Sulfate Aerosol Nucleation in the UTLS using WACCM/CARMA

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NCAR Chemistry-Climate Meeting
March 2, 2009

Why do we care about UTLS Sulfates?

- Impact clouds, climate, and stratospheric chemistry
- Geo-engineering, volcanic eruptions, anthropogenic sulfur emissions

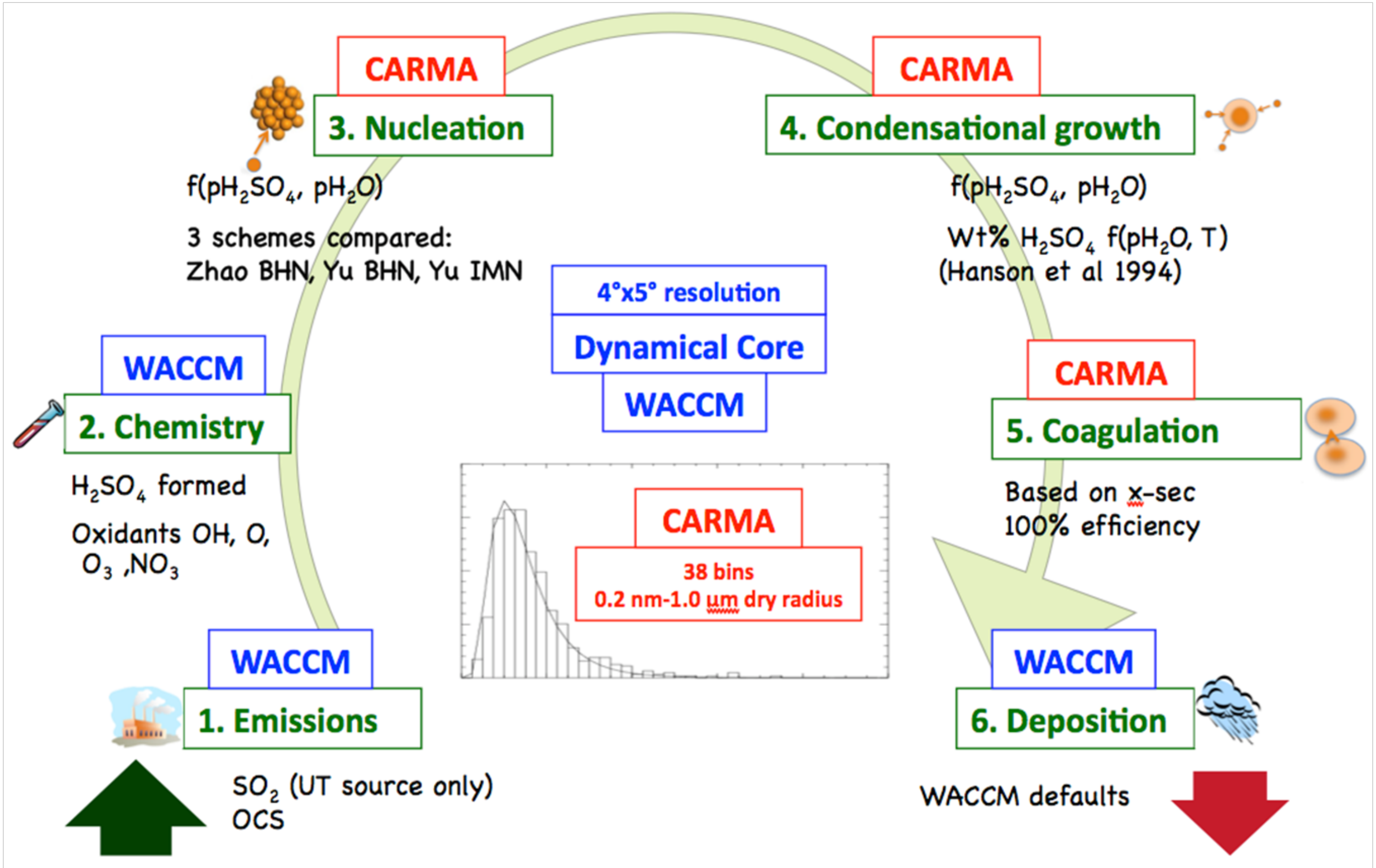
What do we know about the UTLS?

- Freshly nucleated sulfate aerosols observed *(Lee 2004)*
- Pure sulfate, sulfate w/organics, sulfate w/micrometeorites observed *(Murphy et. al. 2007)*
- Aerosol processes not well understood

Research Objective

Investigate the relative impact of various nucleation mechanisms to UTLS sulfate aerosol

Modeling Approach



2 year simulations; 2^{nd} year averaged

Study Design – 3 Nucleation Schemes

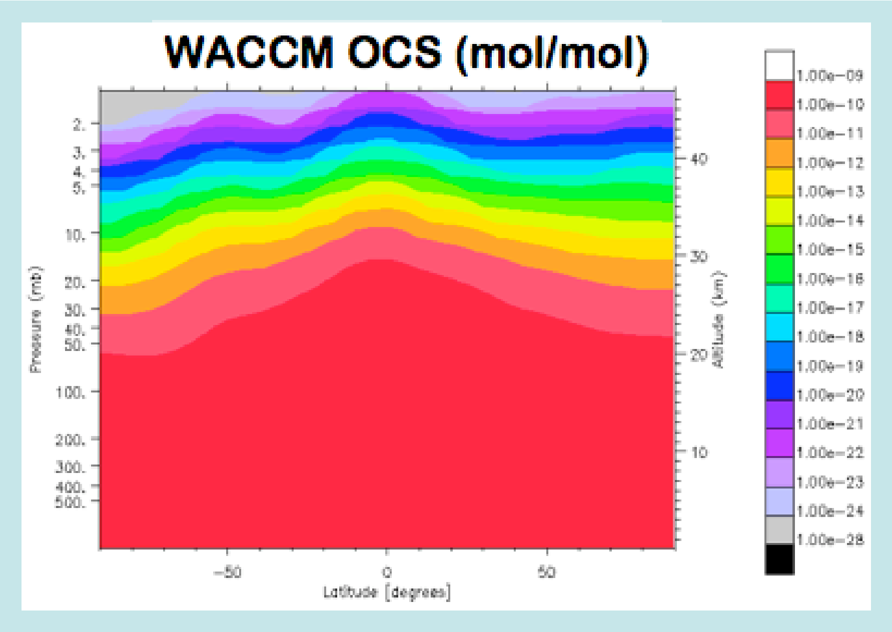
Simulation	Description
Zhao BHN	Critical Gibbs free energy calculated via % H_2SO_4 and cluster volume *(Zhao and Turco 1995)*
Yu BHN	Lookup tables based on quasi-unary nucleation of H_2SO_4 in equilibrium w/H_2O vapor *(Yu 2008-1)*
Yu IMN	Lookup tables based on Yu BHN plus IMN, 10 ion-pairs/cm^3 *(Yu 2008-2)*

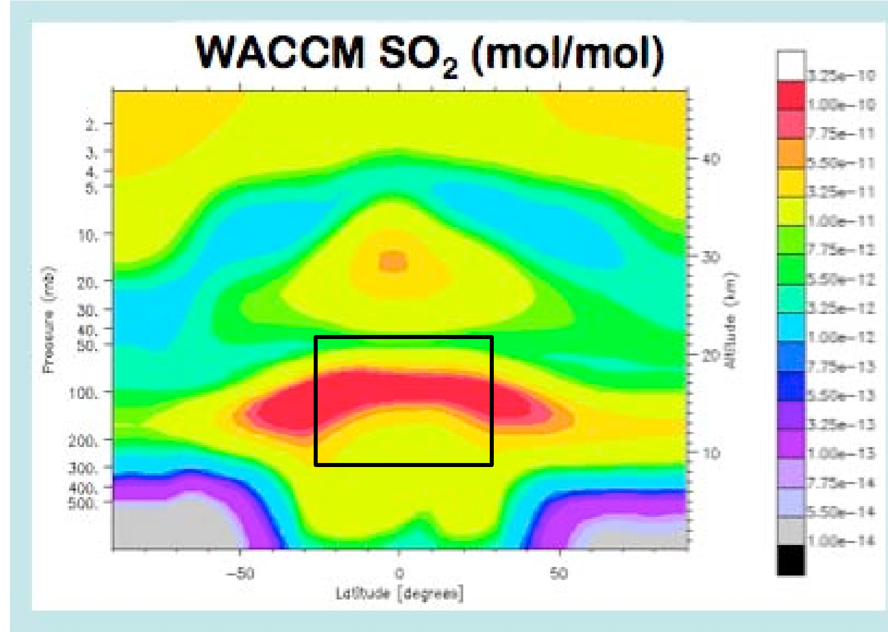
BHN=Binary Homogeneous Nucleation of sulfuric acid-water
IMN=Ion-Mediated Nucleation of sulfuric acid-water

Sulfur Emissions

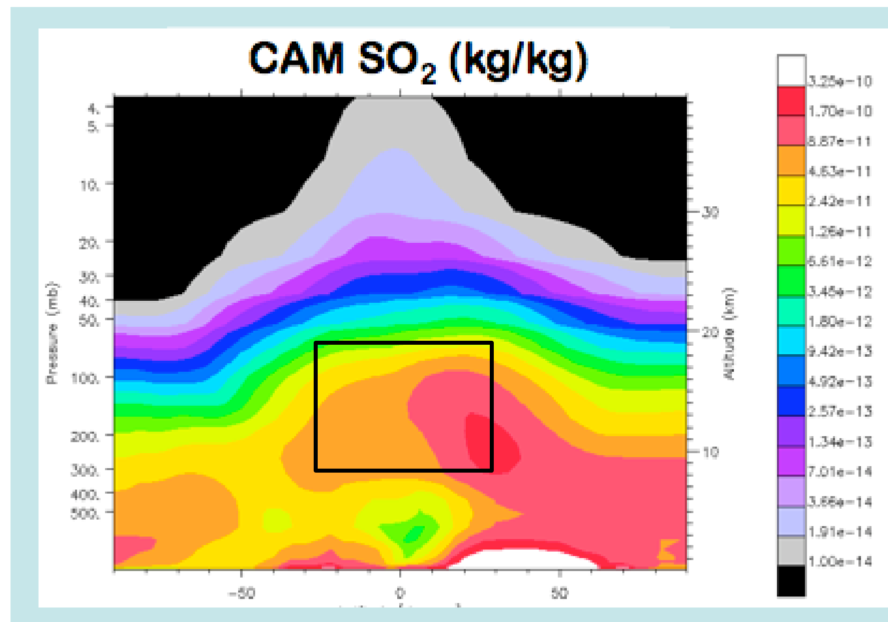
OCS emissions: 510 pptv in troposphere
SO_2 emissions: 800 /cm^3/s at <30° lat >100 hPa,
no surface emissions

Sulfur emissions reasonable (SO_2 too wide)



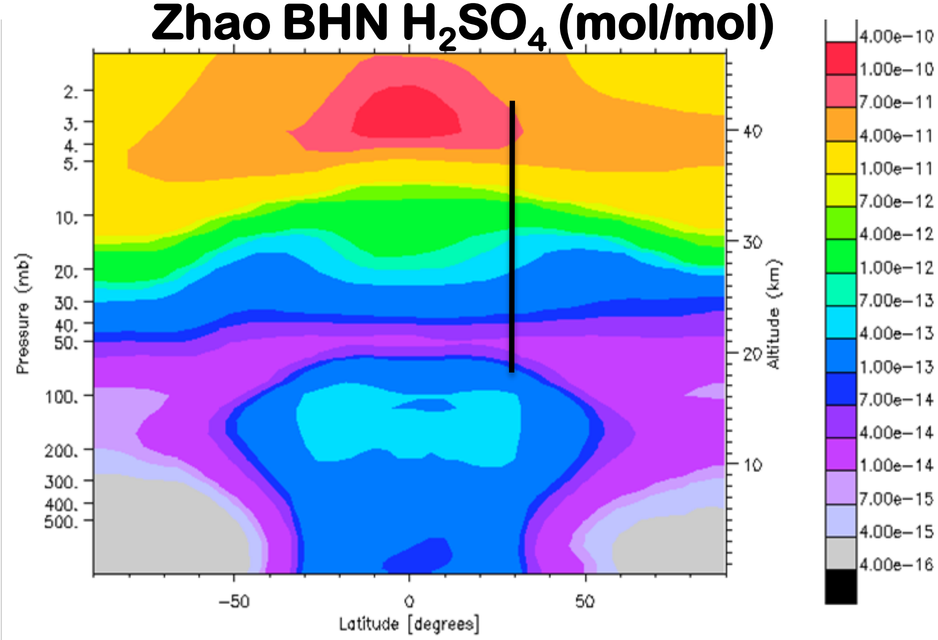


- WACCM OCS reasonable
- WACCM SO_2 reasonable in most regions but too high in the extratropics

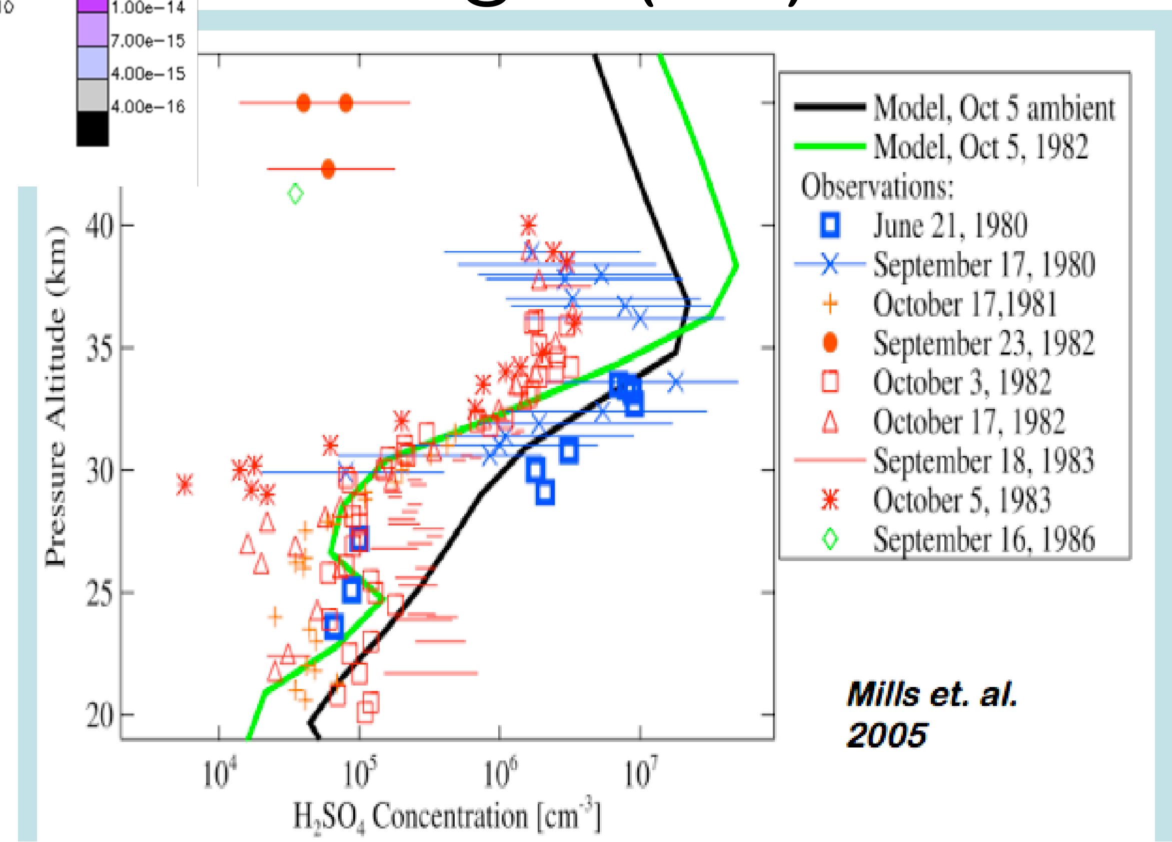


Sulfuric Acid Profiles match obs well

Zhao BHN H_2SO_4 (mol/mol)

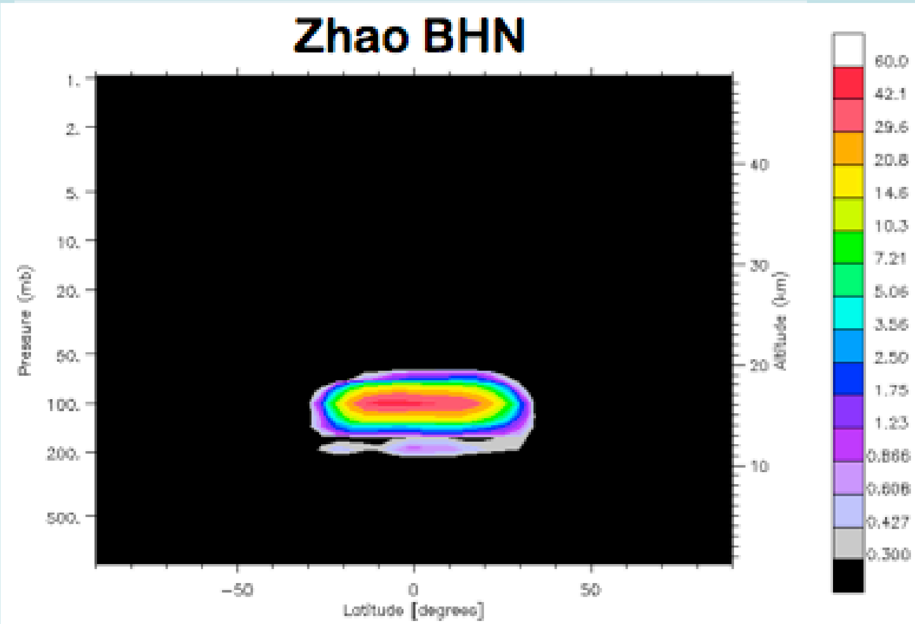


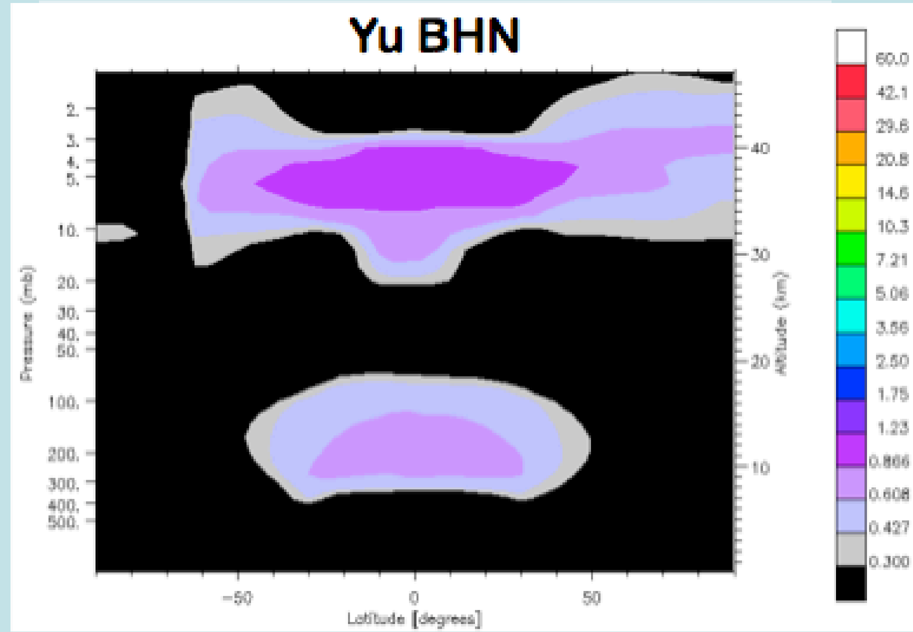
Obs @ 43 N (#/cm^3)



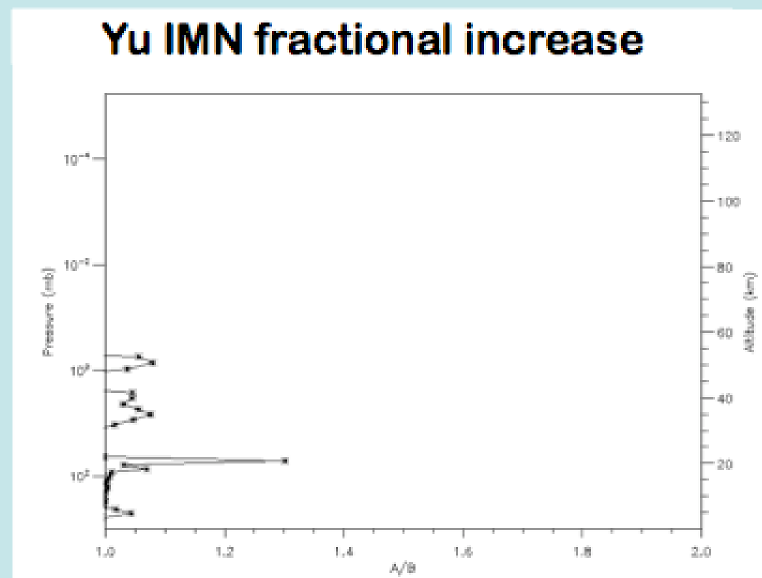
Mills et. al. 2005

Nucleation Schemes predict different rates



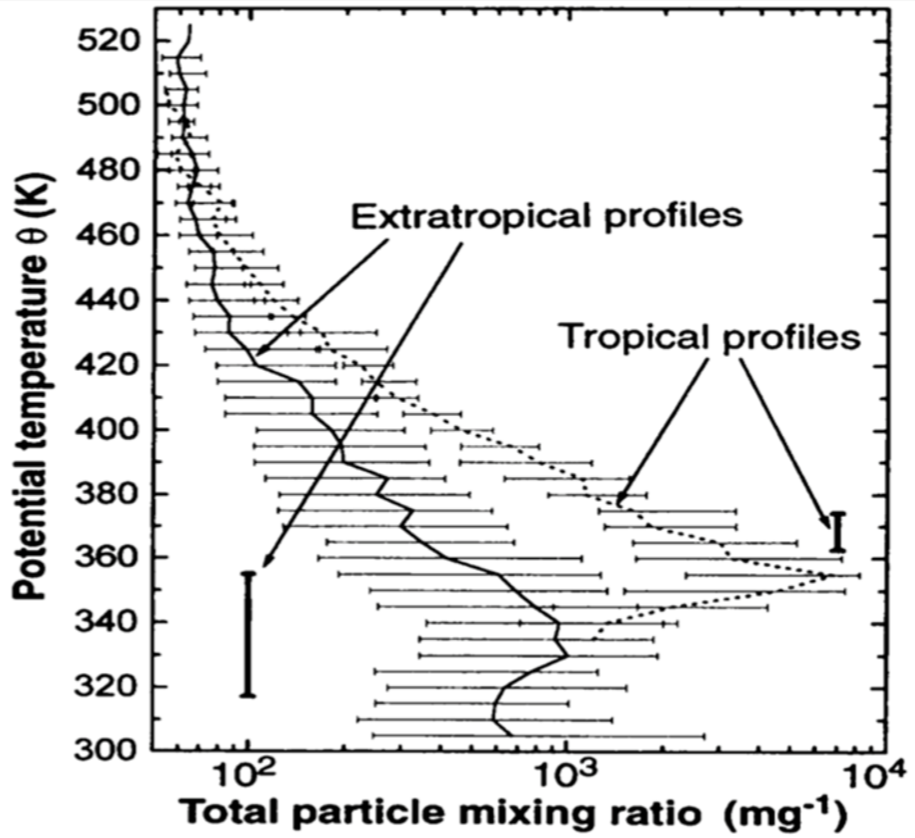


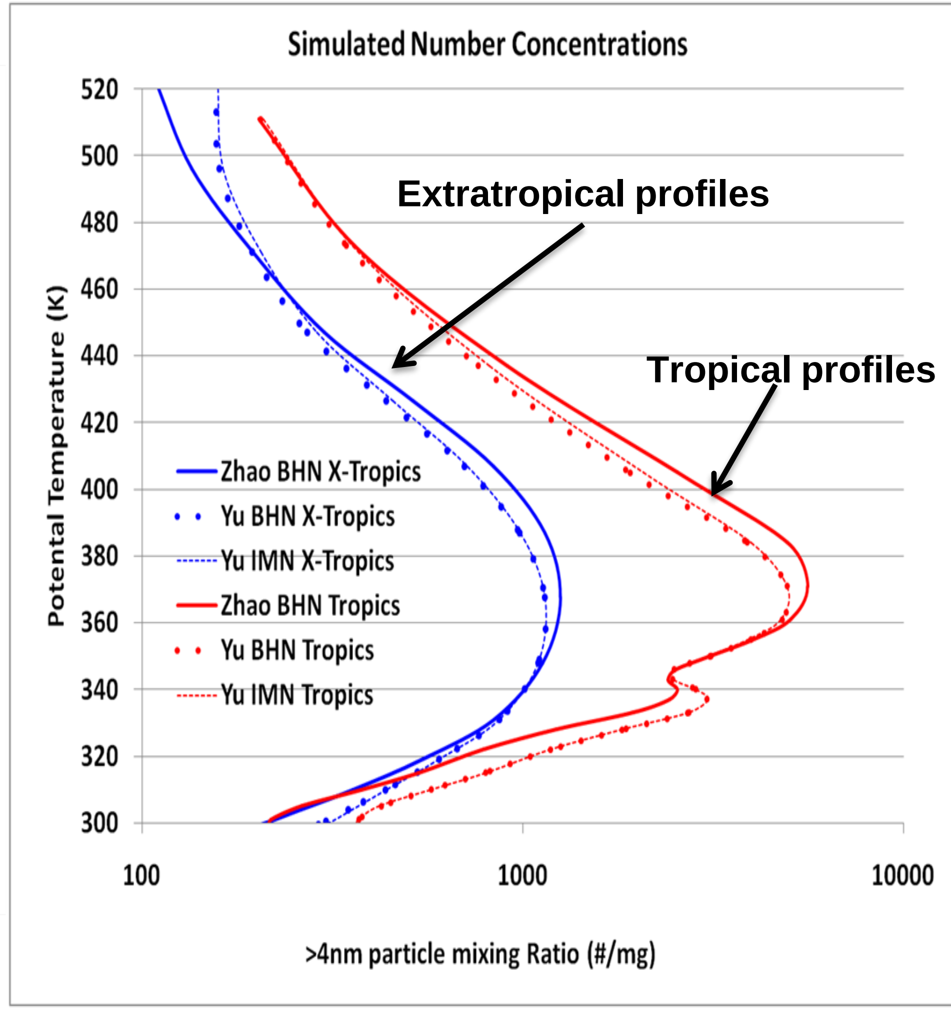
- Zhao BHN rates 100 times higher than Yu BHN in tropical UTLS
- Yu BHN extend to mid-troposphere and upper stratosphere
- Yu IMN increases nucleation by up to 30% over Yu BHN



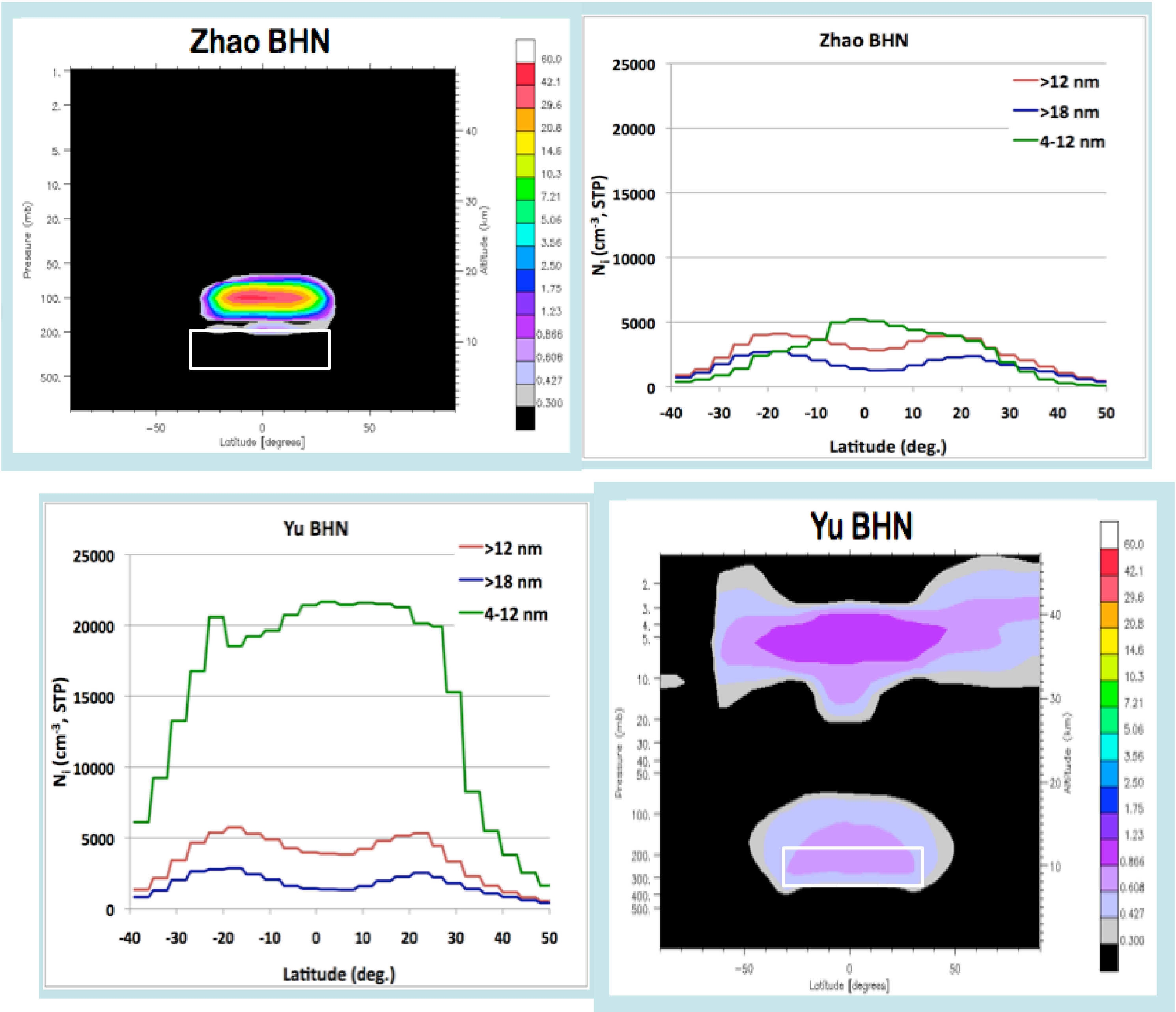
Vertical Profile of # Conc. looks good

Obs – Brock 1995



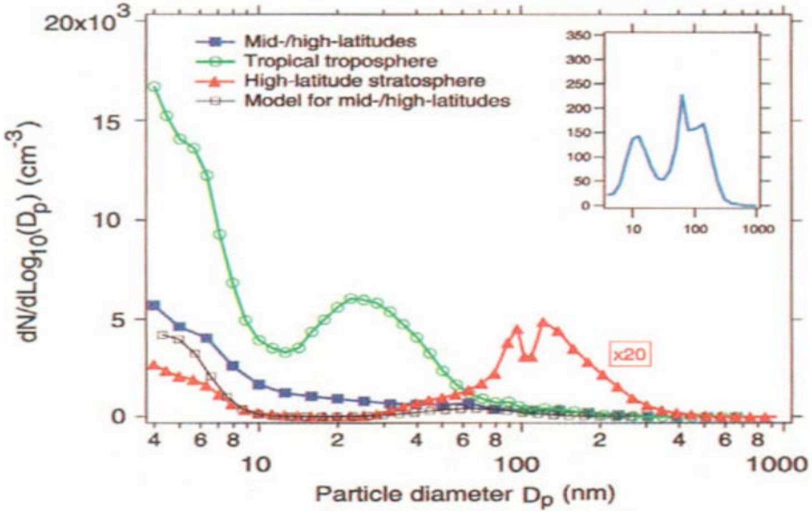


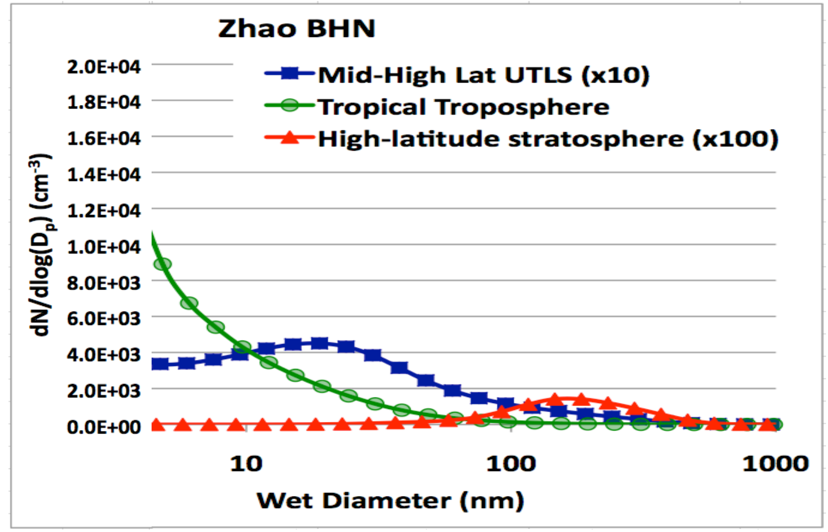
Zonal Profile of # varies, and too wide

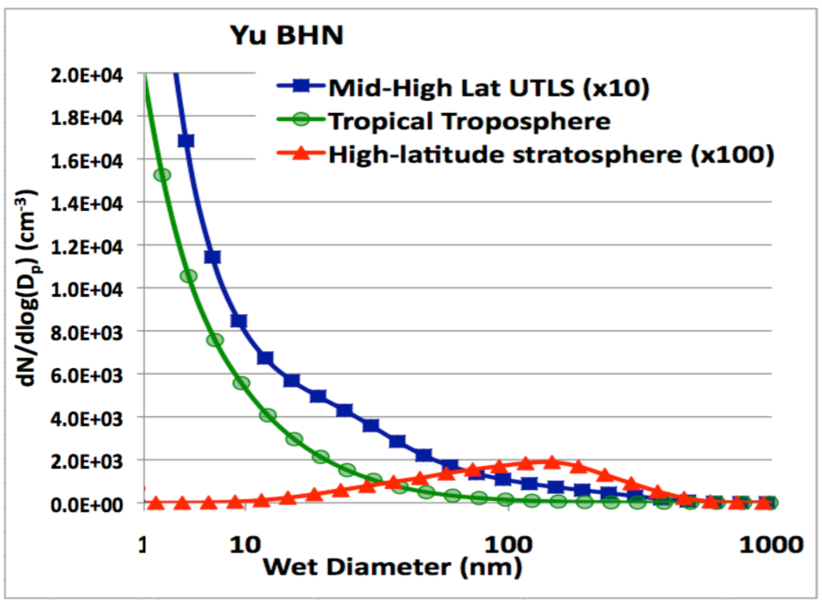


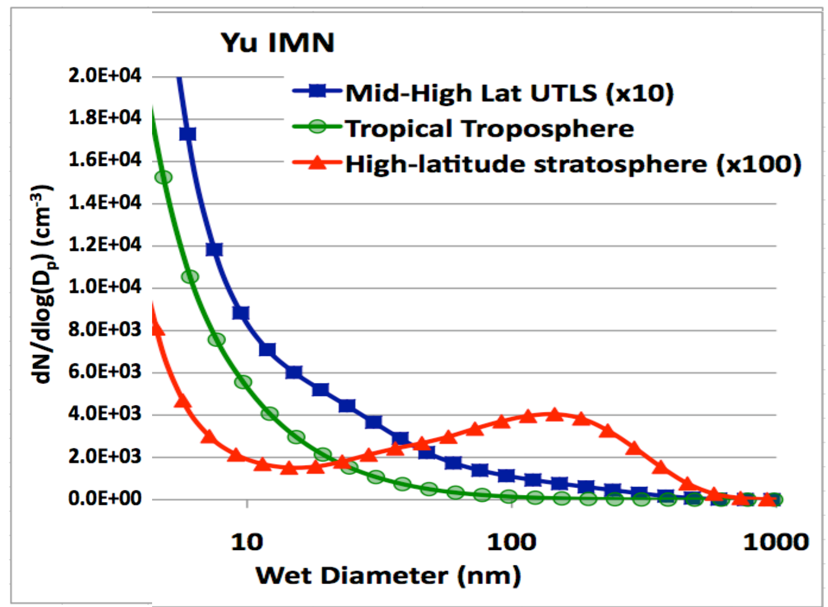
Size Distribution Shapes good

Obs – Lee 2003









Summary

- Three sulfate nucleation schemes successfully incorporated into coupled microphysical dynamical model

- Differences in nucleation schemes:
  - UTLS: Zhao BHN 100 times higher than Yu BHN
  - Mid-trop & Upper Strat: Yu BHN higher
  - Yu IMN rates +0-20% over Yu BHN

- Size distributions, zonal averages, and vertical concentrations match obs reasonably well

Next Steps

- Add sulfur and organics surface sources to model

- Add micrometeorites to model

- Add dust and sea salt aerosols (L. Su, T. Fan)

Acknowledgments

- Brian Toon
- Mike Mills
- Fangqun Yu
- Chuck Bardeen

Aerosols in the UT

•Evidence: cirrus clouds, obs, models, LS aerosols

Significance: aerosol indirect effect; long-range tropospheric transport; stratospheric chemistry

Req'ments to reach UT: high emissions and long lifetime

MUCH uncertainity due to challenges
Observations - low AOD and high altitude make measurements difficult *(SPARC 2006)*
Modeling - difficult to constrain aerosol MMR due to transport, new particle formation, evaporating clouds, wet deposition

Sulfur Chemistry in MOZART

- Photolysis

jh2so4 (73) $H_2SO_4 + hv \rightarrow SO_3 + H_2O$ rate = ** User defined **

- Reactions

(131) $SO_2 + OH + M \rightarrow HSO_3 + M$ troe : ko=3.00E-31*(300/t)**3.30 (208)
ki=1.50E-12 f=0.60

(132) $HSO_3 + O_2 \rightarrow SO_3 + HO_2$ rate = 1.30E-12*exp(-330./t) (209) usr13

(133) $SO_3 + H_2O \rightarrow H_2SO_4$

- Equation Report

d(SO2)/dt = j75*SO3 + r120*OCS*OH + r124*SO*OH + r125*SO*O2
+r126*SO*O3 + r127*SO*NO2 + r128*SO*CLO + r129*SO*BRO
+r130*SO*OCLO - j74*SO2 - r131*M*OH*SO2

d(SO3)/dt = j73*H2SO4 + r132*HSO3*O2 - j75*SO3 - r133*H2O*SO3

d(HSO3)/dt = r131*M*SO2*OH - r132*O2*HSO3

d(H2SO4)/dt = r133*SO3*H2O - j73*H2SO4

Aerosols in the UT/LS

CRYSTAL-FACE: aircraft obs in Florida, analyzed by TEM

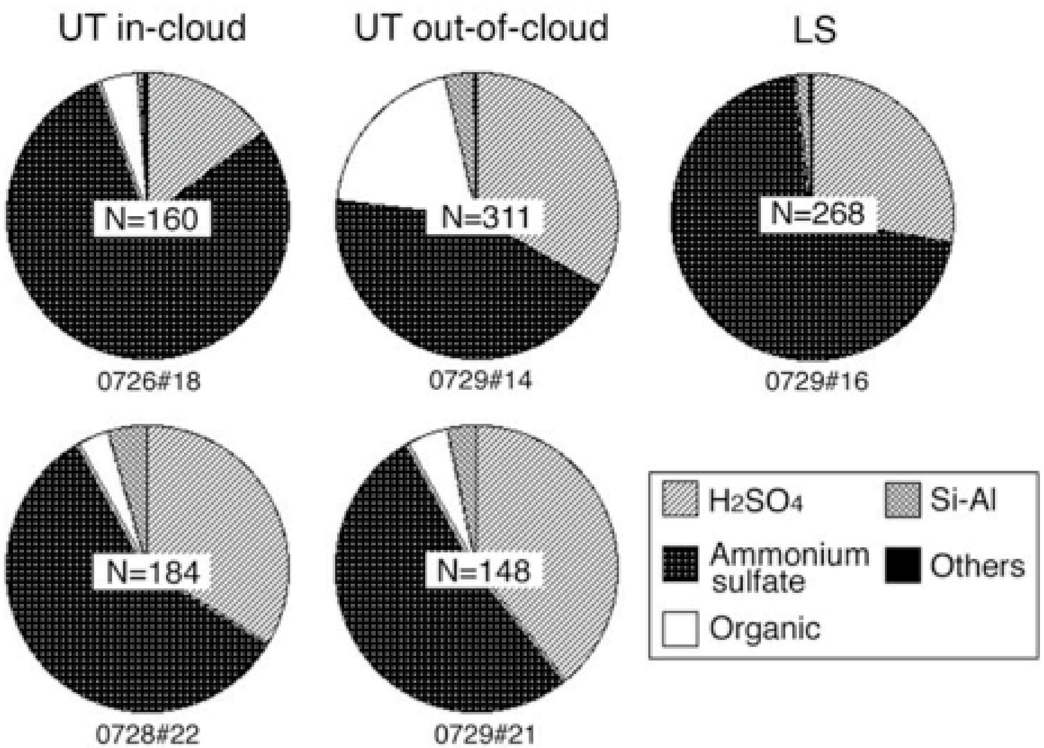


Figure 6. Proportions of UT in-cloud (0726#18 and 0728#22), UT out-of cloud (0729#14 and 0729#21), and LS (0729#16) aerosol samples. The number of analyzed particles is shown in each pie chart. Zn-rich particles are not included in the pie charts. The abundances of organic and Si-Al-rich particles decrease with altitude.

Sulfates dominate everywhere (neut. and unneut.)
Sulfates cross tropopause but organics do not

Kojima et al (2004)