Primary particles largely overlooked in terms of CCN budget

CCN Formation Probability

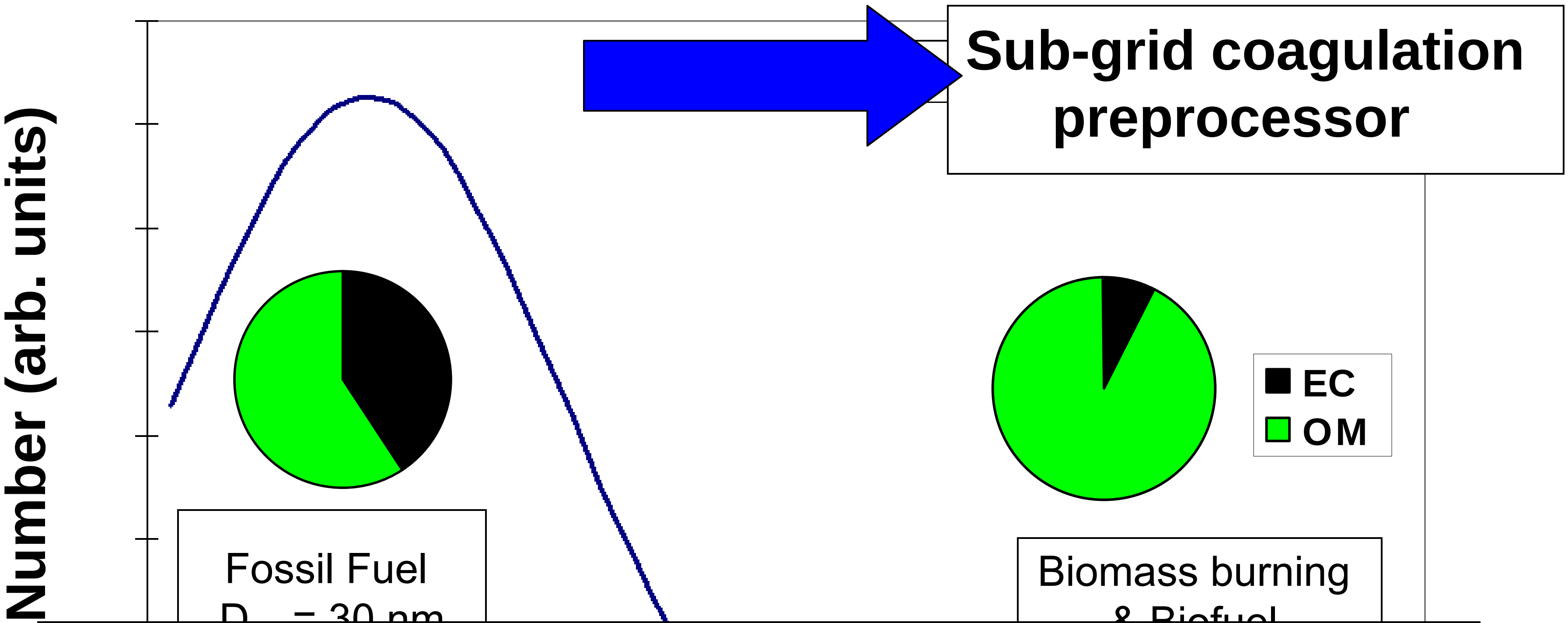


- For any UF particle, the probability of becoming a CCN is a competition between
  - Condensational growth
  - Coagulational scavenging
- Small nuclei suffer compared to primary particles
  - Takes longer to grow
  - More diffusive → higher collision probability
  - A greater number of larger particles to scavenge them

Black Carbon Reductions

- Many have suggested that BC reductions are a fast way to slow global warming
- But… BC controls will
  - Reduce primary particle emissions
  - CCN concentrations
  - Reduce the indirect effect (-0.3 to -1.8 W/m^2)
- *Will BC reductions slow global warming??*
- Collaborators: John Seinfeld/Anne Chen (Caltech); Thanos Nenes (GaTech); Yunha Lee (CMU)

BC Reductions: Primary Particles



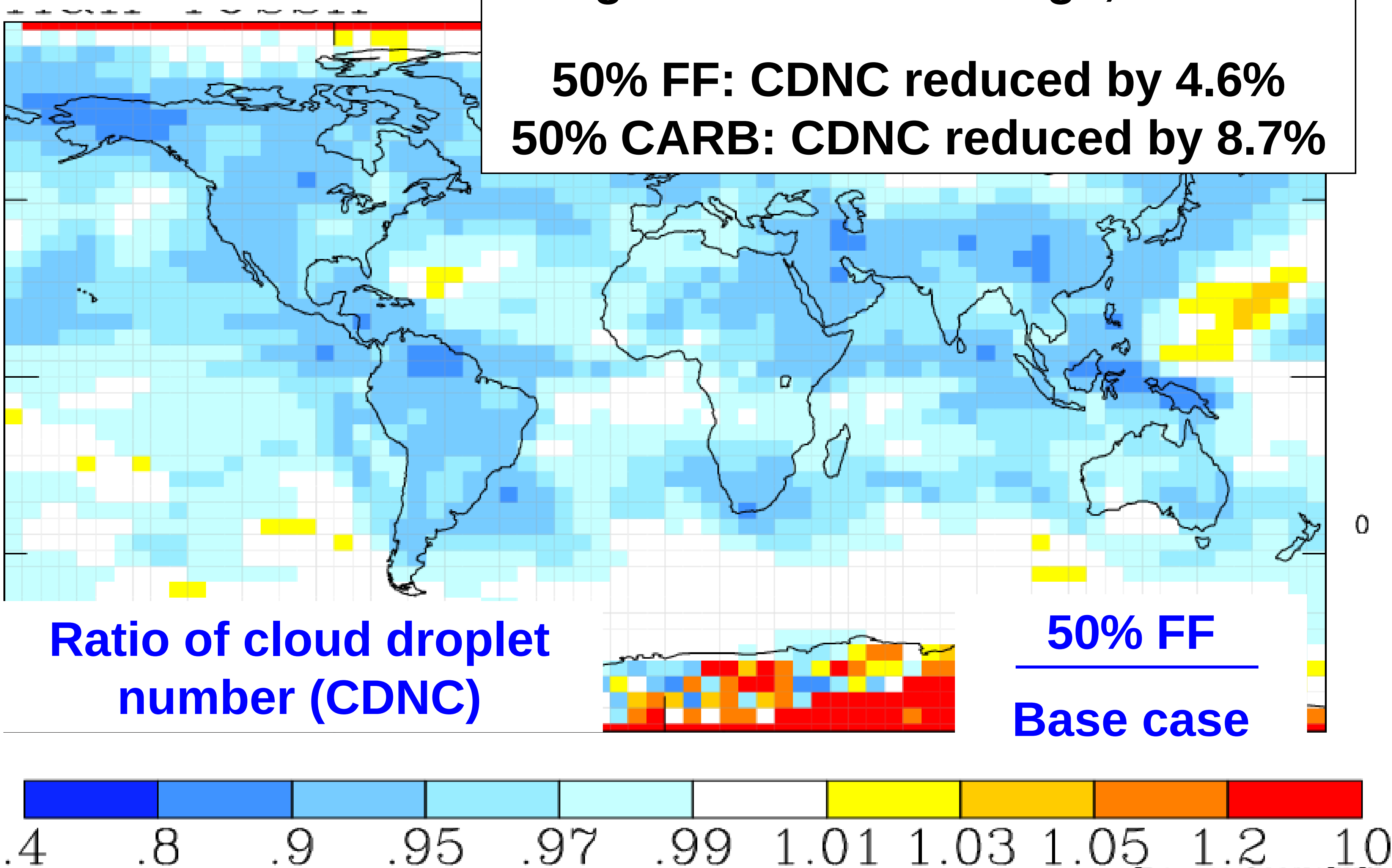
Scenarios:

- **Base case**
- **50% FF: reduce fossil fuel emisions by 50% (EC,OM,N)**
- **50% CARB: reduce all carbonaceous emissions by 50%**

BC Controls Reduce CDNC

In global annual average,

50% FF: CDNC reduced by 4.6%
50% CARB: CDNC reduced by 8.7%



Ratio of cloud droplet number (CDNC)

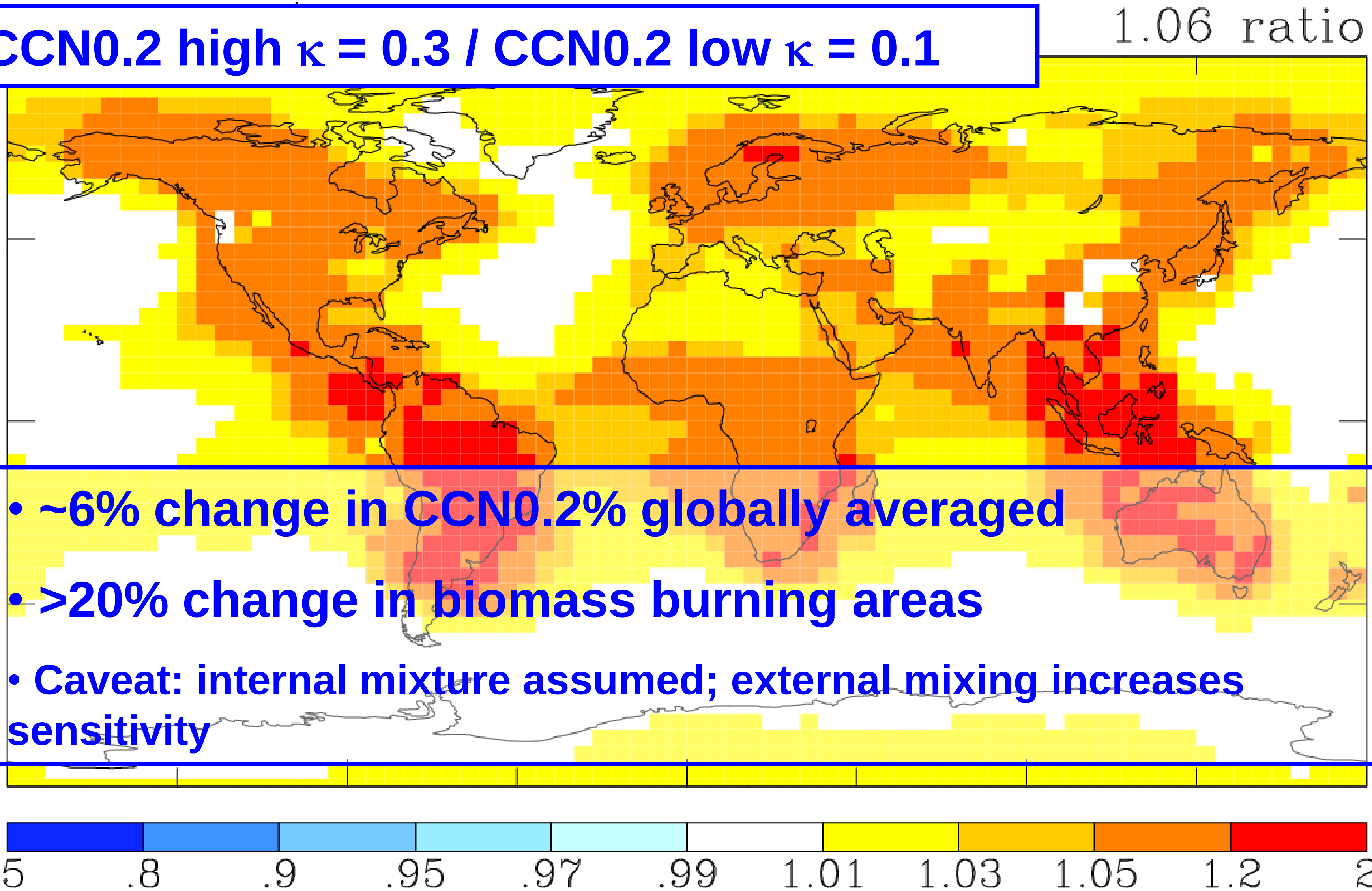
50% FF / Base case

BC Reductions: Forcing Assessment

- For a 50% reduction in fossil fuel EC/OC/N:
  - FF-BC absorption: 0.2 W/m^2 $\rightarrow$ 0.1 W/m^2 = -0.1 W/m^2
  - Semi-direct: 0.3 W/m^2 $\rightarrow$ 0.23 W/m^2 = -0.07 W/m^2
  - Snow albedo: 0.1 W/m^2 $\rightarrow$ 0.07 W/m^2 = -0.03 W/m^2
  - Net: -0.2 W/m^2 (reduced global warming)
- But…
  - Reduced indirect effect (this work) = +0.22 W/m^2
  - Reduced OC cooling = ???
  - Are BC reductions approx. climate neutral?
- Caution: preliminary results
- CCN impacts of reducing black carbon appear to largely (completely?) offset climate benefits

Organic Aerosol Hygroscopicity

CCN0.2 high κ = 0.3 / CCN0.2 low κ = 0.1



- **~6% change in CCN0.2% globally averaged**
- **>20% change in biomass burning areas**
- **Caveat: internal mixture assumed; external mixing increases sensitivity**

SOA Source

•SOA production is major uncertainty on condensational growth of UF to CCN

Compare CCN0.2% predicted in:

•Base SOA Scenario: 19 Tg/yr (traditional biogenic)

•High SOA Scenario: 64 Tg/yr (+45 Tg/yr generic "anthropogenic")

Generic SOA "mechanism":

•45 Tg/yr generic precursor emitted

•Co-located with SO_2 (and ultrafine particles)

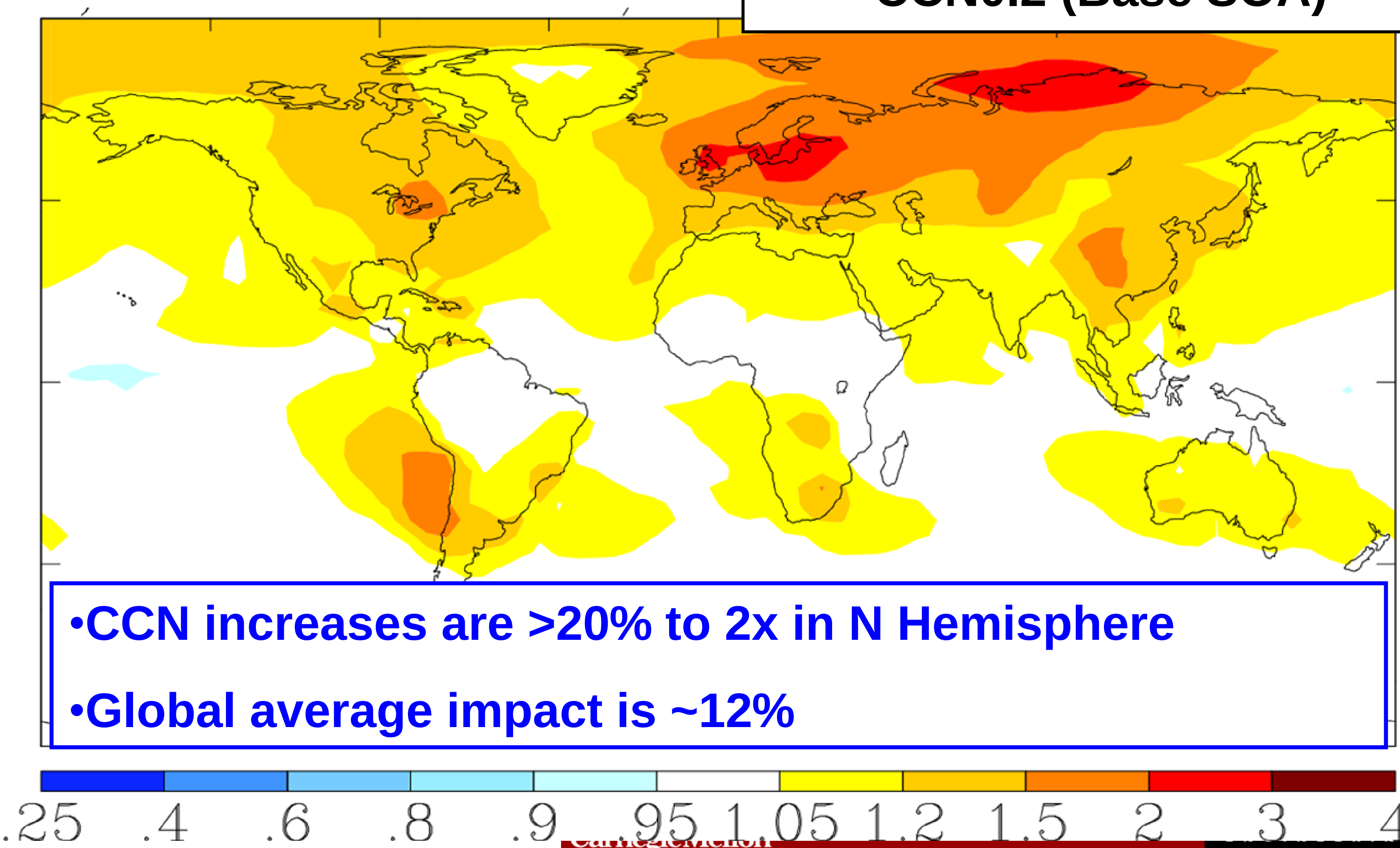
•Oxidation timescale: 12 hours and 100% yield

•Condensation only (non-volatile)

SOA Source

Plot shows ratio:

$$\frac{\text{CCN0.2 (High SOA)}}{\text{CCN0.2 (Base SOA)}}$$



- **CCN increases are >20% to 2x in N Hemisphere**
- **Global average impact is ~12%**

Conclusions

- CCN Sensitivity to nicrophysics and chemistry

Factor	Bounds	CCN Sensitivity
Primary particles	3x	27%
SOA production	19 to 64 Tg/yr	12%
Nucleation	Binary/ternary	11%
OC hygroscopicity	κ from 0.1 to 0.3	6%

- CCN budget coupled to ultrafine mode
  - Large fraction of CCN derived from growth of UF particles
  - CCN very sensitive to primary particles (including BC reductions)
  - …but coagulational scavenging of UF particles also essential (e.g. relatively low sensitivity to nucleation)