

Aerosol Chemical Composition at ESRL

Emphases:

New capabilities

Careful calibrations

Process studies, mostly from aircraft

Build understanding of the entire atmosphere

Major instruments:

SP2: Single particle black carbon

Insights into climate forcing

AMS: Bulk, size-resolved composition

Fast, quantitative data to build correlations

PALMS: Single particle composition

Fundamental understanding of particles



Aircraft



UND/NASA DC-8

0.2-12 km

PALMS



NASA WB-57

5-19 km

PALMS, SP2



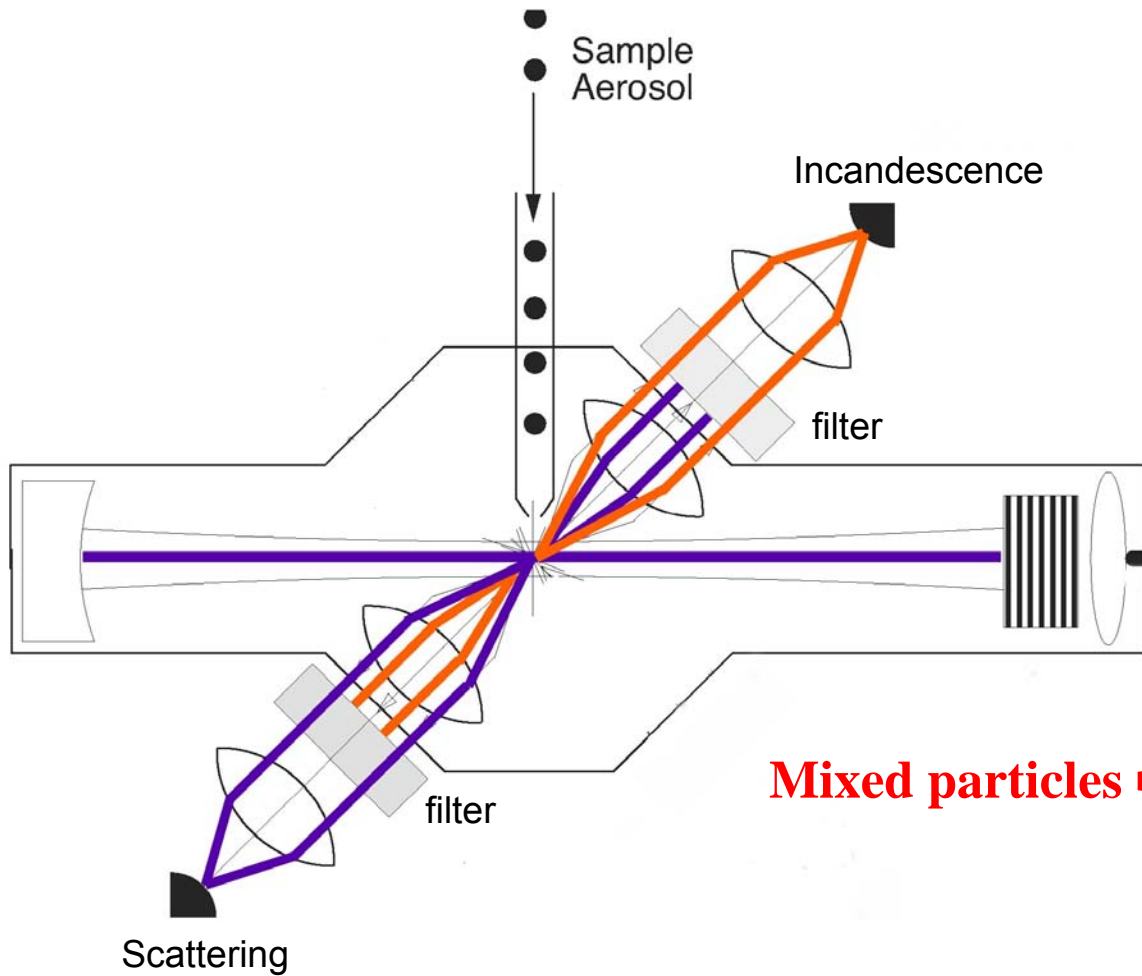
NOAA P-3

0.1-7 km

AMS, PALMS, SP2

Also many gas-phase instruments on each aircraft

Soot Photometer (SP2) Instrument

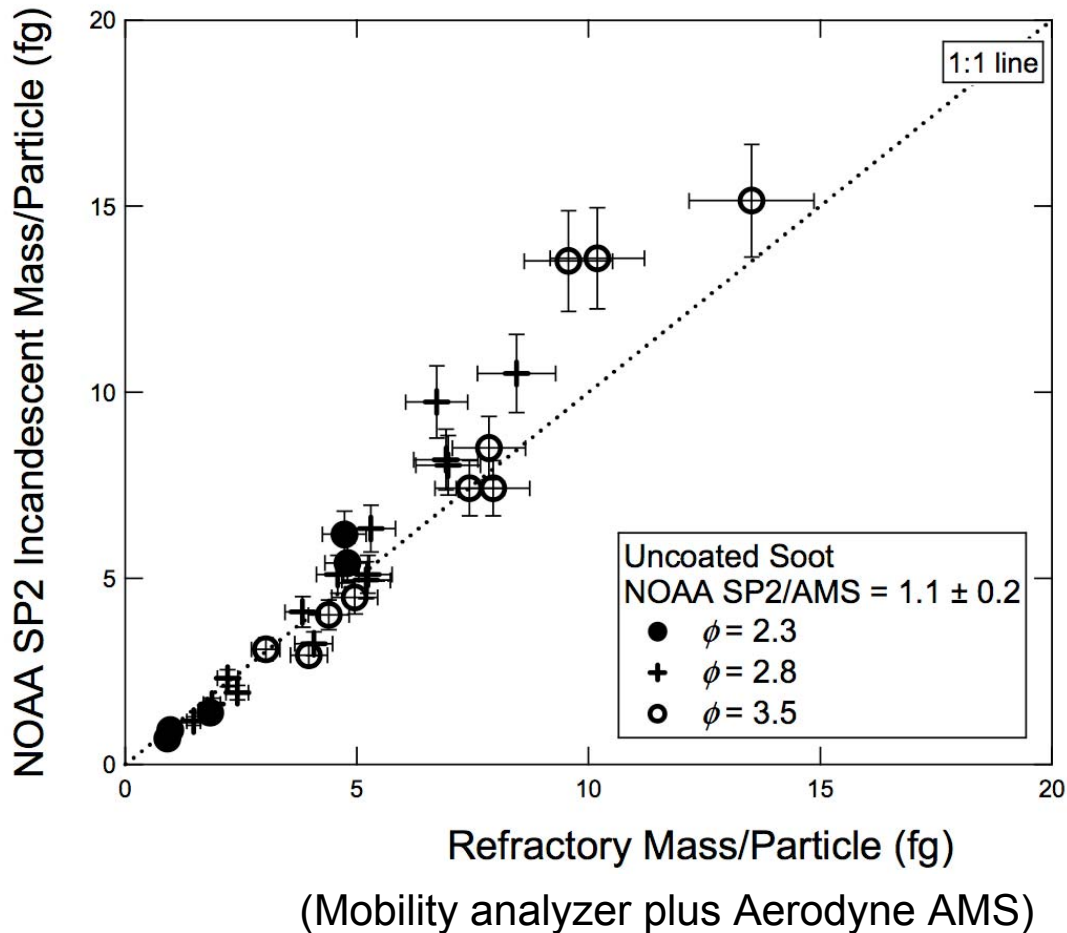


- Particles pass through intense laser beam
- Black particles heat to incandescence. Pure scatterers don't.

Improvements:

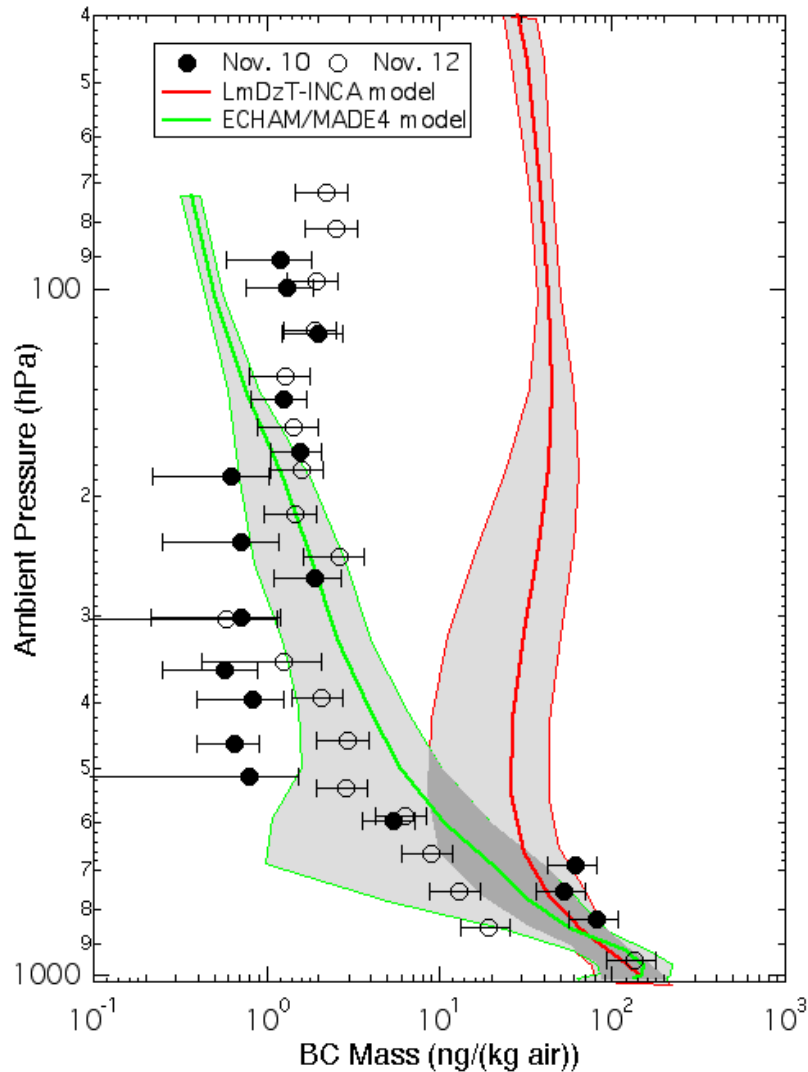
- 1) Scatter signal hardware
- 2) Data processing
- 3) Aircraft operation

SP2 Calibration



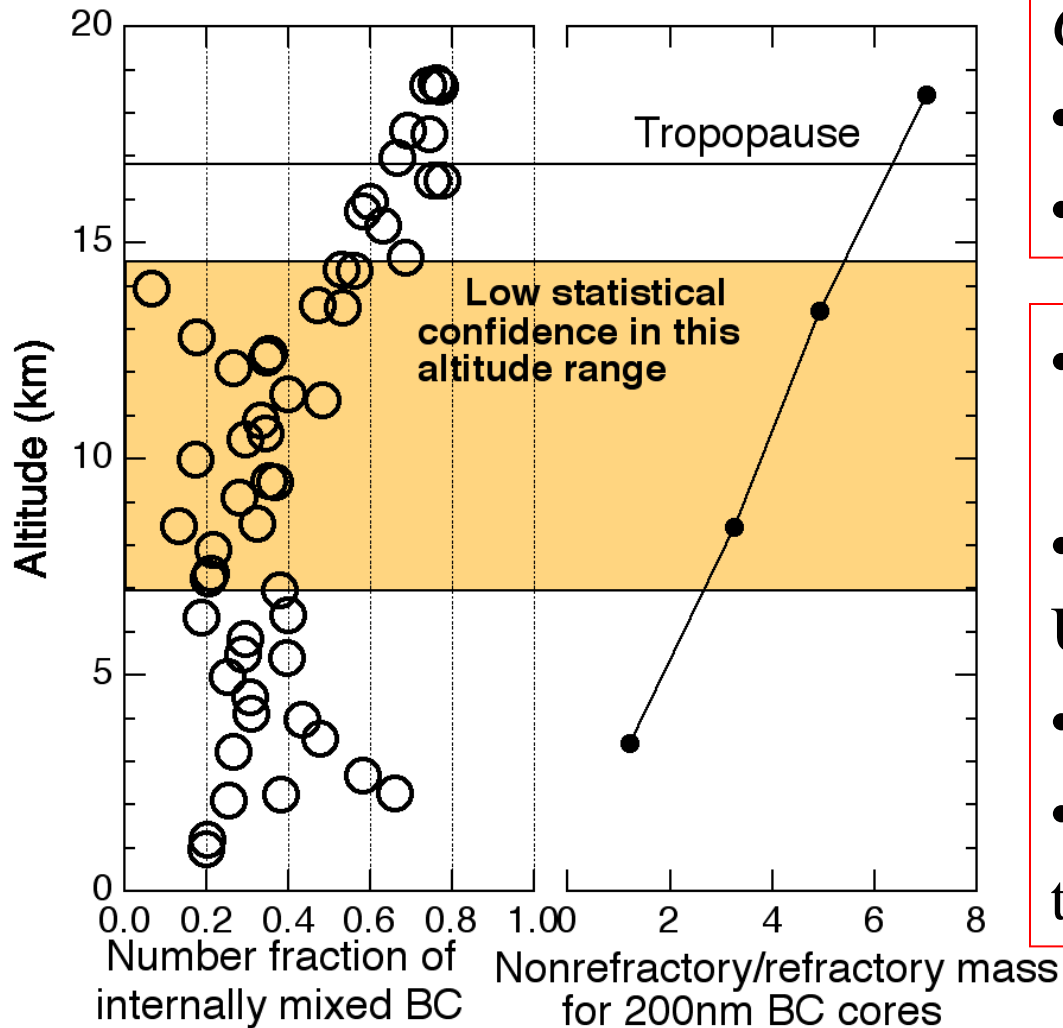
- SP2 response is independent of soot morphology
- Good comparison to other techniques

SP2 Black Carbon Vertical Profile



- Here:
 - NASA WB-57F
 - Two flights, Nov. 2004
 - Over south-central US
 - Accuracy of 30%
- New measurements constrain global models
- Mass-based detection compares well to emission inventories

New Capability: Black Carbon Mixing State



Coatings on black carbon:

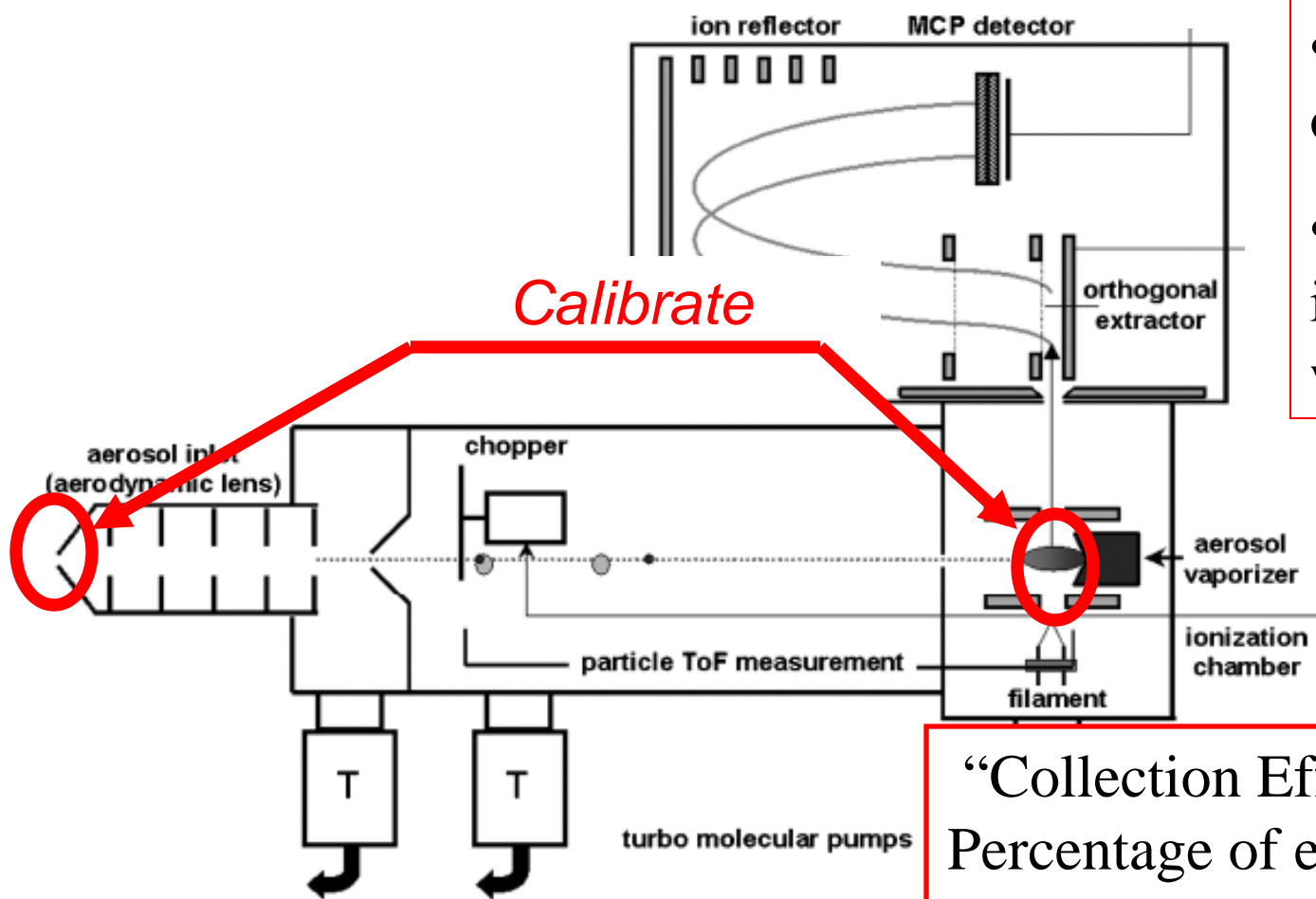
- Enhance absorption
- Influence lifetime

• Here:

3 flights; Costa Rica in January

- Most black carbon in the UT/LS was coated
- More variable lower down
- High altitude particles had thicker coatings

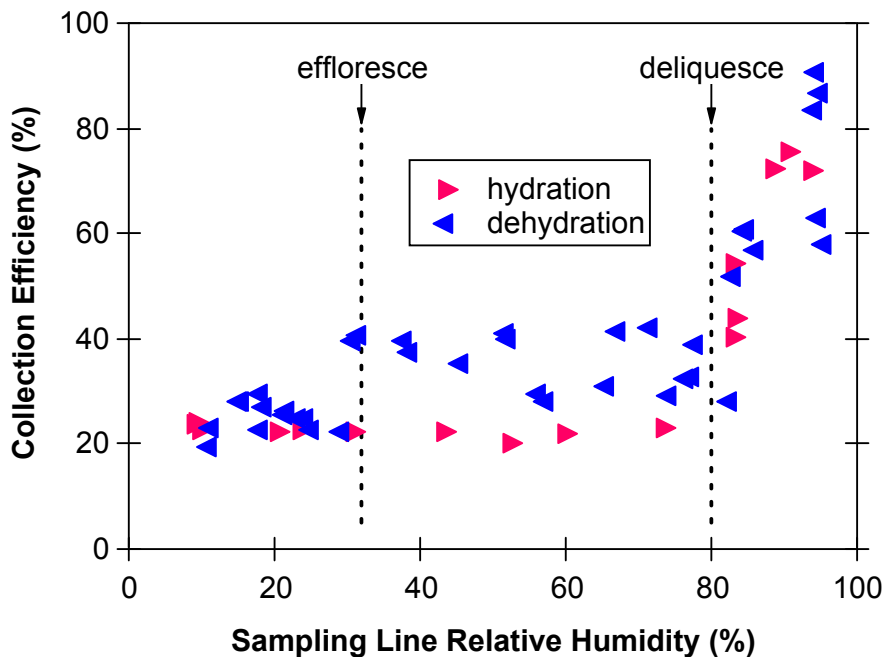
Aerosol Mass Spectrometer (AMS)



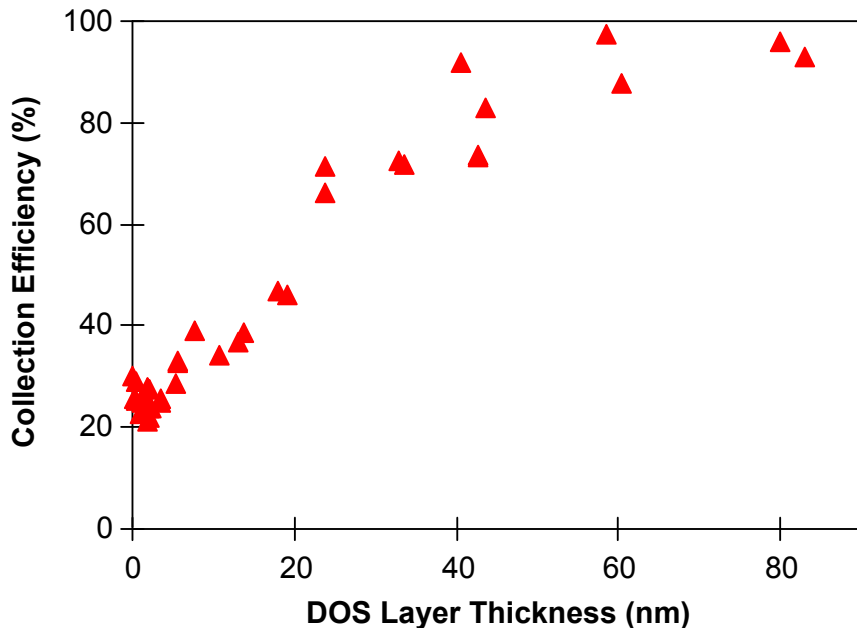
- C-TOF version on NOAA P3
- Over 50 similar instruments worldwide

“Collection Efficiency”:
Percentage of entering particles that hit and evaporate on hot target

AMS calibration

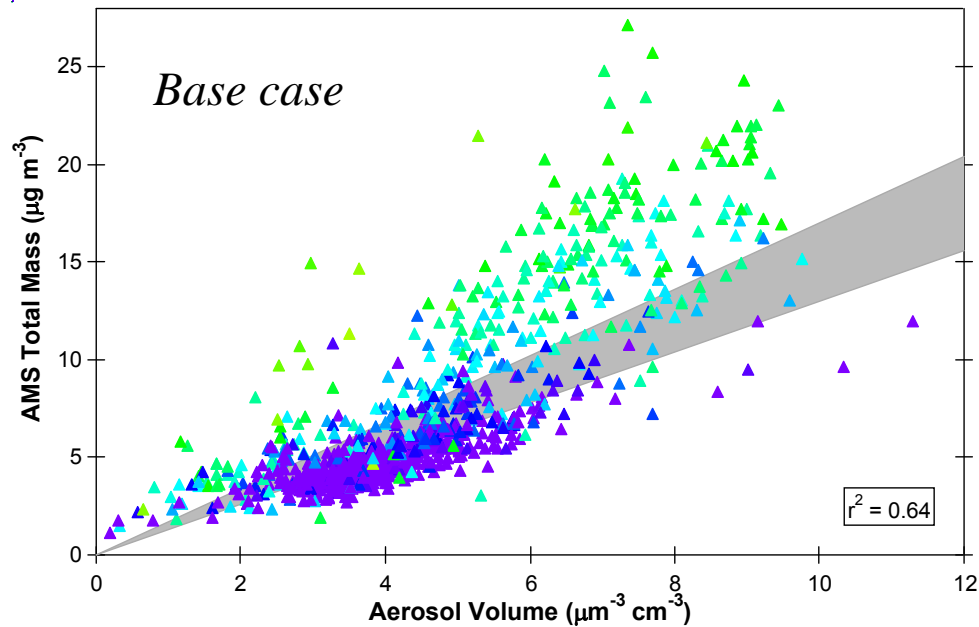


Here: 200 nm ammonium sulfate

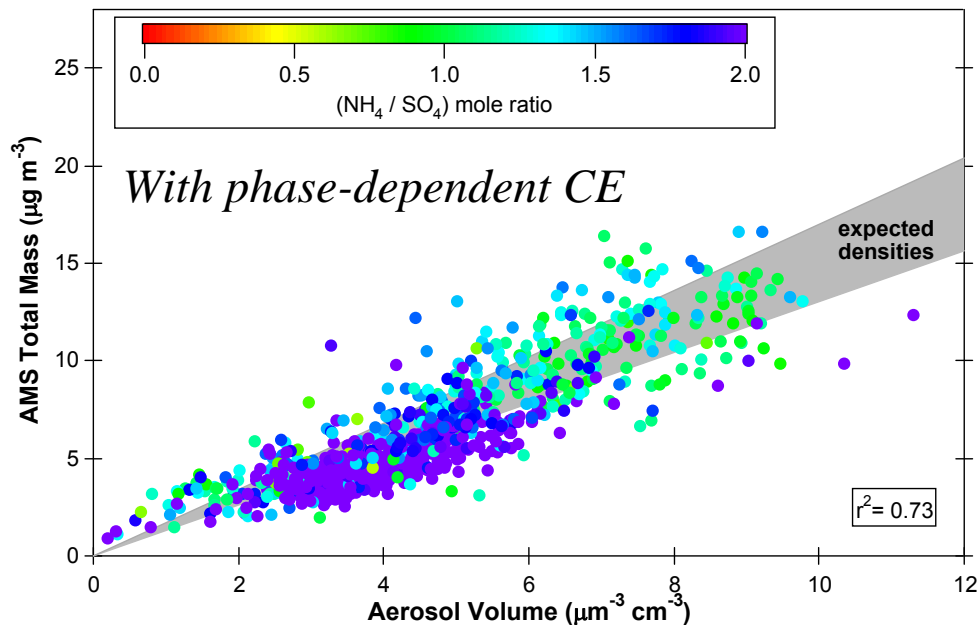


- Phase (liquid or solid) changes AMS collection efficiency up to a *factor of 4*
- Measured mass loadings depend directly on collection efficiency.
- CE often assumed constant $\approx 50\%$

Apply to field data: (2006 Houston TexAQS)



Using $\text{CE}=0.5$ for all data results in large scatter, systematic changes with acidity.

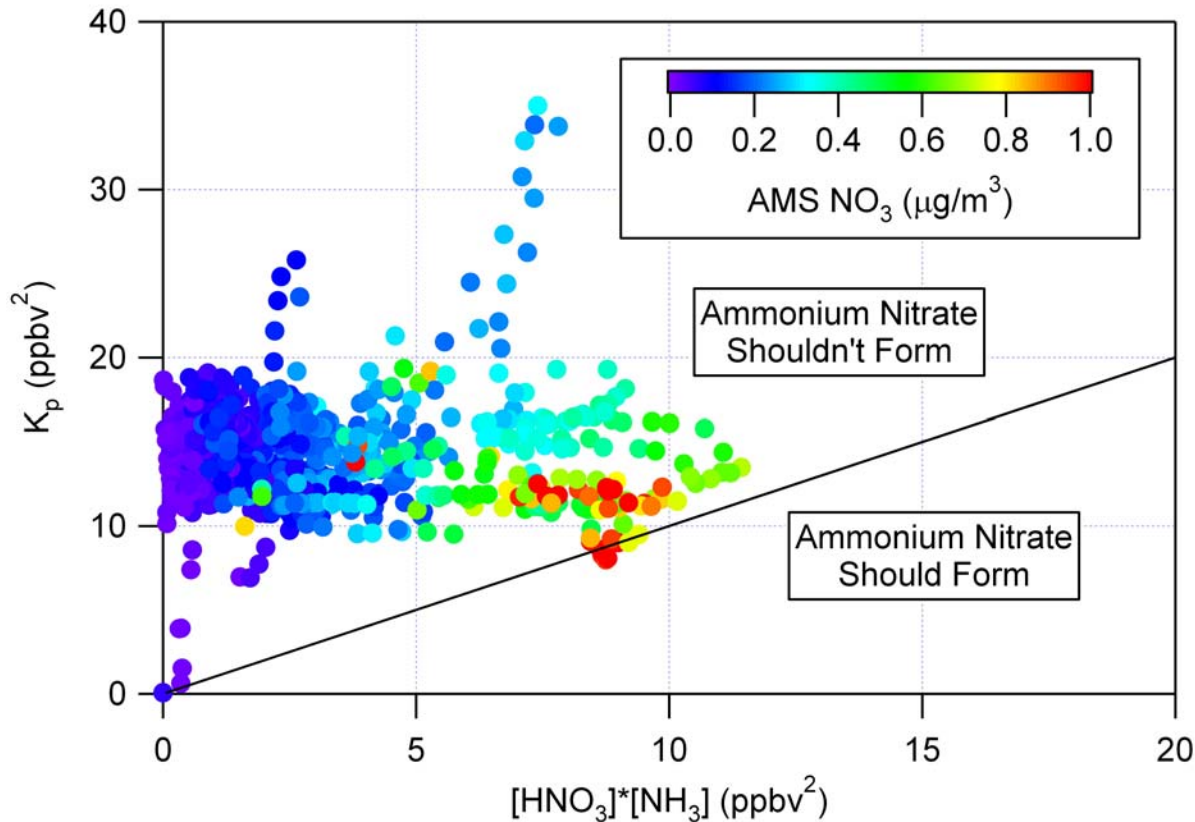


Using phase-dependent CE reduces scatter and systematic errors.

Agreement between independent measurements of volume and mass. (!!!)

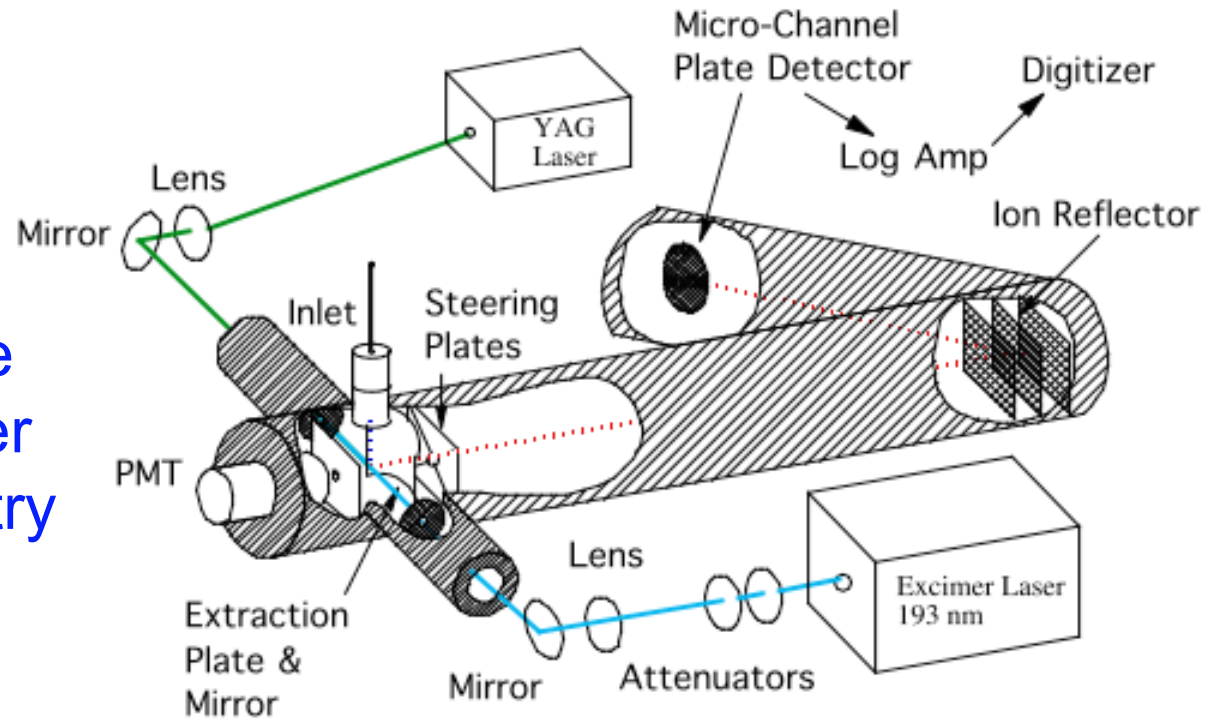
Ammonium nitrate

In the future, ammonium nitrate is likely to become more important for climate.



- Houston 2006
- Gas-phase HNO_3 , NH_3
Aerosol nitrate
- ✓ Excluded region
- ✓ More nitrate near stability region
- Work in progress

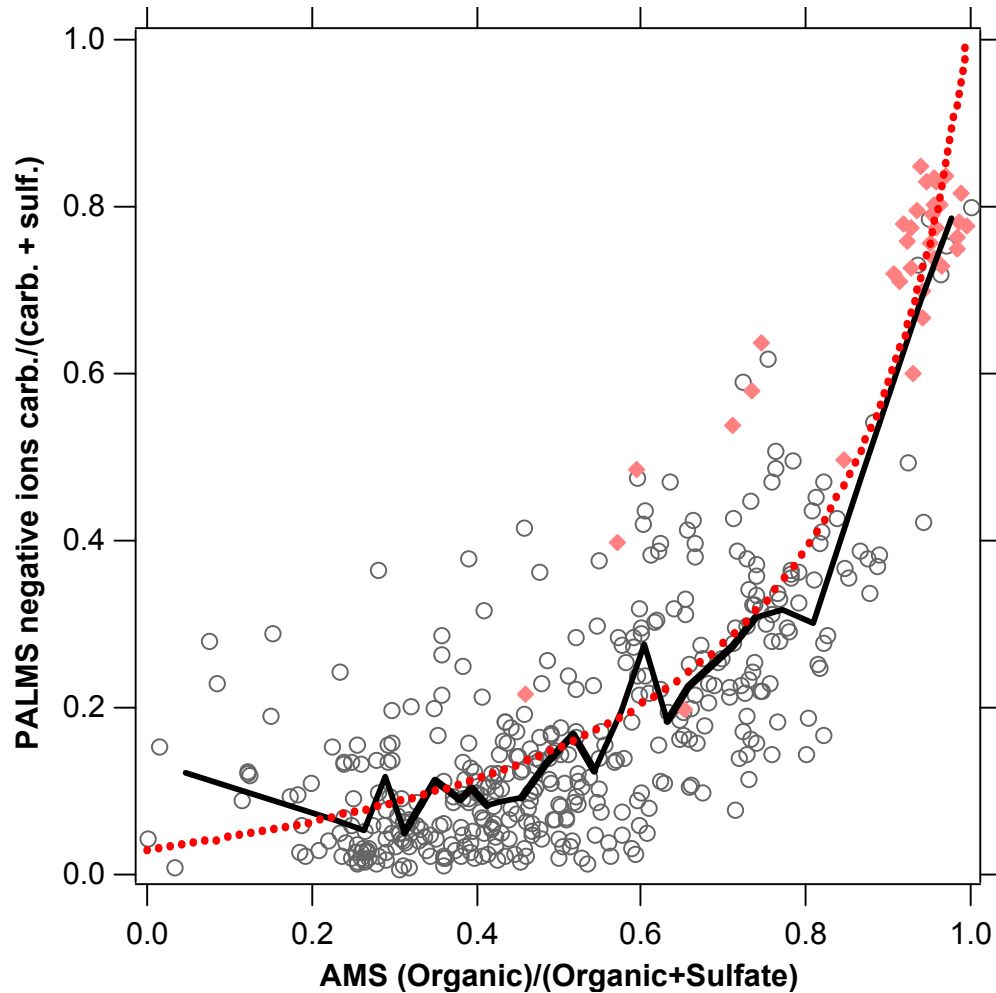
PALMS: Particle Analysis by Laser Mass Spectrometry



- 1) Particle enters vacuum. Trigger from light scattered from continuous laser.
- 2) Excimer laser beam hits particle.
- 3) Positive or negative ions analyzed with TOF mass spectrometer.
 - Size range about 0.25 to over 3 μm diameter

*Most of the mass and light scattering
Minority by number*

PALMS - AMS in-flight comparison

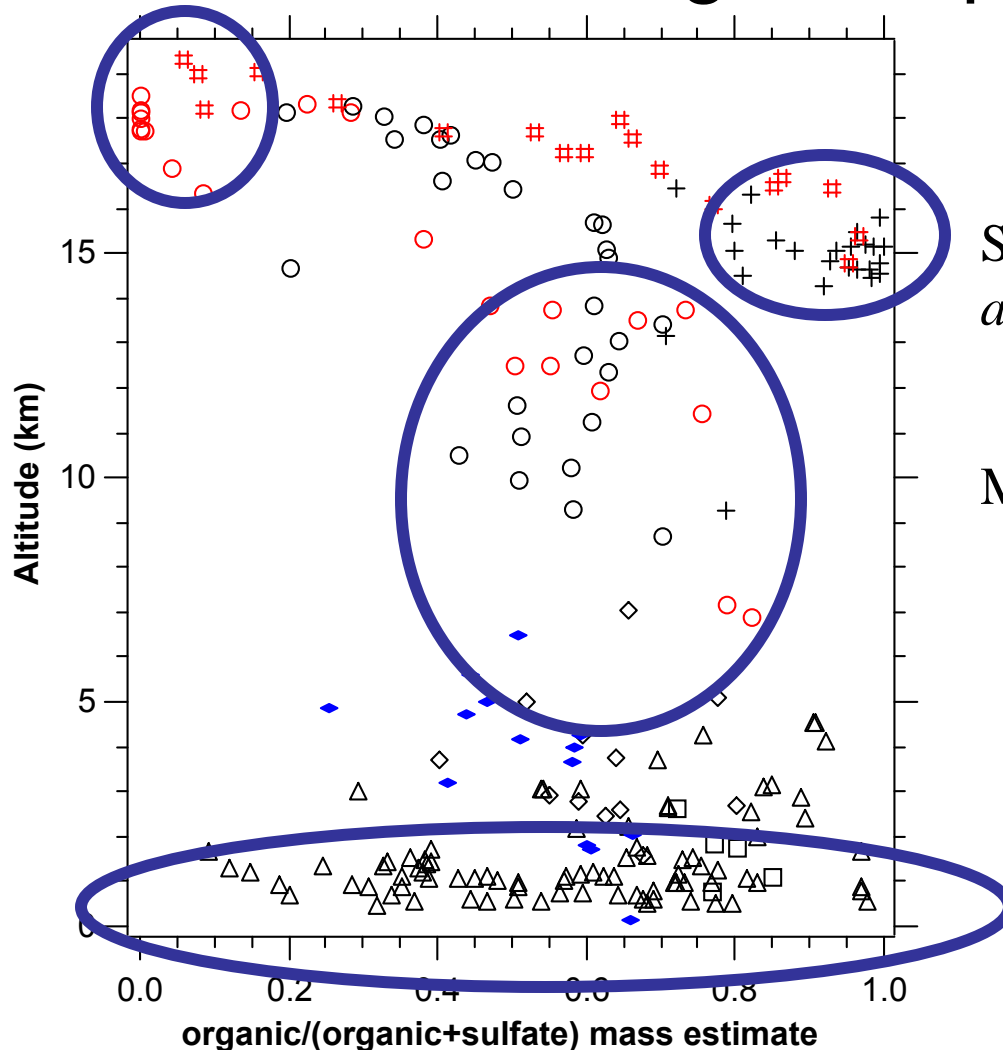


Challenge: Compare instruments without a good laboratory model for organic aerosols.

Solution: Use ambient data.

- Average (black) is captured by simple PALMS relative sensitivity (red)
- Biomass burning plumes identified from gas-phase acetonitrile (diamonds)

Average composition



The stratosphere

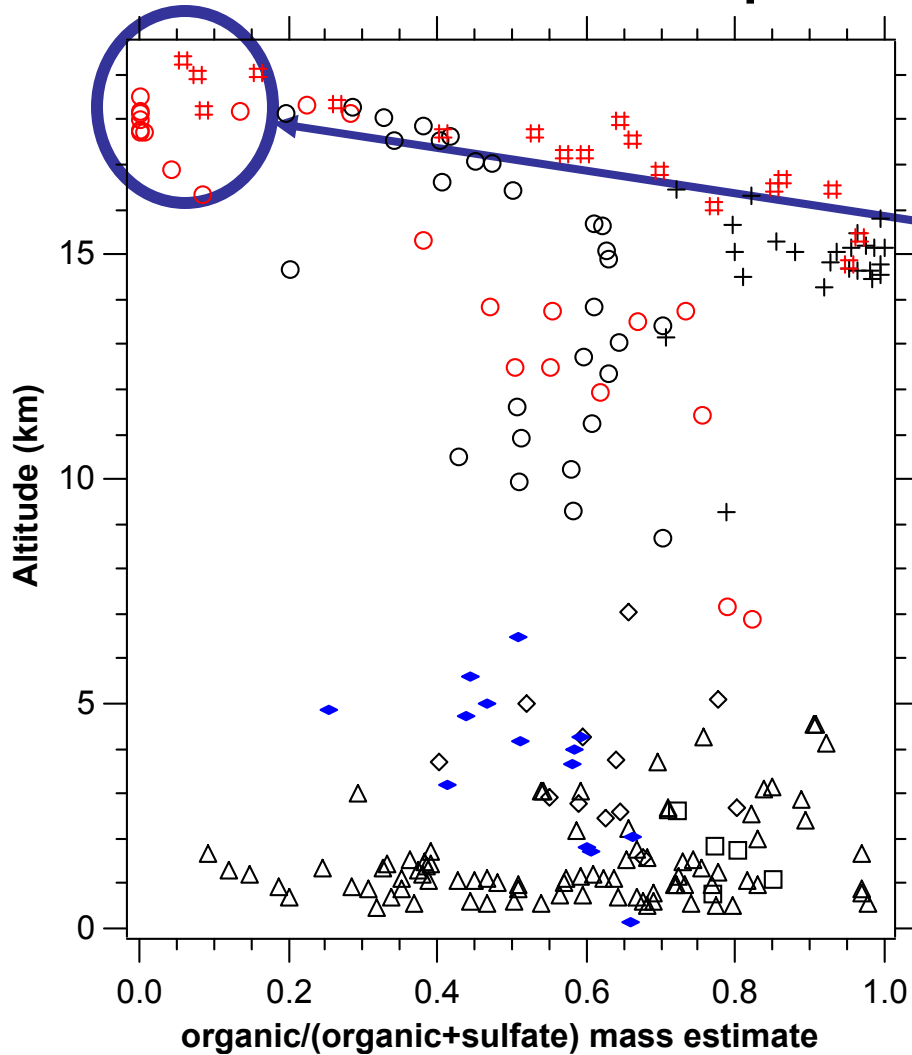
Smoke from forest fires
at 50000'!

Majority organics 20,000-45000'

Full range near the ground

pure sulfate <-----> pure organic

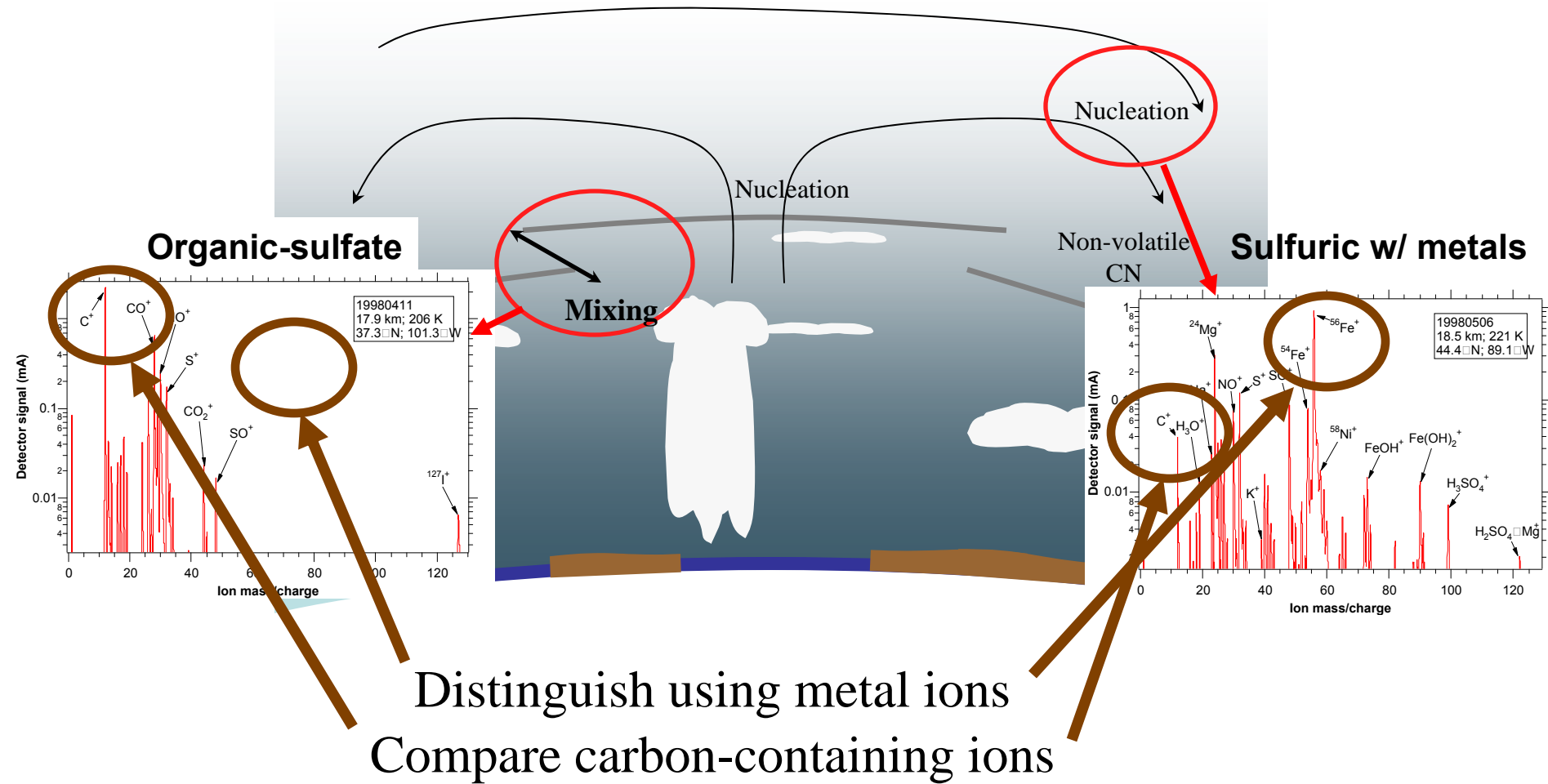
The stratospheric laboratory:



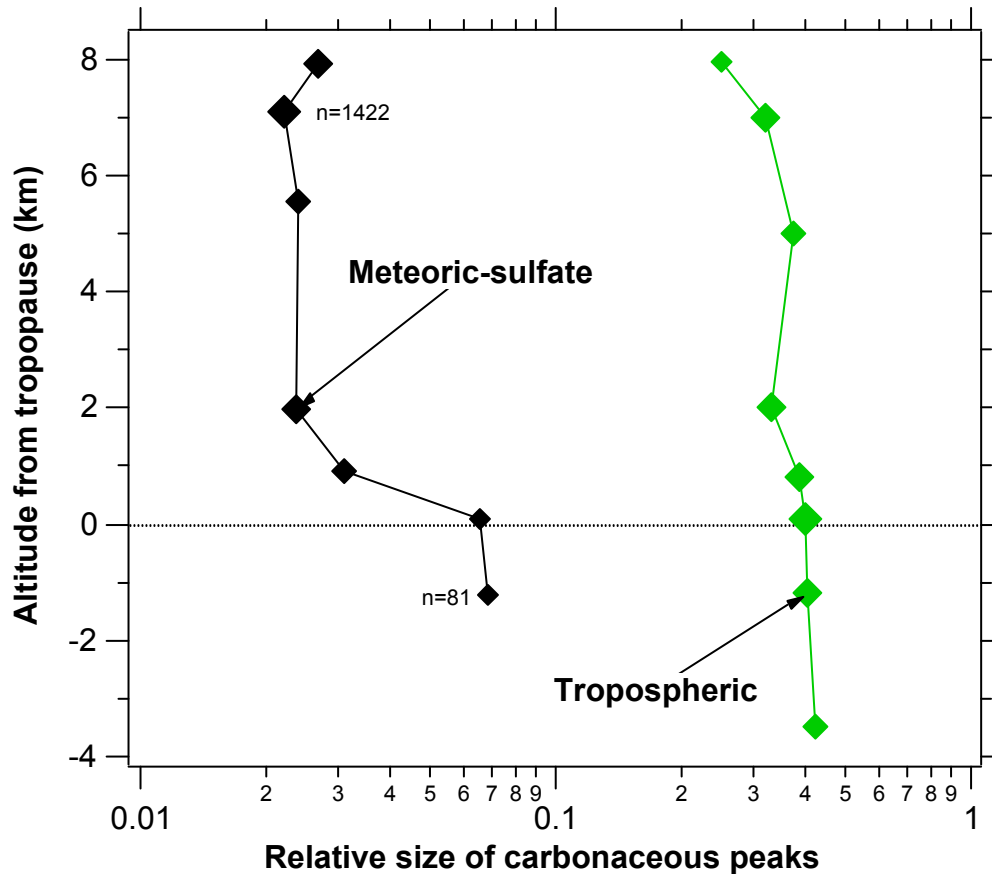
- >95% of particles fall into 3 categories
- Limited gas-phase organic chemistry
- Long residence time

=> Understanding of processes

Stratospheric particles



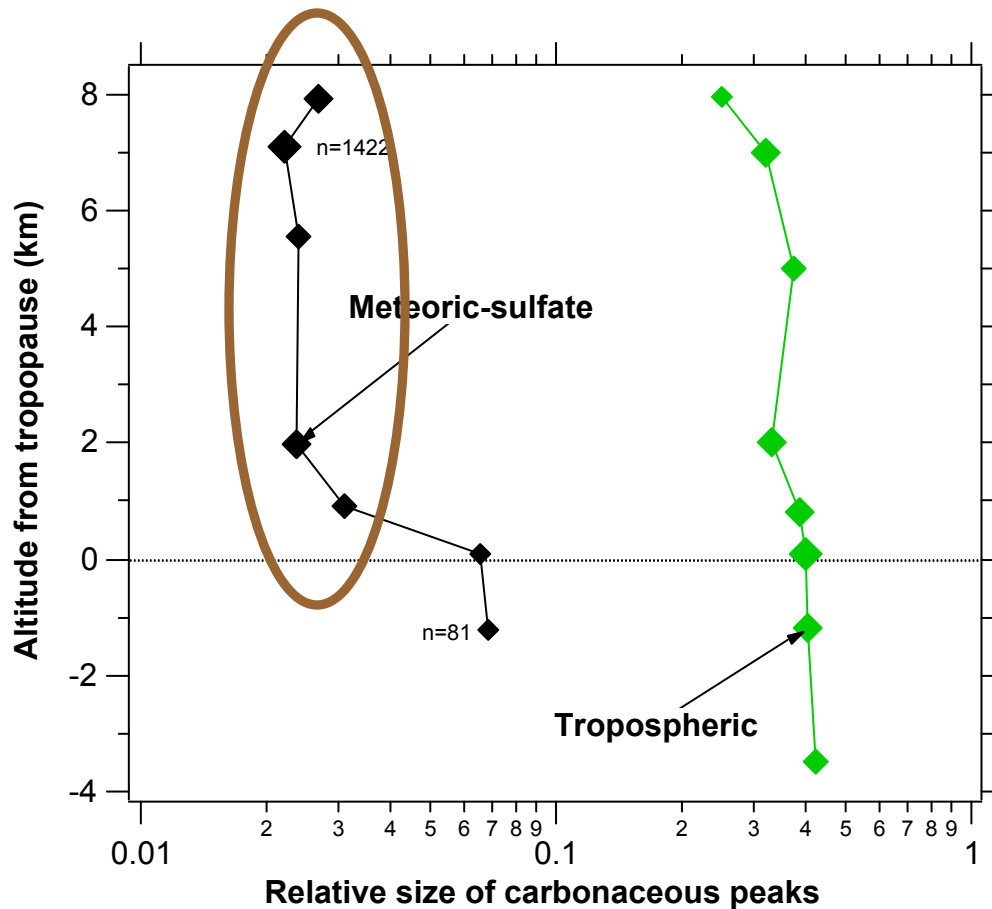
Learning from the stratosphere



Obvious:

Particles formed in the troposphere have more carbon content.

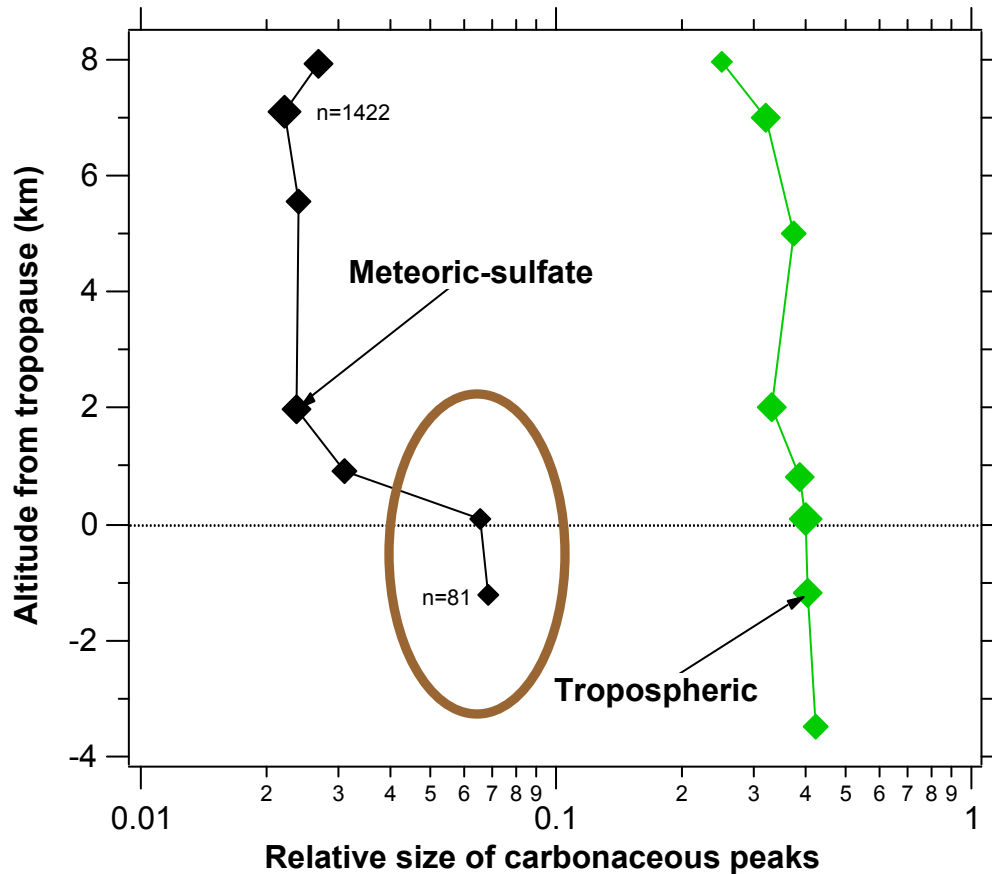
Learning from the stratosphere



Lesson 1:

Limited acid-catalyzed polymerization with small organics like formaldehyde

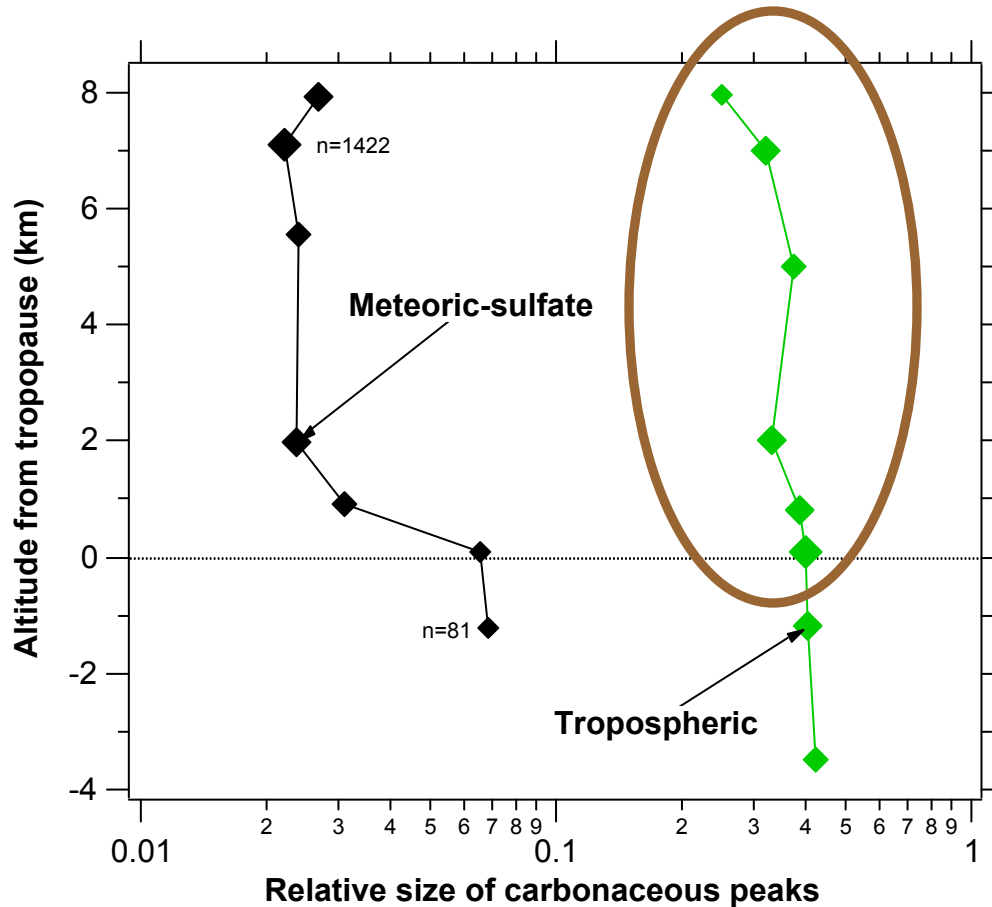
Learning from the stratosphere



Lesson 2:

Secondary organics even in the upper troposphere

Learning from the stratosphere



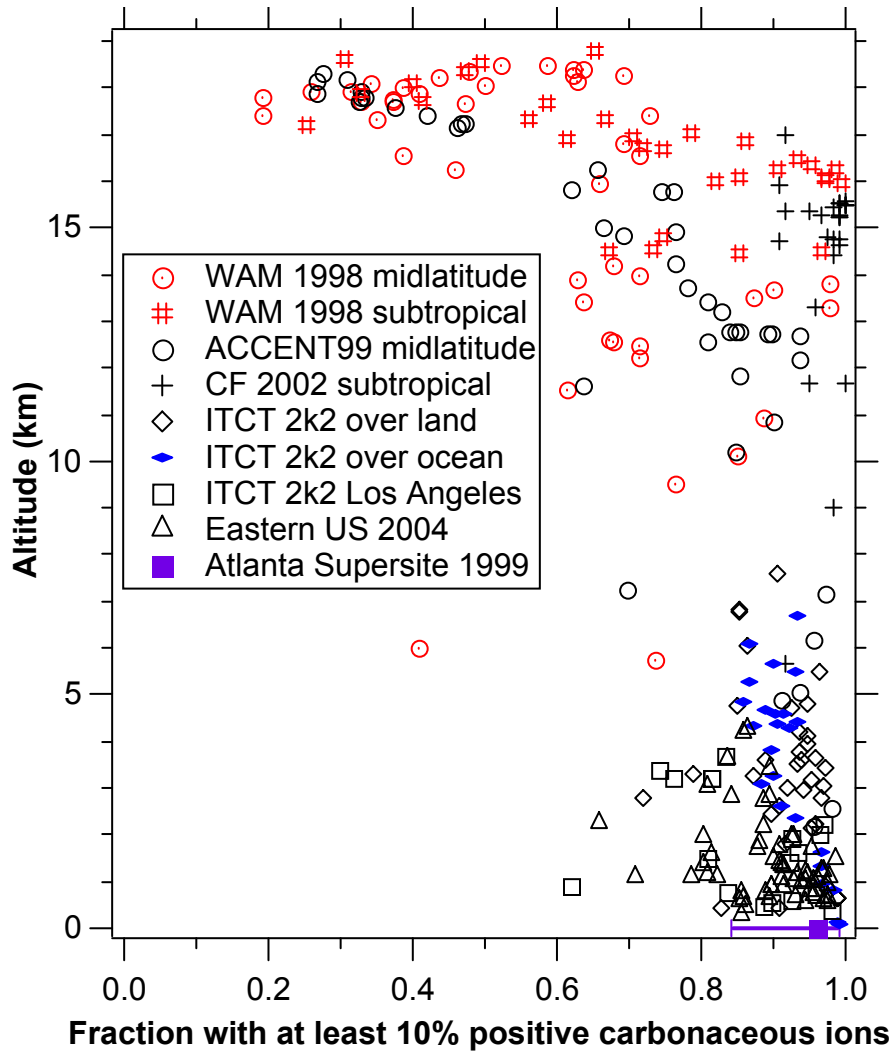
Lesson 3:

Aerosol organics persist in the stratosphere against:

- loss of semi-volatiles

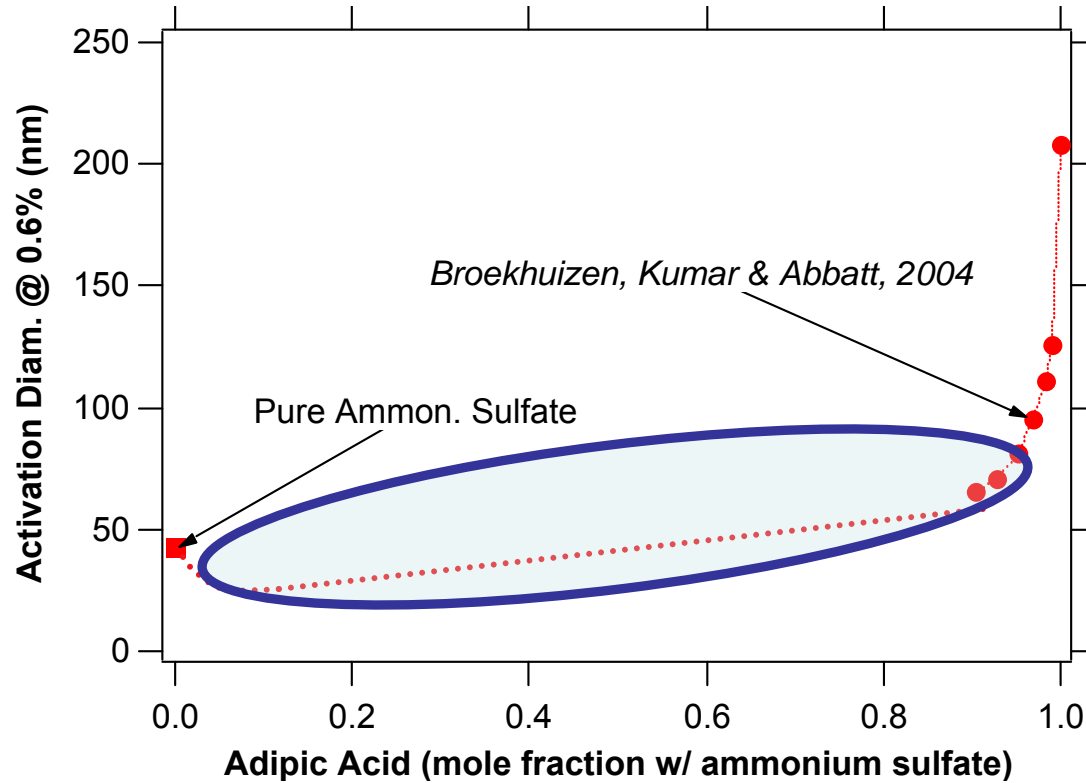
- loss of organic mass due to heterogeneous reactions with OH and O₃.

Mixed organic-sulfate particles



- Most particles contain at least a little organic material
- Almost all also contain some sulfate (not shown)

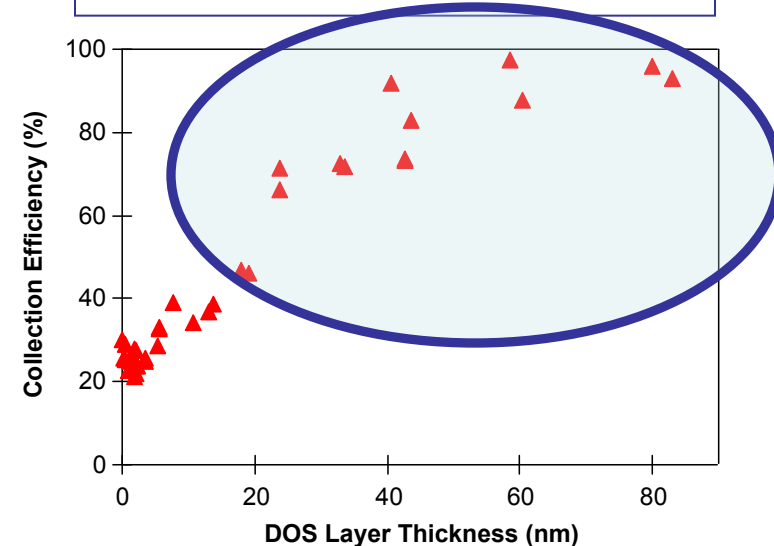
Importance of mixed particles for cloud activation



<10% organic is enough to get out of steep part of curve.

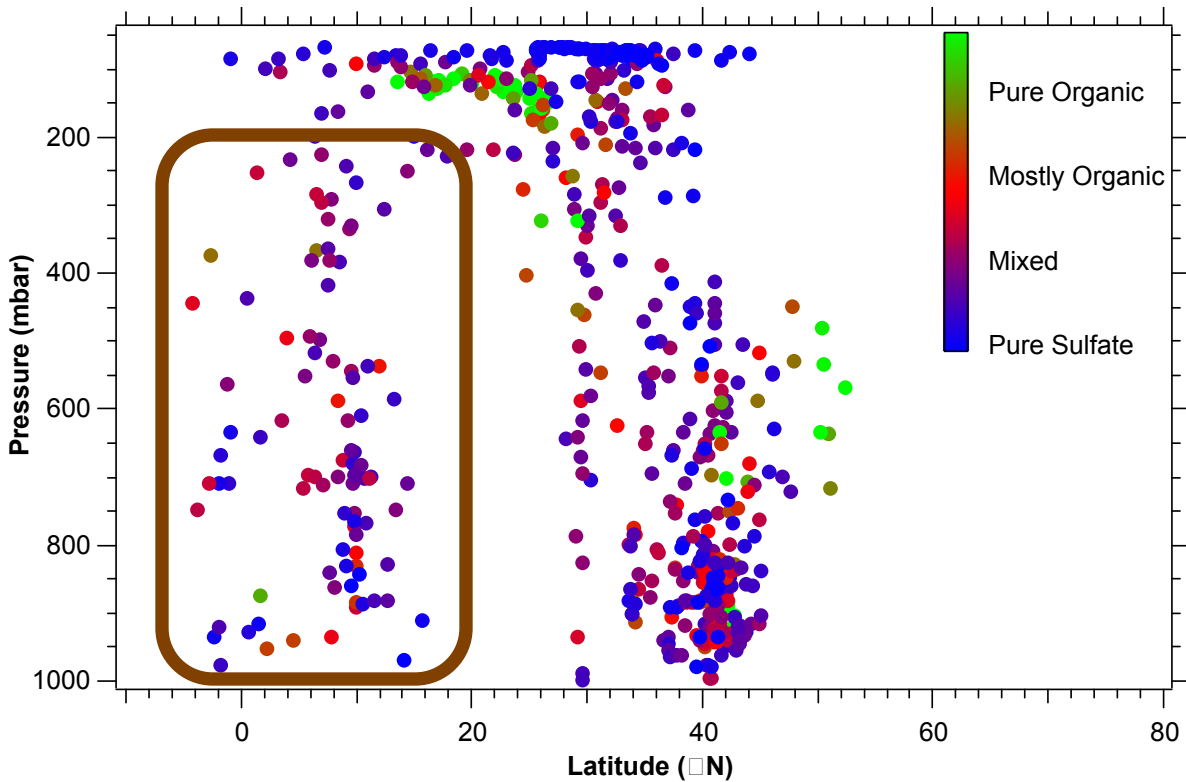
Great simplification for global models!

Mixing also helps AMS calibrations:



McFiggans et al., ACP, 2006

Field Studies for Specific Climate Issues



PALMS on B57, DC8
SP2 on B57

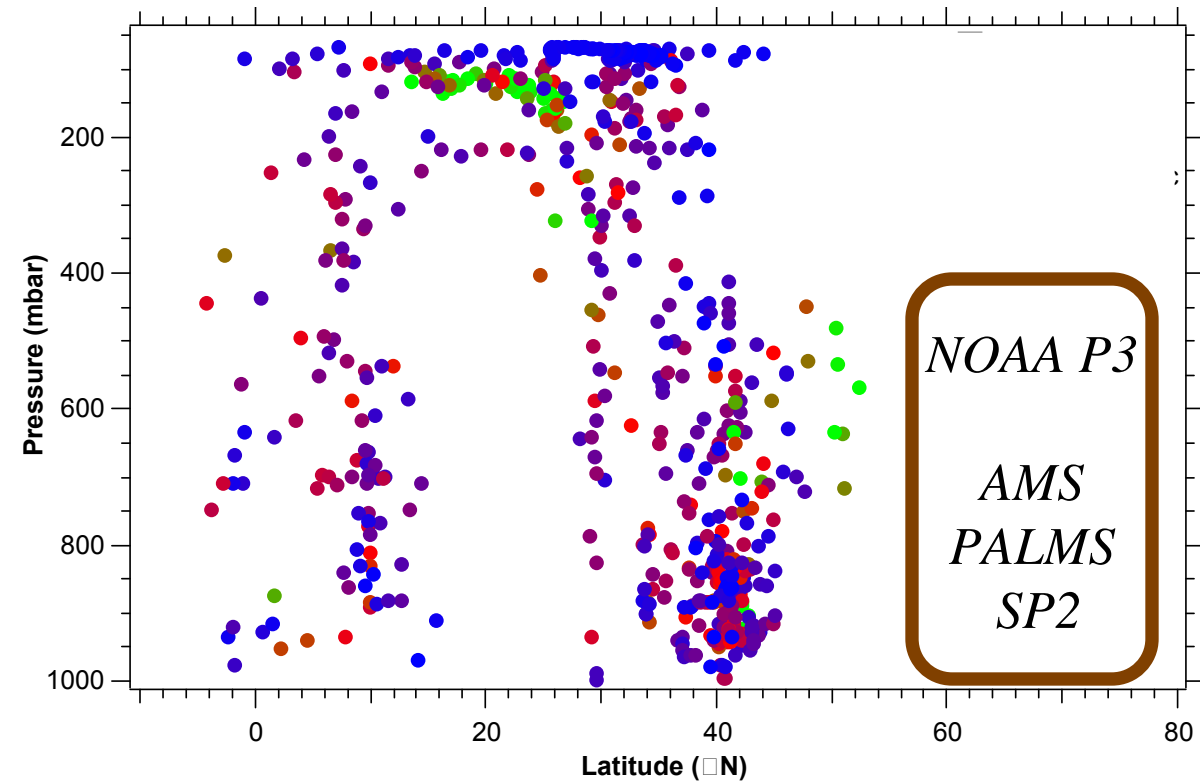
Why: (2006 - 07)

- Tropics are important
- Redistribution by deep convection

Some results:

- Black carbon profiles
- Organic content is different for outflow from continental or maritime convection

Field Studies for Specific Climate Issues



Why: (2008)

- Rapid Arctic changes
- Black carbon over snow and ice
- Different cloud feedbacks

Some results:

- ?
- ?

(end)