Efficient In-cloud Removal of Aerosols by Deep Convection

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Major Scientific Points

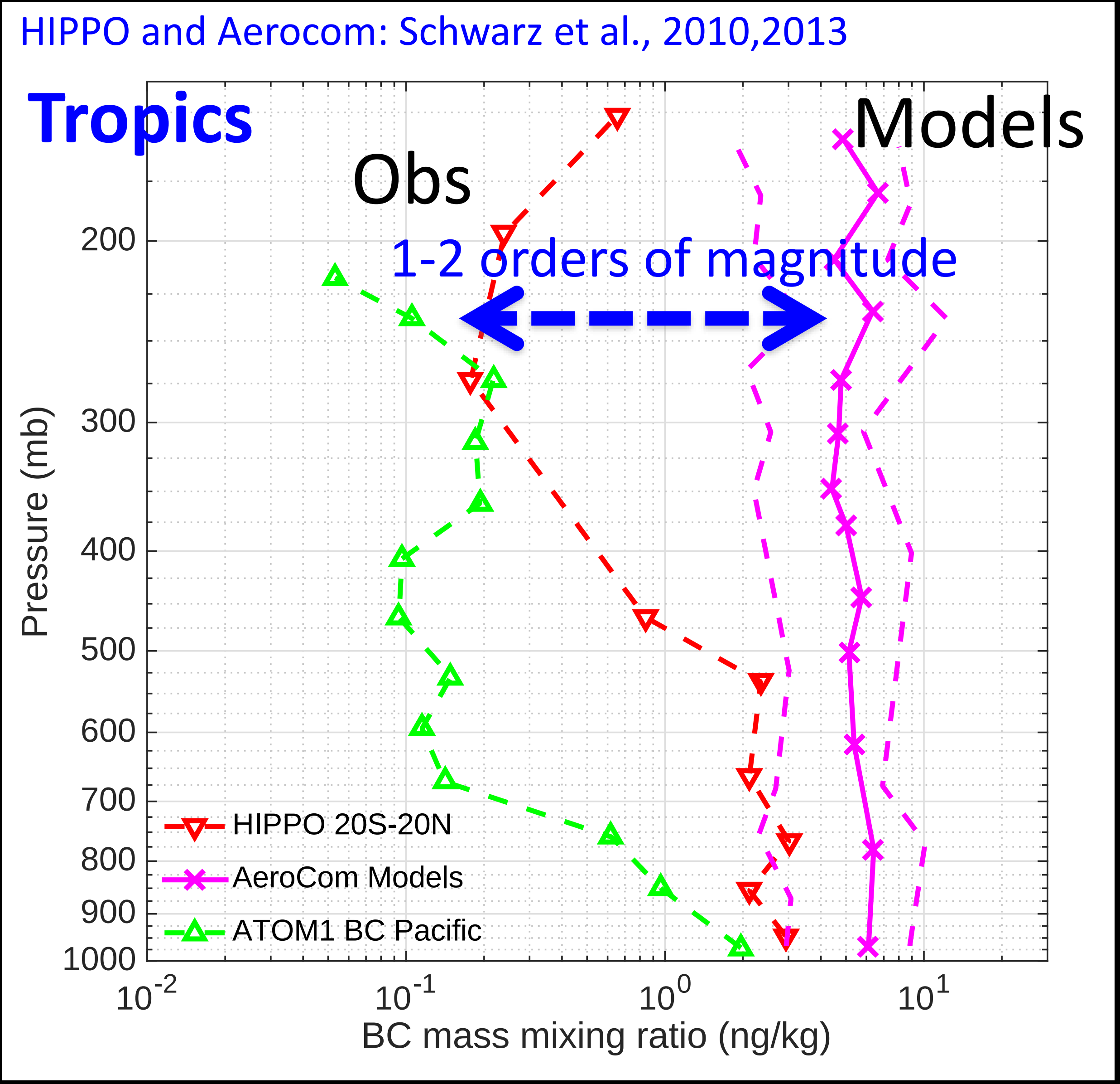
1) ATom data suggests that global aerosol mixing ratio drops by 1-4 orders of magnitudes from surface to the upper troposphere (salt, black carbon, dust).

2) Deep convections can efficiently remove aerosols in-cloud through secondary activation from the entrained air.

3) In the middle and upper troposphere, secondary formed particles including sulfate and organics dominate

BC is efficiently depleted while lifted, it is well-known that global models generally overestimate BC in UT

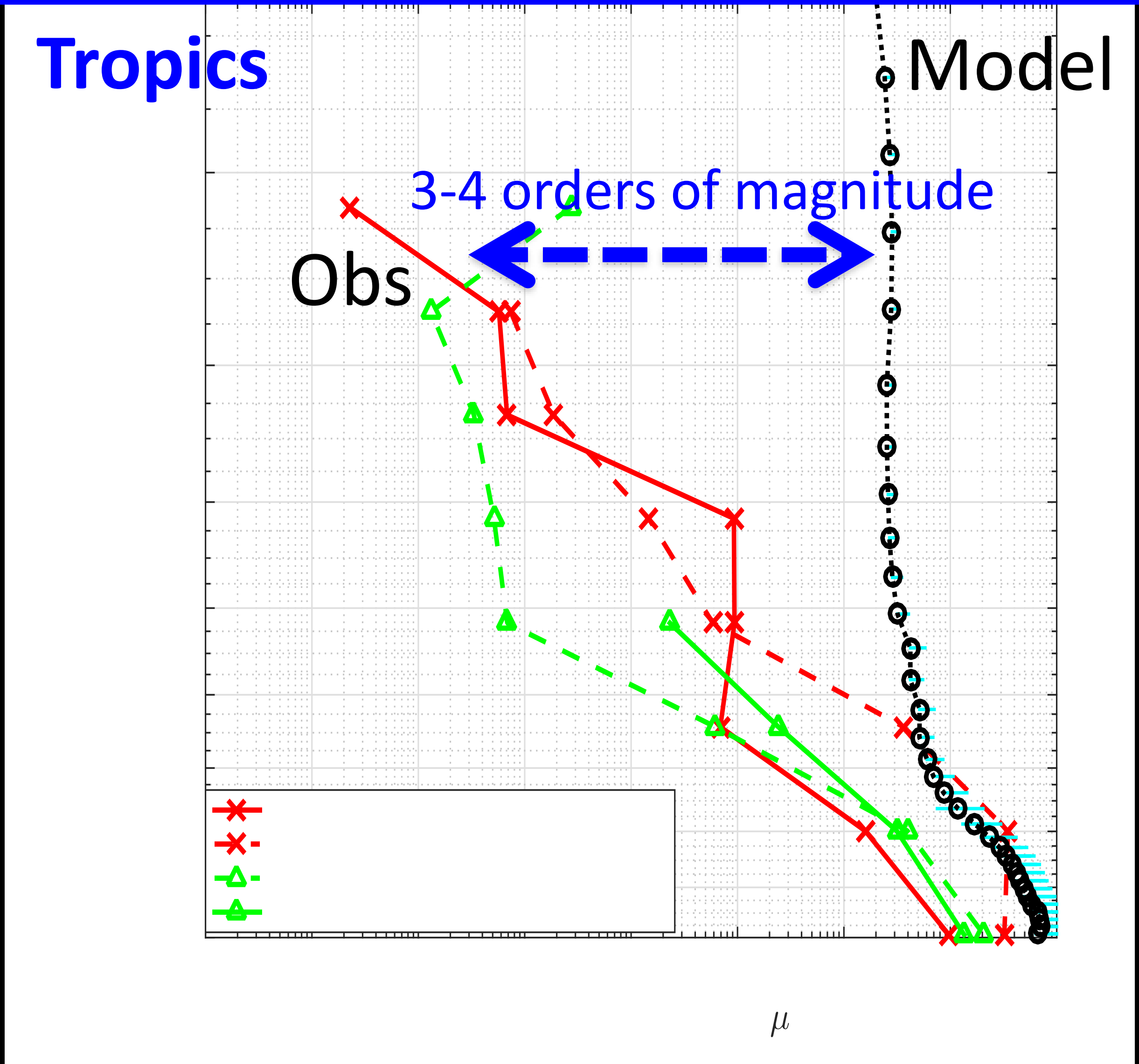
BC



Sea salt is efficiently depleted, and global model (CESM) overestimates by 3-4 orders of magnitude

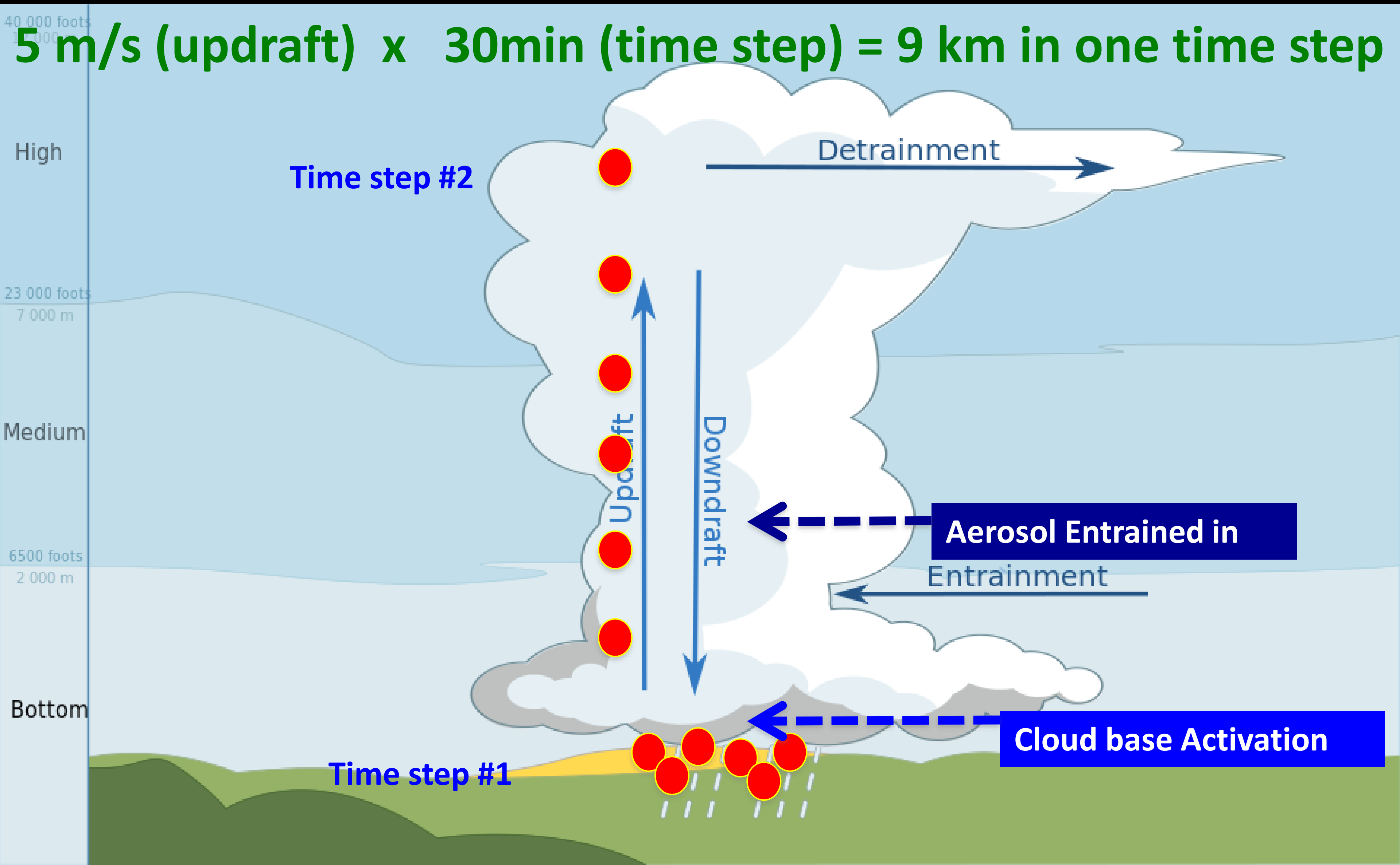
Atom Salt data: PALMS, Murphy et al., 2018, ACPD

SALT



Default Convective Transport in CESM: “Equilibrium State”, no sub-grid scale loss at all!

5 m/s (updraft) x 30min (time step) = 9 km in one time step



Convective Transport in GEOS-Chem (Wang et al., 2014): exponential decay (sub-grid)

$$f = 1 - e^{-\alpha k \Delta z} \quad (1)$$

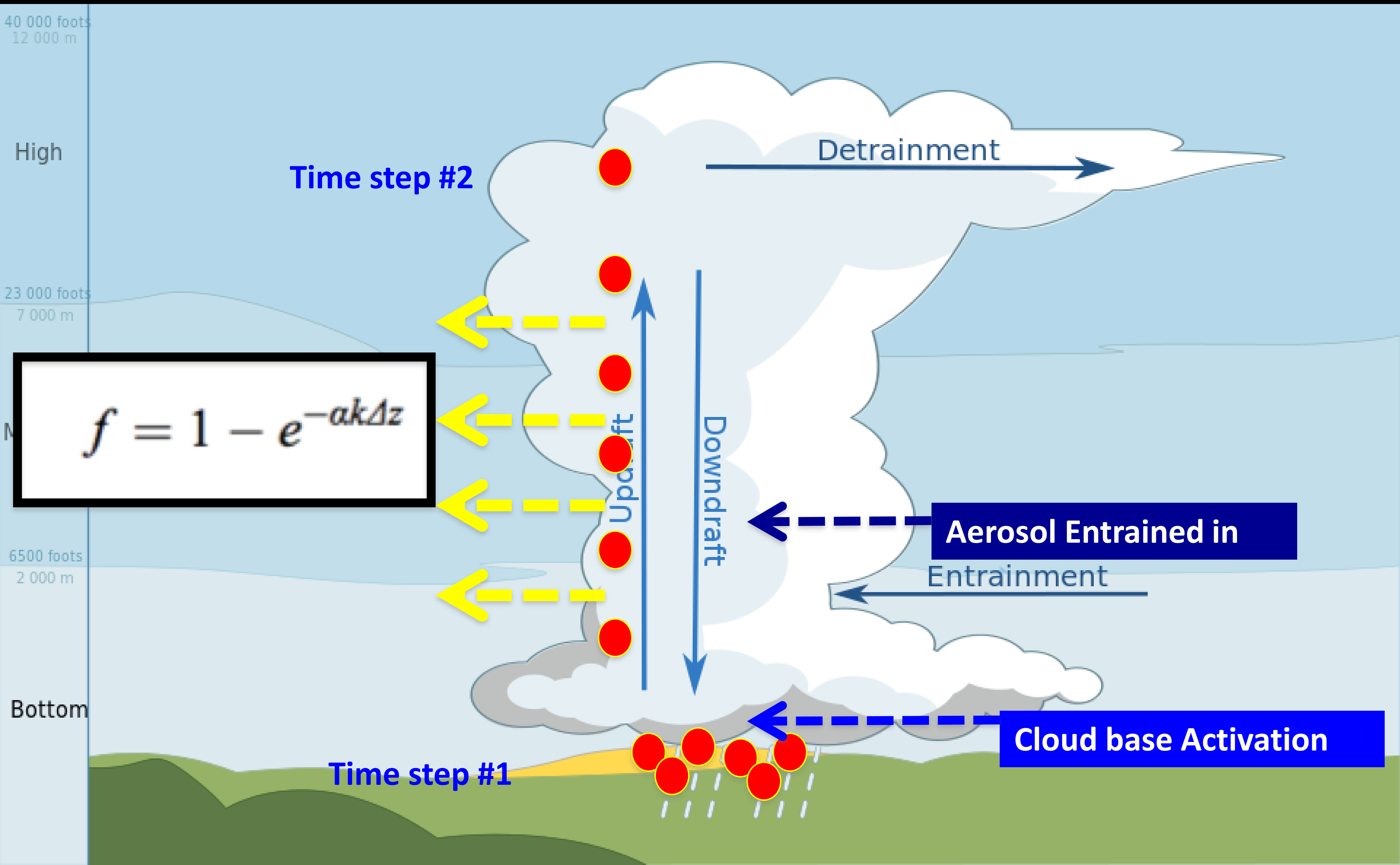
Wang et al., 2014, ACP

where k is a coefficient for conversion of cloud water to precipitation with values of $5 \mathrm{x} 10^{-4}\,\mathrm{m}^{-1}$ over land and $10^{-3}\,\mathrm{m}^{-1}$ over ocean, and α is the fraction of aerosol mass incorporated

Constant k is 1 for hydroscopic aerosol: e.g. salt
Constant k is 0 for hydrophobic aerosol: e.g. dust

Constant k is 0.5 for hydrophobic BC (Wang et al., 2014)

Convective Transport in GEOS-Chem (Wang et al., 2014): exponential decay (sub-grid)



CESM/CARMA: Sectional Aerosol Model

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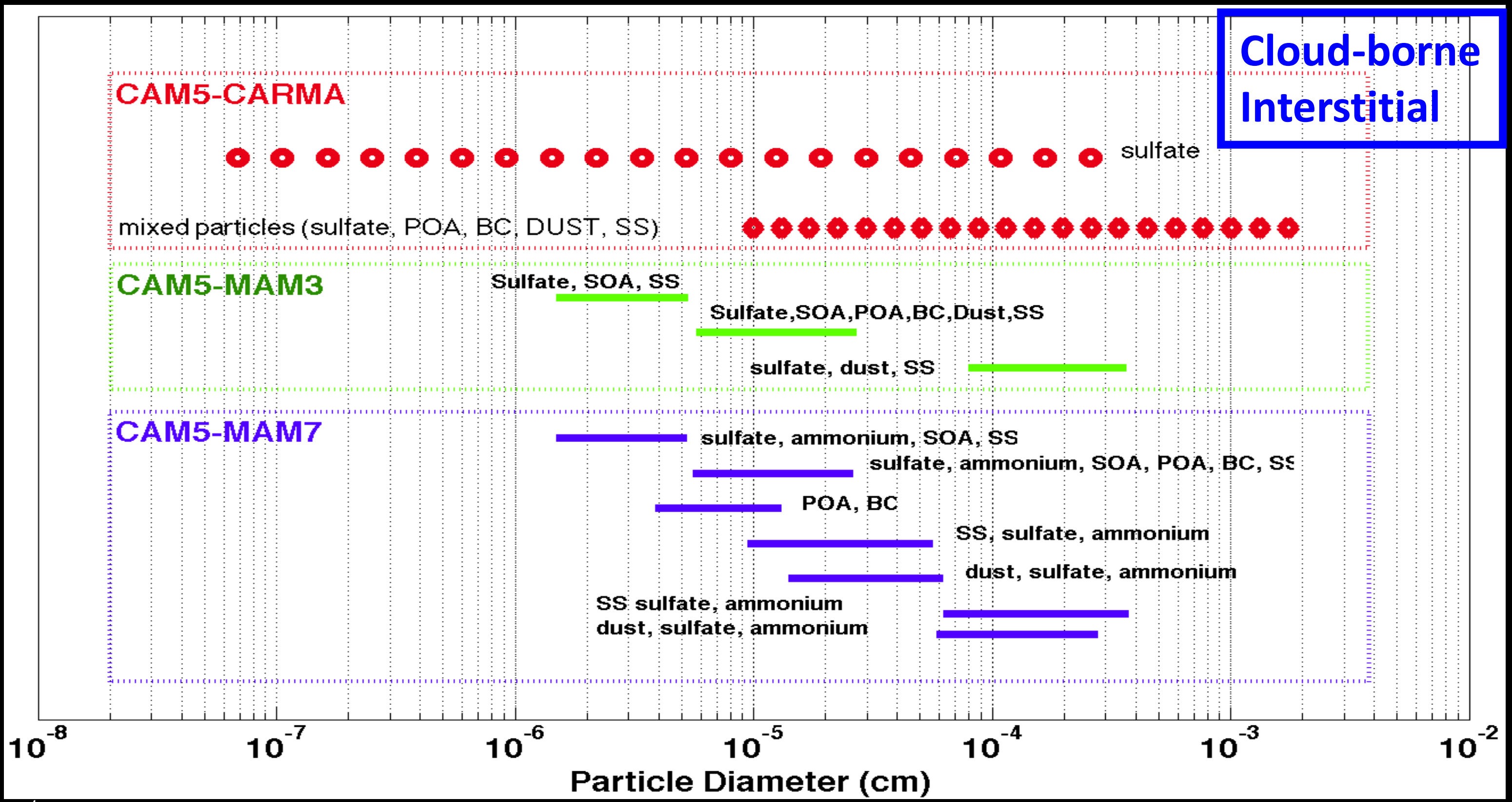
University of Colorado Boulder

Pengfei Yu et al., 2015, *JAMES*
Bardeen et al., 2013, *JGR*
Brian Toon et al., 1988, *JGR*

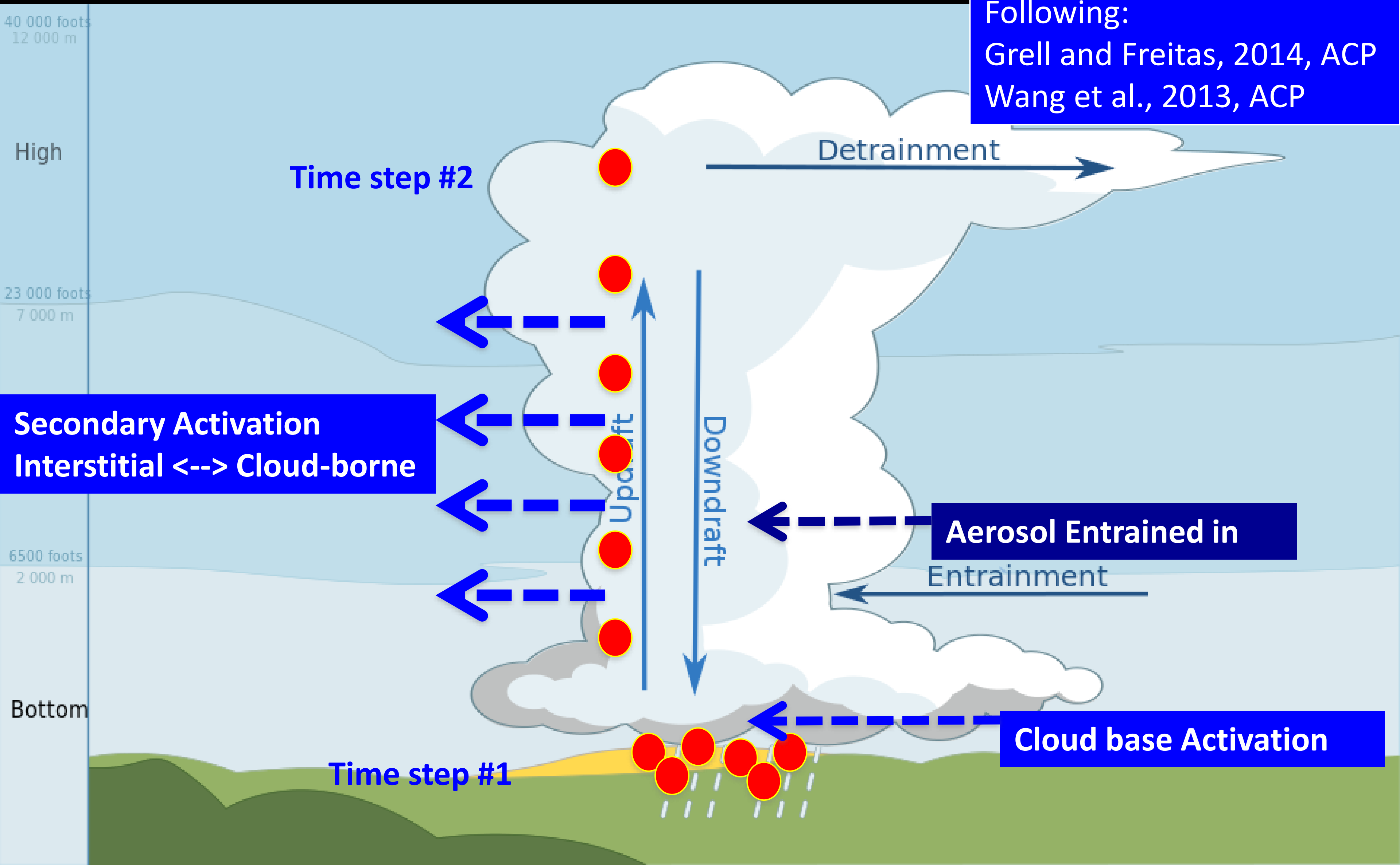


Some features:

Sulfur Chemistry (WACCM, Mike Mills), Heterogeneous Chemistry
SOA Chemistry (4-Bin VBS), Coupled with MG Water Cloud, Coupled with RRTMG
Including Marine organics, biological organics

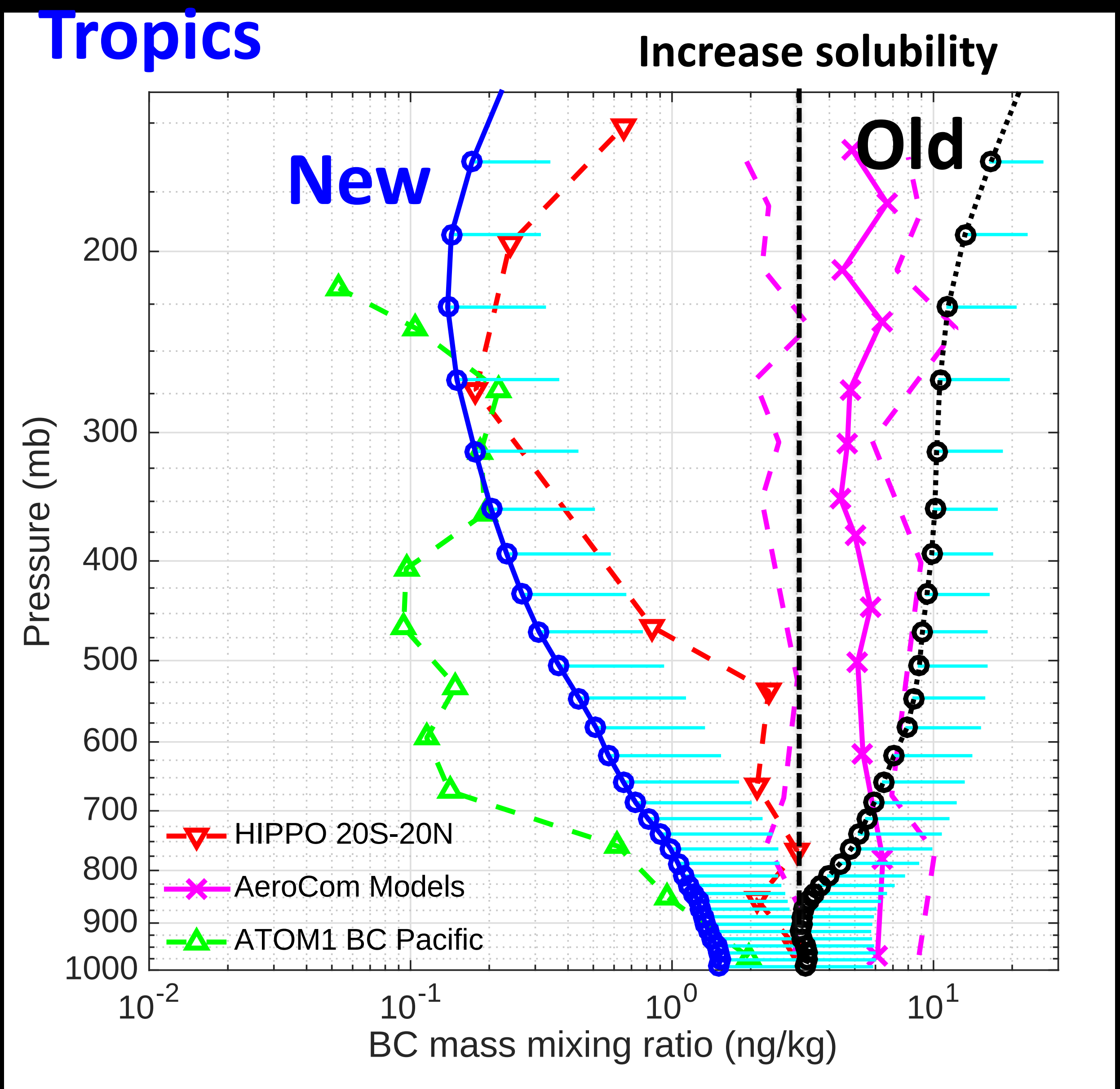
Modified Convective Transport in CESM-CARMA: Secondary activation

Following:
Grell and Freitas, 2014, ACP
Wang et al., 2013, ACP



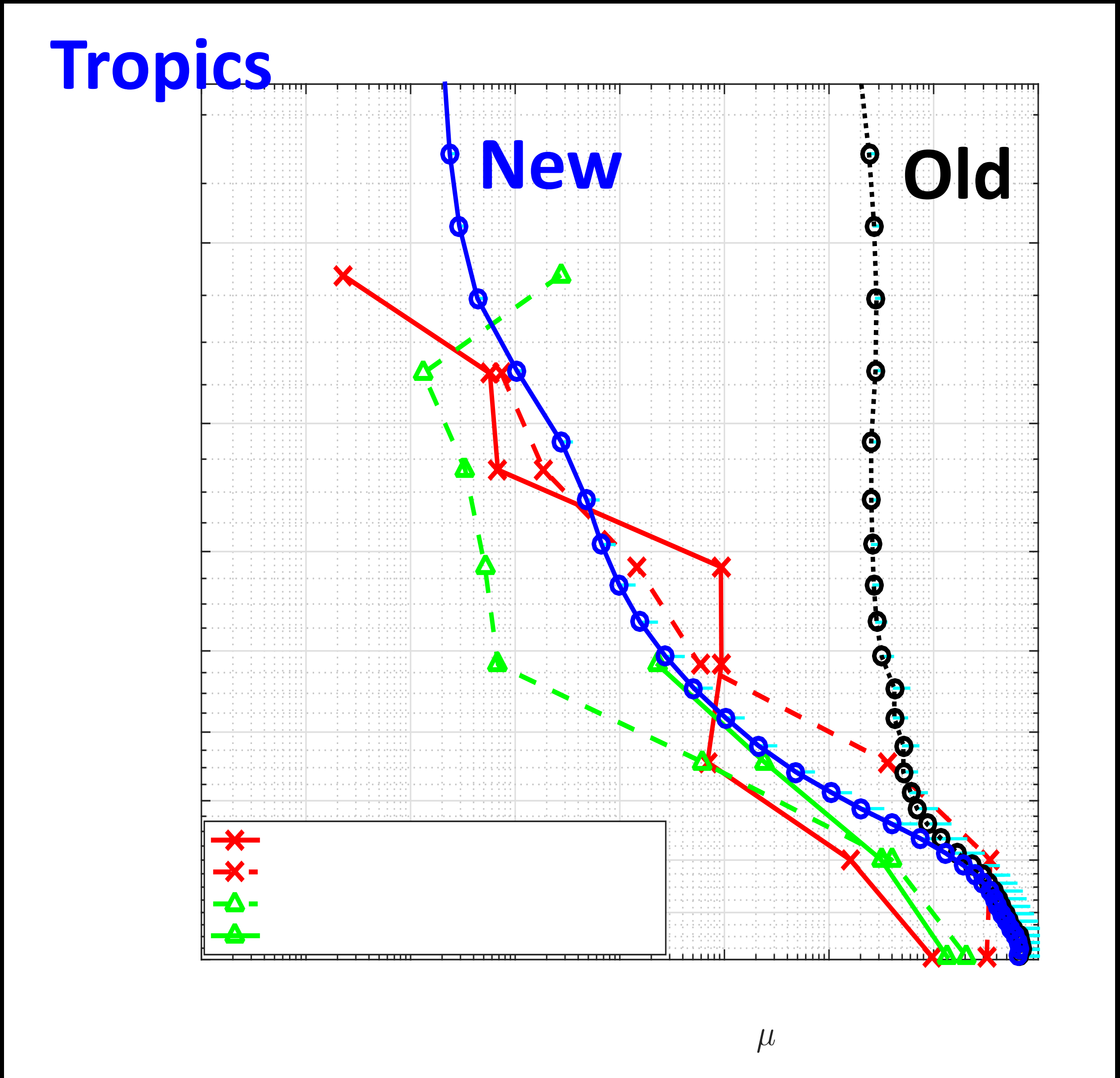
We applied the model fix in CESM-CARMA

BC

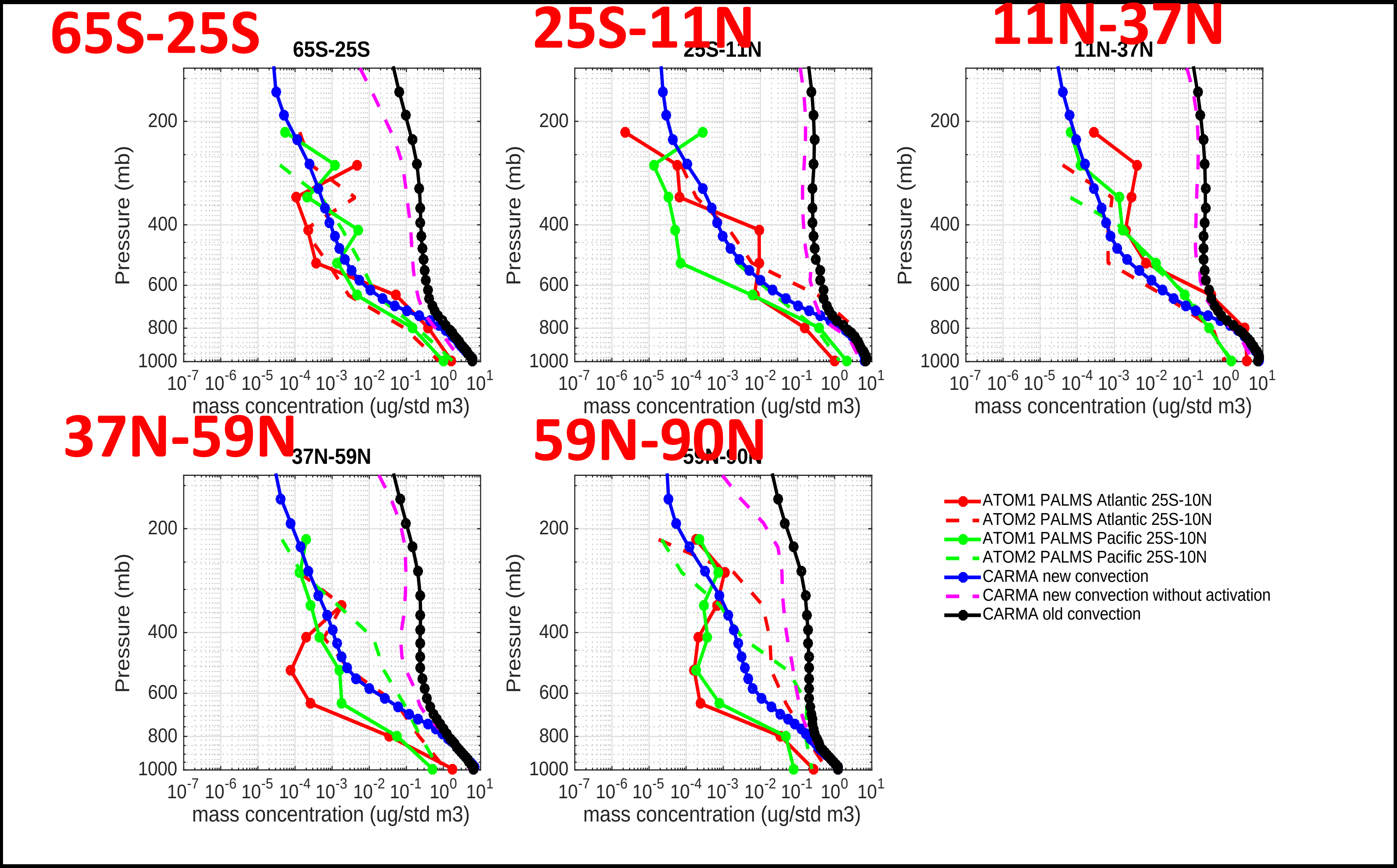


We applied model fix in CESM-CARMA

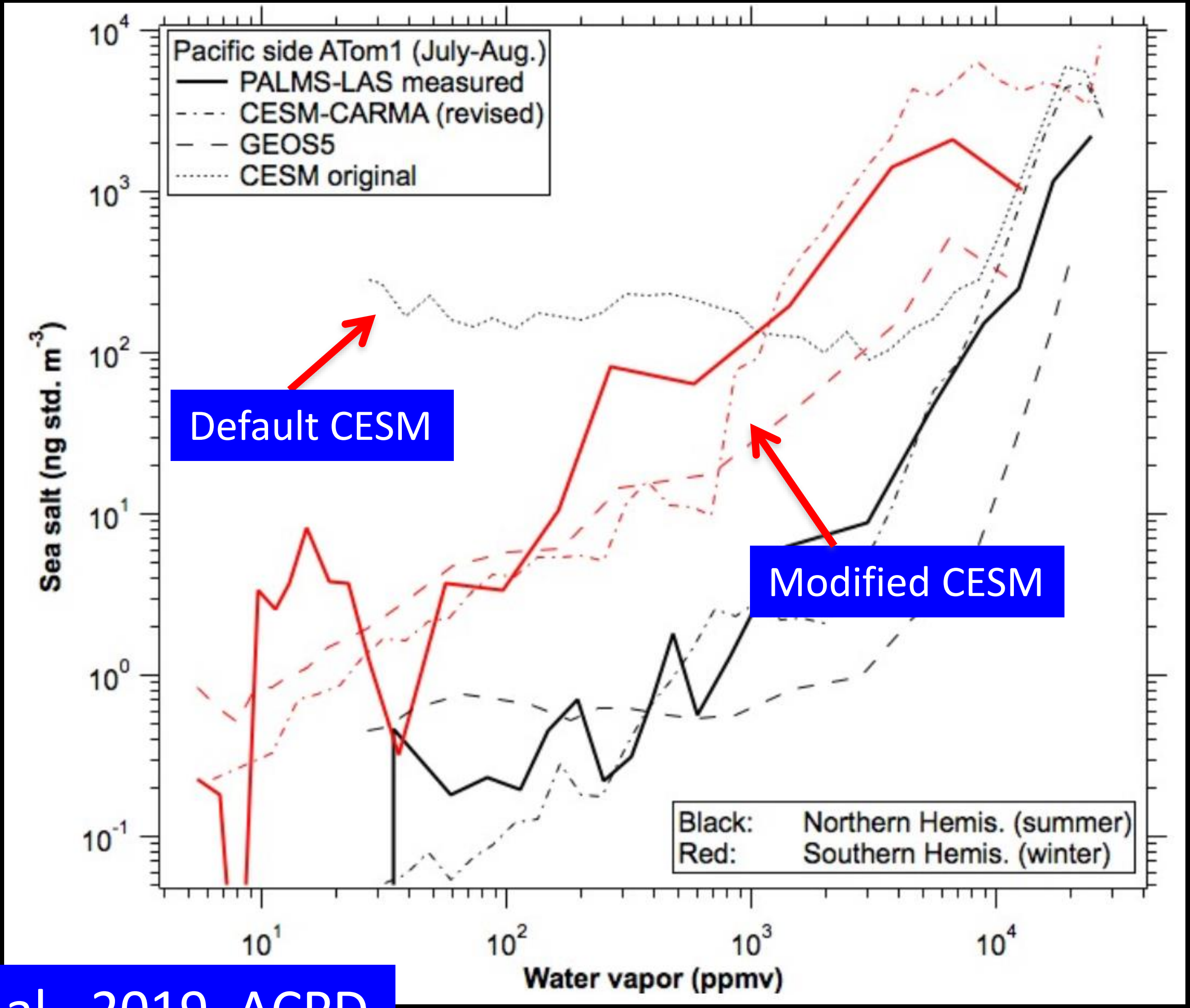
SALT



Modified scheme improves Salt in all latitude bands



Modified convective transport scheme improves Salt vs. H_2O relation



Murphy et al., 2019, ACPD

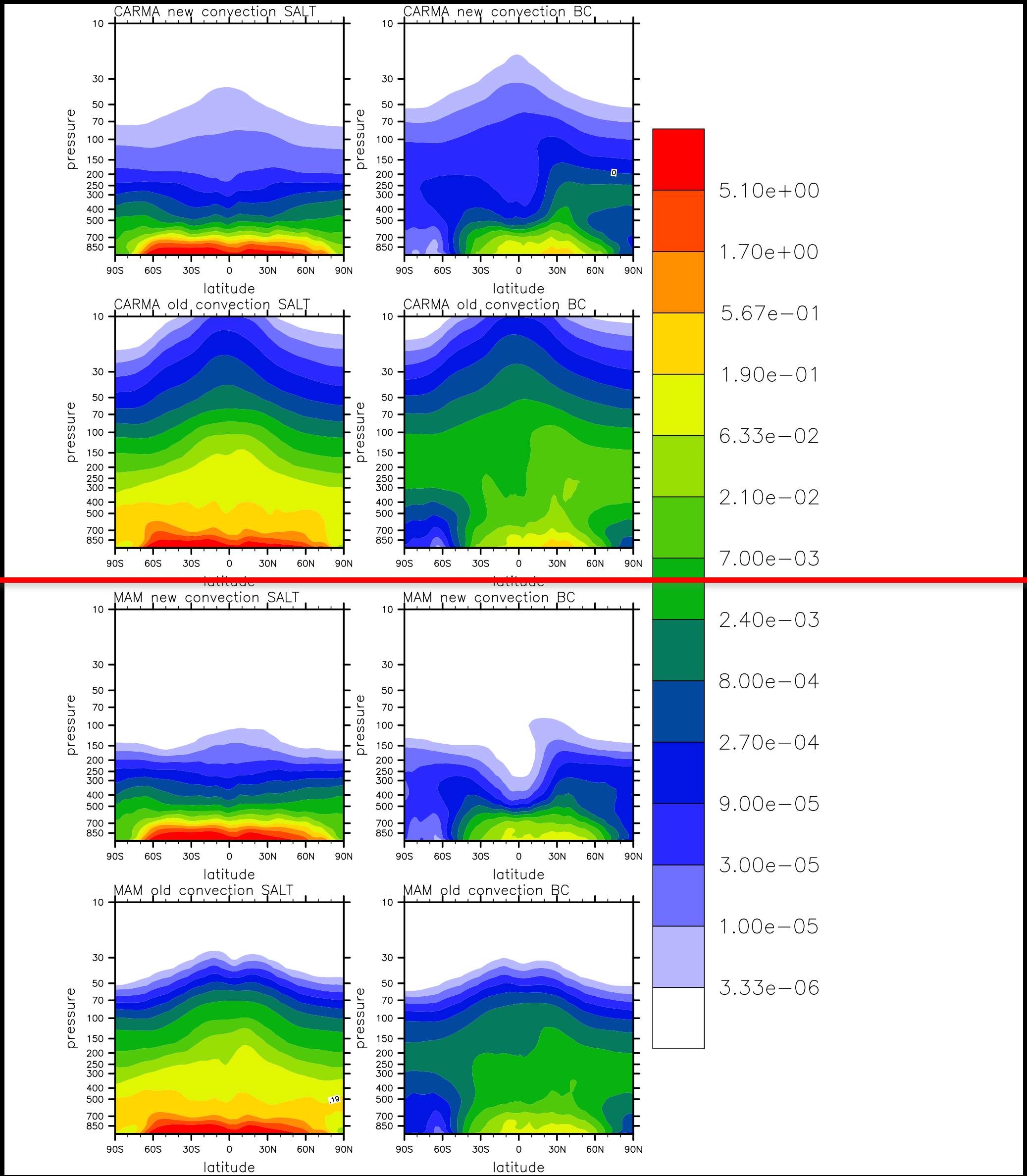
Modified Convective Transport scheme improves both CARMA and MAM

New

Old

New

Old



CARMA

MAM

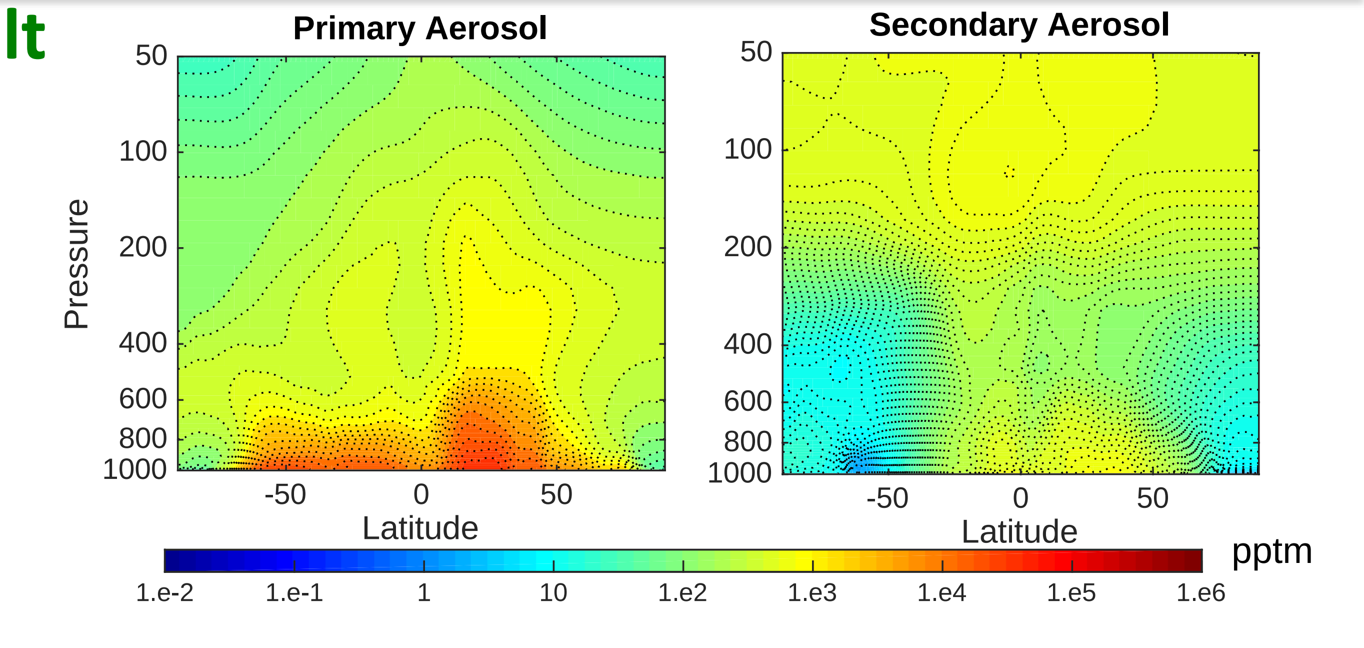
Chris Maloney has applied the fix to MAM

The modified convective transport scheme affects primary aerosol budget in the middle-upper troposphere

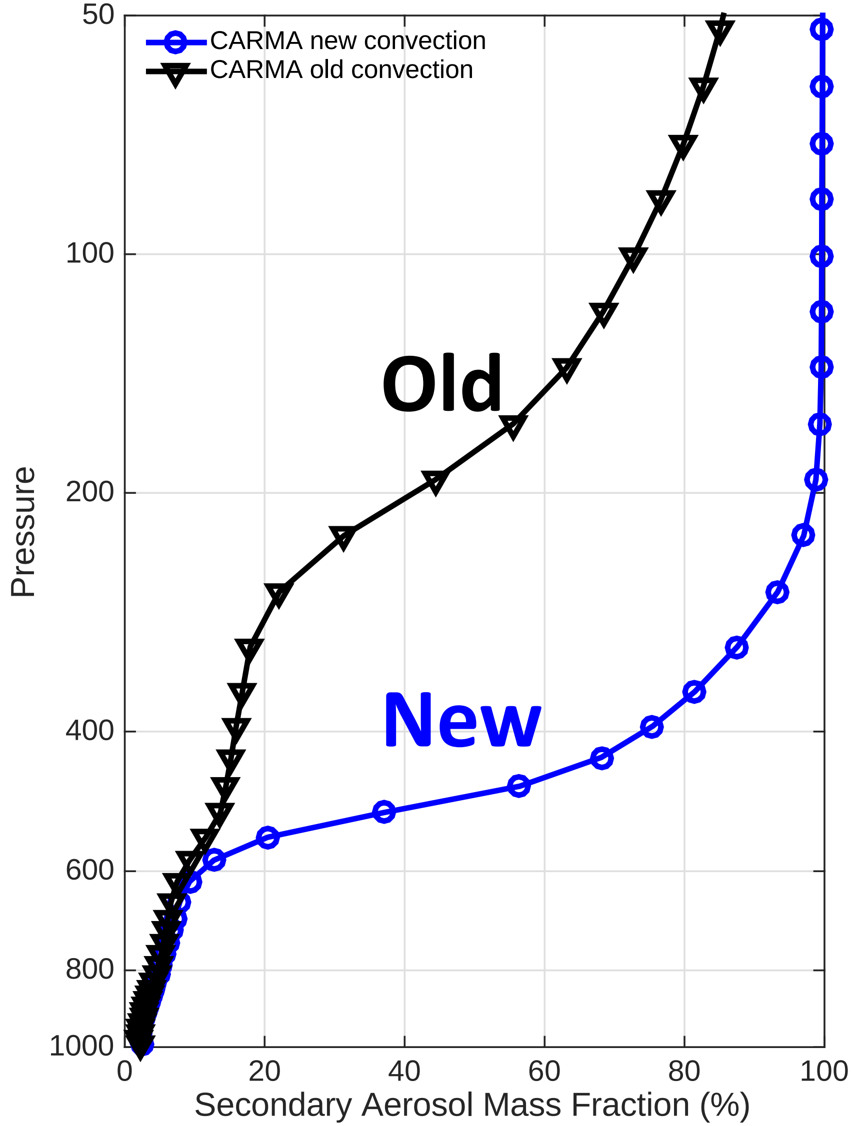
Primary Aerosol:

POA, BC, Dust, Salt

Old Convective



In the modified scheme: secondary particles (sulfate and organics) dominates in the middle-upper troposphere



Secondary Aerosol Mass Fraction (%)

Mathematically…

Convective transport: ensembles of convective plume

Sub-grid, Old:

$$-\frac{\partial(M_u q_{uI})}{\partial p} = E_u q_e - D_u q_{uI}$$

Sub-grid, New:

$$-\frac{\partial(M_u q_{uI})}{\partial p} = E_u q_e - D_u q_{uI} + Act * q_{uI}$$

Act denotes Activation Efficiency

$$Act = ract * M_u$$

M_u denotes updraft

Activation Efficiency is updraft dependent, should be also dependent on particle size, composition as well… need more measurements

Seems a consistent story on missing secondary activation in CESM

In Tropical UT

- Sea Salt… for sure (Murphy et al., Yu et al.) ~10^4
- BC are subject to convective removal (Yu et al.) ~10-100
- Dust is removed efficiently (Froyd et al.) ~10^2-10^3
- POA is removed (Alma, Pedro, Jose, Schill)

- Effect on SOA and sulfate: within a factor of 2

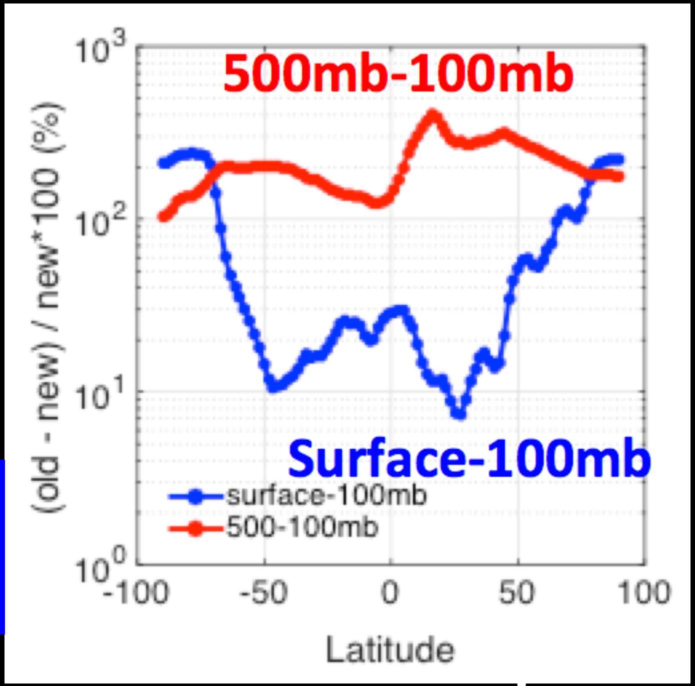
Not yet solved

- Convective Removal efficiency (secondary activation)?
  - Particle size dependent?
  - Particle composition dependent?
  - Time scale of aging of hydrophobic species? Days? Hours?

Scientific Implications

- Aerosol budget in Polar regions, cloud, radiative effects?
- Primary aerosol lifetime?
  - How far they can transport?

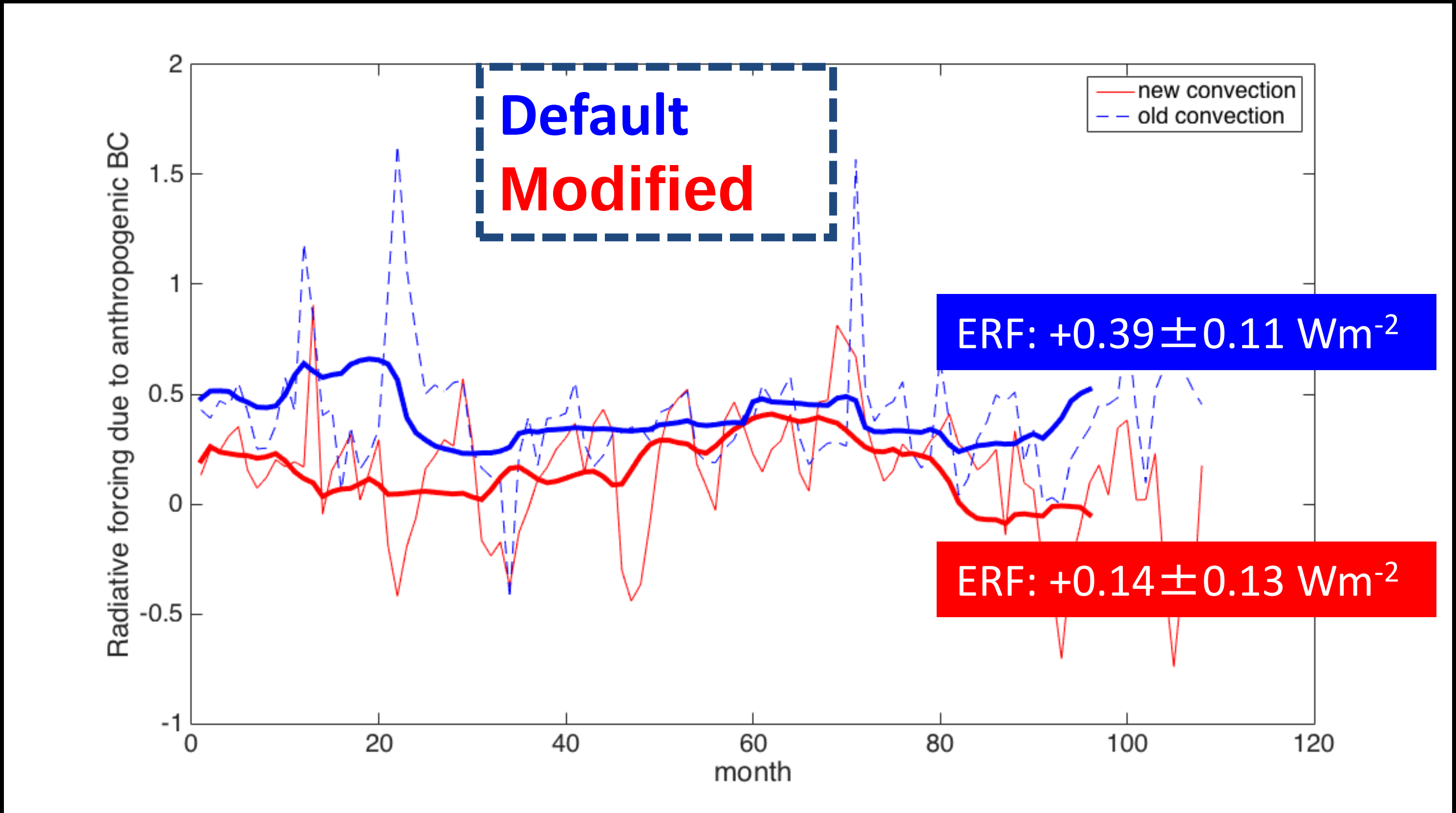
Percentage overestimation
(old-new)/new*100



- Dust budget in TTL, Asian summer monsoon: ice cloud formation/budget, radiative effects?

Scientific Implications

Global Black carbon's radiative forcing overestimates by over a factor of two.



Summary

1) ATom data suggests that global aerosol concentration drops by 1-4 orders of magnitudes from surface to the upper troposphere.
2) Deep convections can efficiently remove aerosols in-cloud through secondary activation from the entrained air.
3) In the middle and upper troposphere, secondary formed particles including sulfate and organics dominate
4) We suggest global models might not need to include insoluble species for BC, dust and organics.
5) More in-situ measurements are needed to quantify the convective transport scheme.
6) Incorrect convective transport scheme overestimates BC's radiative forcing and aerosol budget over polar regions.

The modified convective transport scheme also improves total aerosol--->within a factor 2-3