Closing the BC Gap: Emissions? Optics?

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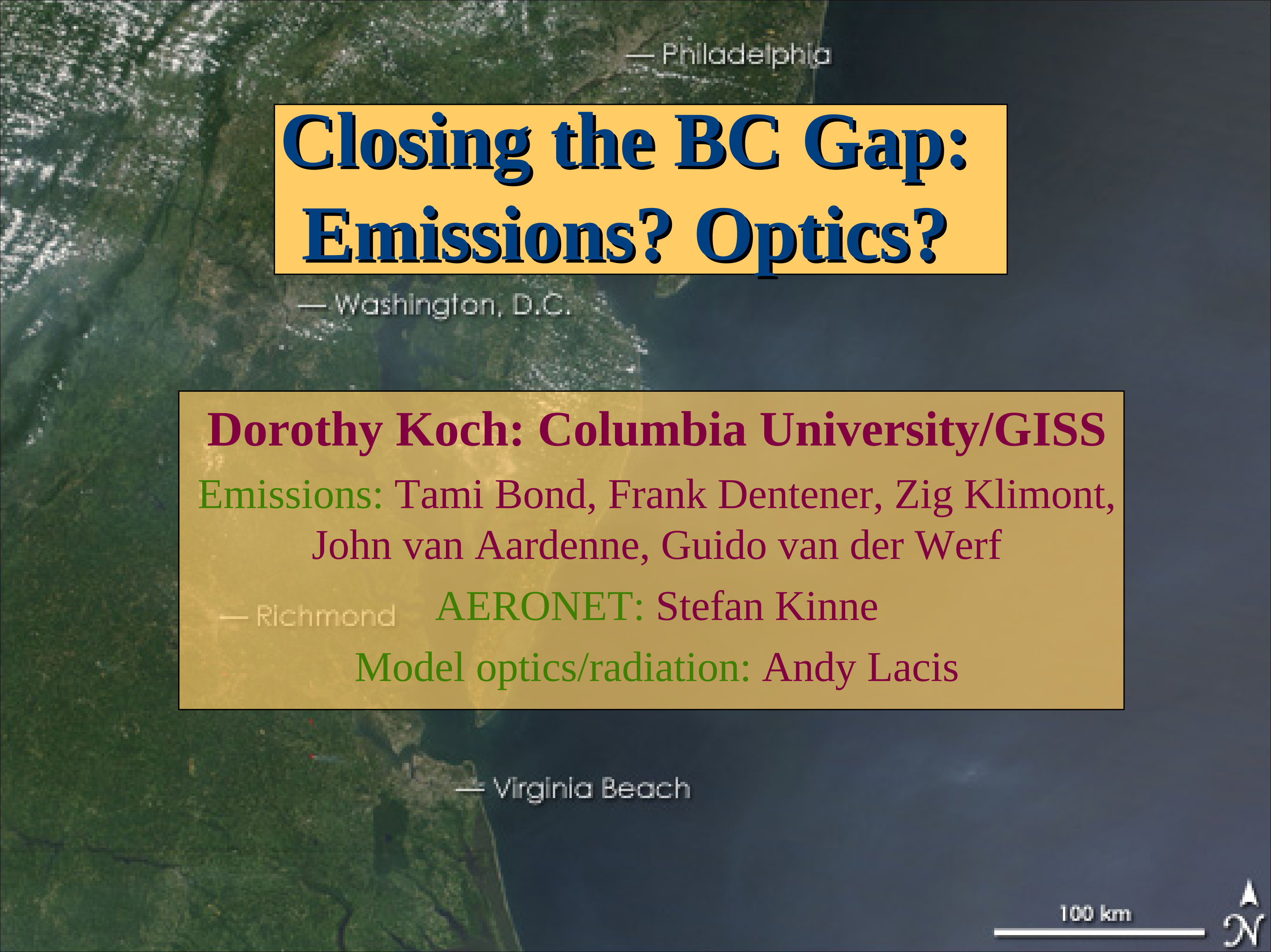
AERONET: Stefan Kinne

Model optics/radiation: Andy Lacis



Why examine BC absorption?

- Carbonaceous aerosols remain a major ‘wild card’ in understanding recent climate change, with highly uncertain direct, indirect and semi-direct effects
- Are GISS carbonaceous aerosol model regional biases relative to observations due to:

  Emissions?

  Optical property assumptions?

  Measurement errors?
- Sato *et al*. (PNAS, 2003) found that GISS and GOCART aerosol climatologies underestimated BC/OC absorption relative to AERONET by a factor of 2-4. These (older) GISS aerosols were therefore enhanced in the Hansen *et al*. climate simulations.

GISS model

- Aerosol mass simulation, external mixture

- Solubility:
  sulfate, sea salt, dust: fully soluble
  Energy BC, OC: soluble after aging
  Biomass burning BC, OC: fixed solubility

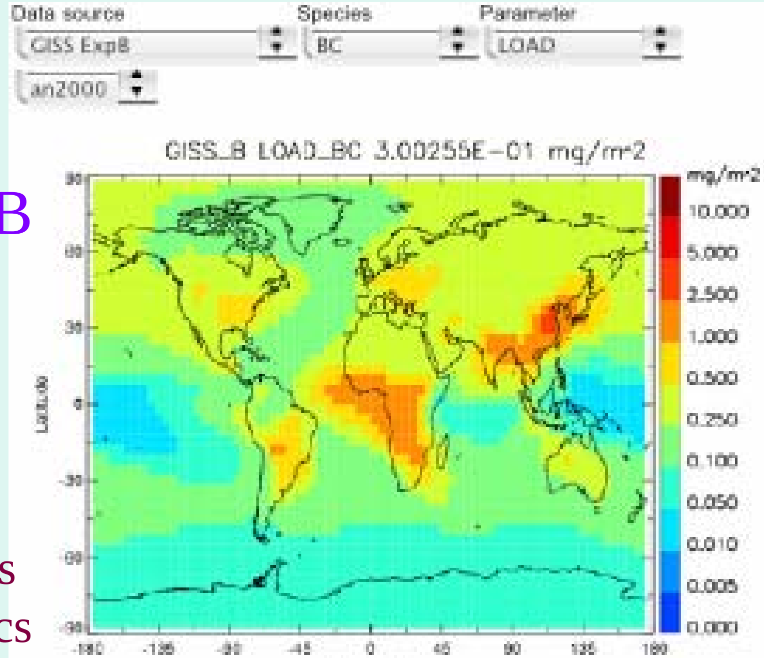
Aerosol Model Intercomparisons

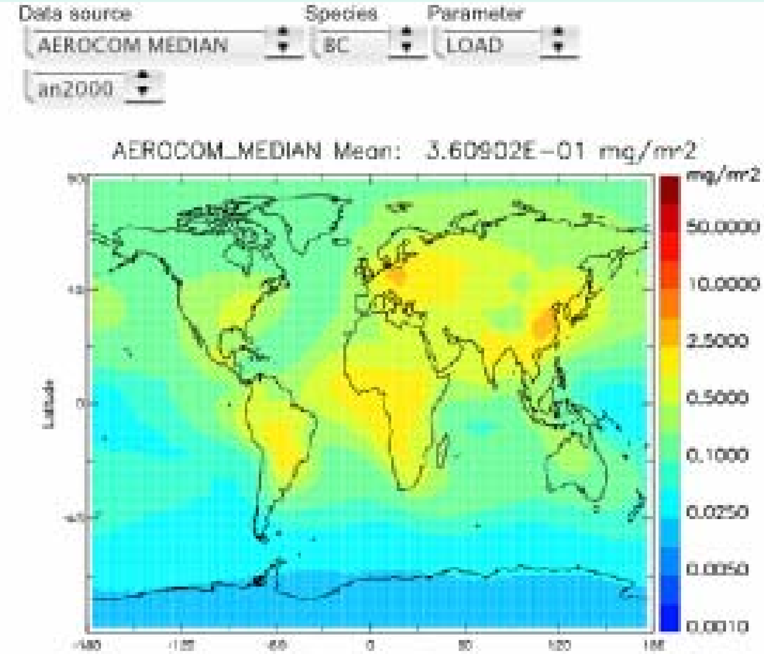
- **VA Beach** 1995 Rn222, Pb210
- Cambridge, England Pb210, sulfate
- COSAM Halifax, Nova Scotia, sulfate, Pb210
- IPCC intercomparison, Hamburg, Germany, all aerosols
- AEROCOM I Paris, France
- AEROCOM II Ispra, Italy
- AEROCOM III New York, USA
- AEROCOM IV Oslo, Norway
- AEROCOM V **VA Beach**, USA

GISS model | Mean of AEROCOM models

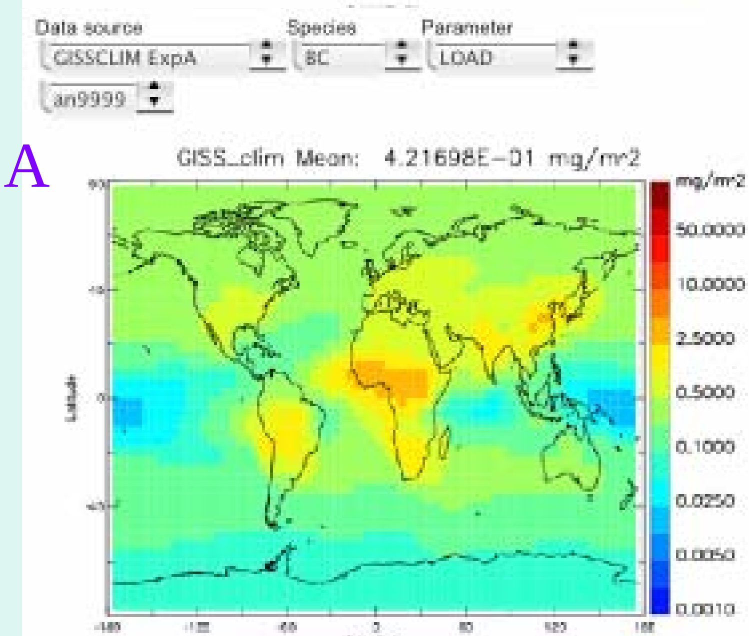
Experiment B

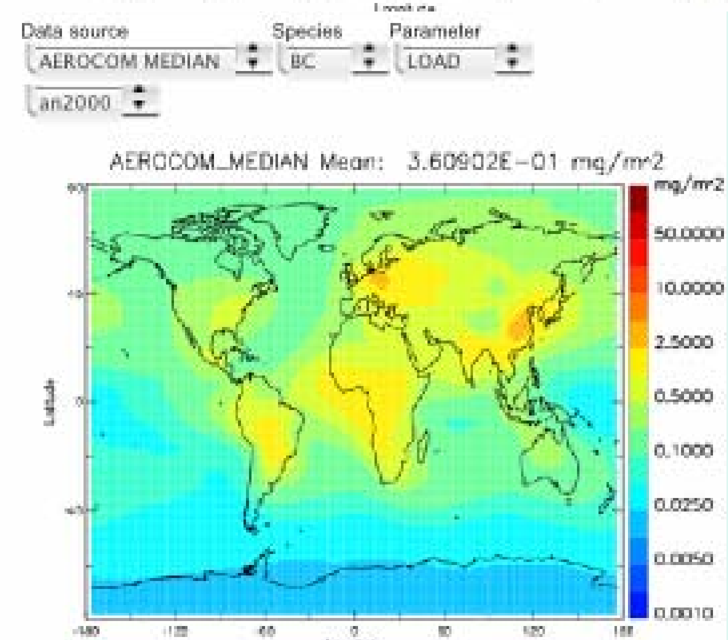
Compared with "Mean AEROCOM":
Total BC load is slightly less
More BC at poles
Less BC in tropics



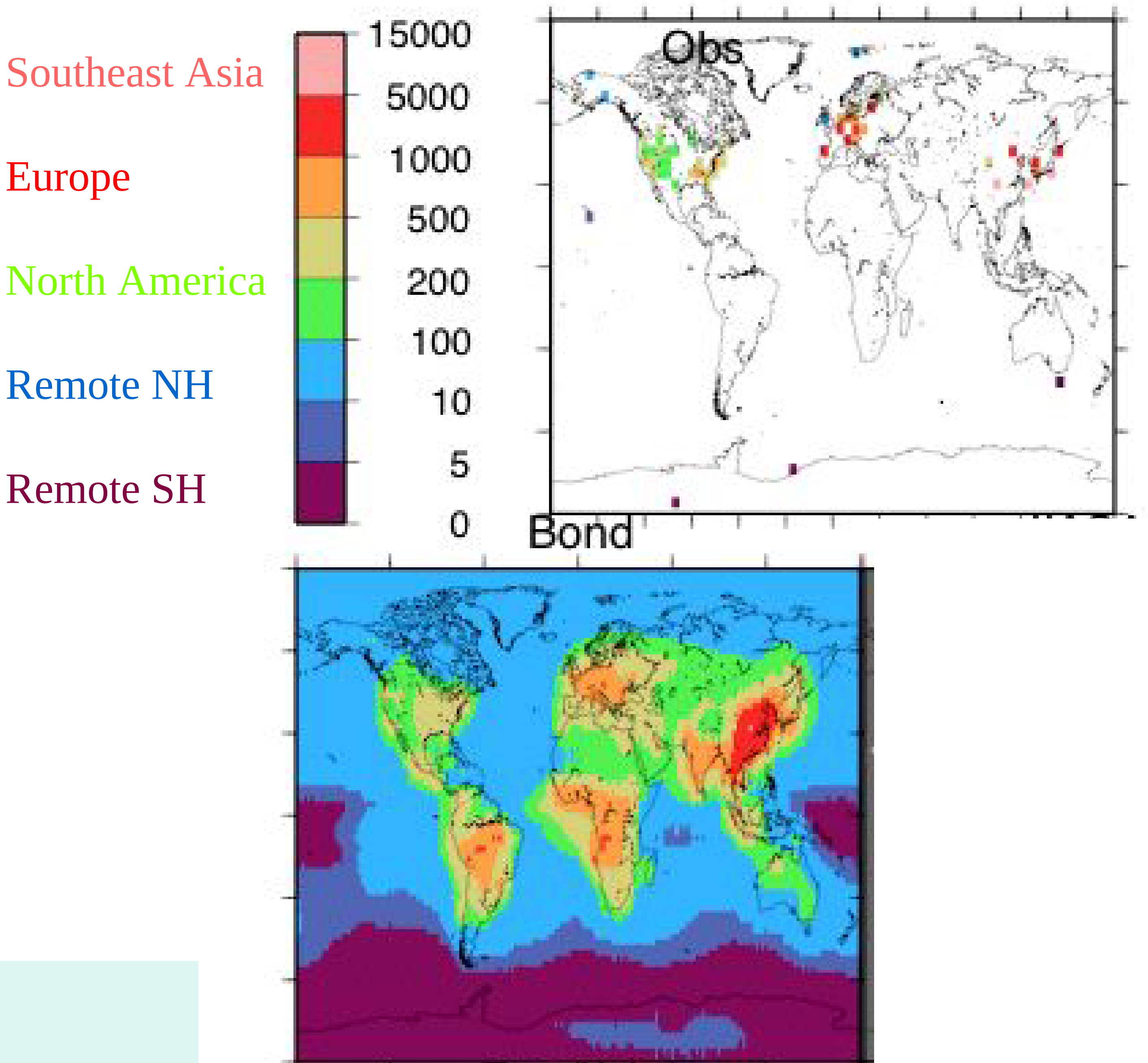


Experiment A

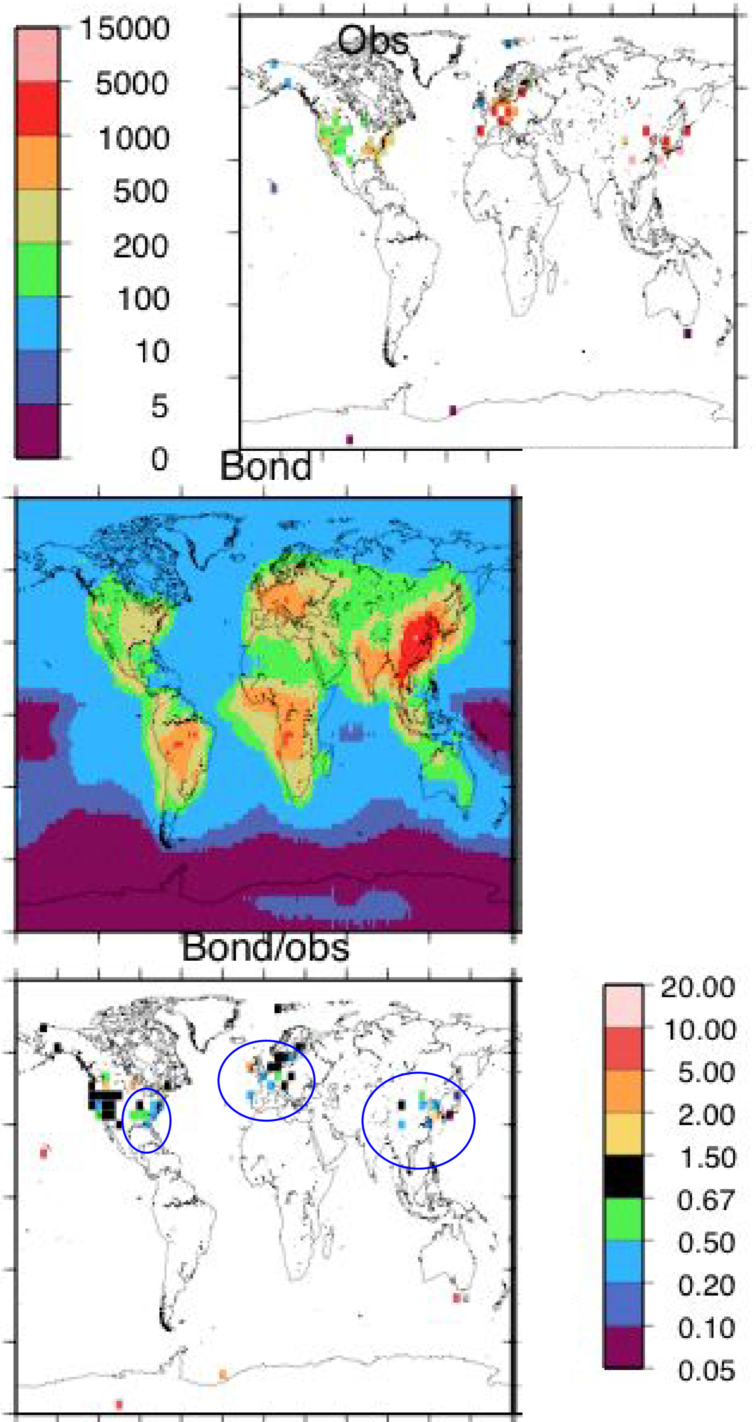




Model comparison with BC surface concentrations



Model comparison with BC surface concentrations



Bond et al emissions inventory

BC < observed in:

Eastern US

Europe

Southeastern Asia

New Present-day Carbonaceous Emission Inventories!!

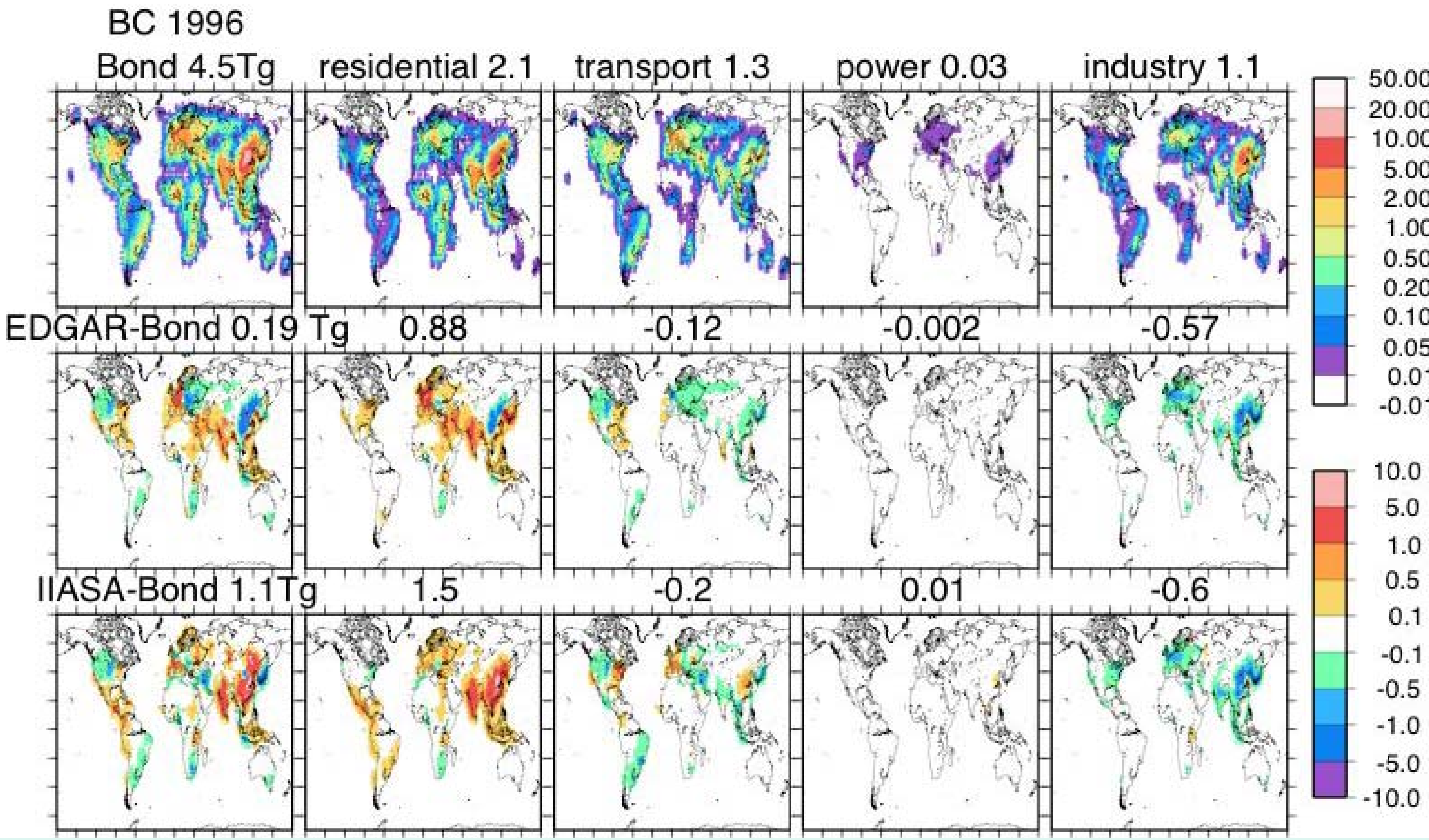
- Energy-related emissions (1995):

1. Bond et al. (2004) (**AEROCOM**)
2. IIASA (Klimont, Amman et al)
3. EDGAR (van Aardenne et al)

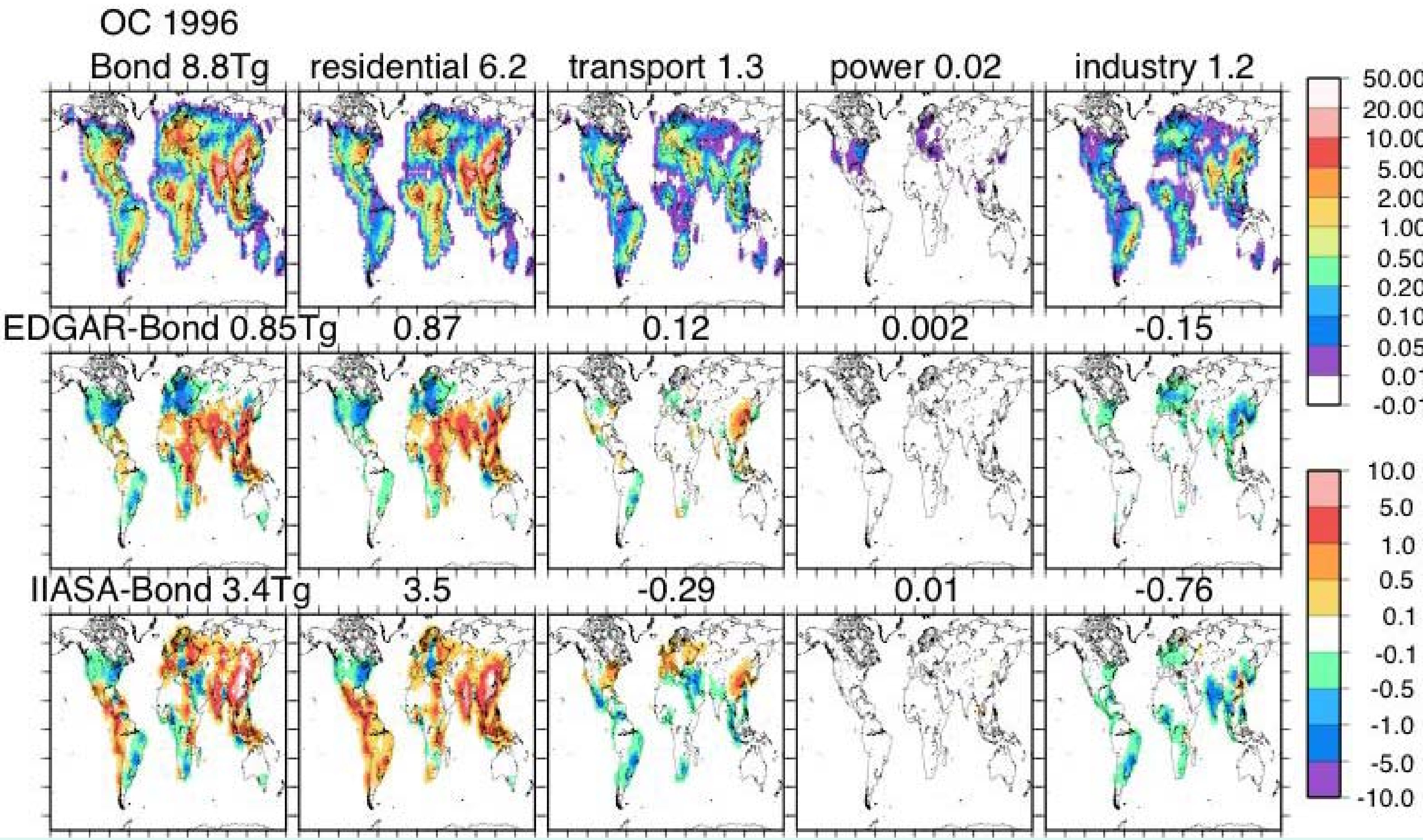
- Biomass Burning

1. GFED v1 1997-2001(**AEROCOM** 2000)
2. GFED v2 1997-2004

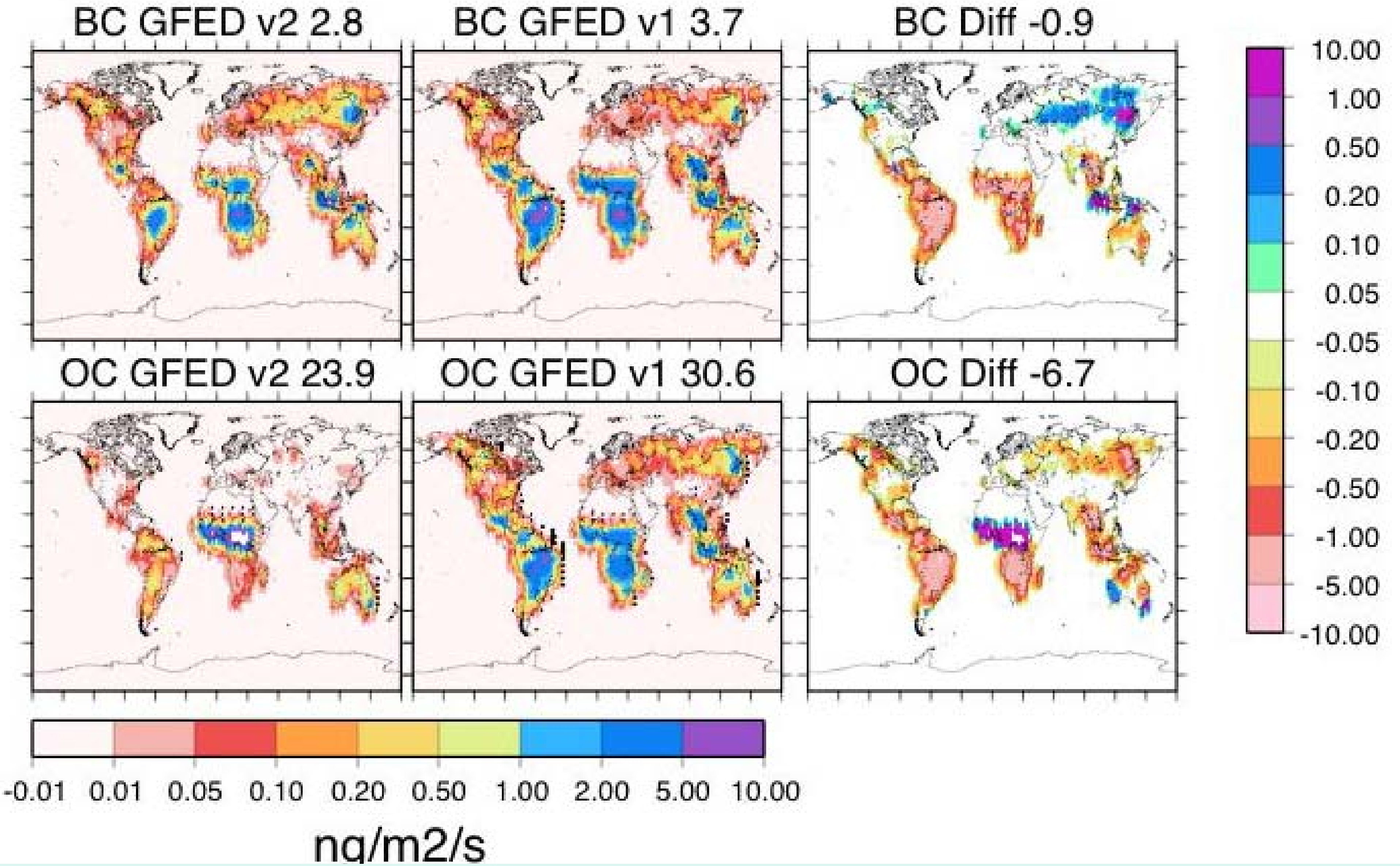
BC Energy-emissions



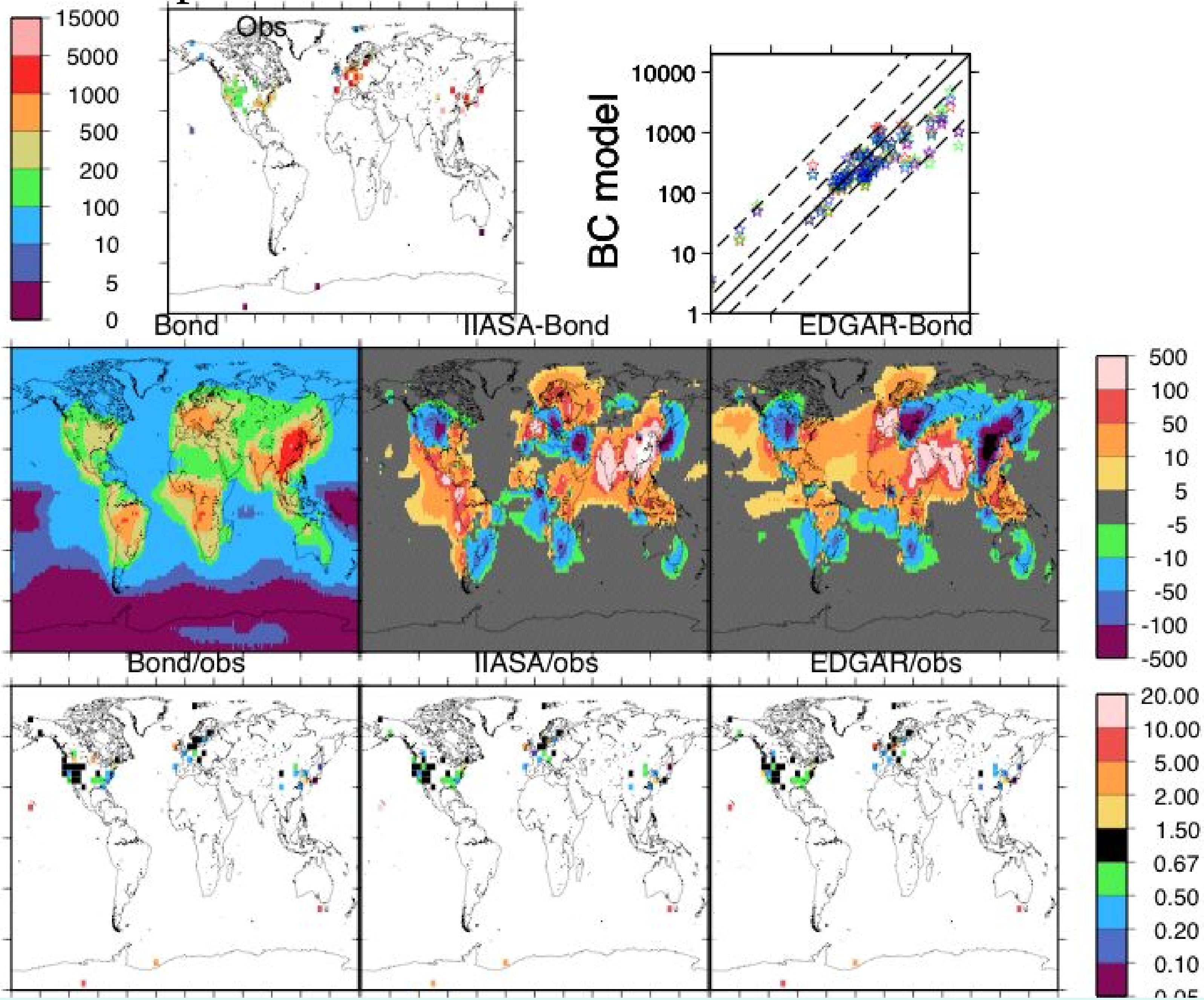
OC Energy-emissions



Biomass burning GFED comparison



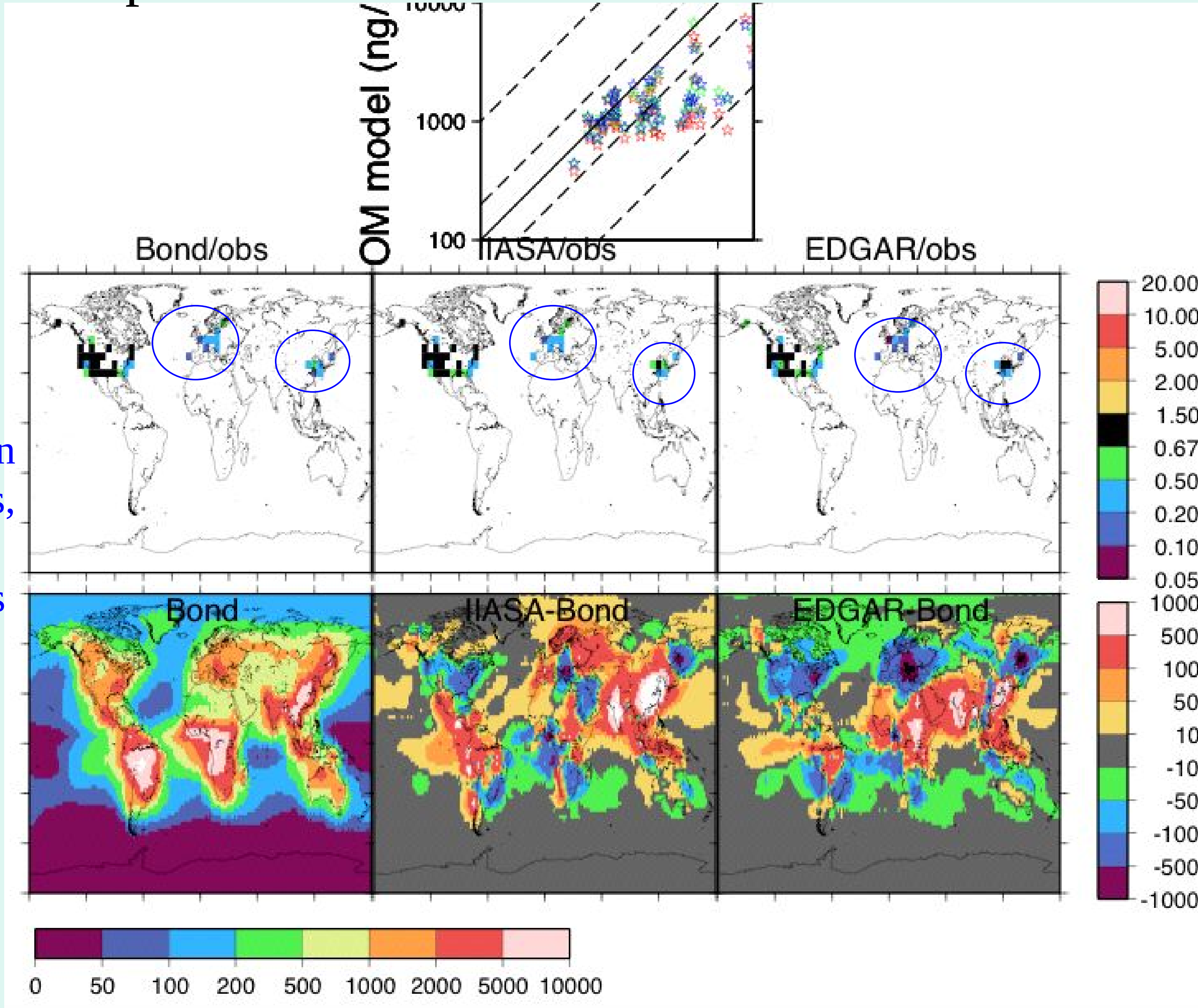
Model comparison with BC surface concentrations



IIASA or EDGAR may improve bias in some locations, however regional biases persist

Model comparison with OC surface concentrations

IIASA or EDGAR may improve bias in some locations, however regional biases persist



GISS model optics/radiation

- Aerosol mass simulation, external mixture
- Assumed effective radii:

  sea salt: 0.44, 1.7 μm

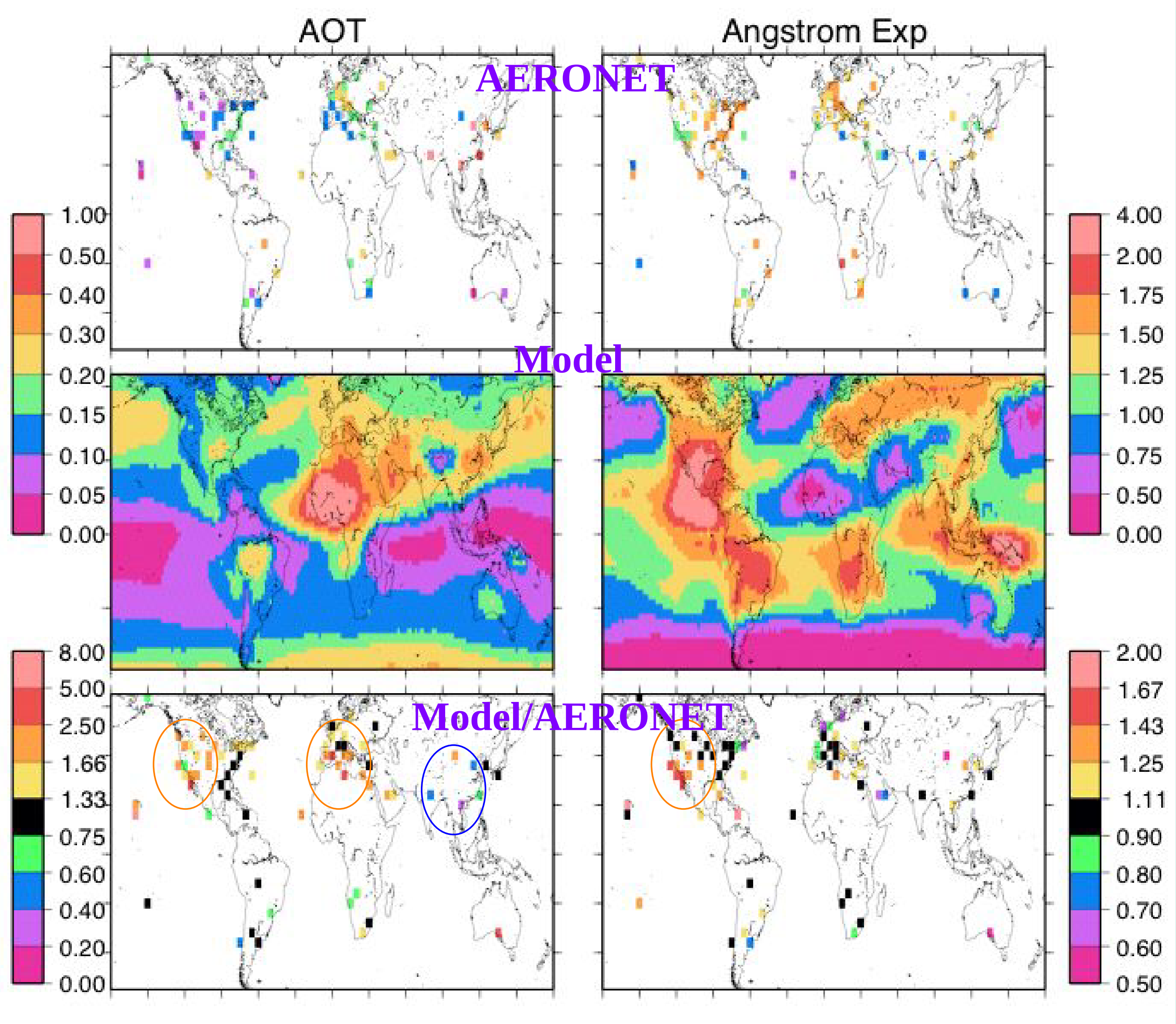
  dust: 0.13,0.23, 0.42, 0.77, 1.39, 2.77, 5.54 μm

  sulfate: 0.15 μm

  OC: 0.2 μm

  BC: 0.08 μm

AOT, Angstrom Exponent



Model AOT:
Too large in North America and Europe
Too small in Asia

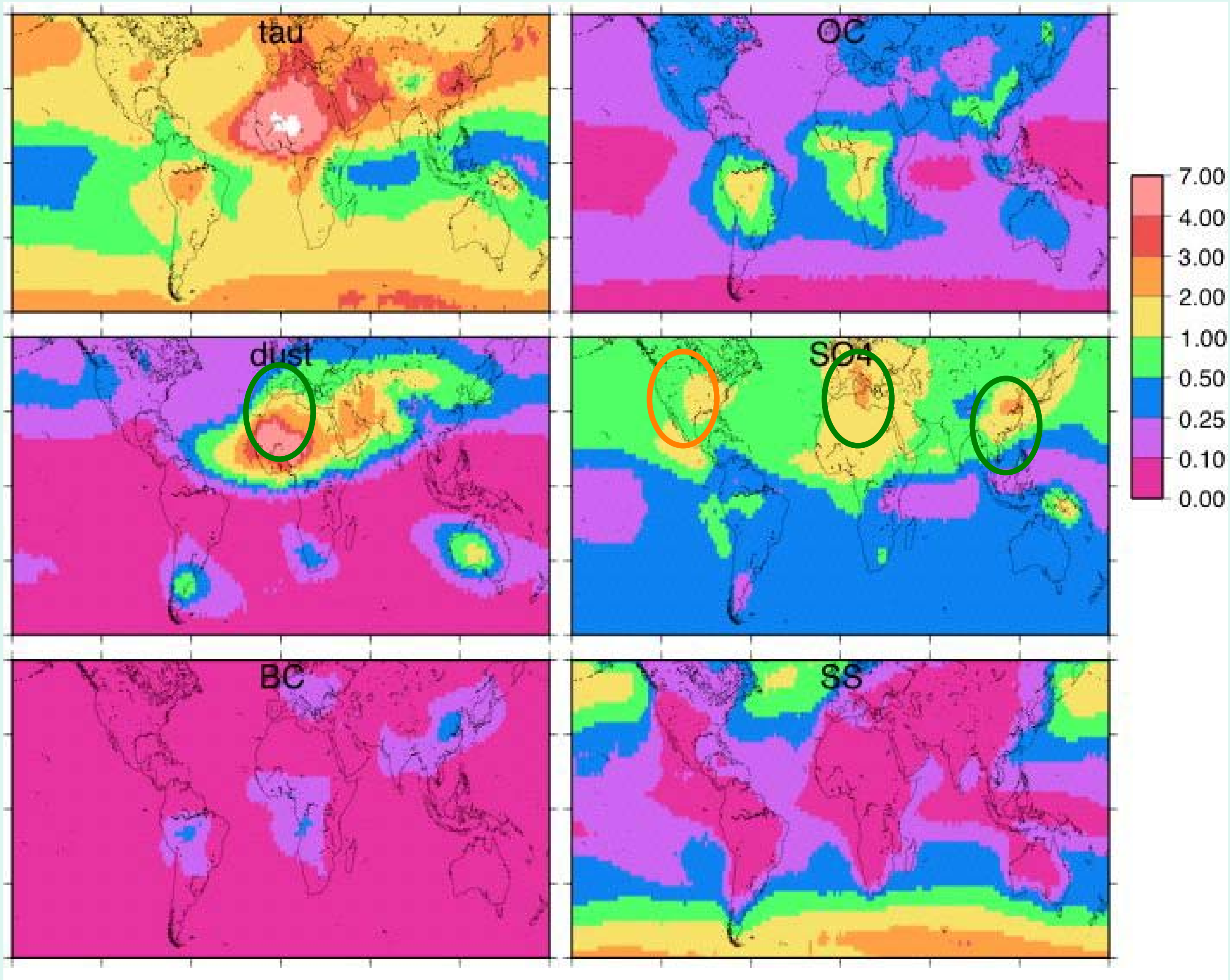
Angstrom Exp:
Particle sizes too small in western US

AOT composition

Model AOT:
Excessive sulfate might explain AOT anomalies in North America

Europe and Asia biases from combination of sulfate, dust and organics??

BC is minor player…



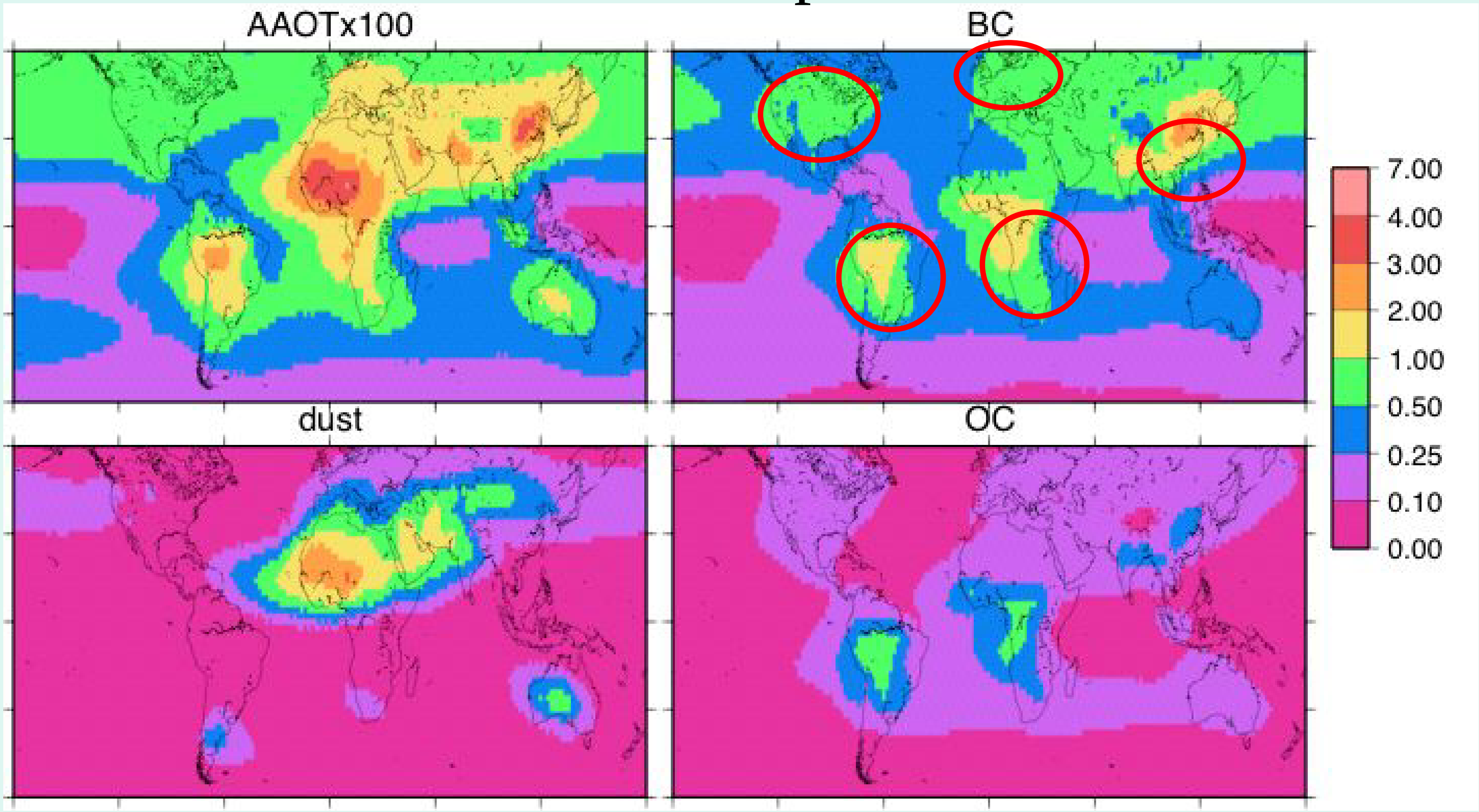
Absorbing AOT (AAOT)

- AAOT is appealing because in regions where BC dominates over other absorbers (dust, OC), it provides a measure of BC amount

- $AAOT=(1-SSA) \times AOT = AOT - AOT_{scattering}$

- SSA(=1-AAOT/AOT) is also sensitive to AOT

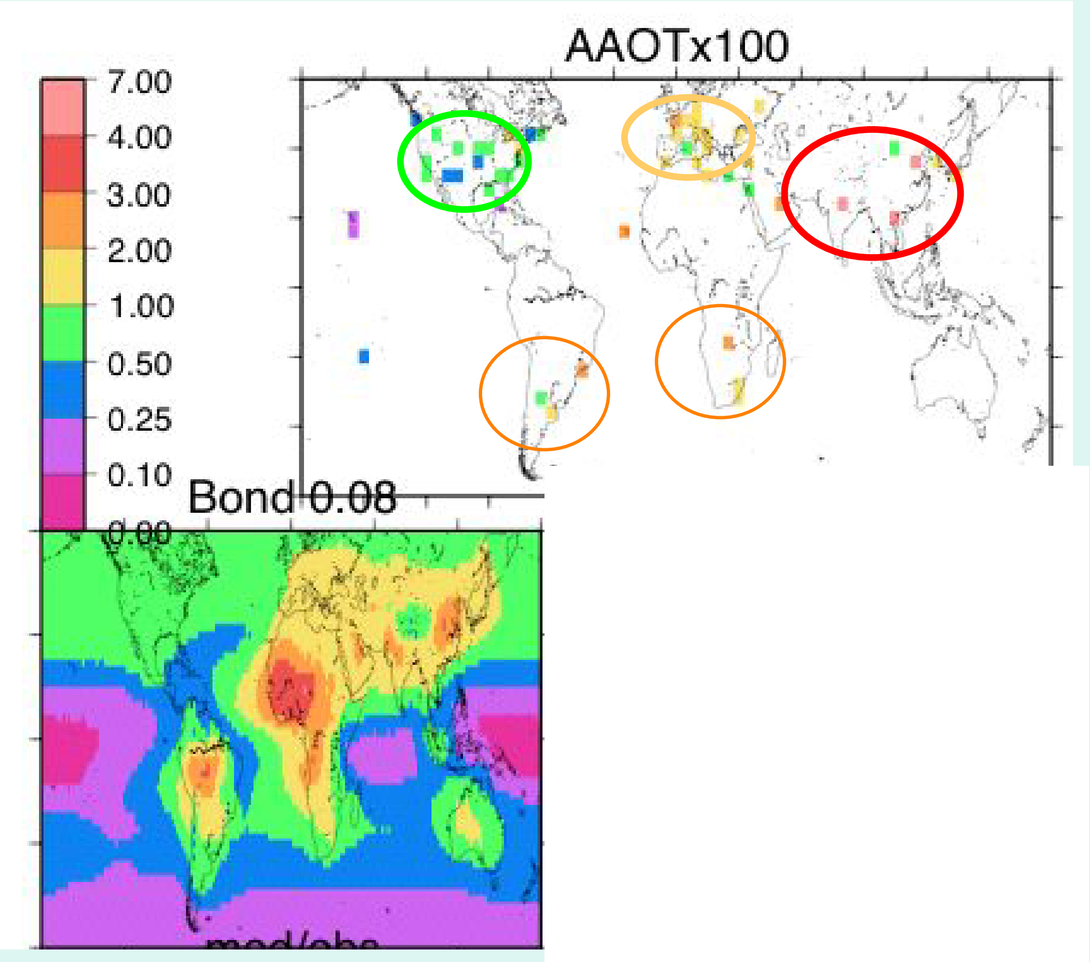
AAOT composition



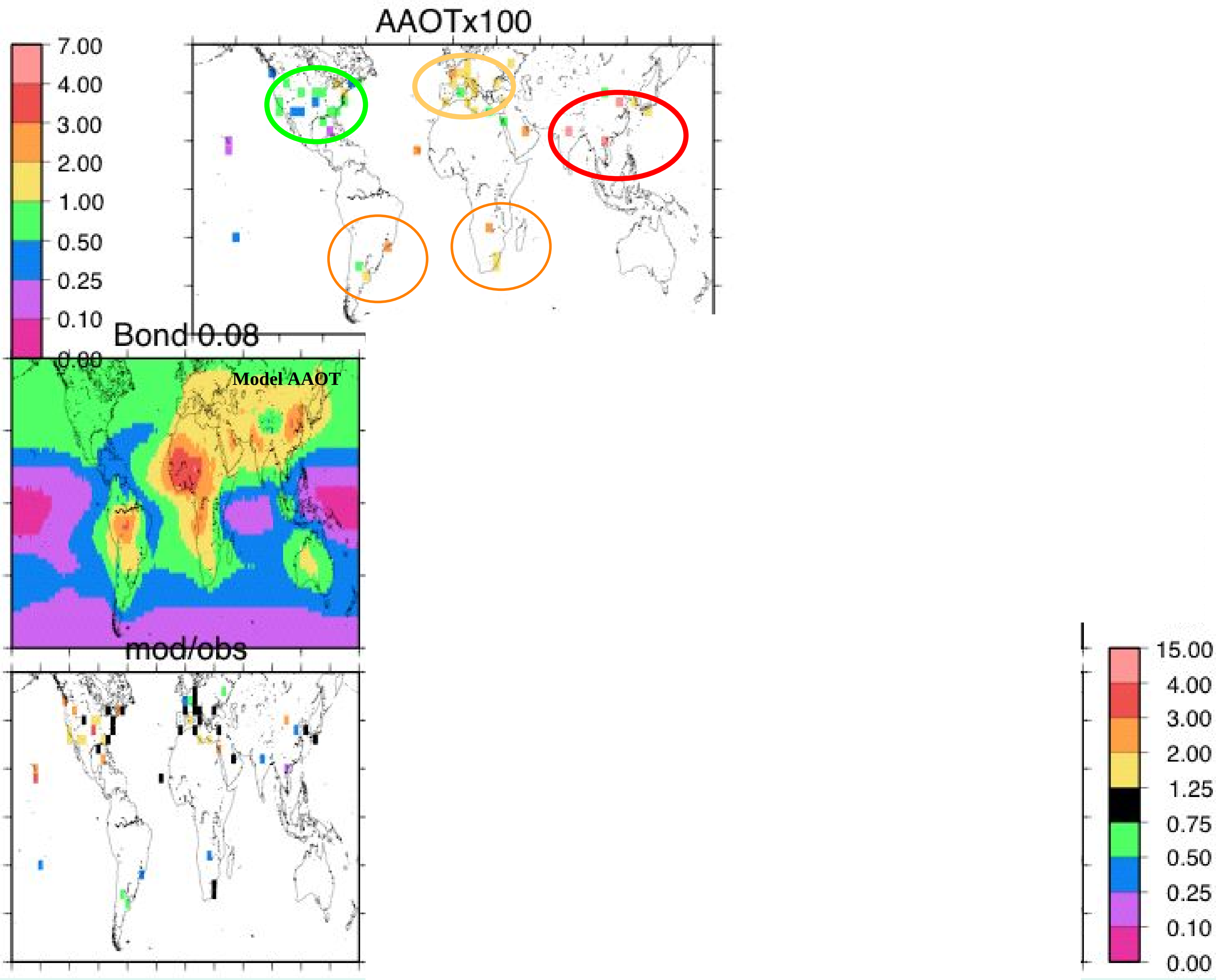
We will focus on regions with
BC AAOT >> dust AAOT

AAOT

AAOT has regional levels



AAOT



Model over-estimates North America and under-estimates Asia?

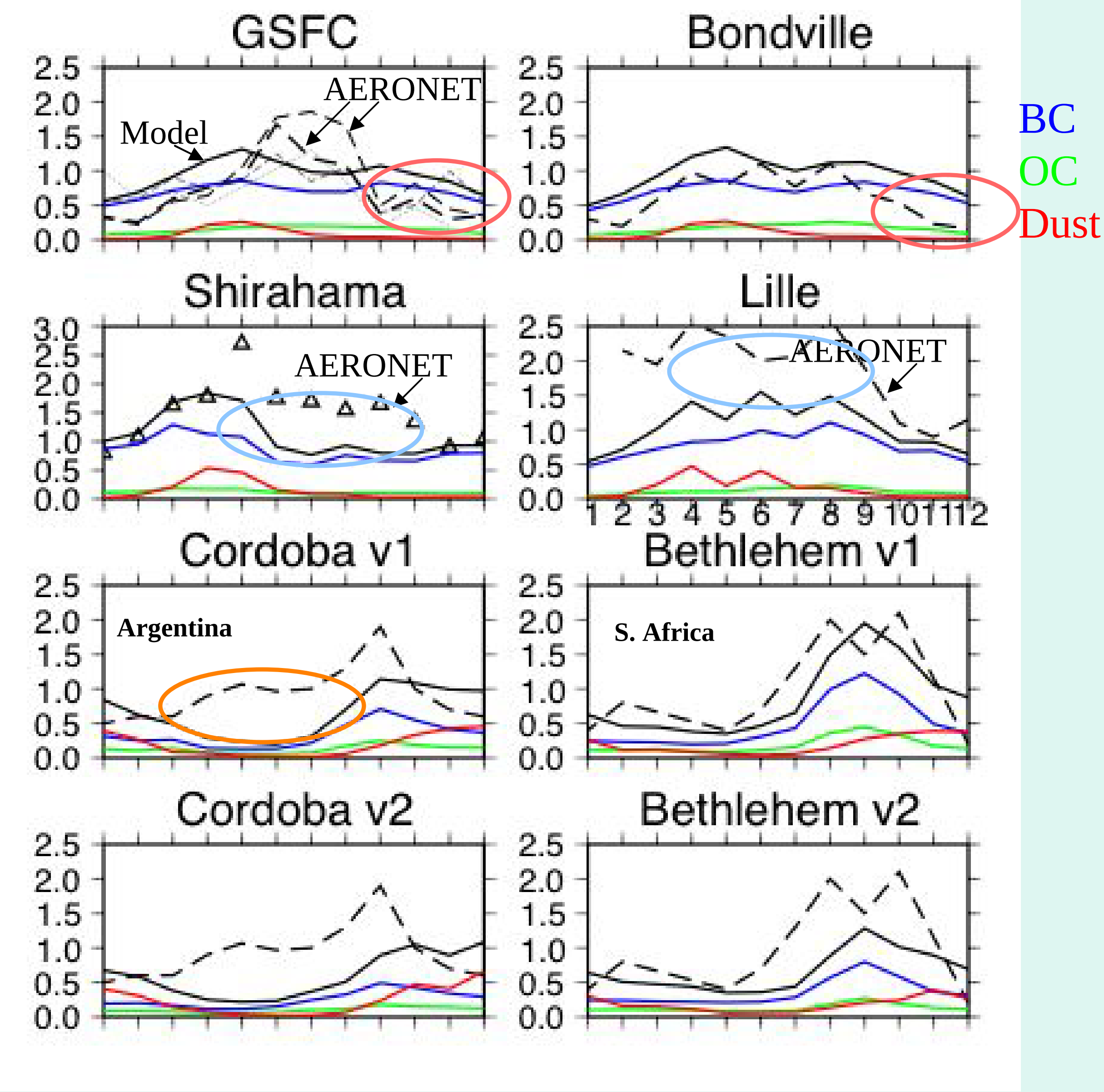
AAOT seasonalities

Model biases

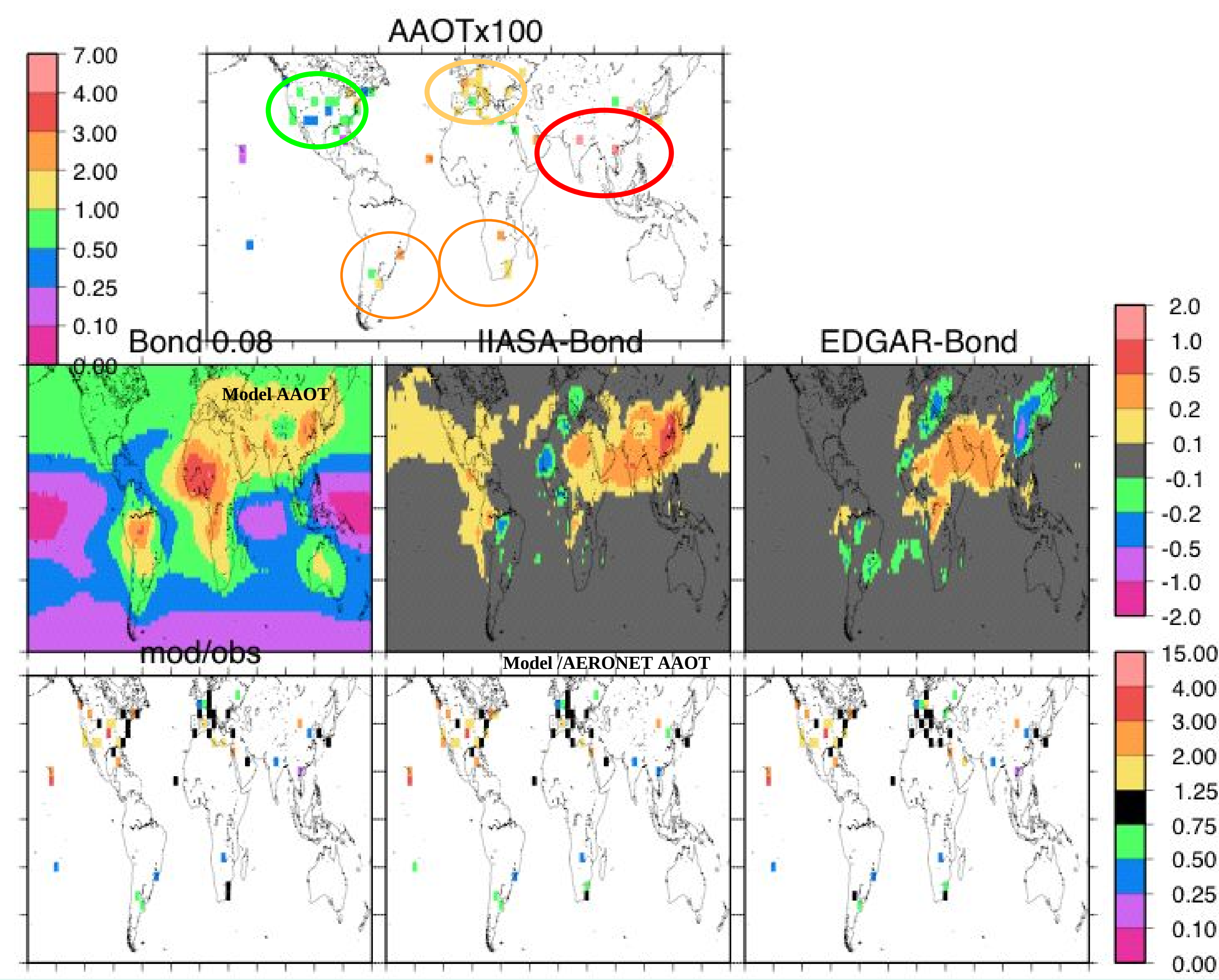
US: over-estimate in winter

Europe and Asia: under-estimate in summer

Argentina: missing urban sources?



AAOT



Overall regional biases persist with different emissions

New BC emission estimates do not help fix model surface concentration or AAOT biases

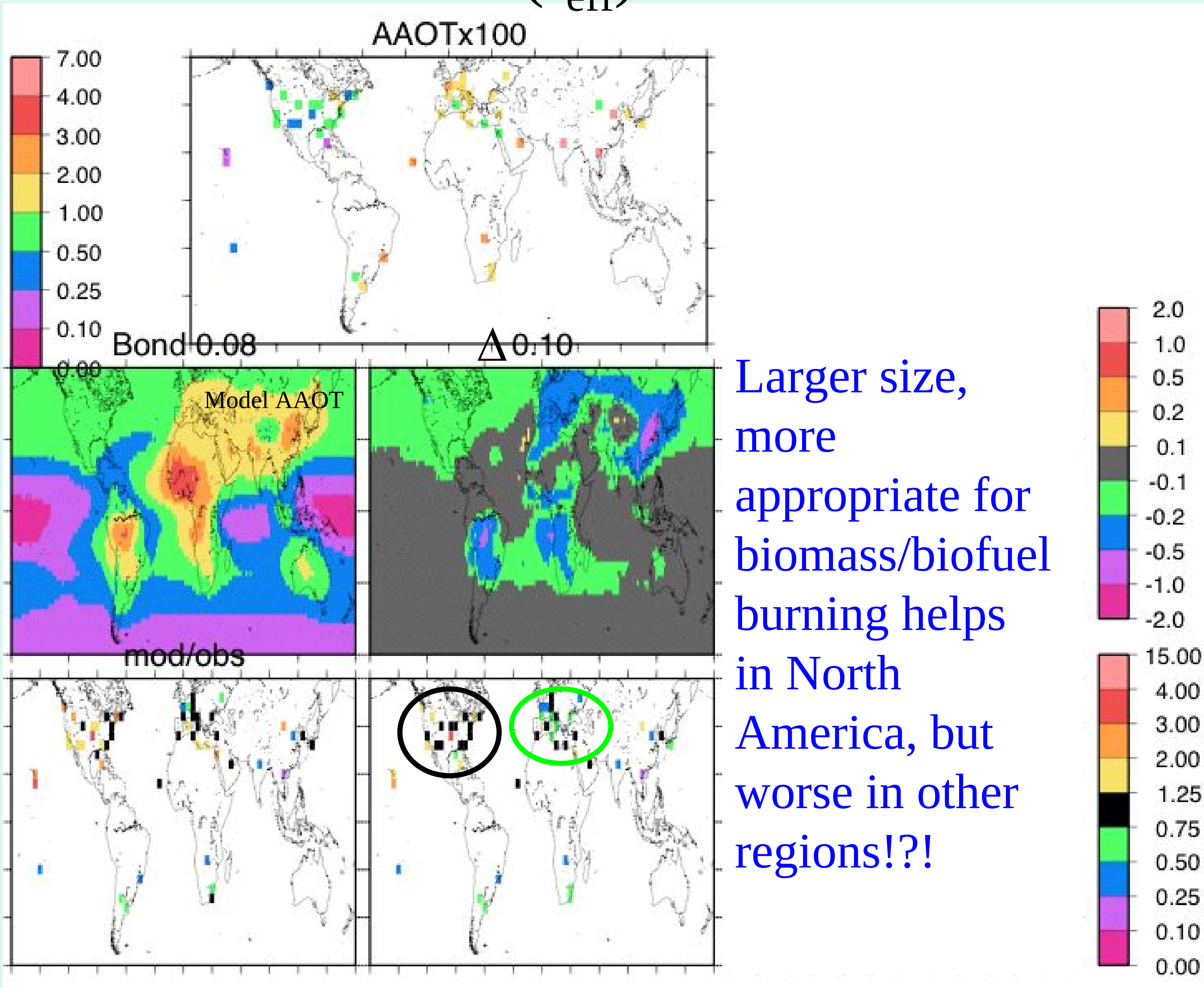
Aerosol Effective Radius Assumptions

- AEROCOM Primary Particle r_{eff}:
  - Biomass and biofuel 0.095 μm
  - Traffic 0.036 μm
  - Industrial 1.66 μm

- We have assumed BC r_{eff}=0.08 μm
  - Now change to r_{eff}=0.06, 0.1 μm

  (At these sizes, absorption decreases as size increases)

AAOT $f(r_{eff})$

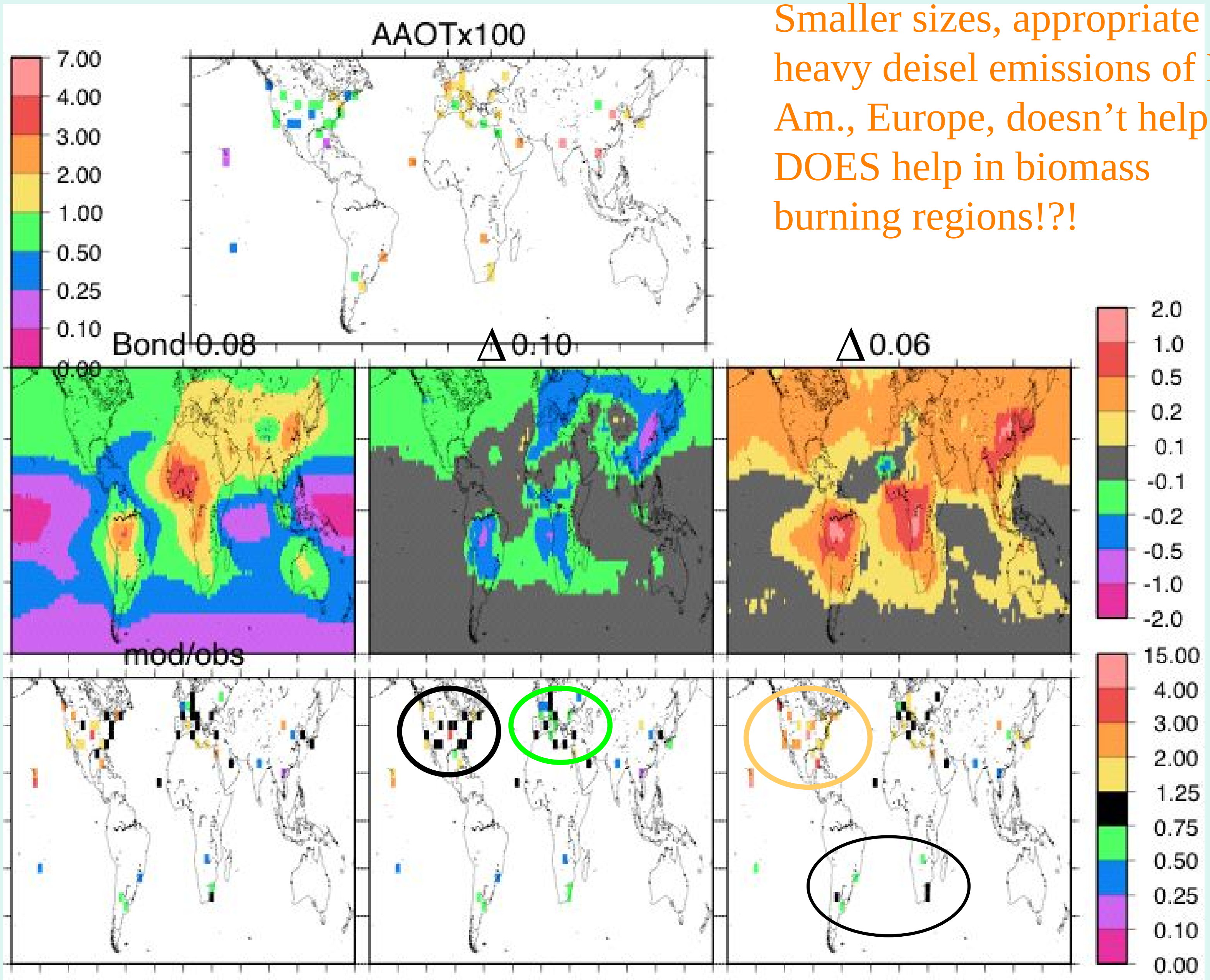


Changing aerosol size has larger impact on AAOT than changing emissions

Larger size, more appropriate for biomass/biofuel burning helps in North America, but worse in other regions!?!

AAOT $f(r_{eff})$

Smaller sizes, appropriate for heavy deisel emissions of N. Am., Europe, doesn't help; It DOES help in biomass burning regions!?!

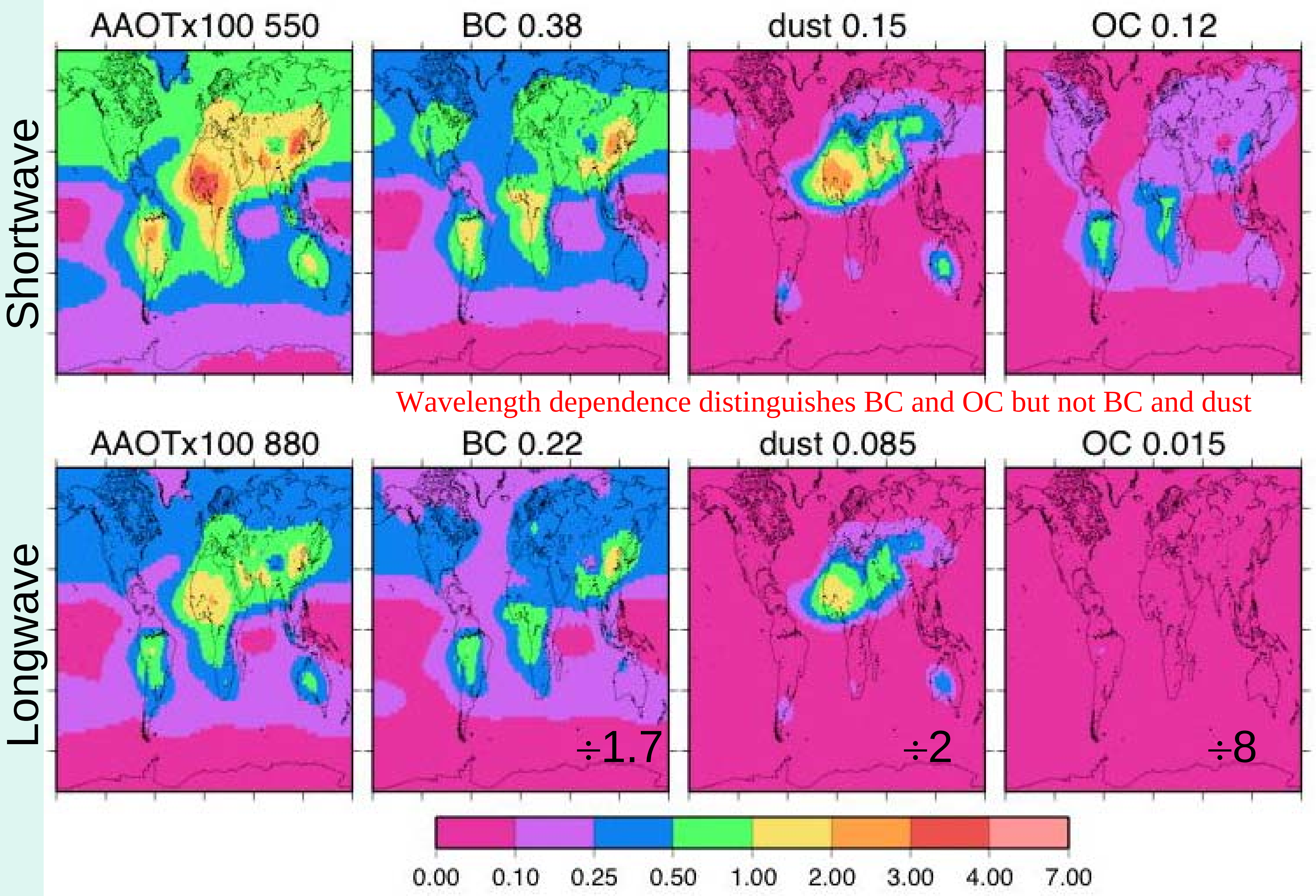


Changing BC effective radius in a logical direction does not help fix model AAOT bias

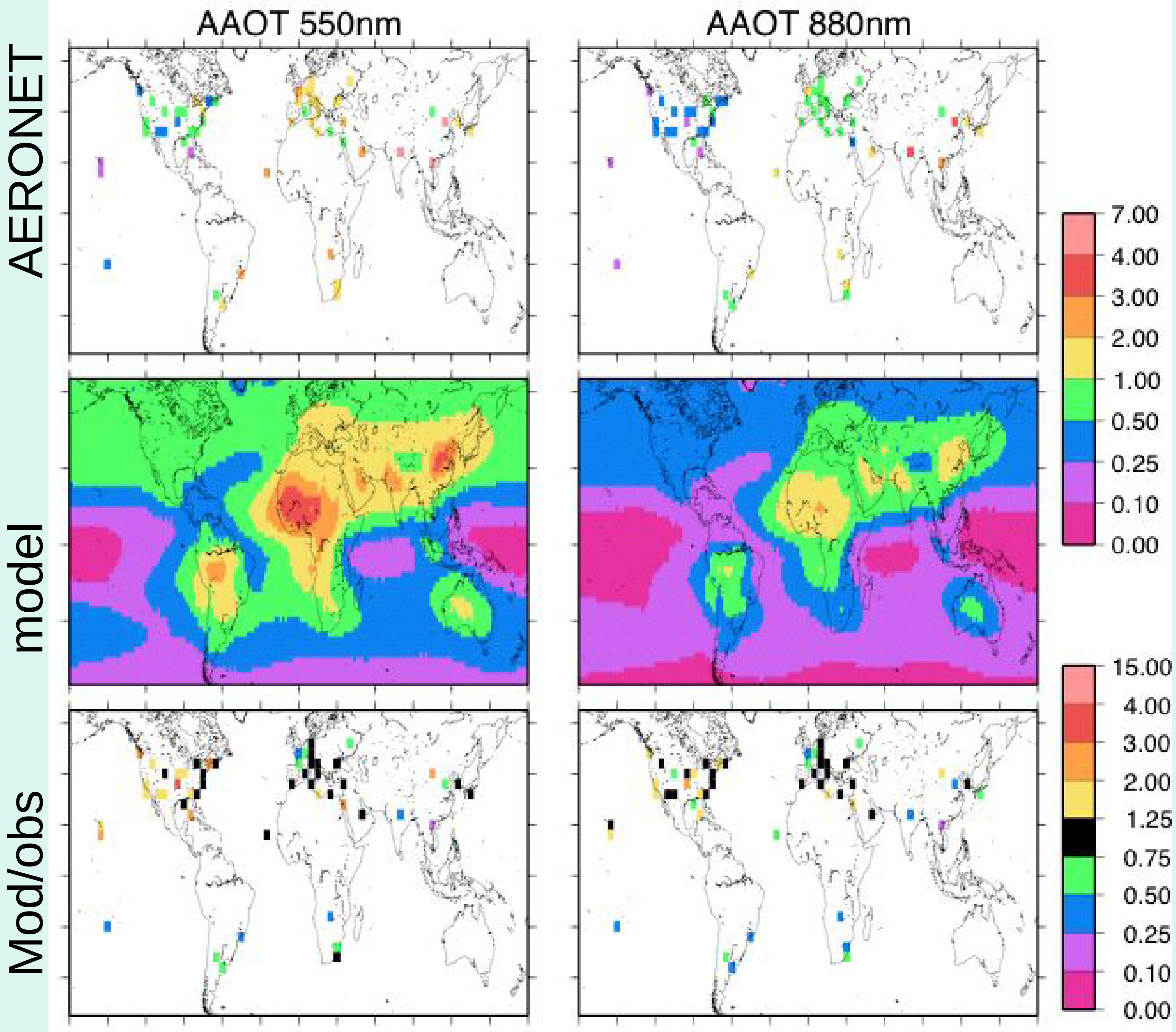
What can we learn from AAOT biases at other wavelengths?

If the bias is less at longer wavelengths then adding absorbing OM would help.

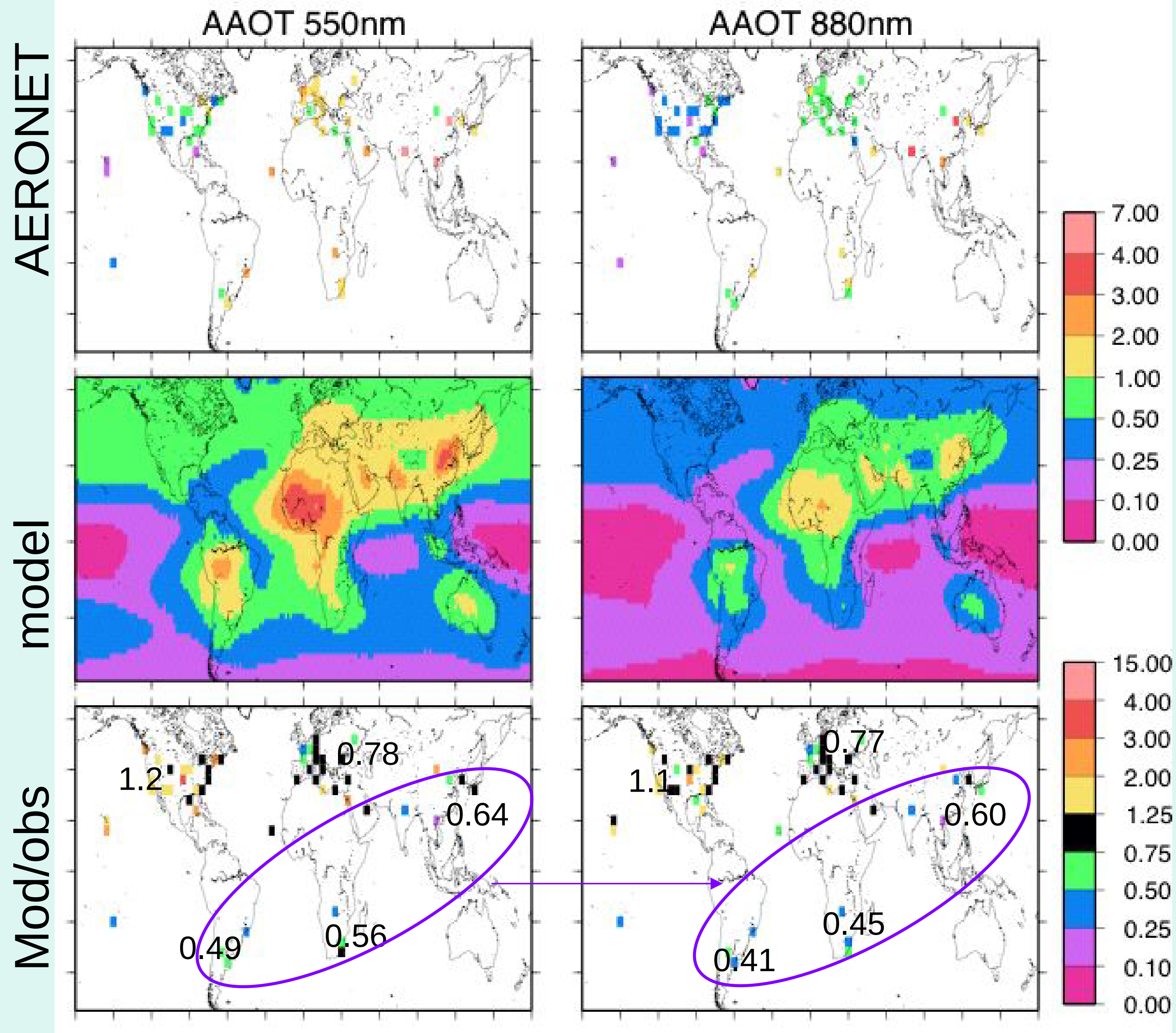
AAOT f(λ)



AAOTx100 550
BC 0.38
dust 0.15
OC 0.12
Shortwave
Wavelength dependence distinguishes BC and OC but not BC and dust
AAOTx100 880
BC 0.22
dust 0.085
OC 0.015
Longwave
÷1.7
÷2
÷8
0.00
0.10
0.25
0.50
1.00
2.00
3.00
4.00
7.00



Do model biases change with wavelength?



Model underestimate is larger at longer wavelengths. **This suggests that BC, rather than OC is lacking.**

Aerosol mixing effects on absorption

Aerosol mixture, coating of BC by sulfate, probably enhances absorption. This will be most important downwind of regions with large BC and sulfate emissions: SE Asia, Europe

It would be less helpful *within* these regions and in biomass burning regions.

And the model already over-estimates absorption in remote regions.

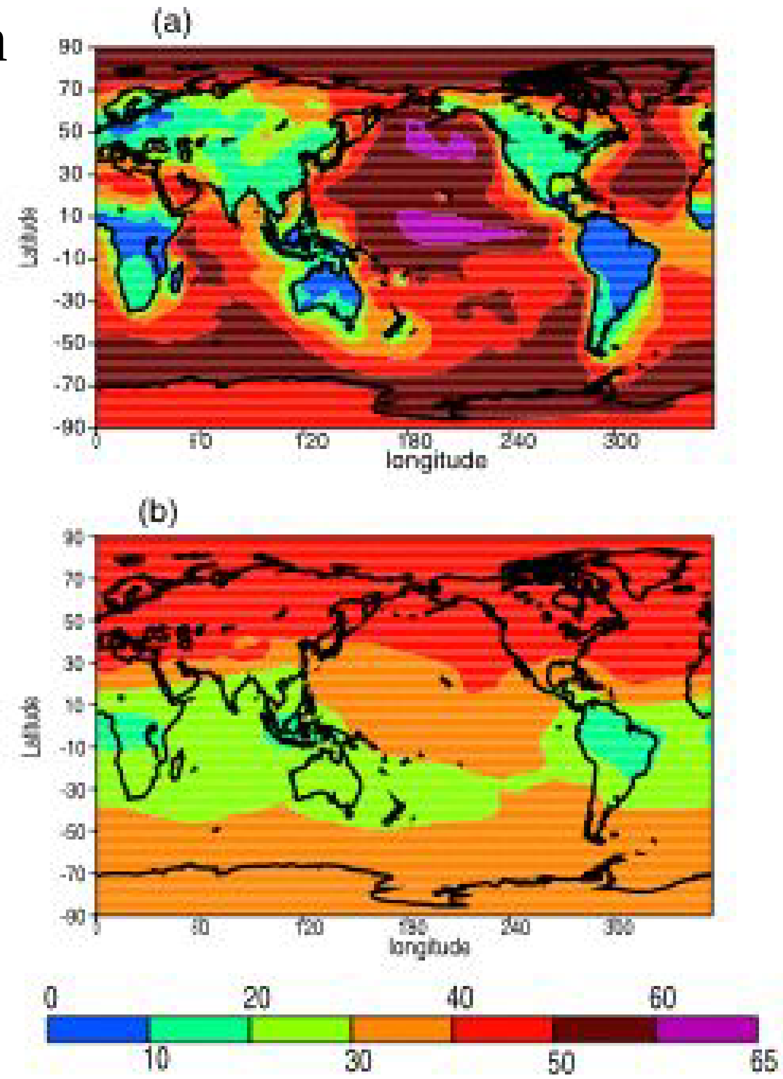


Figure 23. Annual mean mass fraction (%) of sulfate on carbonaceous aerosol (sulfate mass on OM/BC relative to sulfate mass on OM/BC plus OM/BC mass) (a) at the surface and (b) in the middle to upper troposphere (536–187 hPa).

Closing the gap: Some ideas…

1. Particle structure evolution with age:

   Wood-burn particles transform from:

   fresh, fluffy, more absorbing ⟹ **compact, less absorbing**

   Such aging effects on model optics might help explain AAOT biases (under-estimates in Europe, Asia, biomass burning regions).

2. Emission of large (e.g. super-micron) particles near source regions would help explain the model surface concentration biases near source regions.

Conclusions

1. There are broad regional patterns of BC, both surface concentrations and AERONET τ_{abs}:

Asia > Europe, biomass burning regions > North America > remote NH > remote SH

These patterns appear in spite of local variabilities due to urban locations, measurement uncertainties

2. Newest emission estimates hopefully improve our links from sources to climate effects; however these estimates do not greatly change our model biases relative to observations.

3. Our model underestimates BC in SE Asia, biomass burning regions, parts of Europe.

4. The bias is slightly greater at longer wavelengths, suggesting that the deficiency is in black, not organic carbon.

Conclusions, cont’d

5. Adding aerosol size information is not likely to help:

Smaller particles in North America (where diesel sources dominate) would increase absorption there, but absorption is already too large.

Larger particles in biomass burning and residential source regions would decrease absorption, where absorption is already too small.

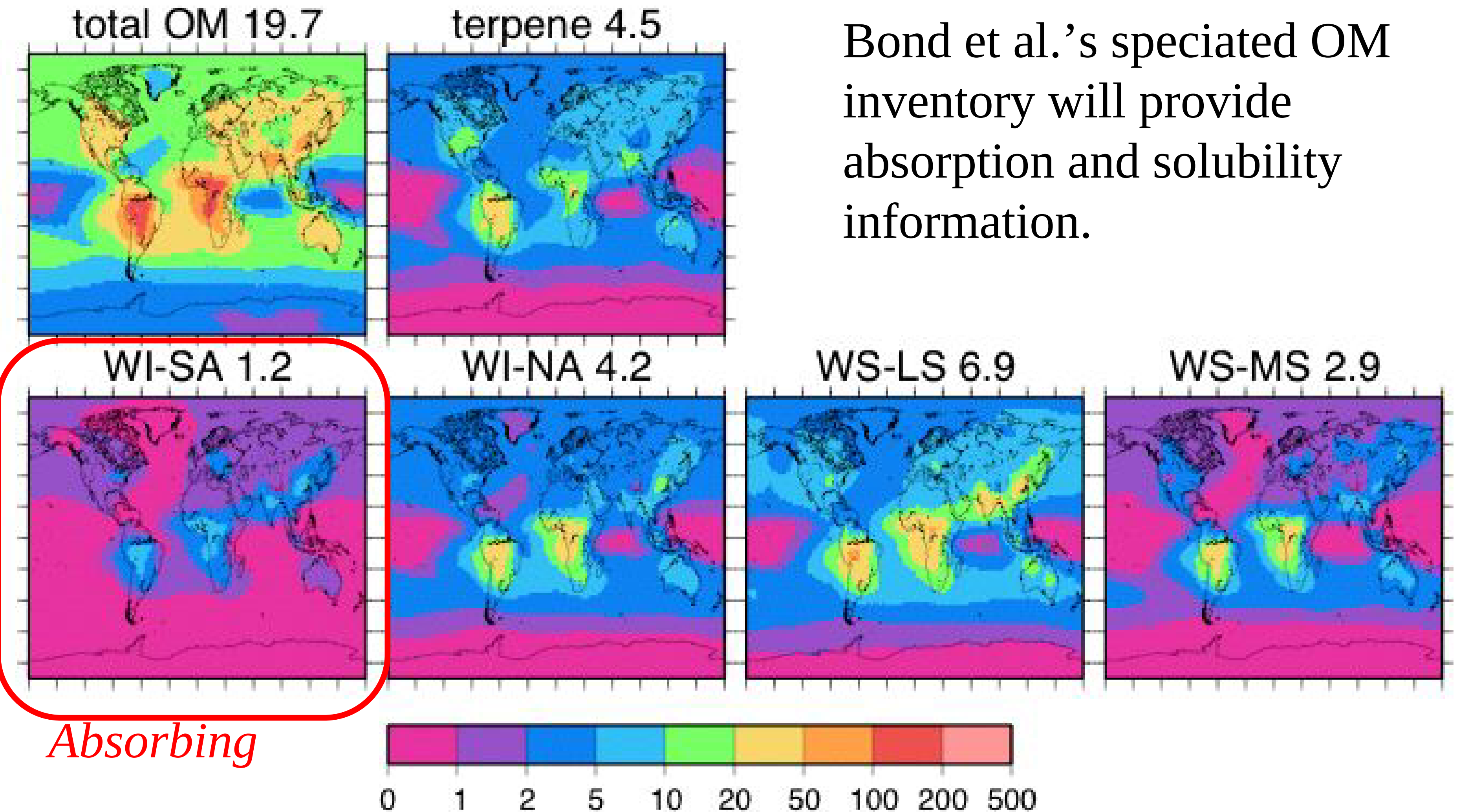
6. Adding aerosol mixing would increase absorption downwind of SE Asia, Europe sources but would not help much close to source regions where biases are largest.

7. **Perhaps wood burning and (Asian) coal burning are more absorbing than our current optical model assumes: particle structural or density effects?**

8. Emission estimates of coarse particles would improve surface concentration bias near source regions.

9. OMI: increased AAOT spatial coverage

Absorbing OM?



Bond et al.'s speciated OM inventory will provide absorption and solubility information.

However this would mostly boost short-wave absorption(?)

Schuster column BC