Sources of Marine Boundary Layer Particles
VOCALS Meeting June 11, 2007

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HIGEAR
Hawaii Group for Environmental Aerosol Research

PACIFIC AIRCRAFT EXPERIMENTS 1990-2006

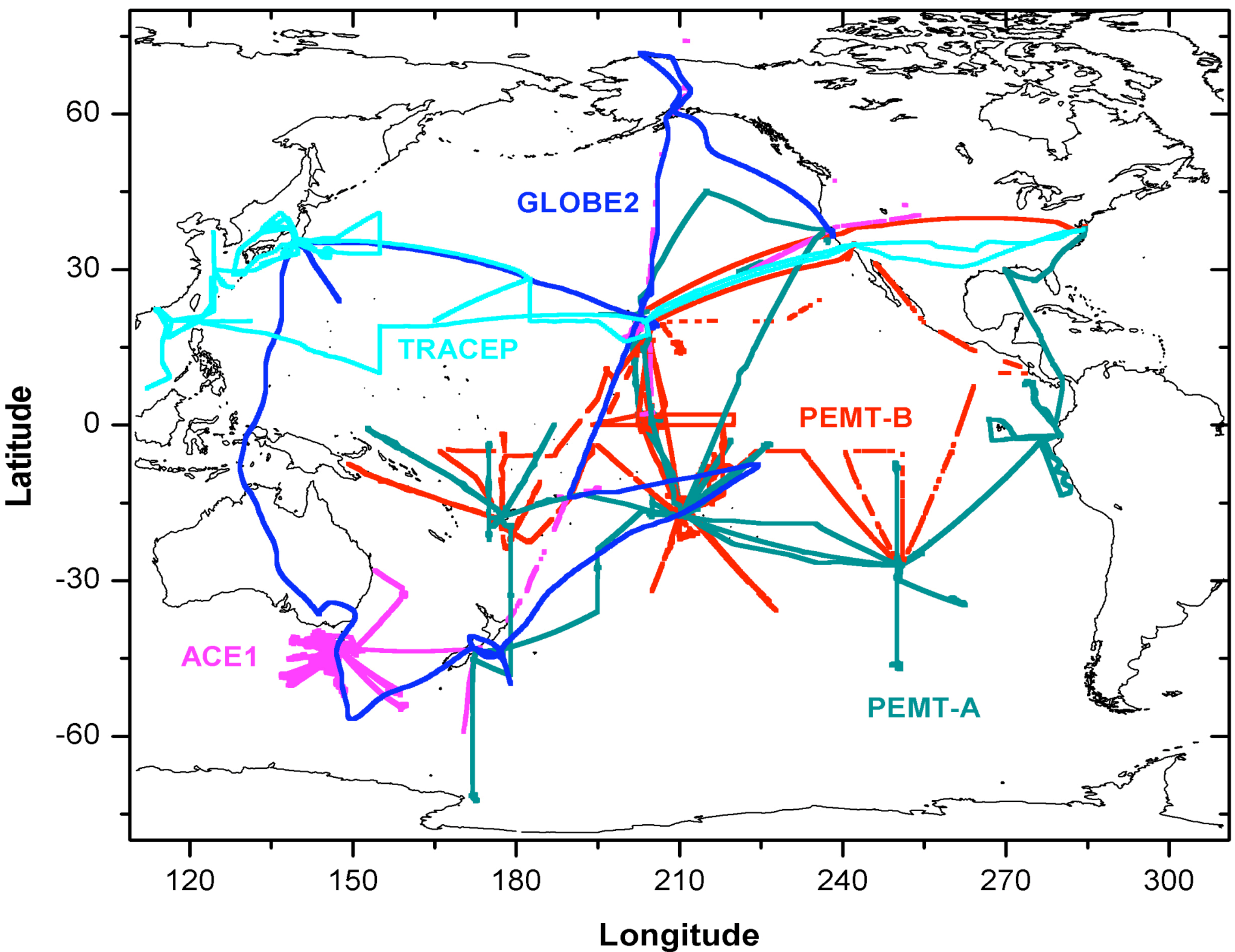
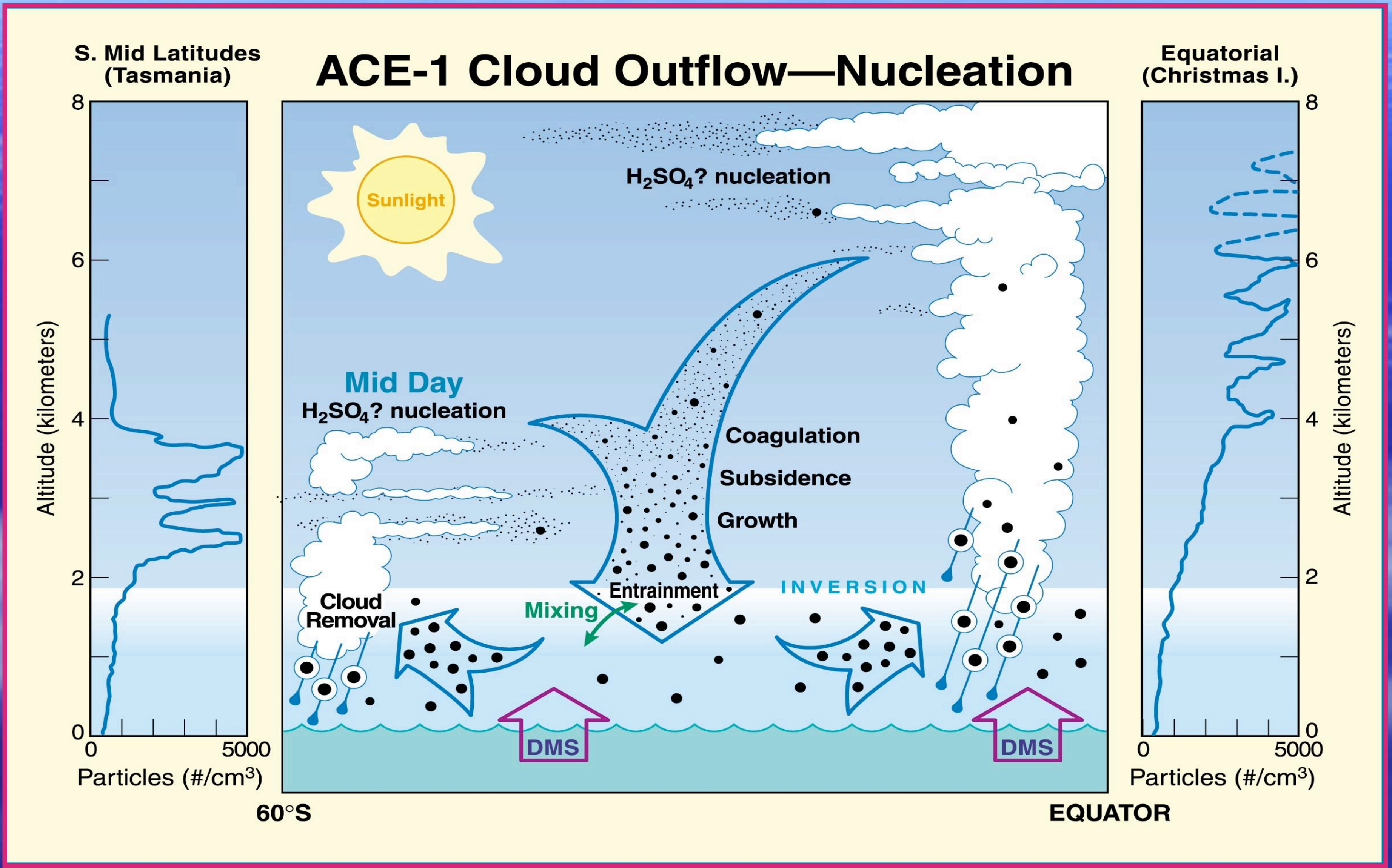
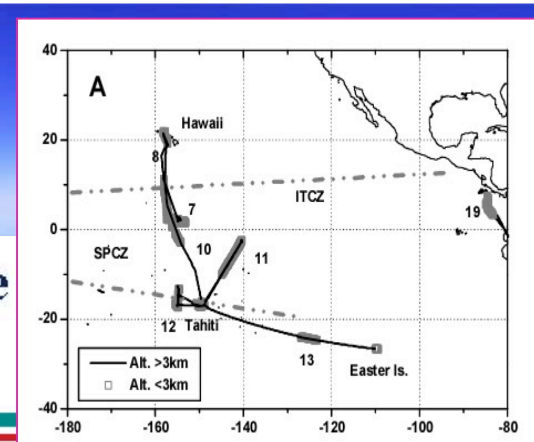


Diagram of Free Troposphere Nucleation Through Cloud Processes

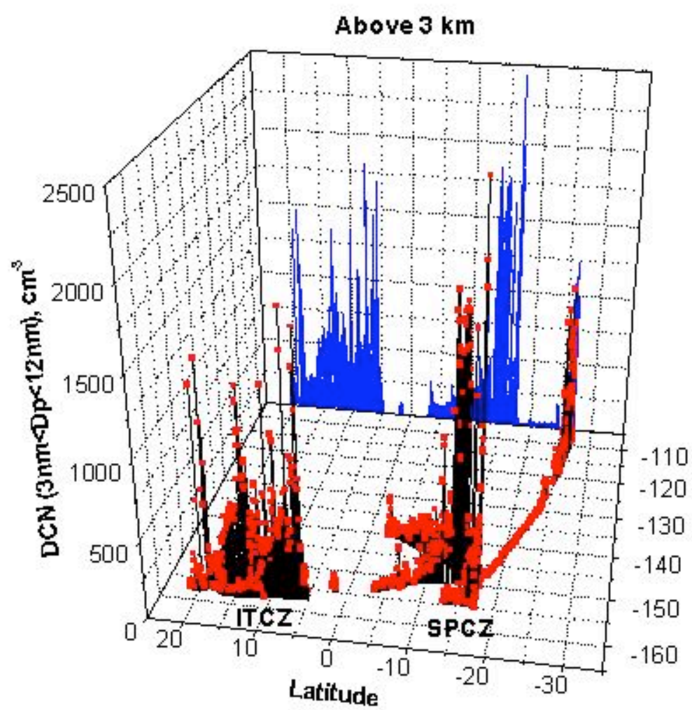


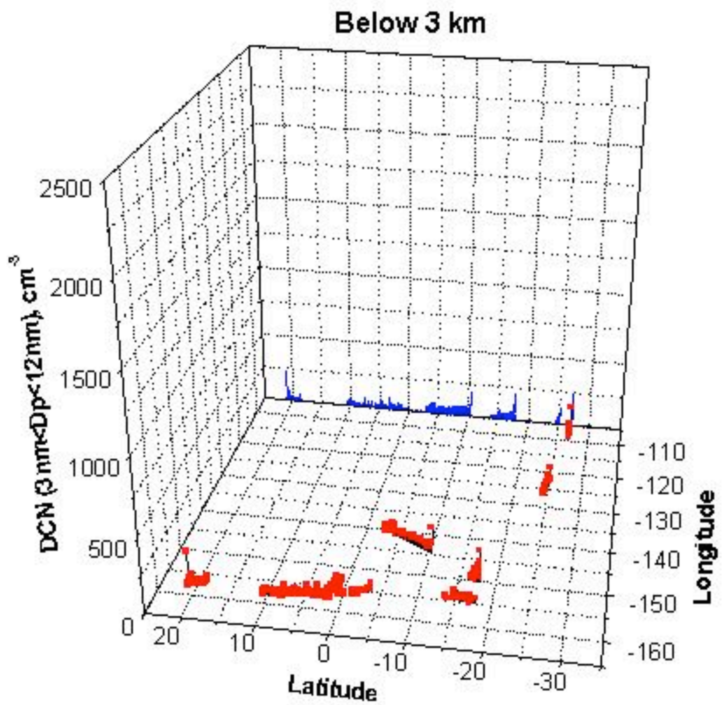
Free Troposphere Nuclei



Number Concentration is Enhanced in FT as the Result of New Particles Production.

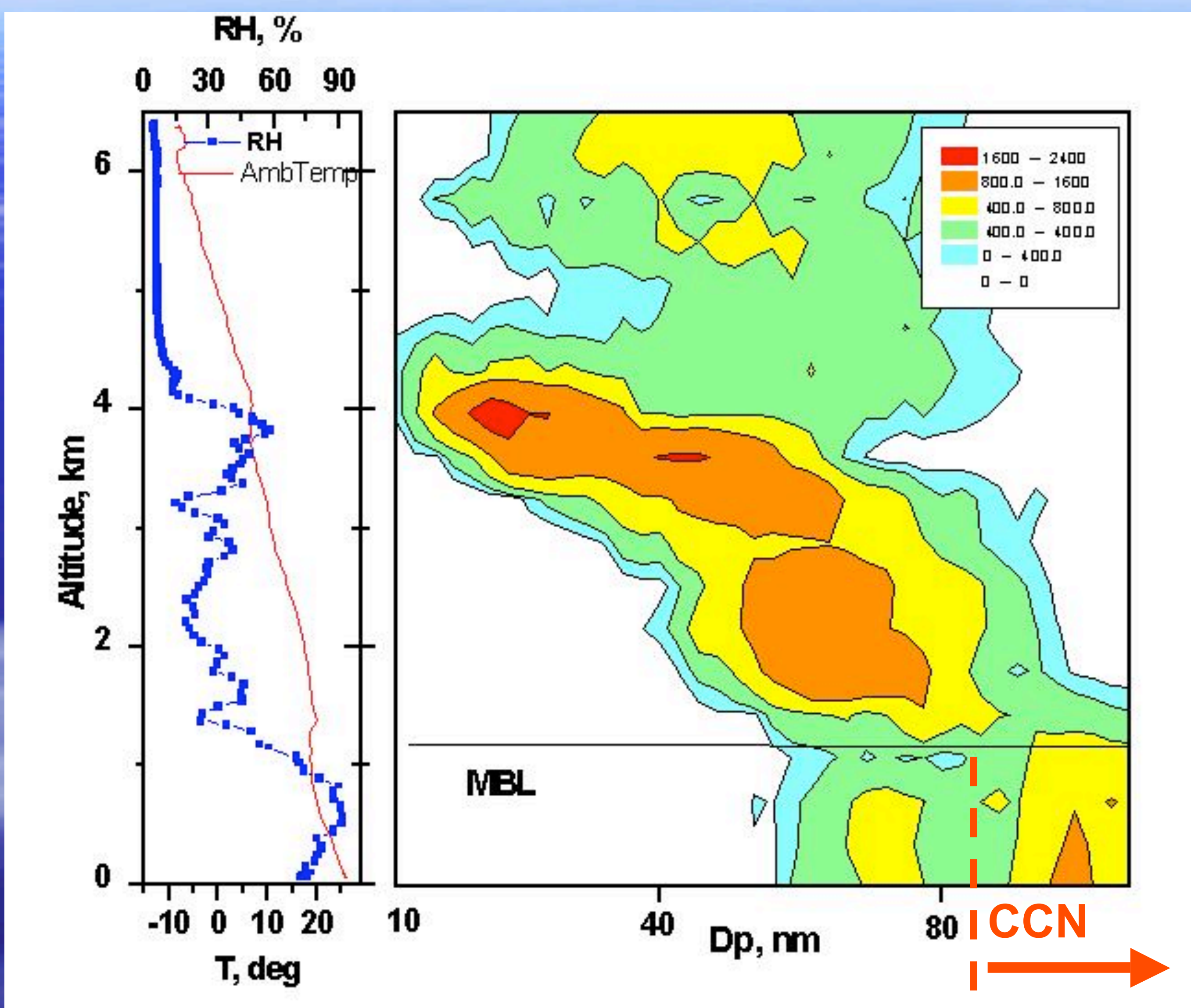
A 3D latitude-longitude distributions of DCN (3nm<Dp<12nm) are highest aloft (above 3km) near ITCZ and SPCZ and almost no DCN particles are present in the MBL (observations from all PEM Tropics flights).



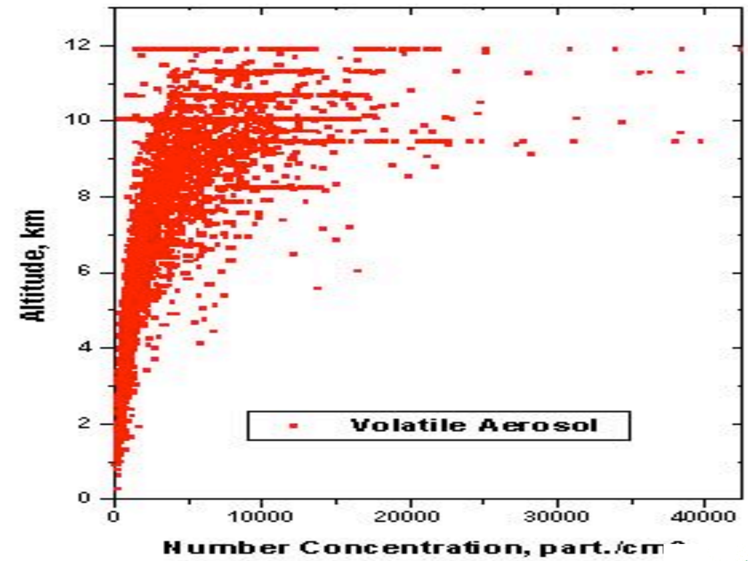


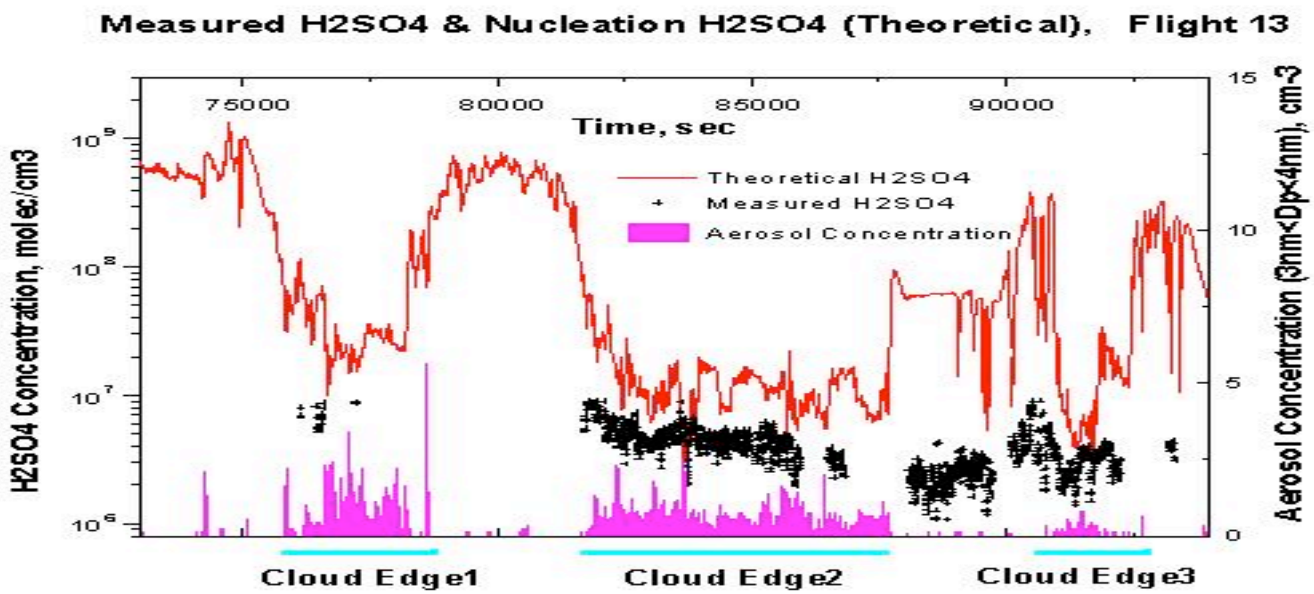
Subsidence from ACE-1 Profile in Tropics.

Note 90nm Hopple minimum through cloud processing in the MBL



Cumulus layers (photo), vertical profile of nuclei layers and nucleation from sulfuric acid at cloud edges

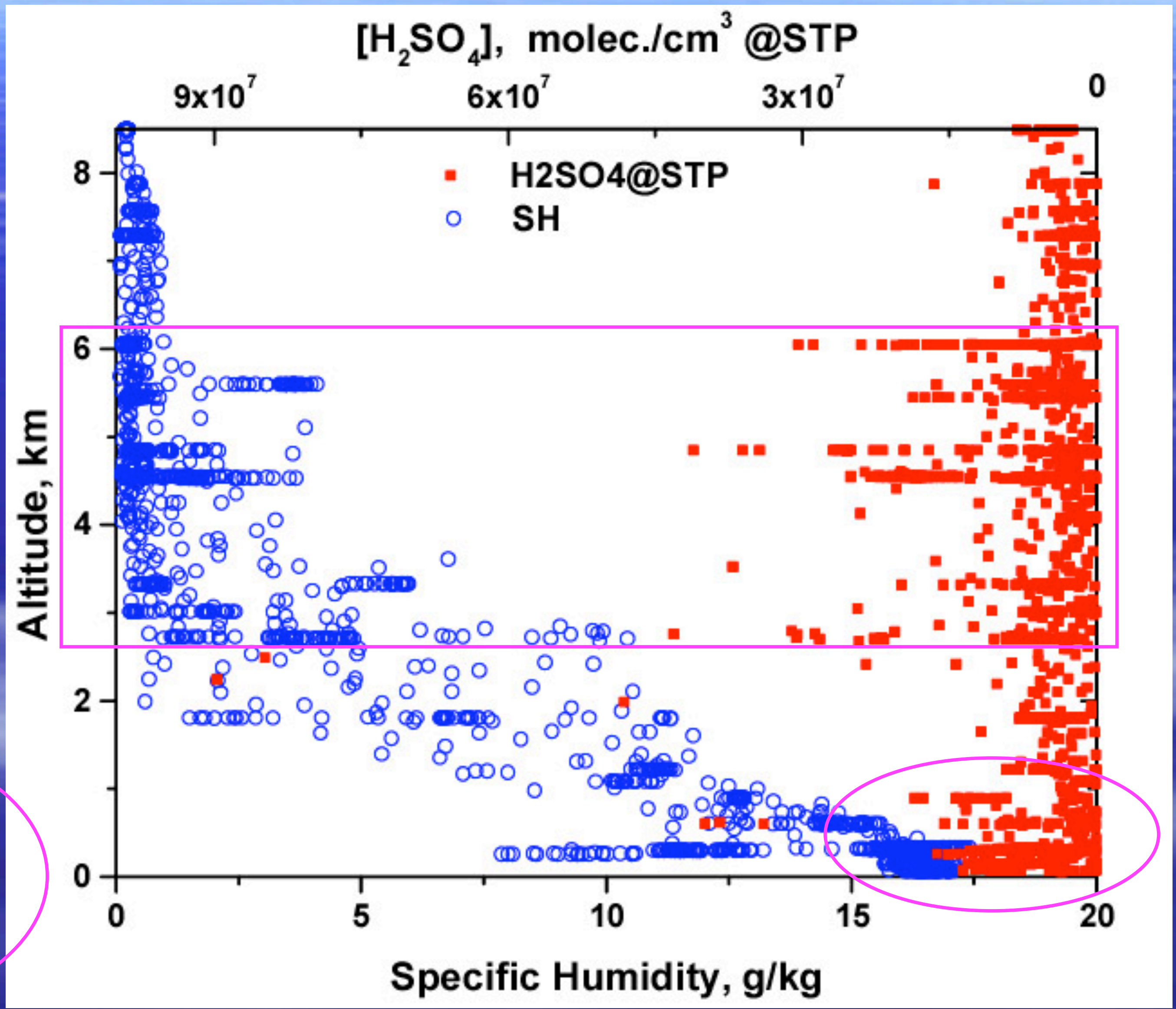




C_{crit} = -.16 exp(0.1T -3.5RH-27.7); criteria for binary nucleation of sulfuric acid, A. Wexler et al. 1994

Vertical Profiles of Specific Humidity and Sulfuric Acid (F. Eisele)during PEMT A&B

Most elevated sulfuric is in 2-6km range with evidence of cloud detrainment into dry air with low surface area.



Some cases of high sulfuric in scavenged low surface area MBL air & inversion criteria for binary nucleation of sulfuric acid

$Ccrit = -.16 \exp(0.1T - 3.5RH - 27.7)$

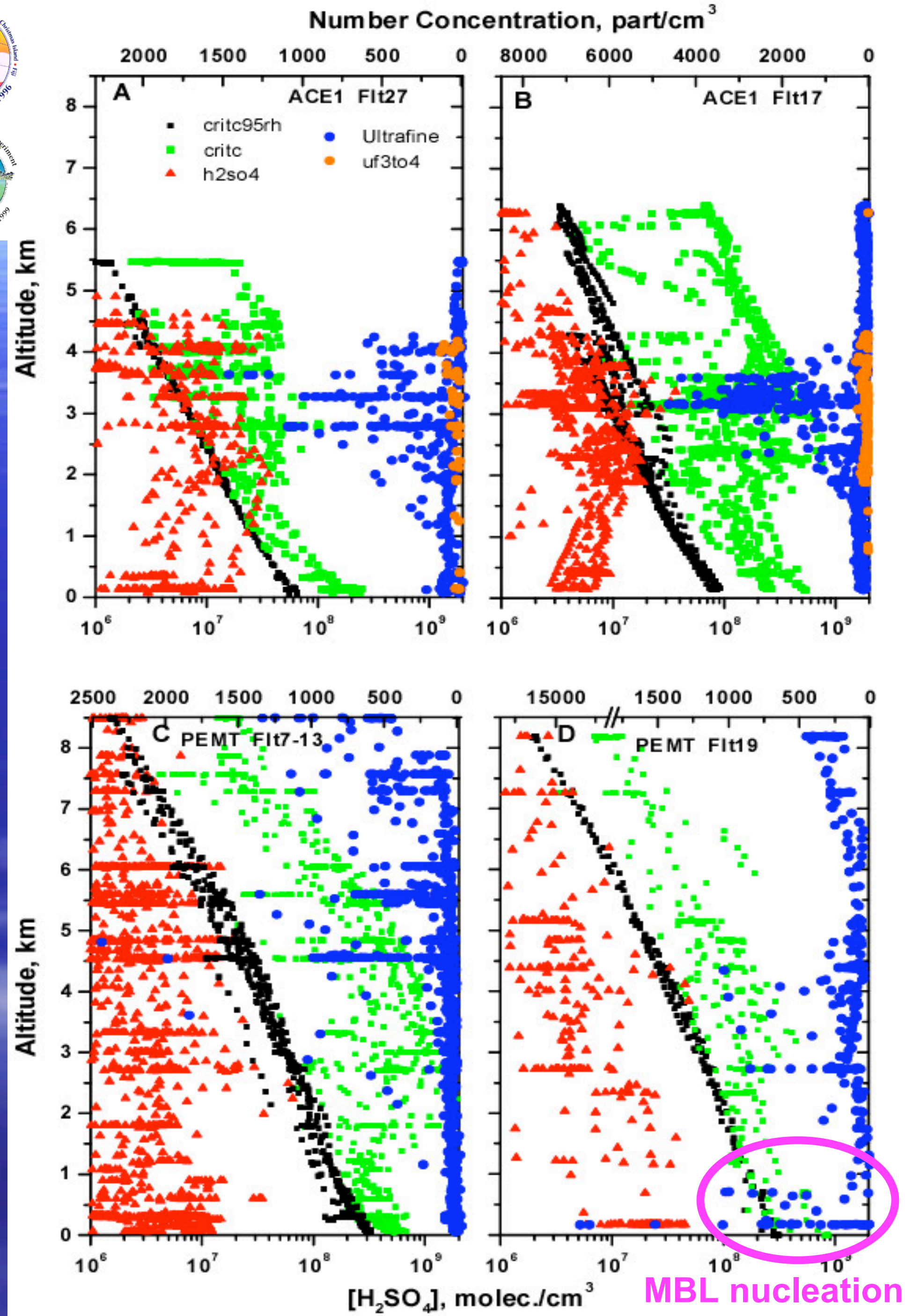
Ccrit95= value at 95% RH

The **Critical concentration** of sulfuric acid required for binary nucleation of sulfuric acid can be parameterized in terms of RH and ambient T (green symbols) (Wexler, 1989).

The critical concentration near cloud can be estimated at 95%RH and ambient T. Crit95 - (black symbols).

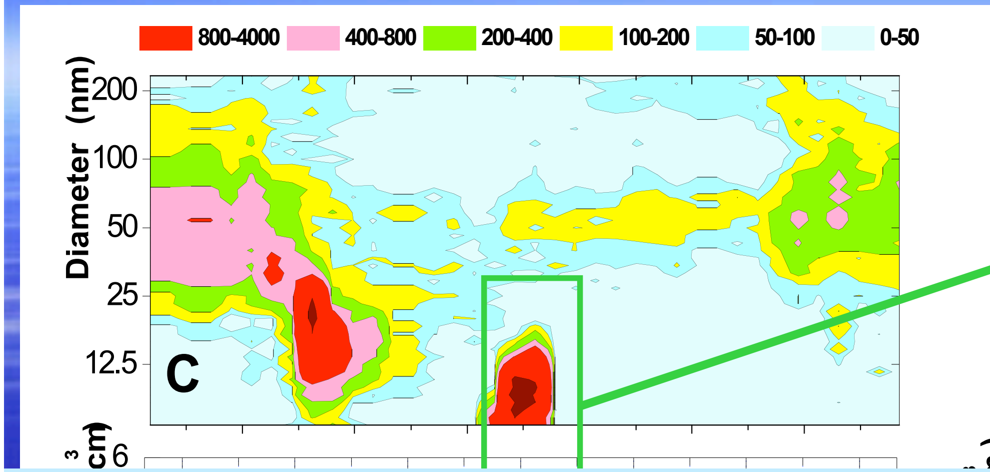
Ultrafine particles and high concentrations of 3-4nm particles (R. Weber) are frequently high when measured sulfuric concentrations (F. Eisele) are close to Crit95

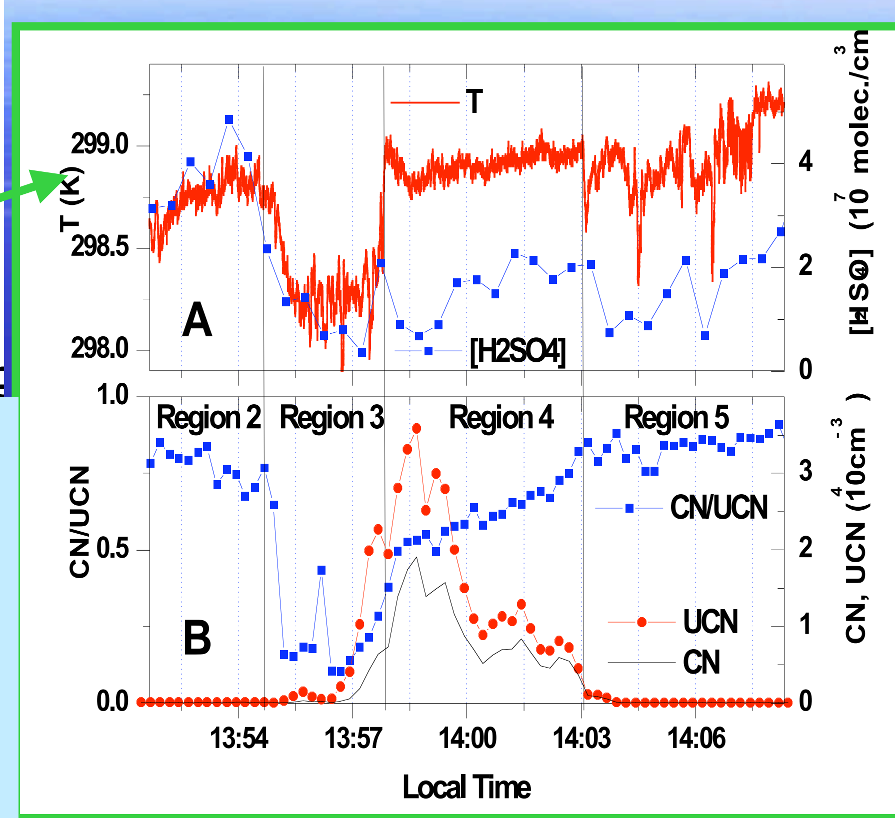
Cloud outflow and low T provide favored regions for nucleation when sufficient sulfuric acid is present.



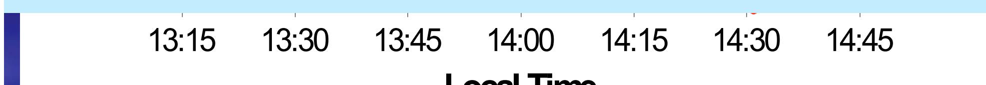
MBL nucleation for over 20km off Equador at 100m alt with 94%RH

Did we measure an example of POC conditions?



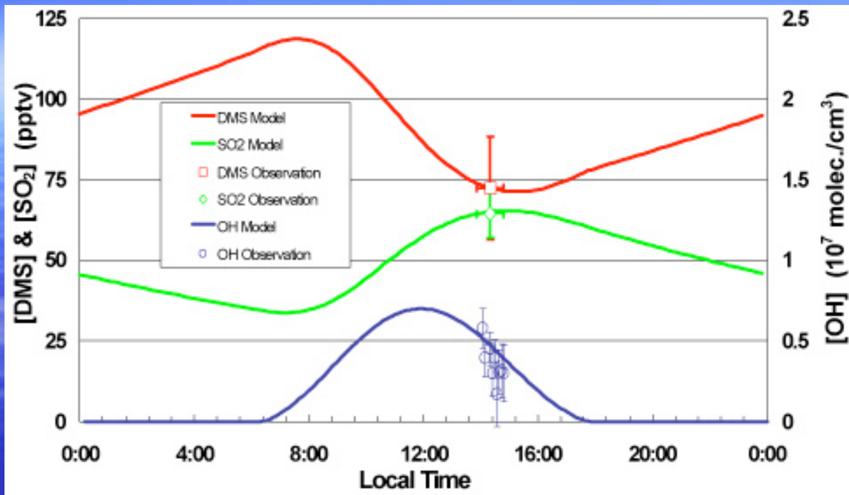


- Air highly scavenged by earlier deep convection
- Highest sulfuric in region of lowest surface area and with no nucleation
- Nucleation evident and sulfuric drops ½
- Onset associated with 0.5 deg temp drop
- As surface area increases - nucleation suppressed
- Other elevated larger nuclei near 0.6 km on descent suggest earlier nucleation



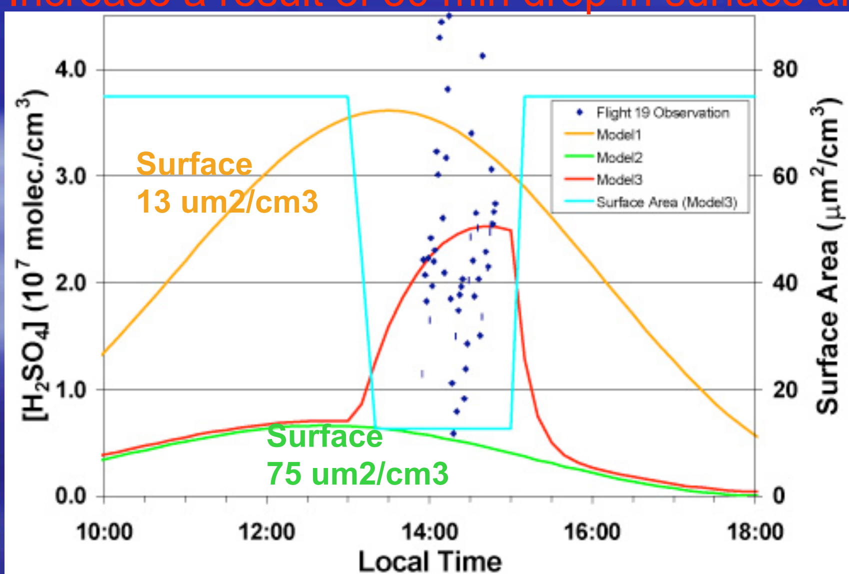
Clarke, A.D., et al., SCIENCE, Oct 2., 1998.

Model Predicts High SO2 and OH just after Noon

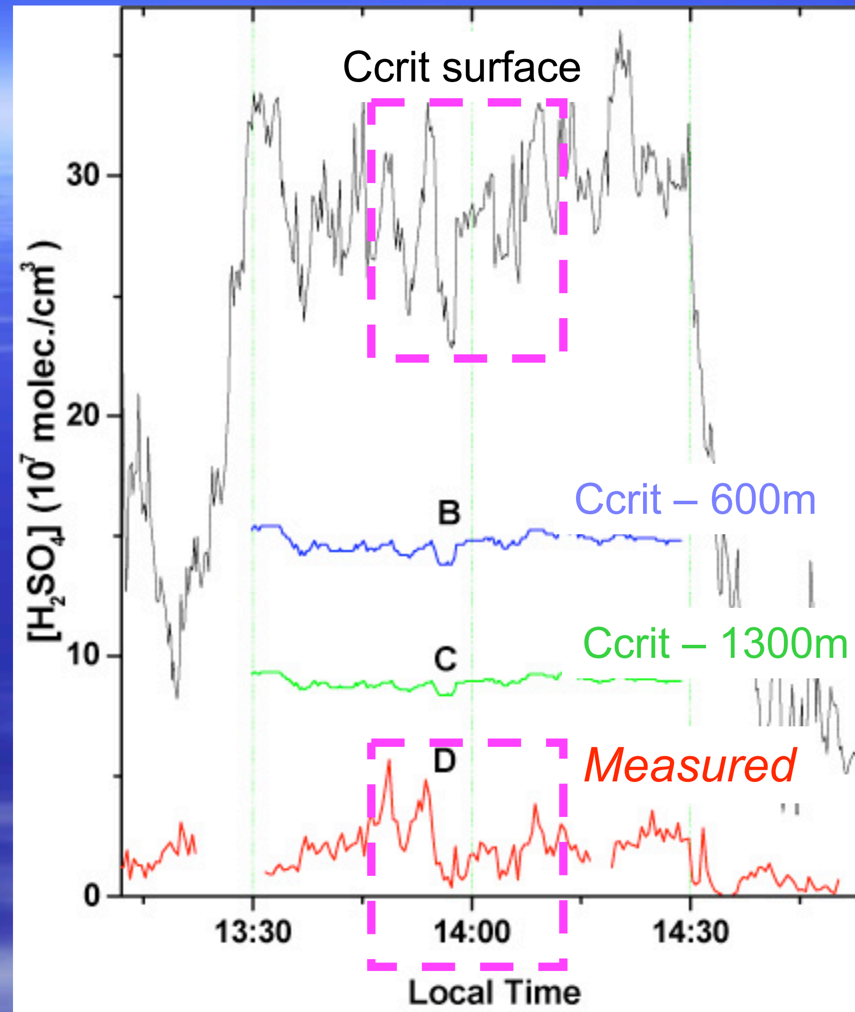


Model sulfuric acid (red line) achieves highest measured concentrations in cloud scavenged air

Increase a result of 30 min drop in surface area



$C_{crit} = -.16 \exp(0.1T - 3.5RH - 27.7)$;

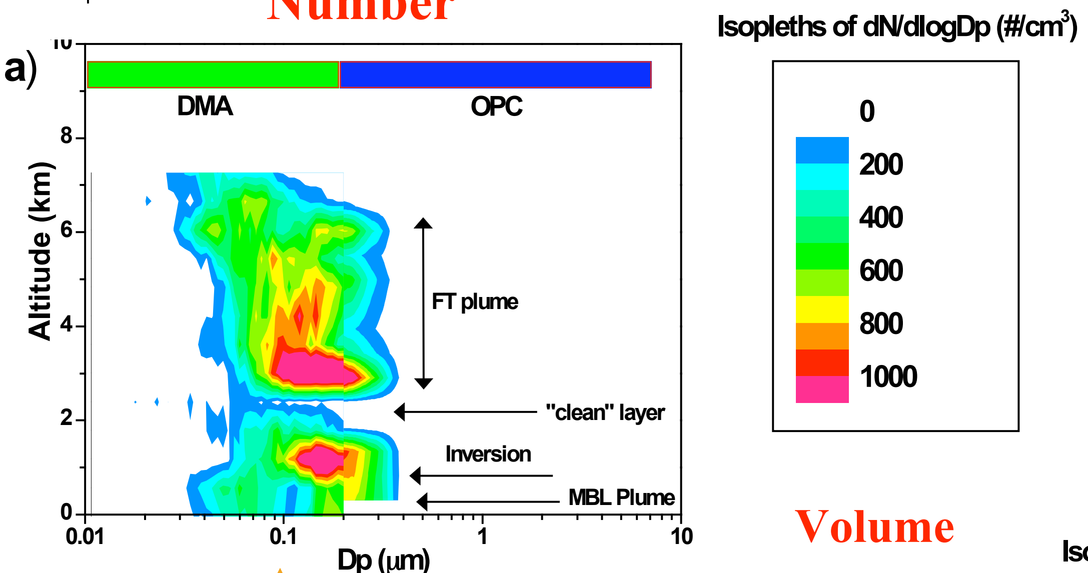


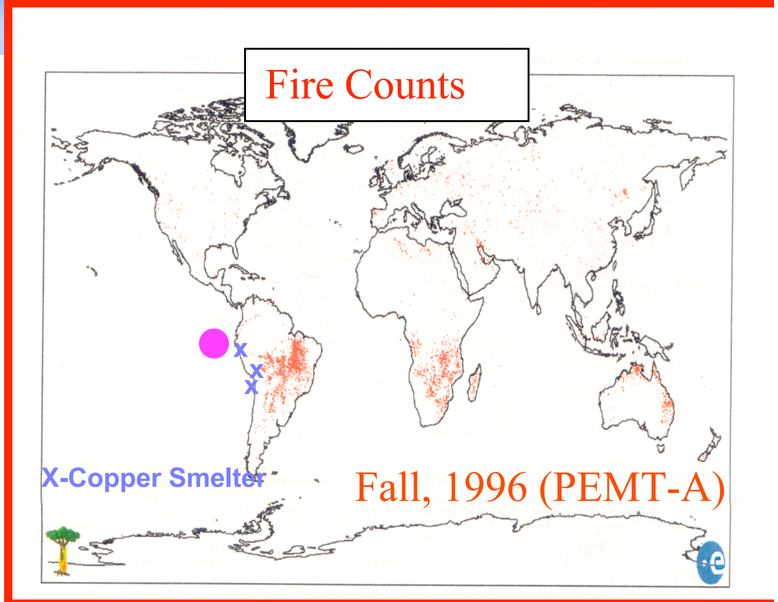
Measured sulfuric acid much lower than values needed for nucleation at surface (A) but closer to values needed aloft near cloud (C)

POLLUTED AIR MASSES

PEMT A - South American Outflow - Fall 1996

Number



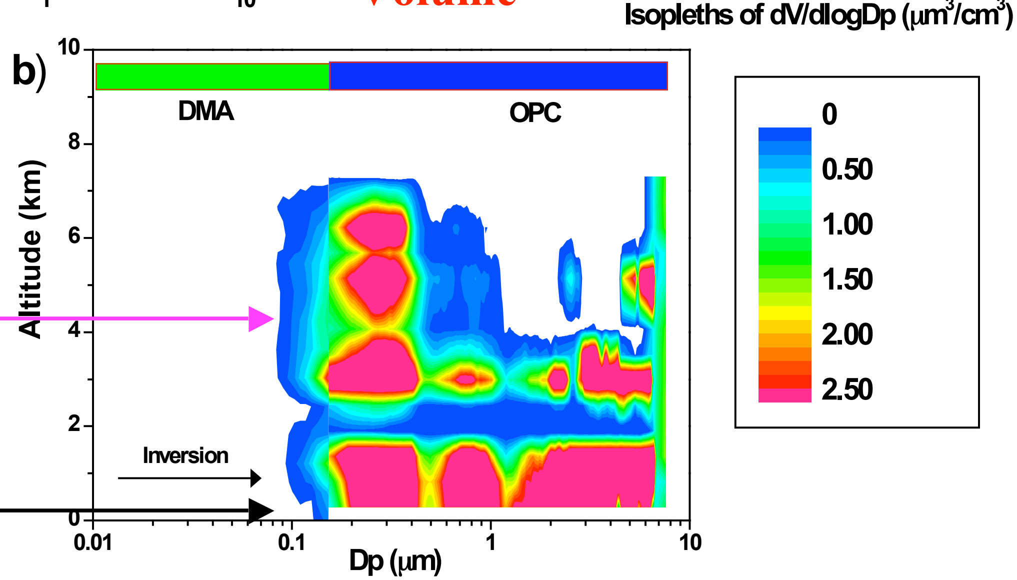


Volume

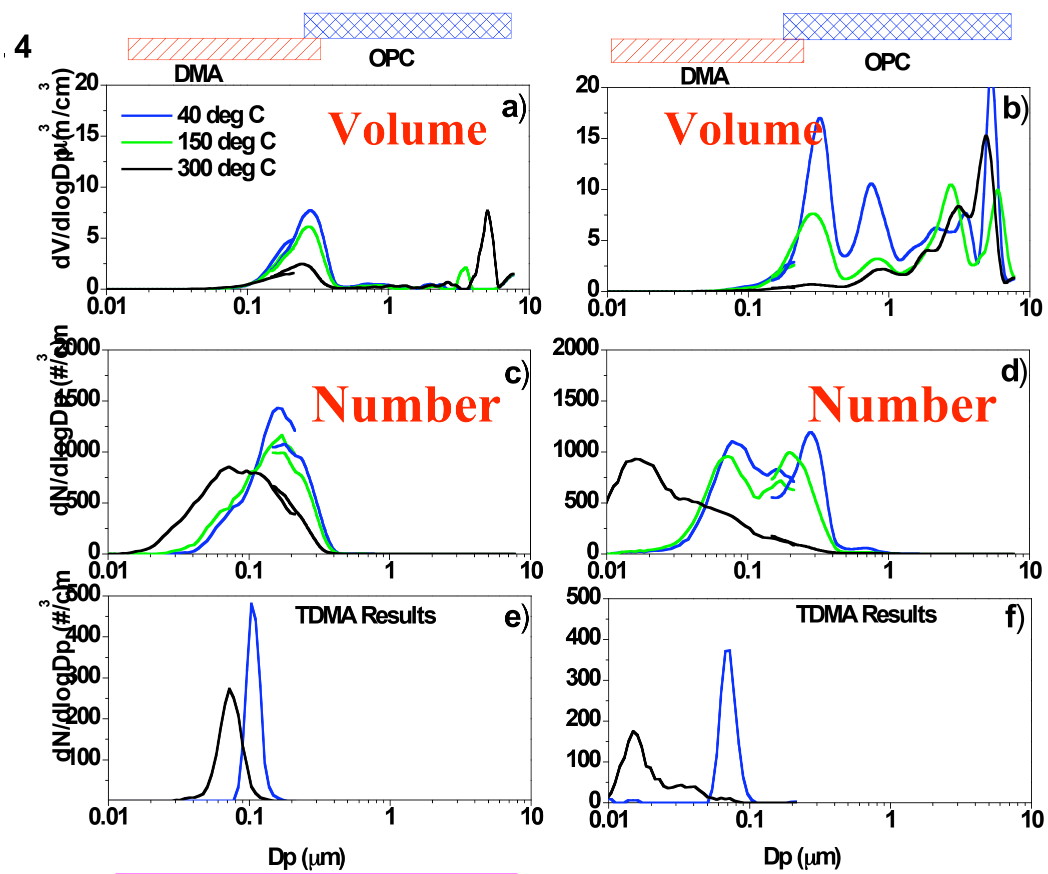
Hopple Minimum

Amazon Biomass Burning Aloft

Coastal Anthropogenic Pollution Above and Below Inversion



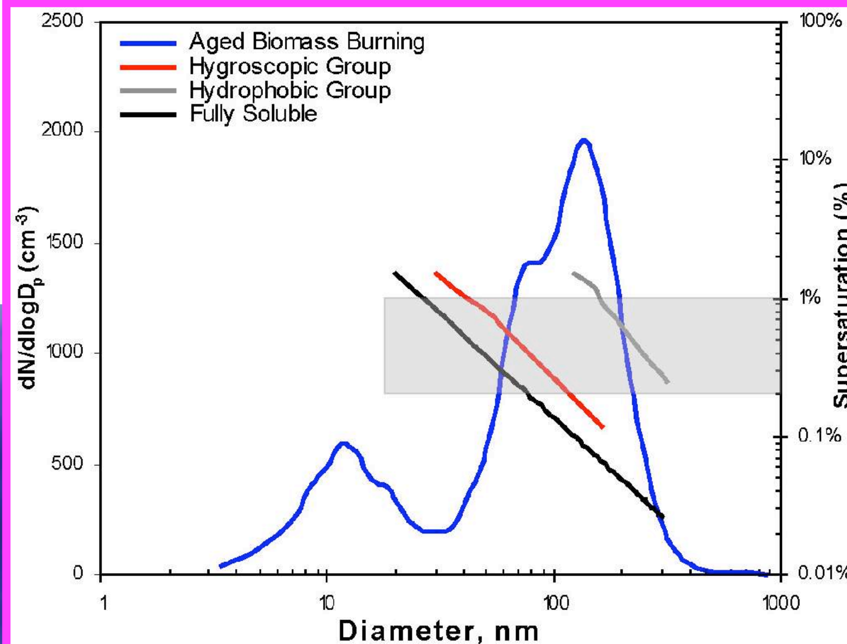
PEMT A - South American Outflow "Polluted"



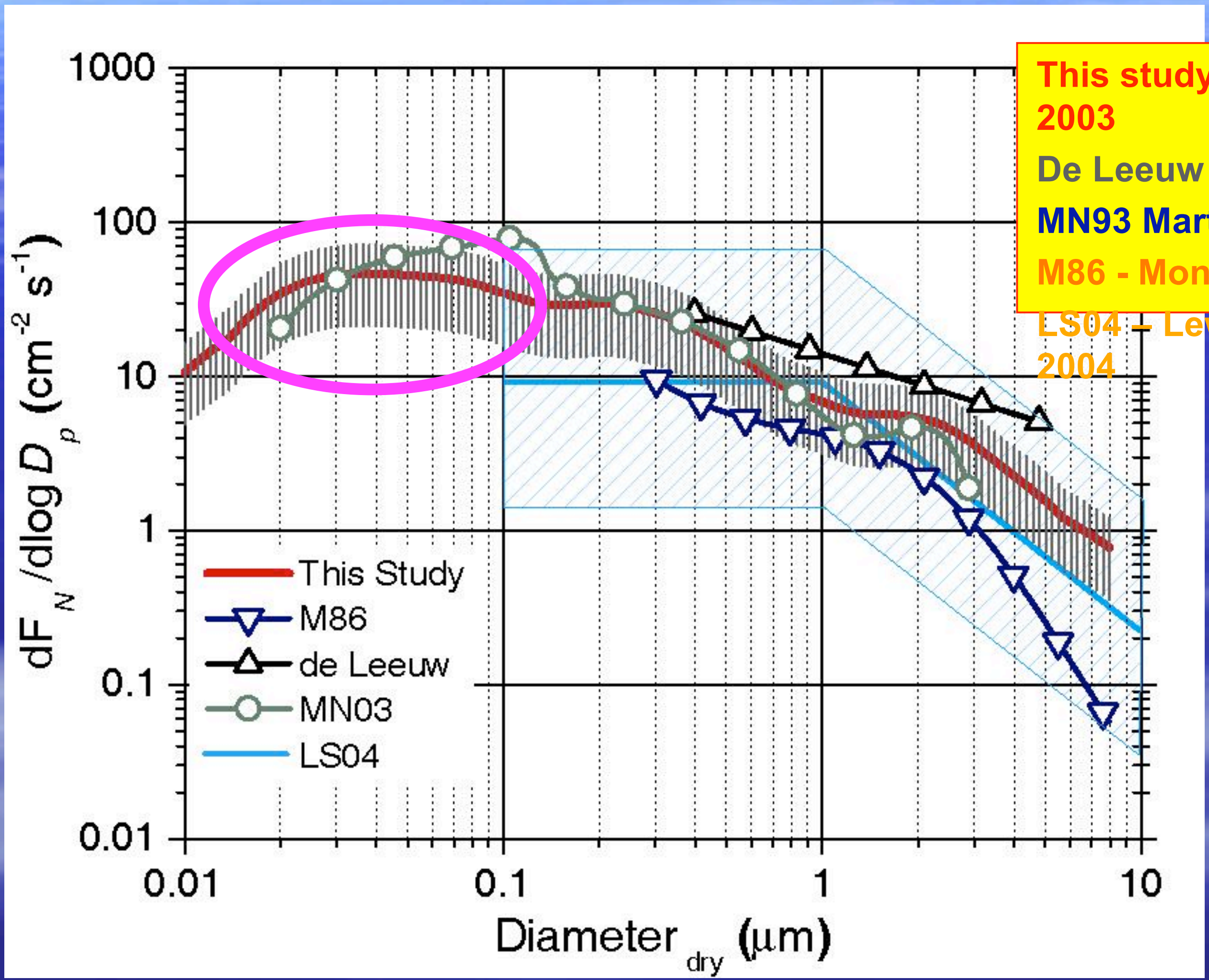
Amazon Biomass Burning Aloft

Coastal Anthropogenic Pollution Below

Size and composition have strong influence on activation as CCN



WHAT ABOUT SEA-SALT?
Our New Sea-Salt Source Function (# /cm2/s) at 9 m/s whitecaps using nominal Monohan whitecap coverage



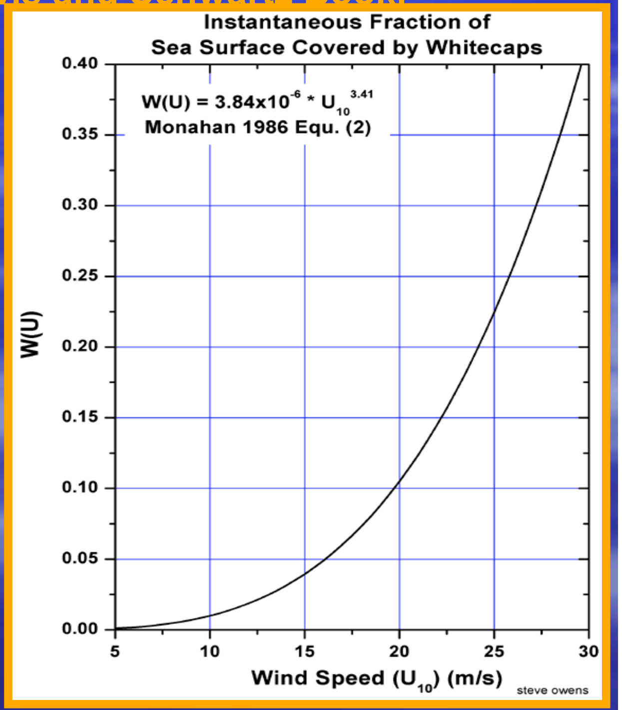
This study - SEAS coastal ocean 2003

De Leeuw – coastal 2001

MN93 Martensen wave tank 2003

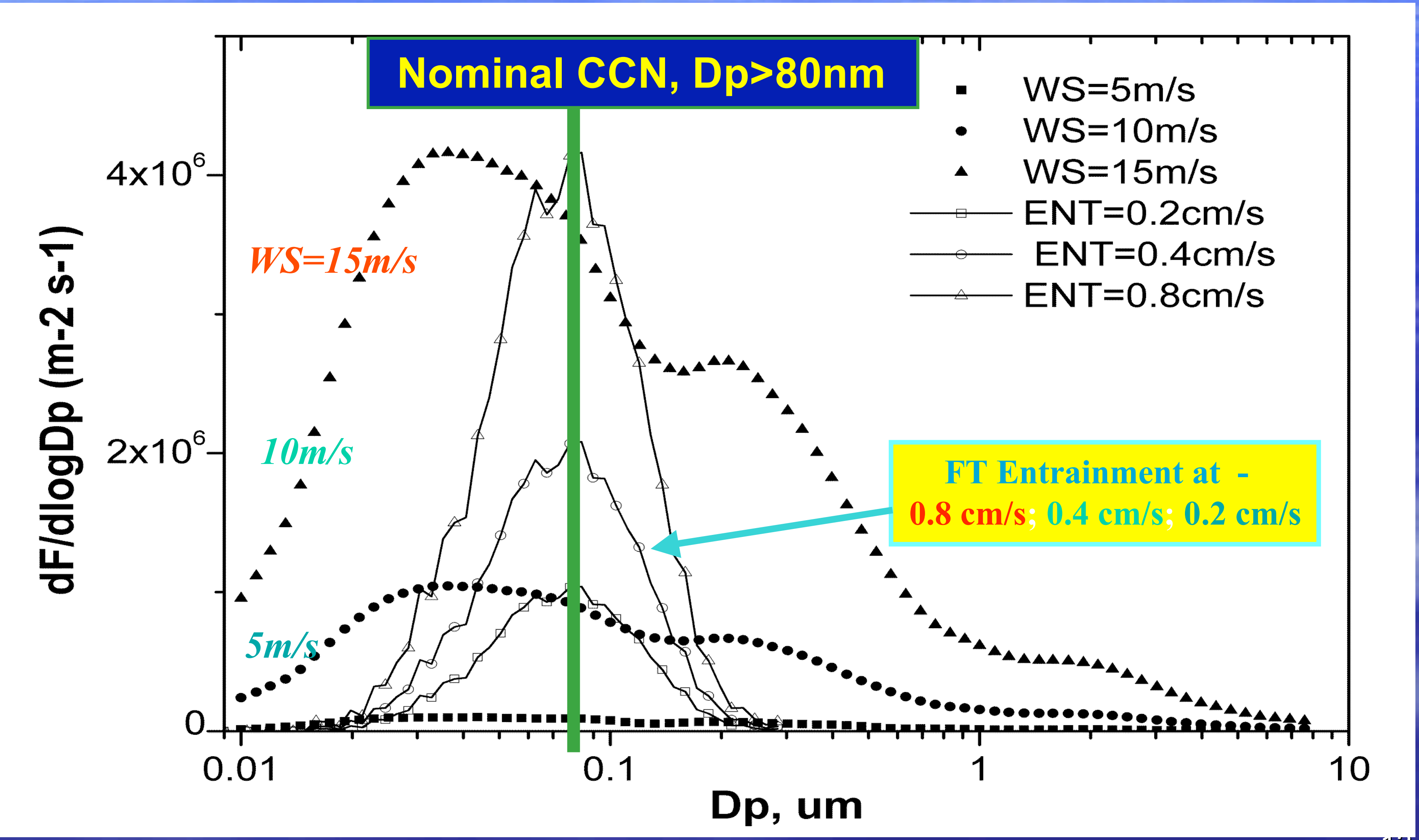
M86 - Monahan wave tank 1986

LS04 – Lewis and Schwartz Book, 2004



Clarke, A., S. Owens and J. Zhou, An ultrafine sea-salt flux from breaking waves: Implications for CCN in the remote marine atmosphere, J. Geophys. Res., 111, D06202, doi:10.1029/2005JD006565.

Comparison of Nominal SS Surface flux and FT Sulfate Flux for various Wind Speeds and Entrainment Rates



Estimated % contribution of Sea-Salt to total CCN flux > 80nm for various wind speeds and entrainment rates

Wind Speed > / FT Entrainment	5 ms^{-1} ($6\ cm^{-2}s^{-1}$)	10 ms^{-1} ($64\ cm^{-2}s^{-1}$)	15 ms^{-1} ($254\ cm^{-2}s^{-1}$)
0.2 $cm\ s^{-1}$ ($23\ cm^{-2}s^{-1}$)	20 %	73 %	91 %
0.4 $cm\ s^{-1}$ ($46\ cm^{-2}s^{-1}$)	11%	58 %	84 %
0.8 $cm\ s^{-1}$ ($92\ cm^{-2}s^{-1}$)	6 %	41 %	73 %

Summary of Potential Sources

- MBL Nucleation [Daylight] - appears favored near top of scavenged MBL (POC?) and growth rates are faster (for fixed source strength) for reduced surface area. Growth to CCN still slow (days) and slower as surface area increases.

  Fully soluble and effective as CCN

  [care needed to remove apparent fresh nuclei caused by droplet shatter]

- Clean FT entrainment [Day and Night] - typical concentrations can replenish CCN size nuclei in MBL in 2-4 days under clean conditions.

  Fully soluble and effective as CCN

- Polluted FT entrainment [Day and Night] – pollution may be possible this time of year from biomass burning (above inversion) and from coastal pollution.

  Partially soluble (pollution) to less soluble (biomass burning)

- Sea-Salt production - an effective source but depend on wind speed over about 7m/s (can be localized near clouds) or precipitation falling on surface.

  Fully soluble and effective as CCN.

Putting This SSA flux in GCM model GISS II – prime

Models SSA concentrations and not just fluxes
Allows interaction with sulfur cycle *(Adams and Seinfeld, 2002).*

Global Evaluation of CCN Formation by Direct Emission of Sea-Salt and Growth of Ultrafine Sea-Salt

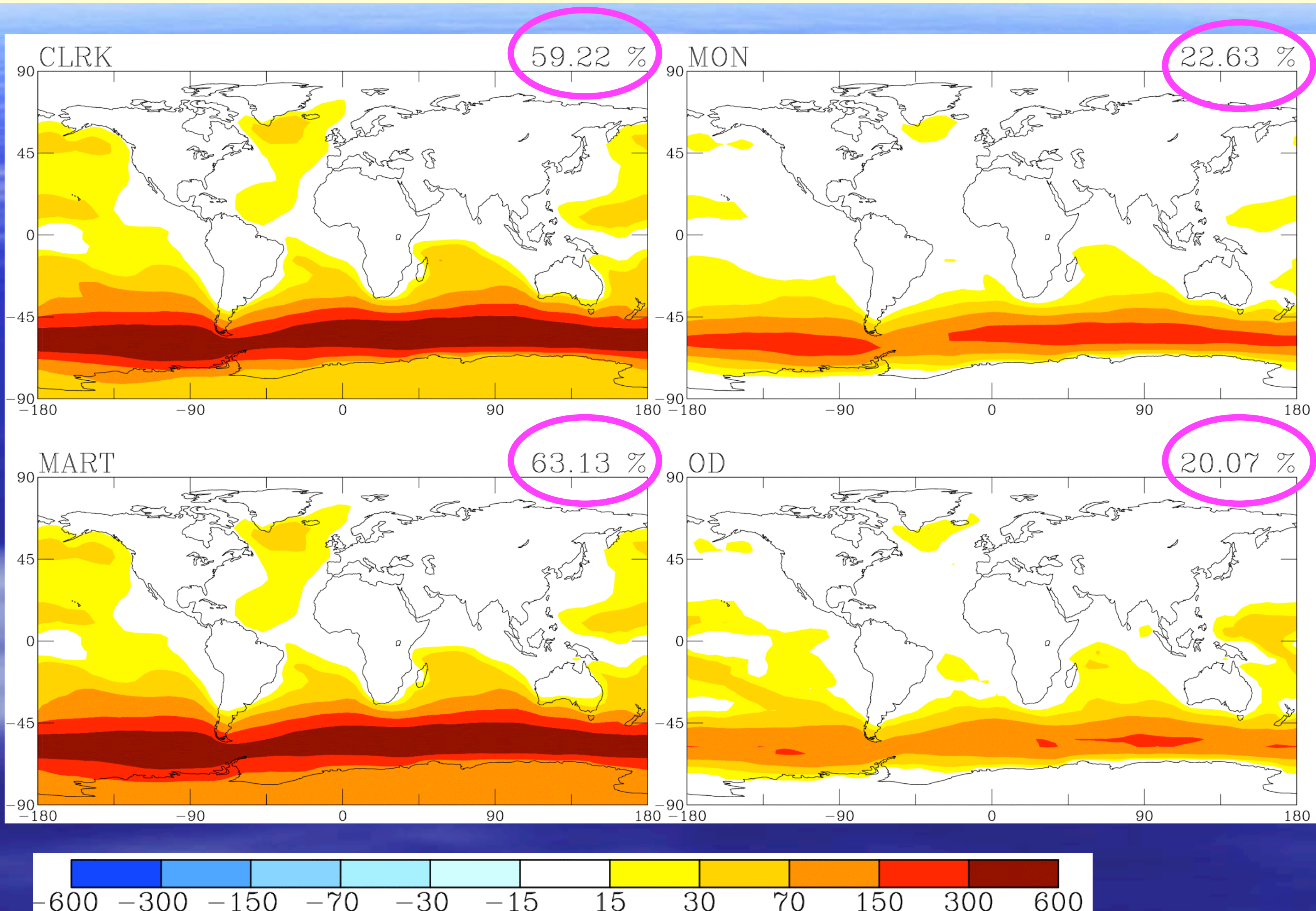
Jeff Pierce and Peter Adams

JGR-Atmospheres, 2006

Model Details

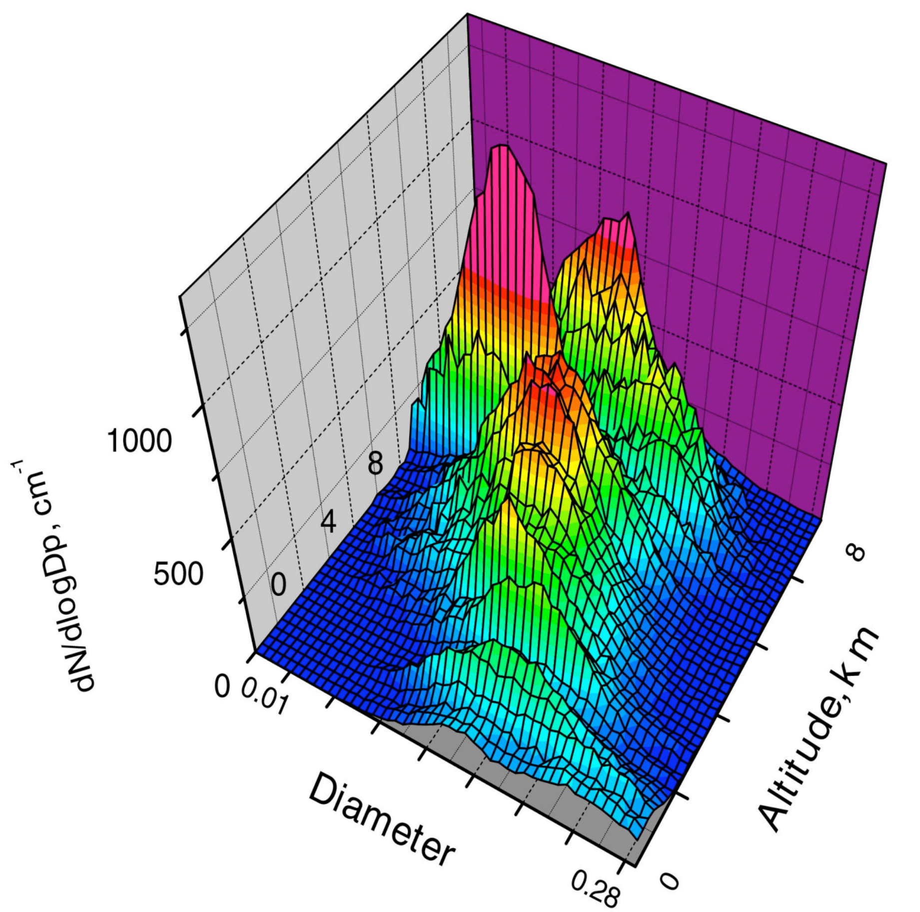
- NASA Goddard Institute for Space Studies GCM II-prime
  - 4° latitude x 5° longitude
  - 9 vertical layers between surface and 10 mb
  - One hour tracer time step
- Two Moment Aerosol Sectional (TOMAS) aerosol microphysics algorithm
  - Explicitly calculates both aerosol number and mass in each size section
  - 30 size sections (~10 nm - ~10 μm)
  - Sea-salt and sulfate aerosols only

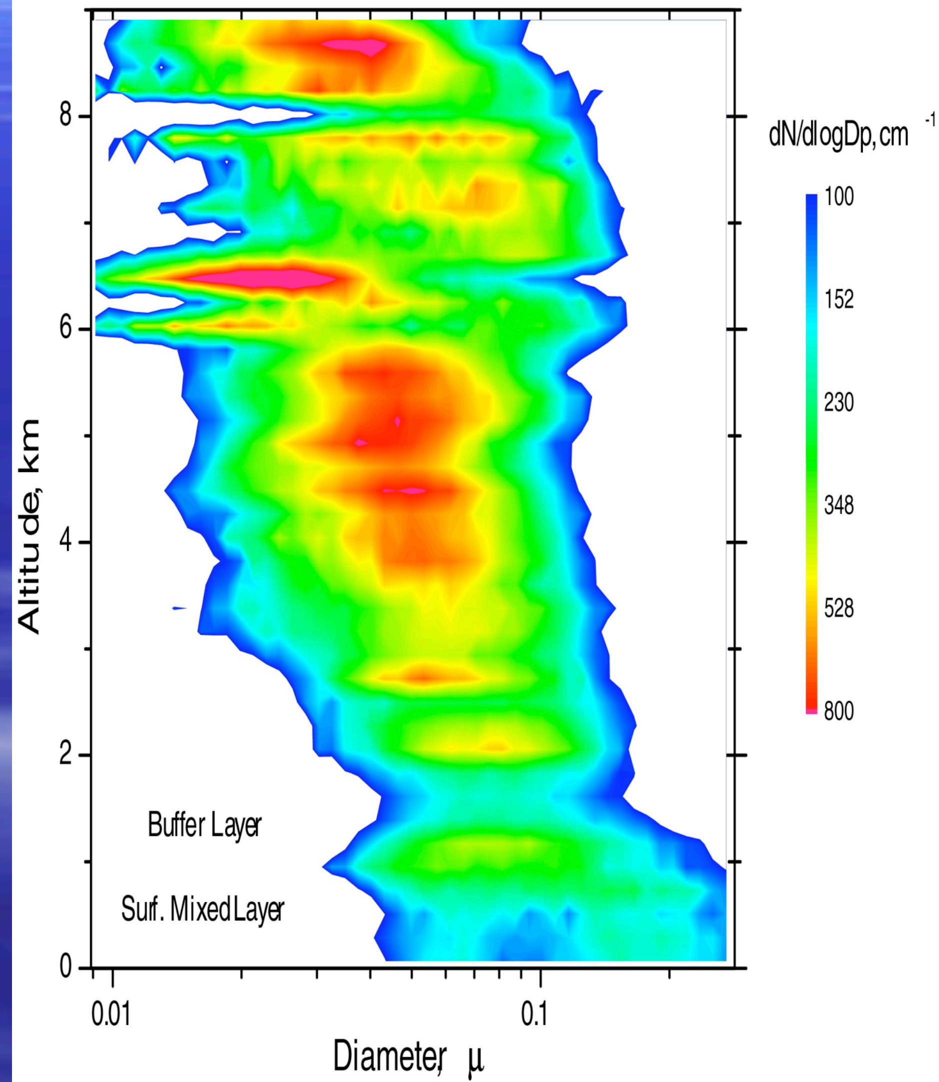
Sea-salt impact on cloud condensation nuclei
All sea-salt CCN(0.2%) - Percent Change relative to sulfate



3-D and 2-D Representations of Size Distribution Growth During Subsidence from Average of 6 PEMT-B Profiles in Tropics.

Note development of 90nm minimum through cloud processing in the MBL



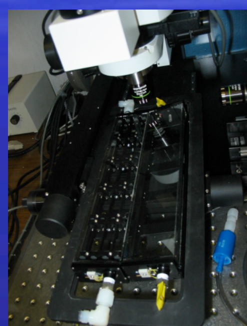


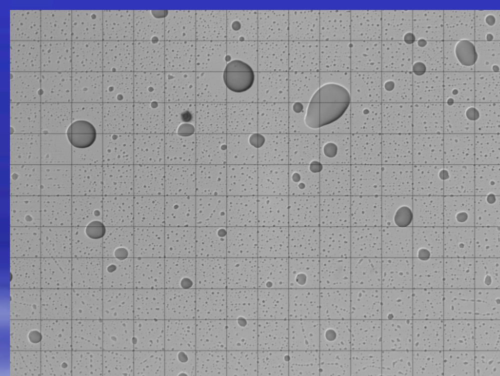
Giant sea salt aerosols

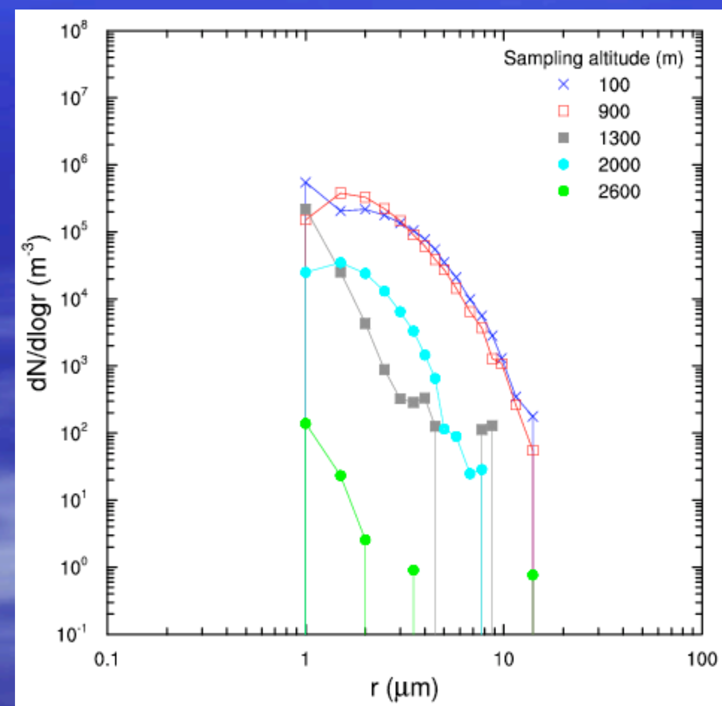
Jorgen Jensen

Are they important for precipitation?
- **In well-mixed stratocumulus**
- **In open-cell cumulus**

Does the open-cell structure lead to changes in sea-salt aerosol amount and size distribution?
- **Due to precipitation scavenging**
- **Due to cold-air downdrafts modifying the surface wind strength**







GNI impactor and optical microscope to provide high sample-volume size distributions. Use as part of cloud model initialization.