Simulations of New Particle Formation in the UTLS using WACCM/CARMA

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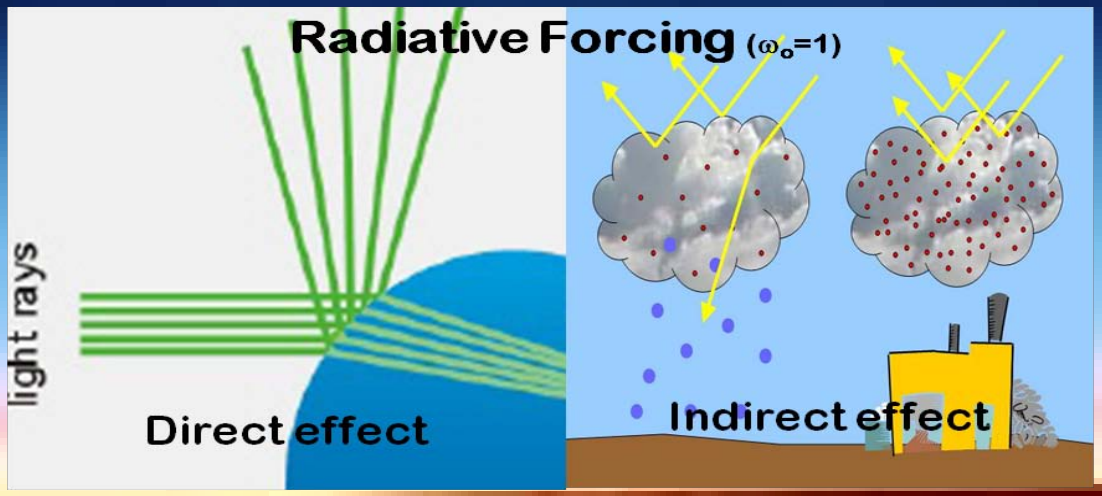
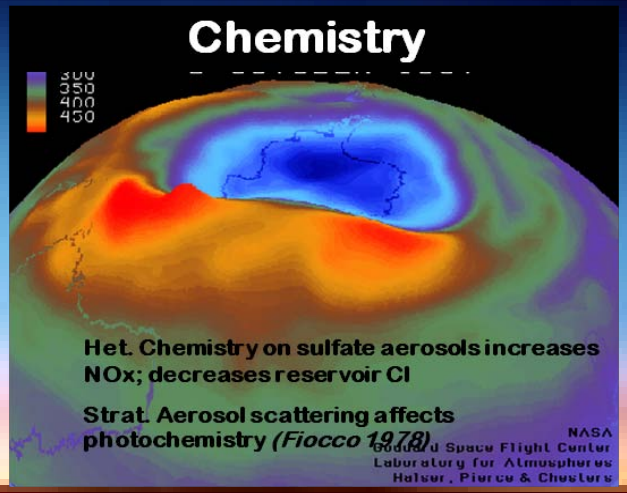
NASA Space Shuttle photo

Why do we care about UTLS aerosols?





Science Questions

- Can binary homogeneous nucleation scheme(s) adequately represent particles in the UTLS?
- Is ion-induced nucleation important in the UTLS? (e.g. is there a cosmic ray-sun-climate link?)

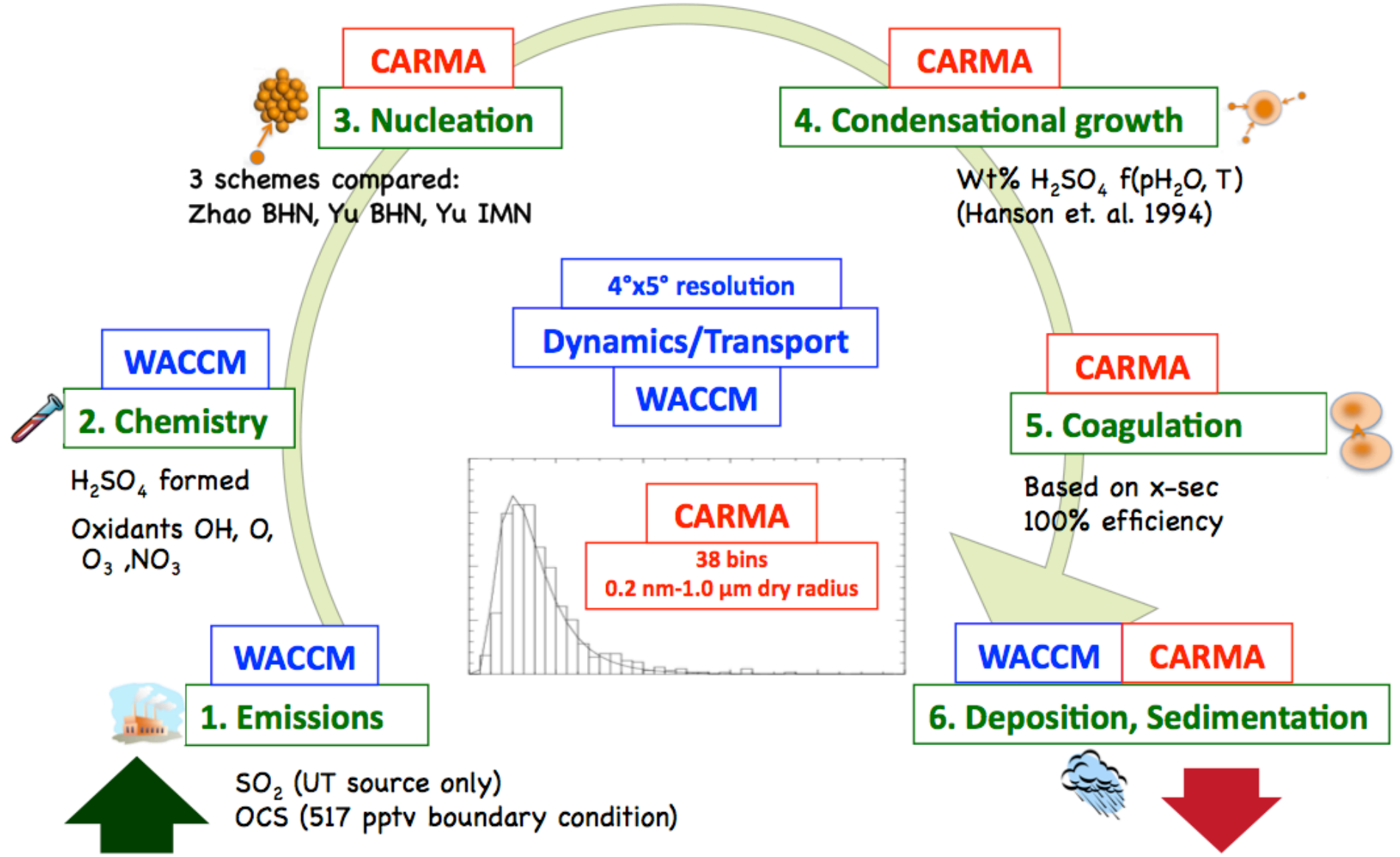
Objective: Model the impacts of BHN schemes and the role of ions on UTLS aerosol properties

Three nucleation schemes simulated:

"Zhao BHN" (Zhao and Turco 1995)	Numerical approximation of nucleation rates based on classical nucleation theory
"Yu BHN" (Yu 2008)	Lookup Tables based on quasi-unary nucleation of H_2SO_4 in equilibrium w/H_2O vapor
"Yu IMN" (Yu 2008)	Lookup Tables based on Yu BHN plus stability from ion-pairs (10 ion-pairs cm^{-3} s^{-1})

BHN = Binary Homogeneous Nucleation of Sulfuric Acid and Water
IMN = Ion-Mediated Nucleation of Sulfuric Acid and Water

Approach – WACCM/CARMA coupled model



2 year simulations; 2nd year averaged

Modified MOZART Chemistry

56 chemical species + electrons + 7 sulfur species: OCS, SO2, S, SO, SO3, HOSO2, H2SO4

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*-------------------------------------
*   photo-ionization
*-------------------------------------
 [jh2so4]    H2SO4 + hv -> SO3 + H2O
 [jso2]      SO2 + hv -> SO + O
 [jso3]      SO3 + hv -> SO2 + O
 [jocs]      OCS + hv -> S + CO
 [jso]       SO + hv -> S + O

* ------------------------------------------------------------
* Sulfur Reactions
* ------------------------------------------------------------
 OCS + O -> SO + CO                                    ; 2.10E-11, -2200.0
 OCS + OH -> SO2 + C + H                               ; 1.10E-13, -1200.0
 S + OH -> SO + H                                      ; 6.60E-11
 S + O2 -> SO + O                                      ; 2.30E-12, 0.0
 S + O3 -> SO + O2                                     ; 1.20E-11
 SO + OH -> SO2 + H                                    ; 8.60E-11
 SO + O2 -> SO2 + O                                    ; 2.60E-13, -2400.0
 SO + O3 -> SO2 + O2                                   ; 3.60E-12, -1000.0
 SO + NO2 -> SO2 + NO                                  ; 1.40E-11, 0.0
 SO + CLO -> SO2 + CL                                  ; 2.80E-11
 SO + BRO -> SO2 + BR                                  ; 5.70E-11
 SO + OCLO -> SO2 + CLO                                ; 1.90E-12
 SO2 + OH + M -> HSO3 + M                              ; 3.0E-31,3.3, 1.50E-12,0.0, 0.6
 HSO3 + O2 -> SO3 + HO2                                ; 1.30E-12, -330.0
 [usr13] SO3 + H2O -> H2SO4
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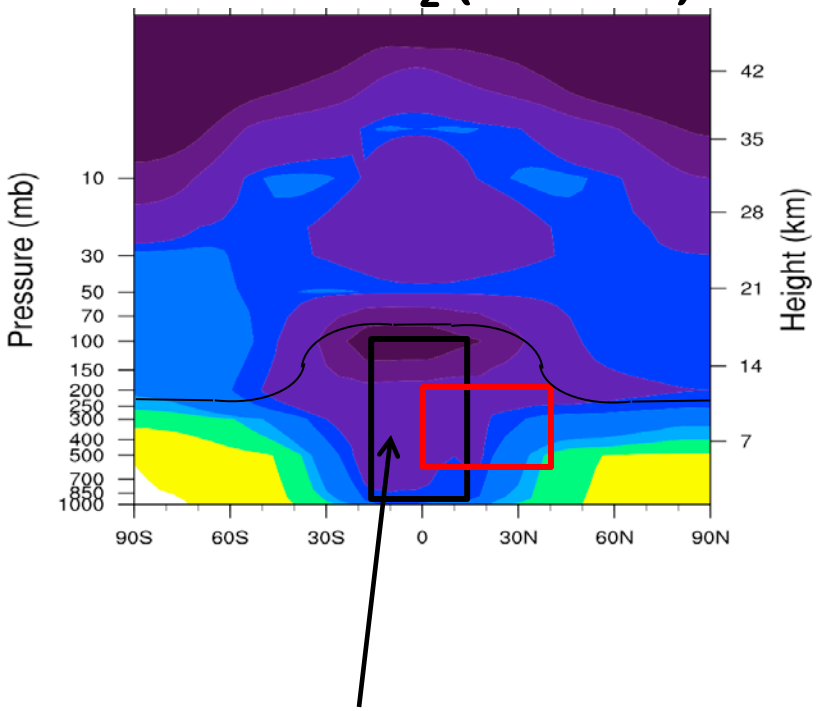
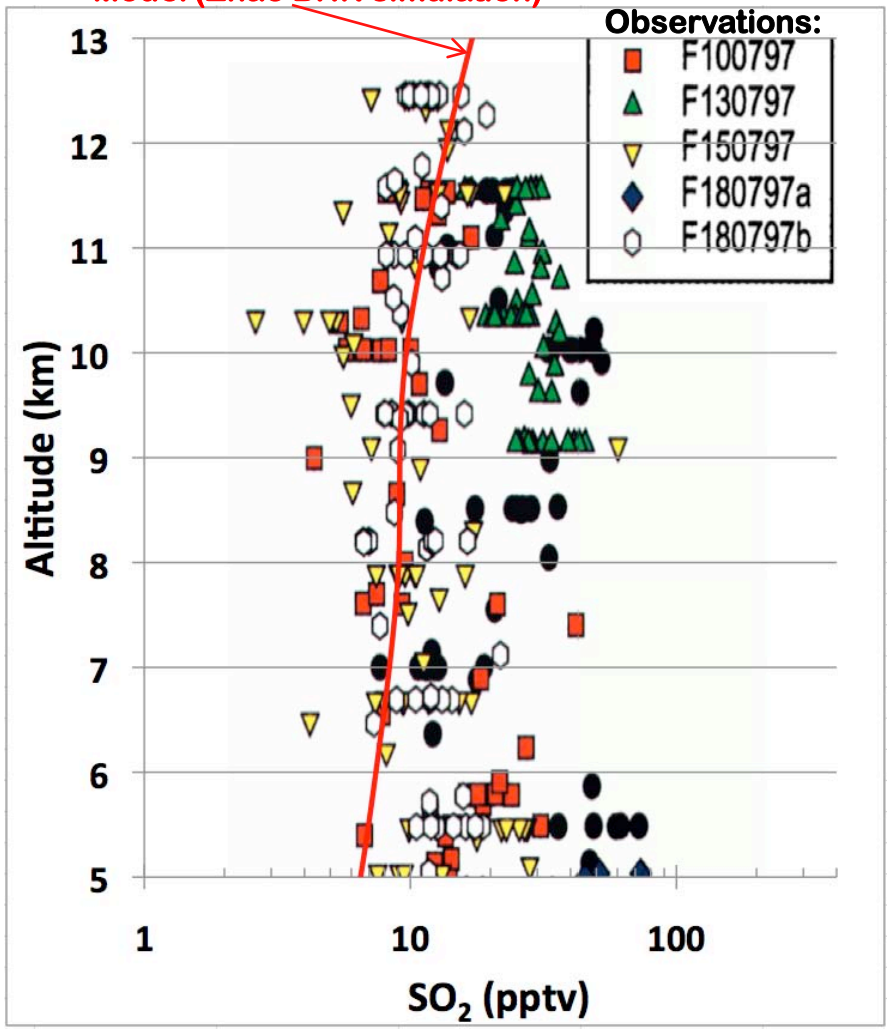
Modeled SO_2 compares well to observations

Modeled SO_2 (mol/mol)



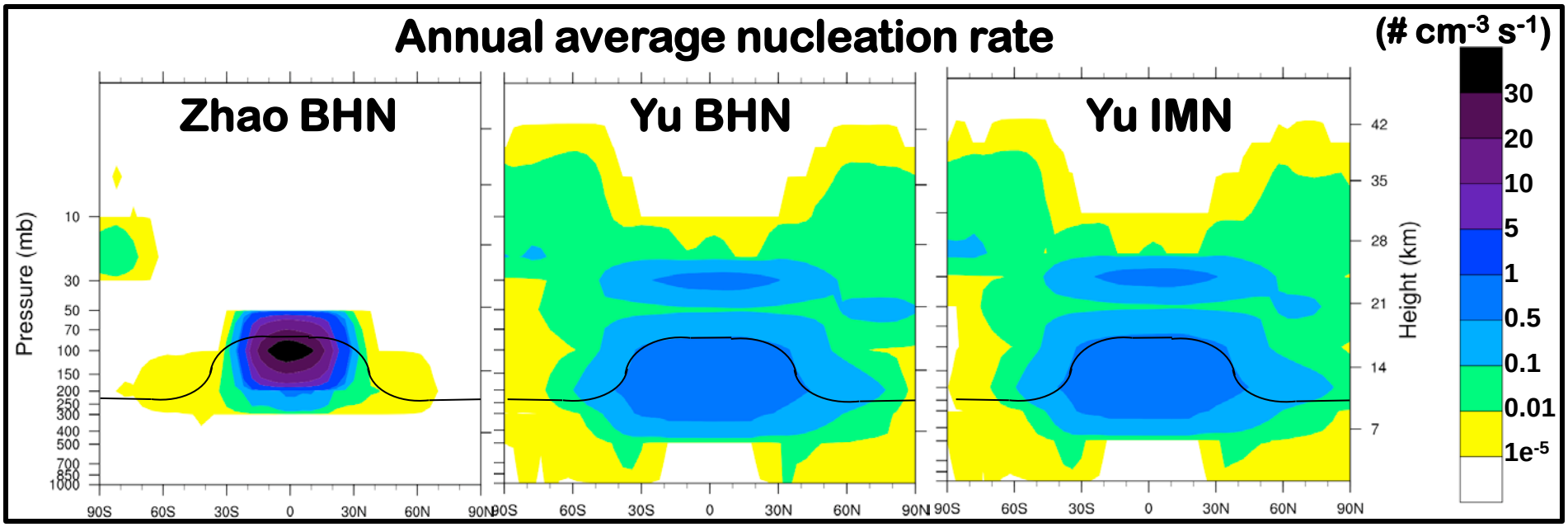
SO_2 emissions:
800 /cm^3/s at <15° lat >100 hPa

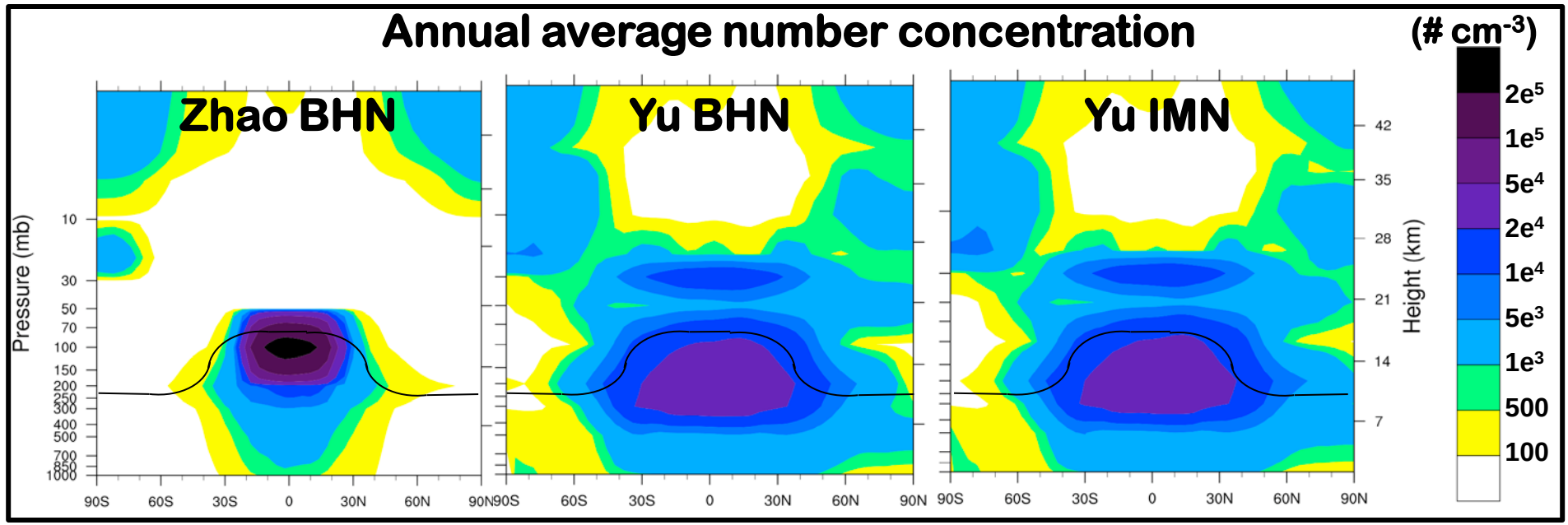


Aircraft observations taken July 1997 between 28° and 30° North

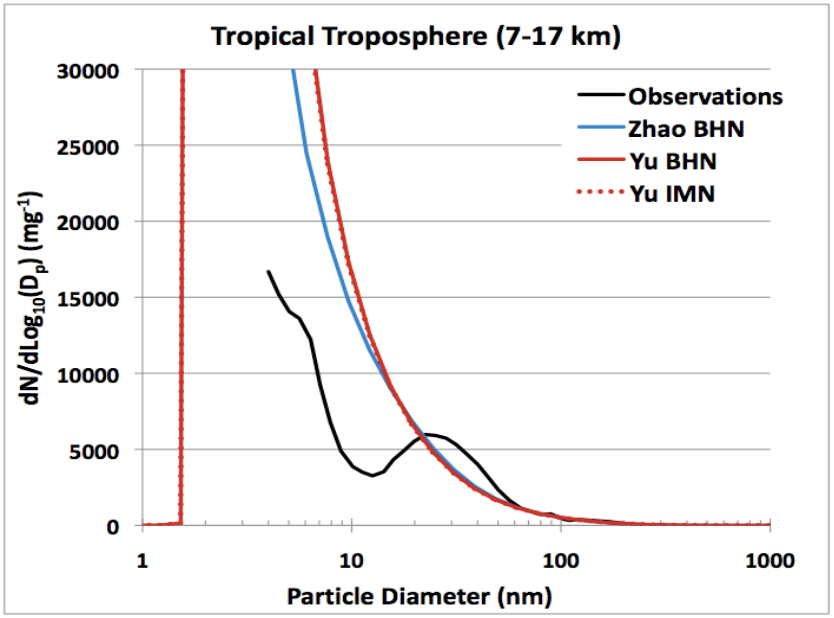
(Curtius et. al. 2001)

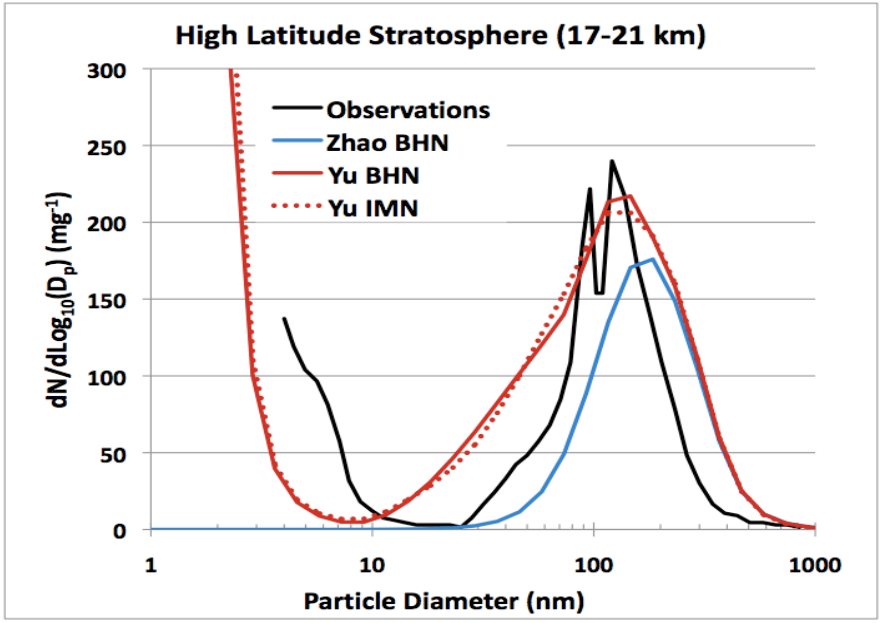
BHN rates differ 100x; ions insignificant

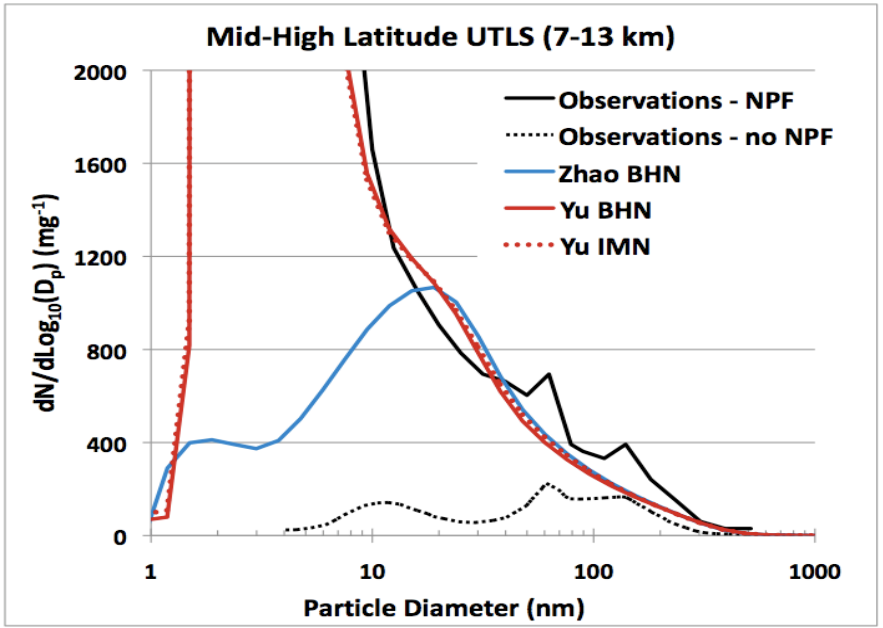




All 3 simulations generally reproduce observed sizes; BHN & IMN schemes similar



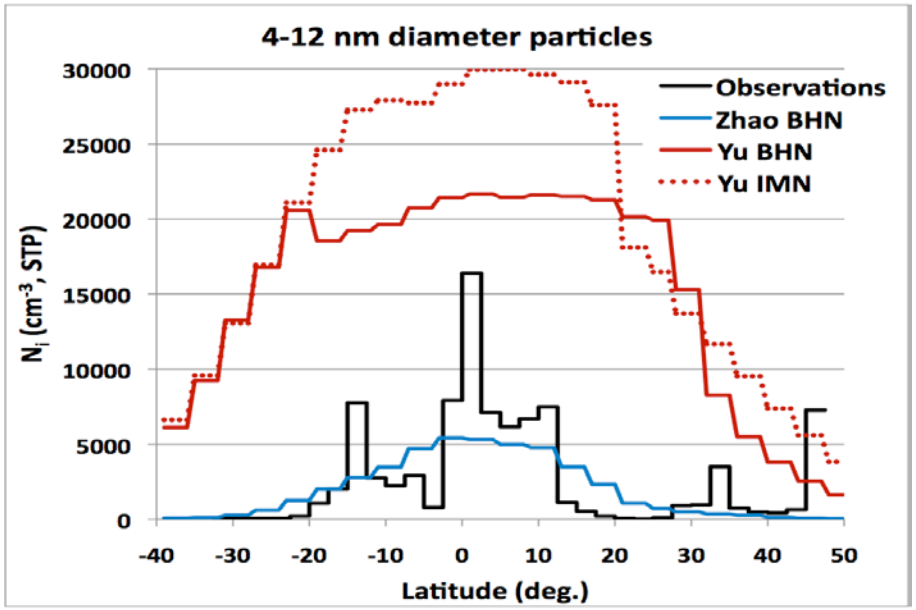


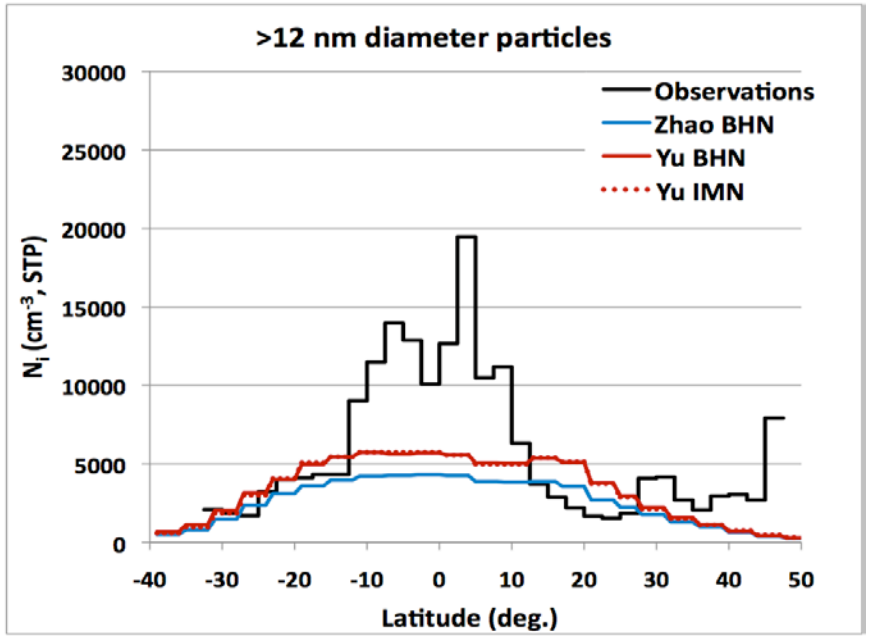


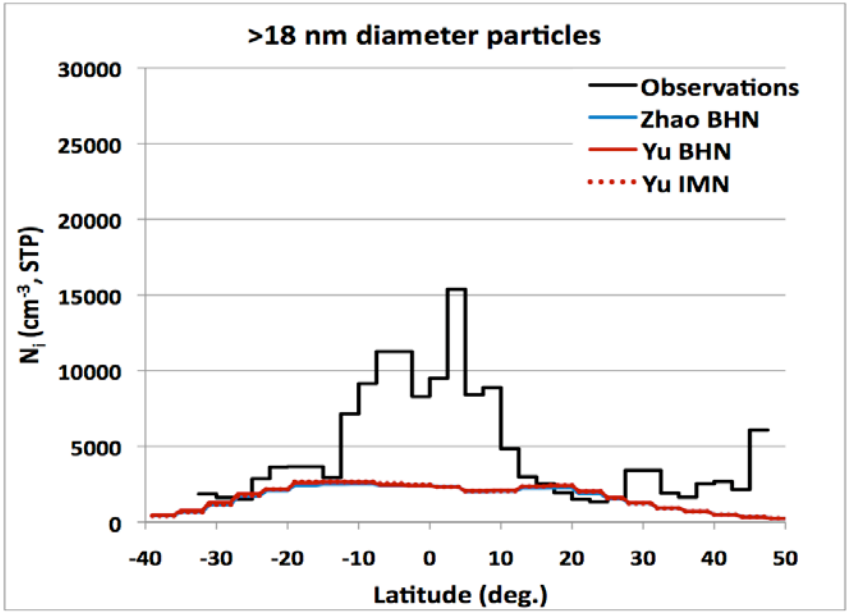
Observations are with 56 NASA aircraft flights 1998-2000

(Lee et. al. 2003)

Comparison to CARIBIC Observations 8.8-11 km

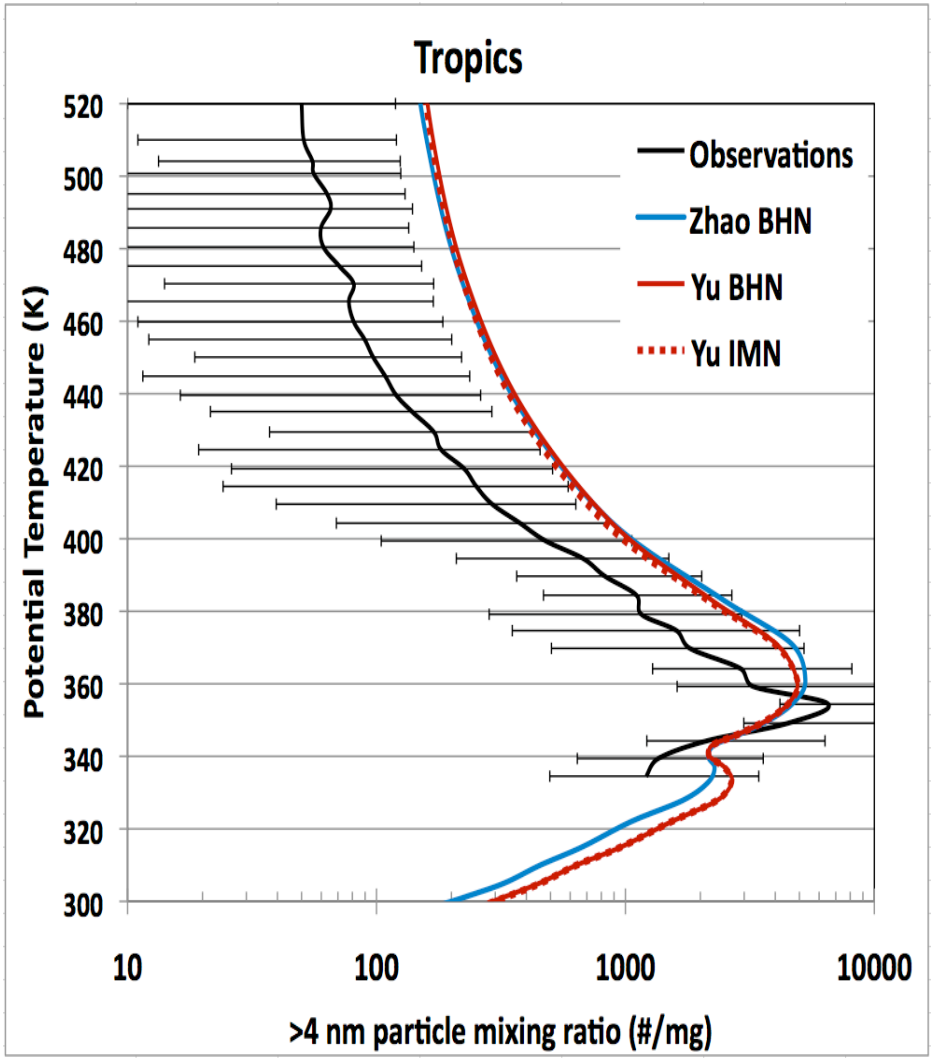


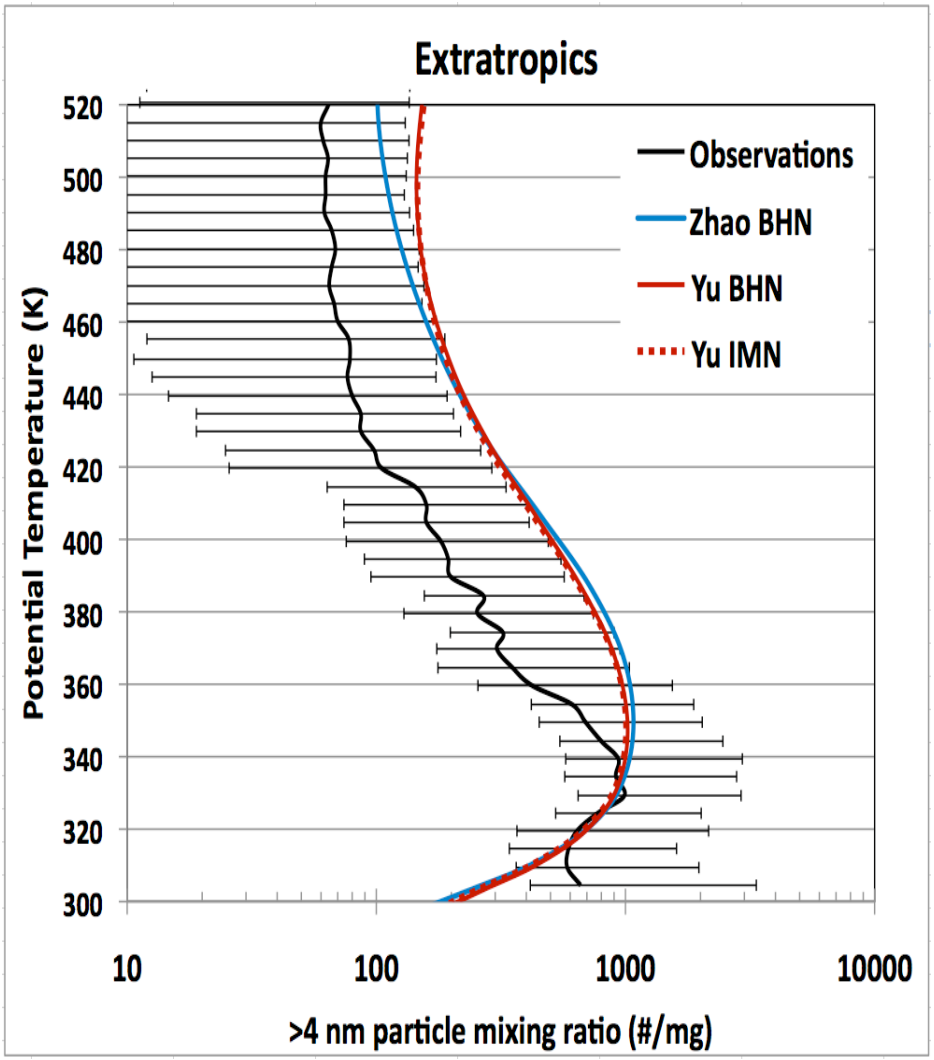




Obs. from CARIBIC 8.8-11 km *(Heintzenberg et. al. 2006)*

All 3 schemes similar and peak # compares well to obs; stratospheric numbers high from excessive nucleation





Assimilation of aircraft obs from 80's & 90's *(Brock et. al. 1995)*

WACCM/CARMA simulations suggest:

- Binary Homogeneous Nucleation of sulfuric acid and water is a key source of UTLS aerosol
- Ions are responsible for little additional nucleation in the UTLS, suggesting a minimal sun-cosmic ray-climate link
- Nuc. Rates for BHN schemes differ significantly but coagulation controls number concentration >10 nm
- All 3 schemes comparable reasonably well to observations of size distribution and number concentration
- Yu lookup tables are designed for troposphere and are not reliable in the stratosphere and above

Next Steps

- Add a surface emissions dataset of SO_2 (J-F Lamarque)
- Add micrometeoritic dust cores as a mechanism for new particle formation via heterogeneous nucleation (M. Mills, C. Bardeen)
- Study stratospheric sulfate geoengineering scheme (M. Mills, S. Tilmes)
- Convert to Fortran 90 version of CARMA (C. Bardeen)