Airborne Observations of Aerosol Optical Properties: Evidence for Cloud Processing and Scavenging

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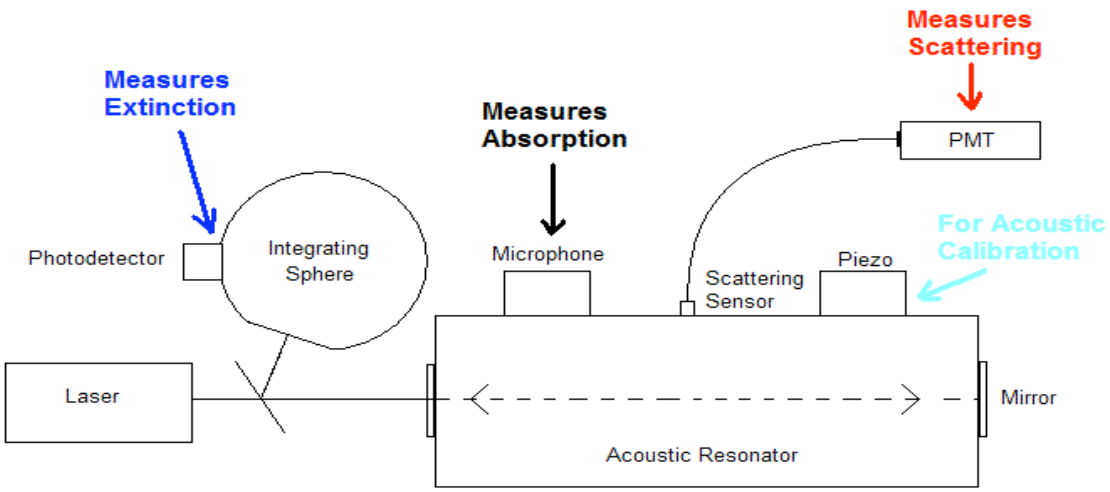
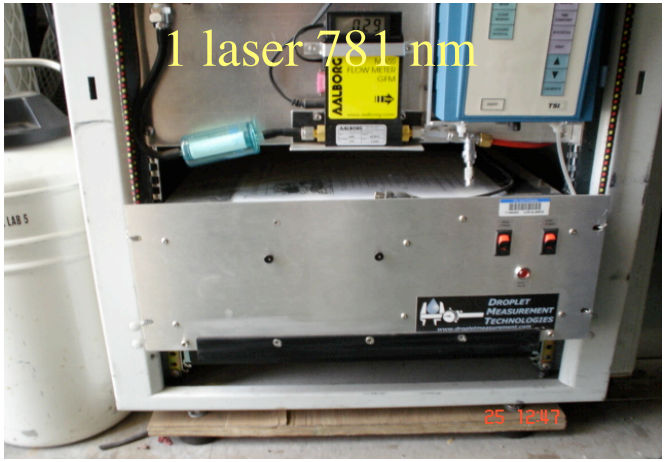
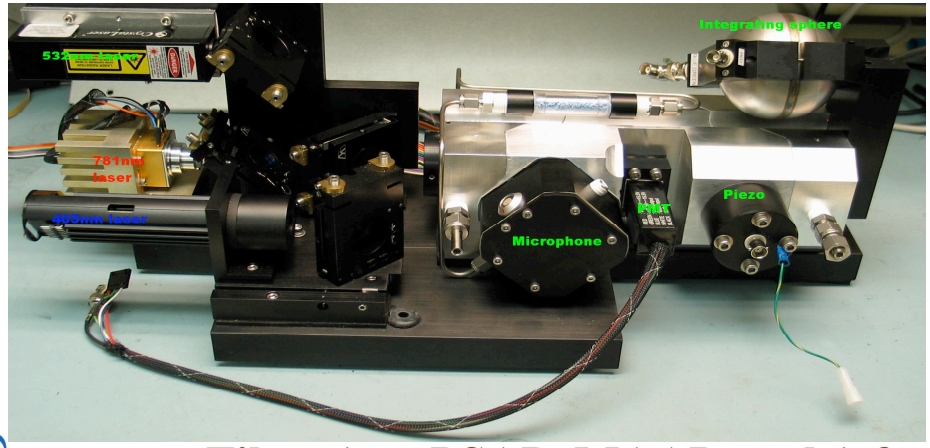
Outline

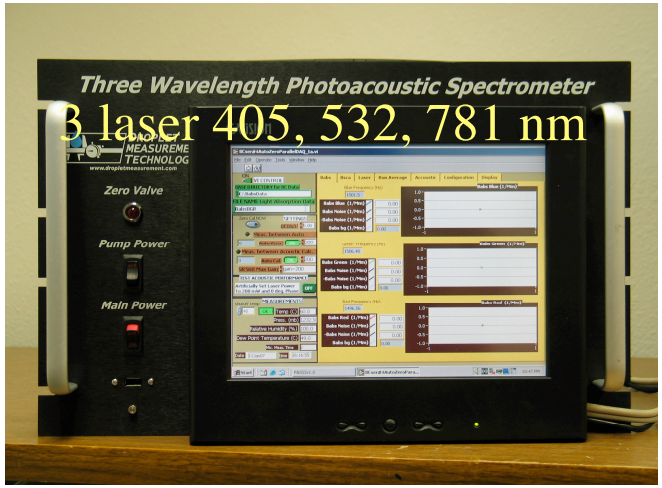
- Photo-acoustic Instruments: 1 laser and 3 laser
- Marine Stratus Experiment (MASE): Monterey/Pt. Reyes CA
  - What is the mixing state of in-cloud soot/SO_4/salt aerosols?
  - Can cloud drop black carbon alter their albedo and climate?
- Gulf of Mexico Atm. Comp. & Climate Study (GoMACCS): Houston TX
- Cumulus Humilis Aerosol Plume Study (CHAPS): SGP/Ponca City
  - Do cloud processes alter aerosol optical properties?
  - Is SO_4 selectively scavenged over soot in fresh urban emission?

Photoacoustic Instrument: Aerosol Absorption, Scattering & Single Scatter Albedo (Dry)









Filter (e.g. PSAP, MAAP, aeth.) free absorption measurement

Scattering, absorption & single scatter albedo on same sample

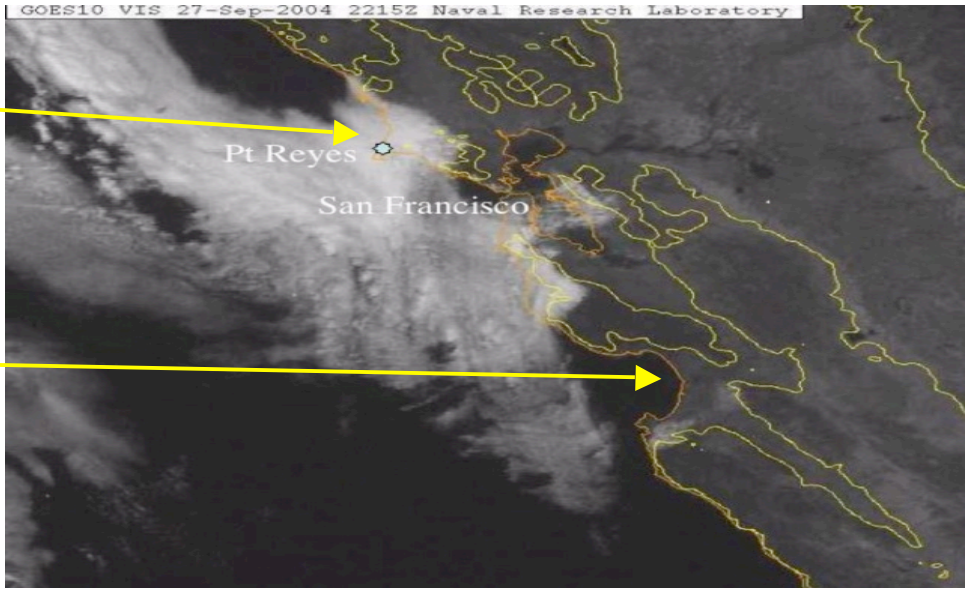
Marine Stratocumulus Experiment (PA, AMS, PILS, size distribution, LWC) : Polluted Cloud Event 6 July '05

Inlet

Navy CIRPAS Twin Otter (Seinfeld et al)

DOE-ARM Mobile

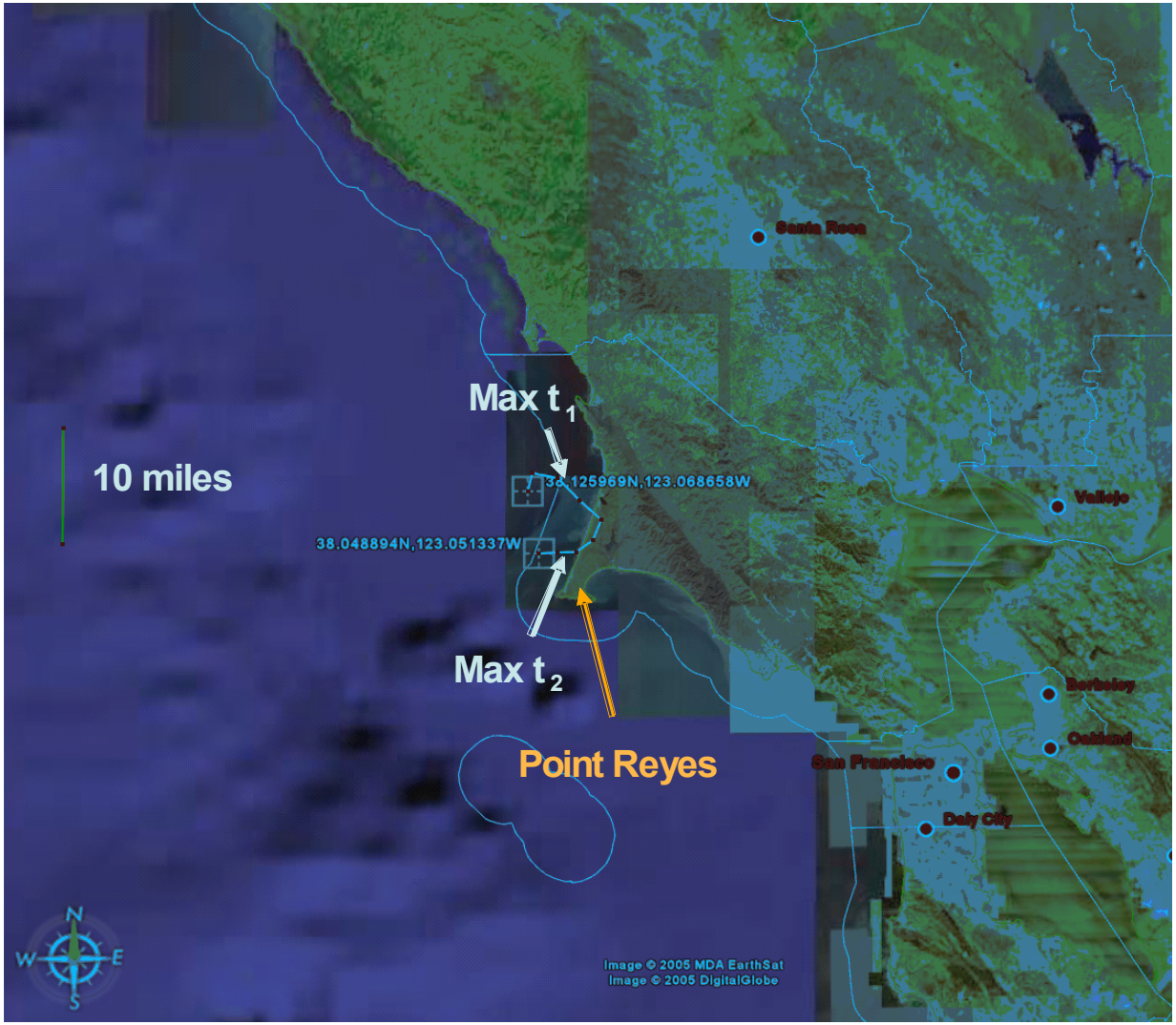
CIRPAS Monterrey



MASE: 6 July 05, In cloud plume

$V^{airplane} \sim 50$ m/s

$\Delta t^{Plume} \sim 10$ min



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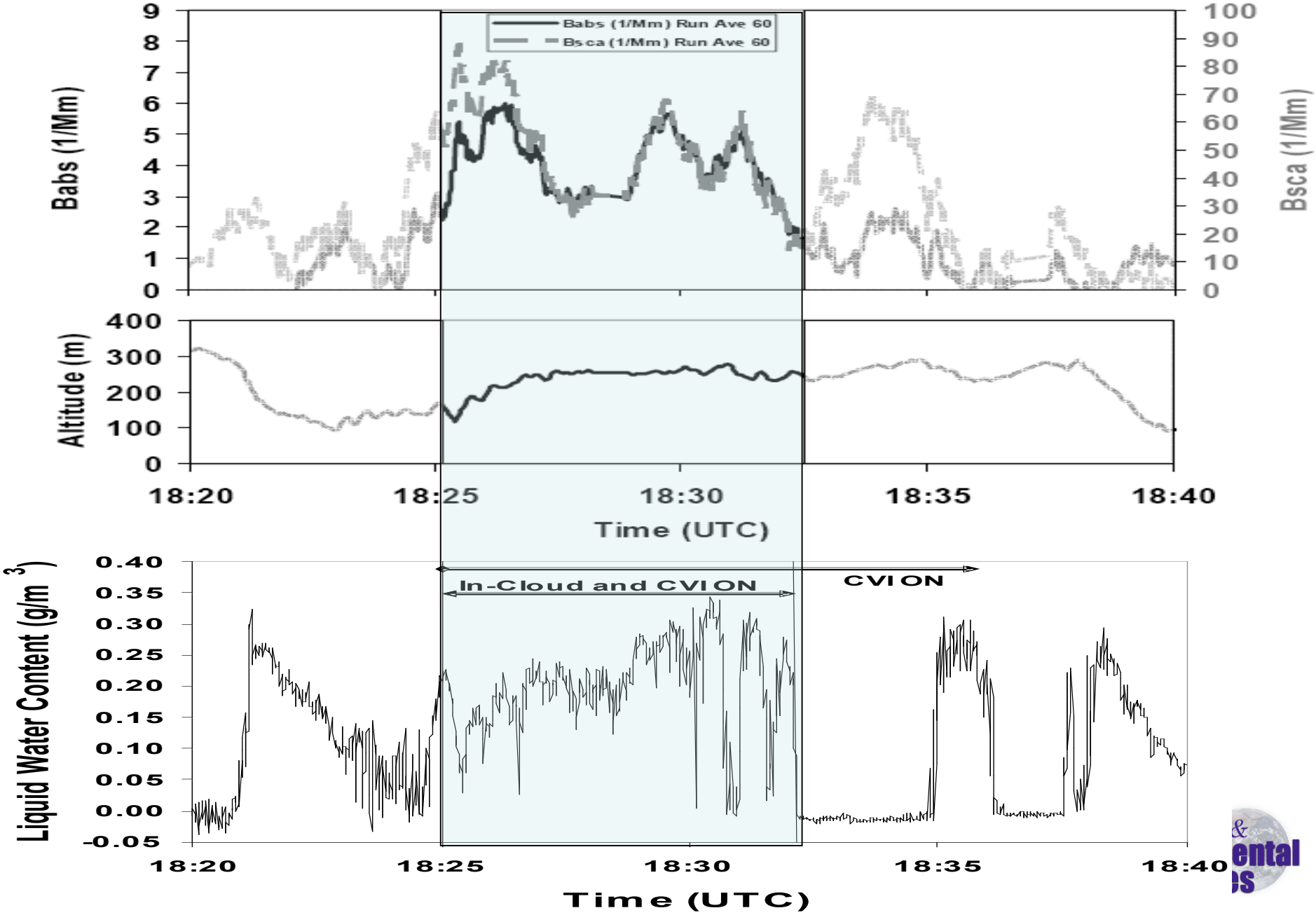
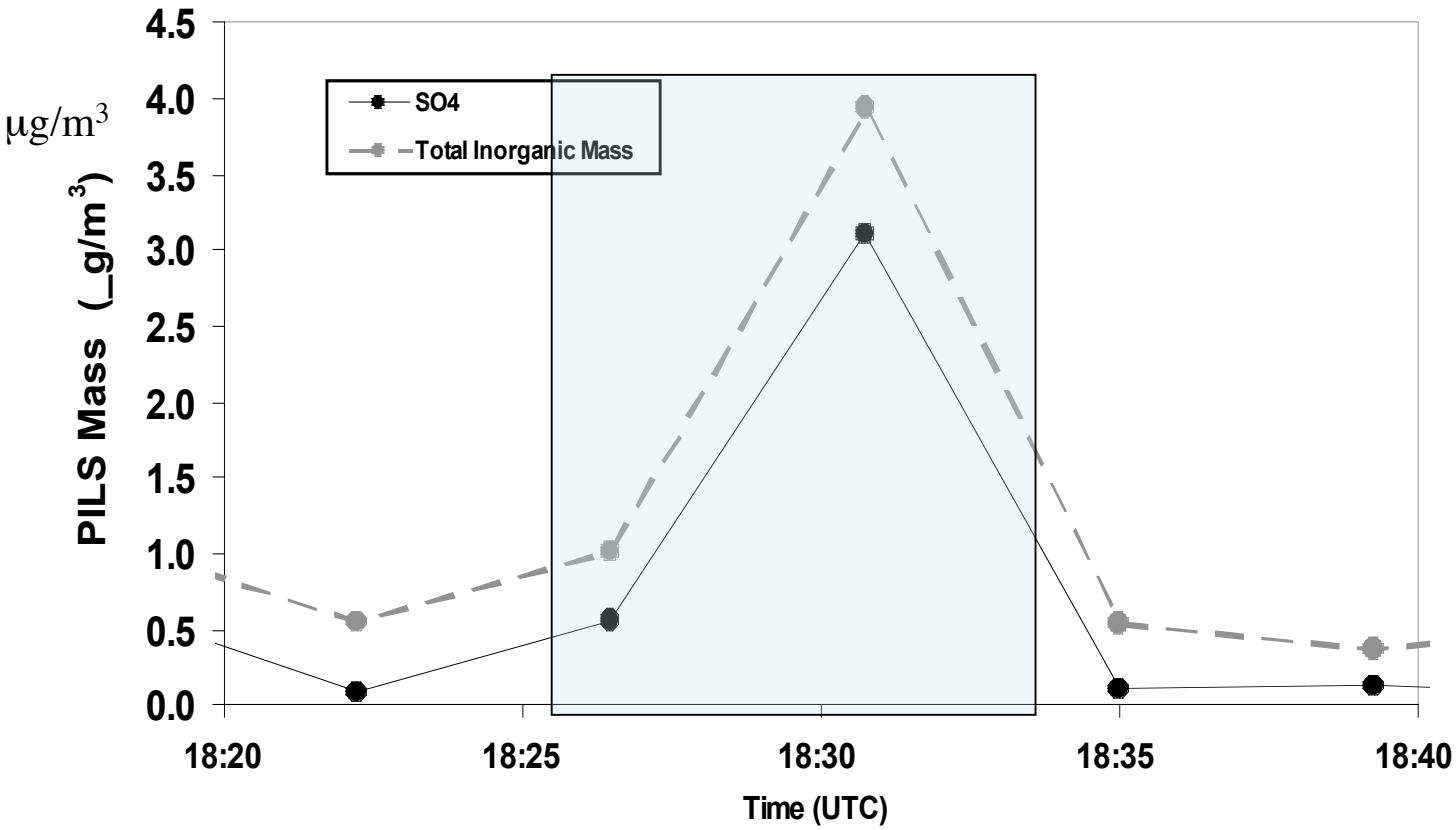
In Cloud Aerosol Plume: Absorption, Scattering, Liquid Water



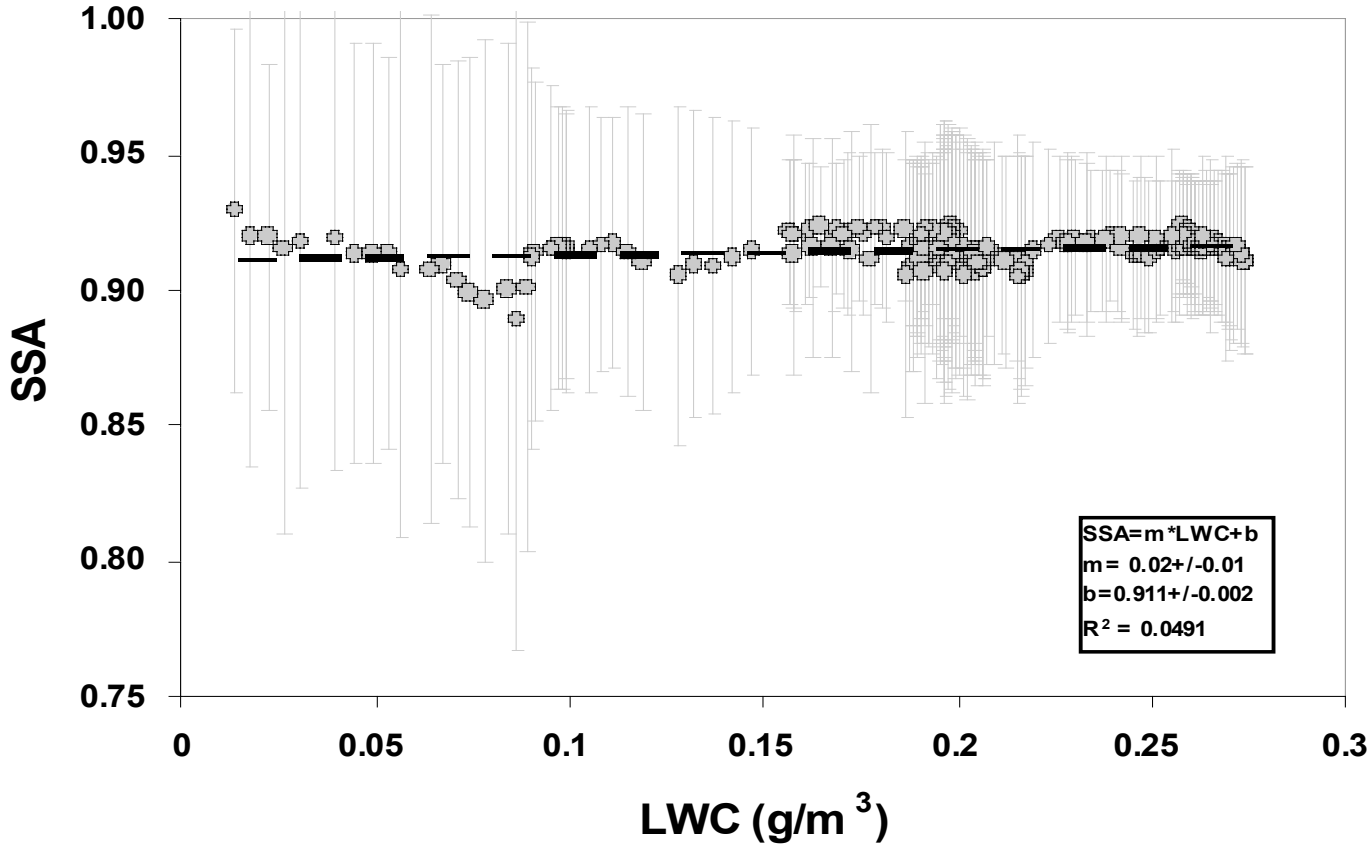
Inorganic and Sulfate (PILS)



Absorbing (soot) mixed with sulfate and inorganics (sea salt), no dust

Are they internally or externally mixed?

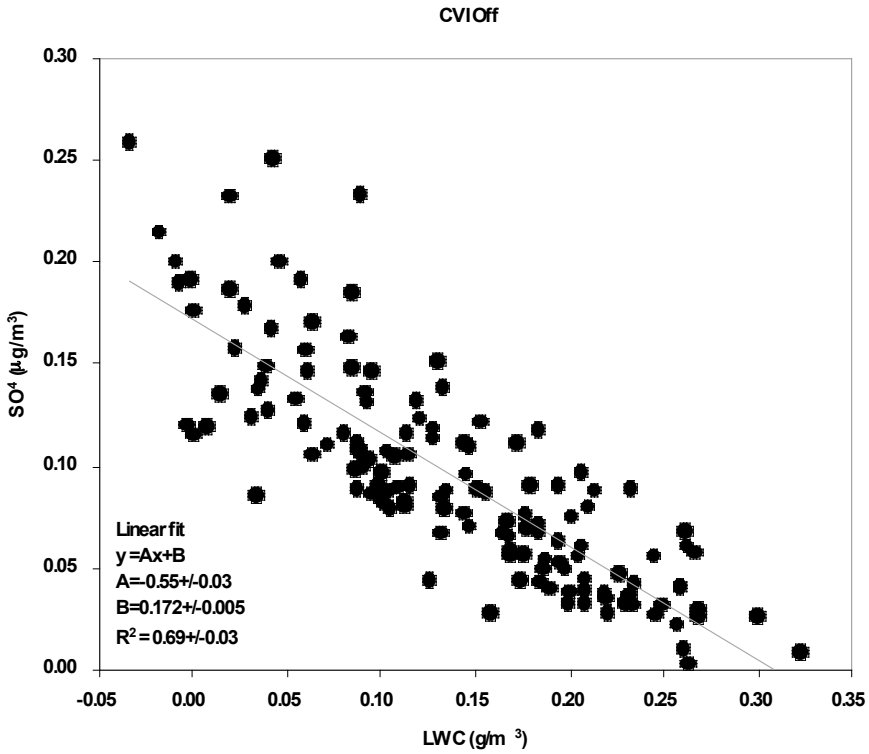
SSA Large and Independent of LWC



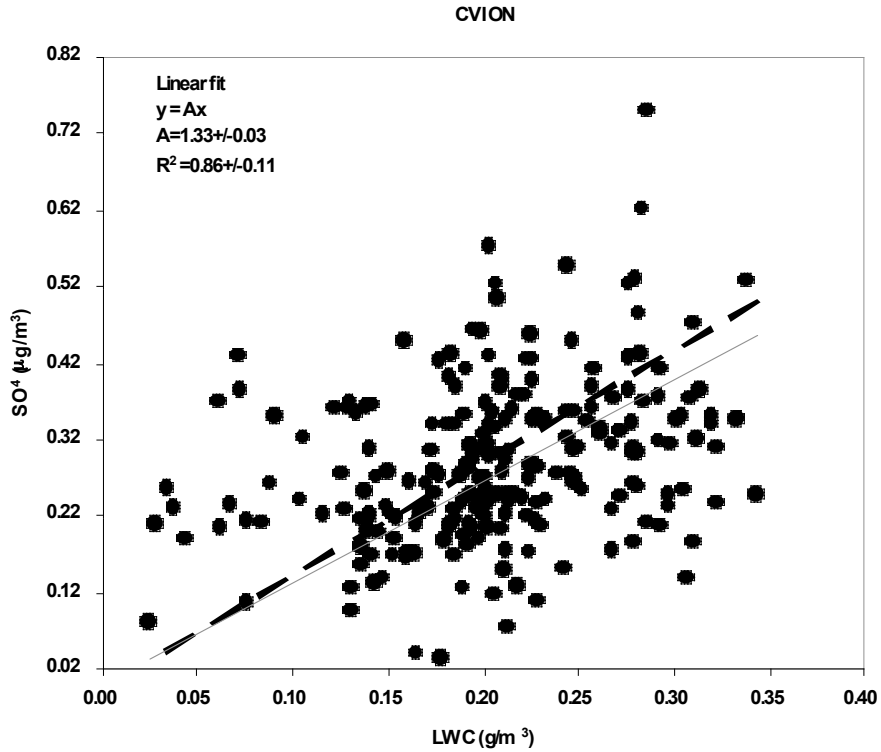
Suggests internal mixing

AMS-Sulfate vs LWC: Scavenging by clouds

Interstitial (CVI-Off)



Cloud Drop (CVI-On)



Nucleation aerosol scavenging efficiency ~ 60-80% (<LWC> ~ 0.2 g/m^3)
High similar to sulfate, suggests internal mixture

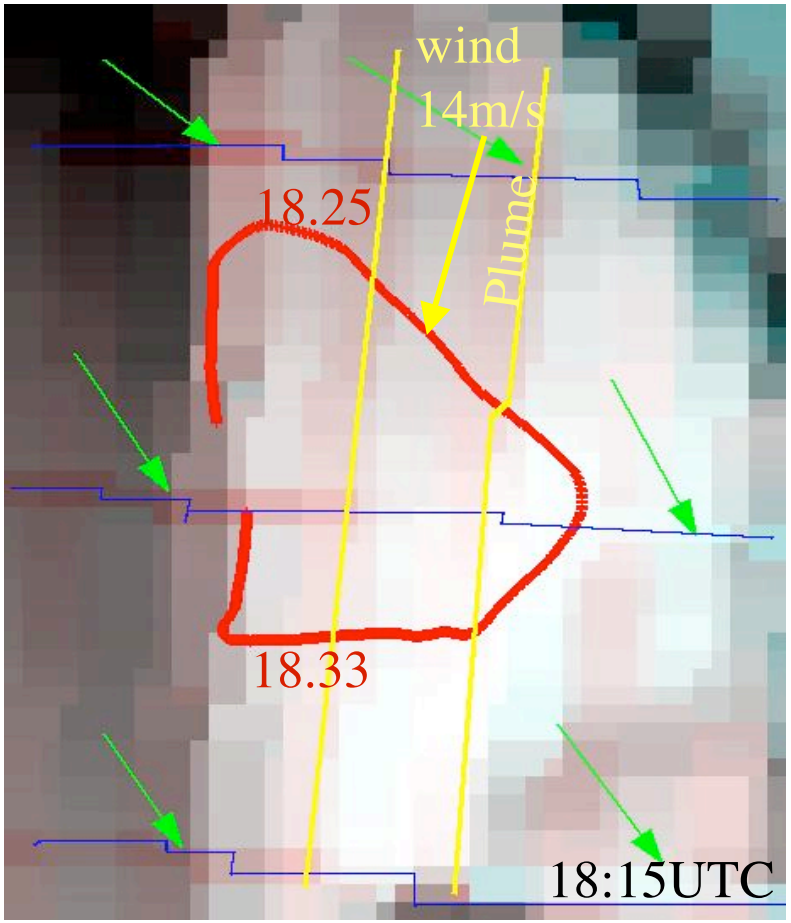
Derived Cloud Drop Soot Volume Fraction

- Constrained by independent data indicating absorption dominated by a small number of larger (1 μ) particles.
  - $PILS_{S04}$ (<1μ)/AMS_{SO4}(<0.7μ) ~ 10
  - BC/SO_4 refractive index + volume mixing
  - Best fits our optical measurements
  - No DMA enhancement of submicron particles
  - Low CPC counts ~200 particles cm^{-3}
- Cloud soot to water volume fraction of **$(1.7 \pm 0.7) \cdot 10^{-6}$**

Aircraft Transect on Satellite Overpass:

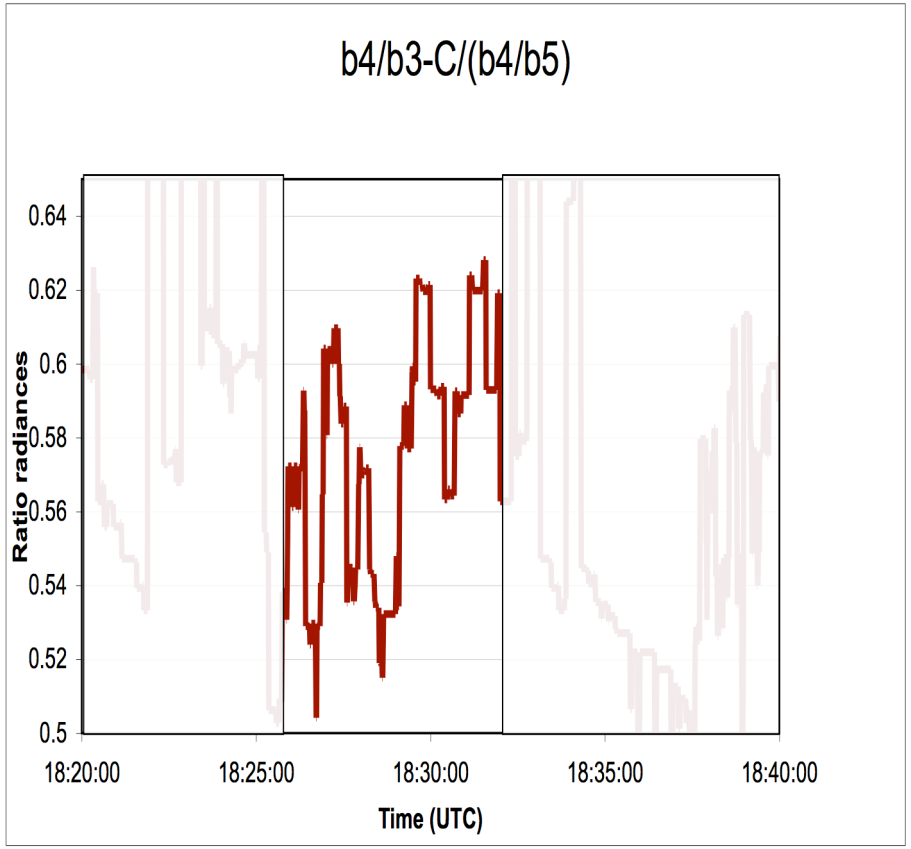
Does MODIS resolve the in cloud absorbing plume?

b3, 459-479nm spectral radiances (W/m2-um-sr)



Aircraft transect on MODIS image

b4(545-560nm), b5(1230-1250nm)



Possibly! Pending corrections due to winds and slight (10 min) time lag

Effect on Cloud Drop Single Scatter Albedo & Climate Forcing

Twohy et al. JGR 1989 ~2.3-7.9·10^{-8}

Hintzenberger 2000 ~ 0.5-3·10^{-6}

Hitzenberger 2001 ~ 0.2-3·10^{-6}

⇒Our MASE inference (1.7±0.7) ·10^{-6}

⇒Most polluted cloud sampled

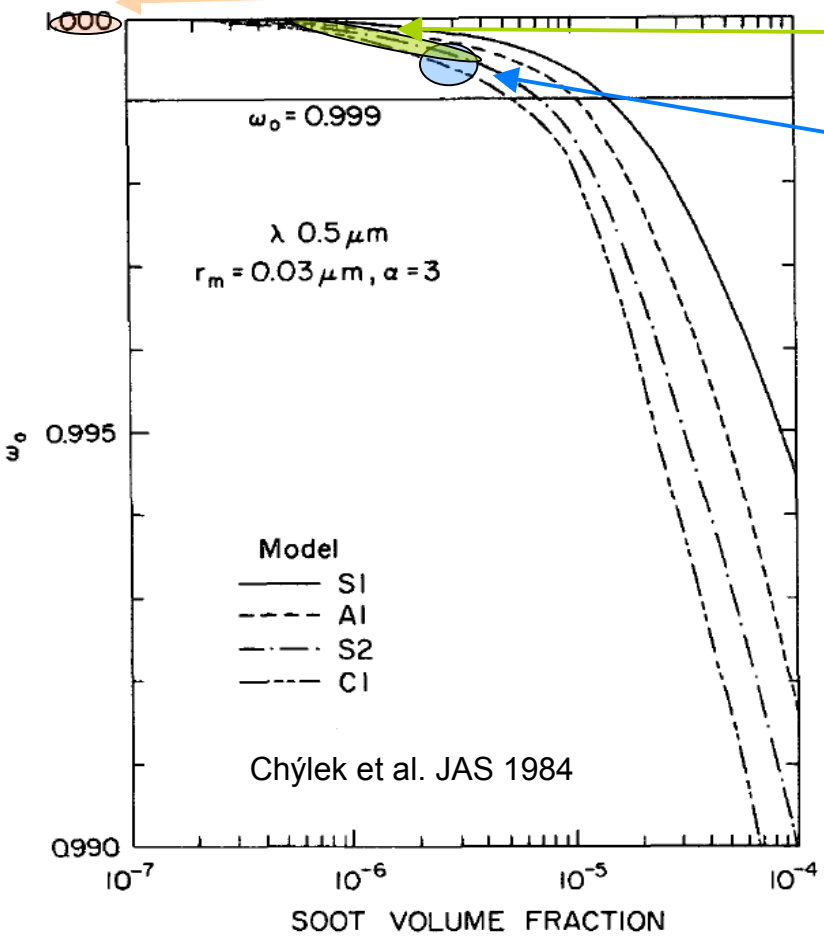


FIG. 5. Single scattering albedo of 4 model cloud drop-size distributions as a function of the soot volume fraction. The value of ω_0 = 0.999 corresponds to the value inferred from observations of albedos of thick clouds. To produce this value, water drops have to contain definite amounts of graphitic carbon within the range of 5×10^{-6} to 1.5×10^{-5} by volume.

Cloud drop ω_o change of 0.999 leads to solar TOA forcing of 0.5 W m^{-2} for low clouds and 1.68 W m^{-2} for all clouds using a 1-D radiative-convective model constrained by MAST data (*Erlick 2003, 2001)*

Our results imply cloud albedo change to 0.9995, which when extended to 10% of global clouds (extreme) yields a climate forcing 0.03 to 0.08 W m^{-2}

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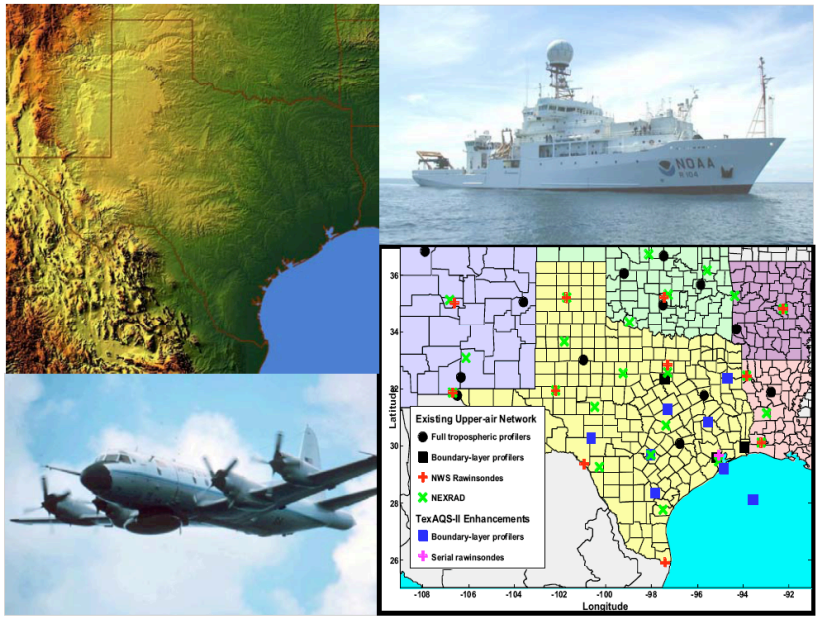
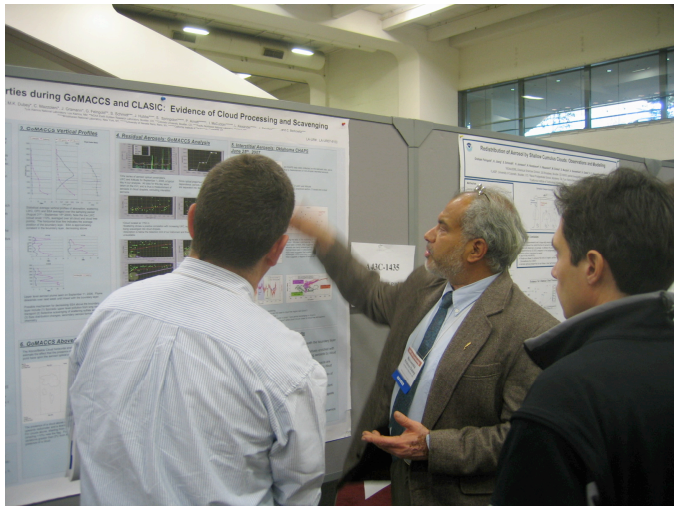
GoMACCS/TEXAQS Houston Aug/Sep 06

2006 TEXAQS/GoMACCS

Science and Implementation Plan





Combining Climate Change and Air Quality Research

March 2005

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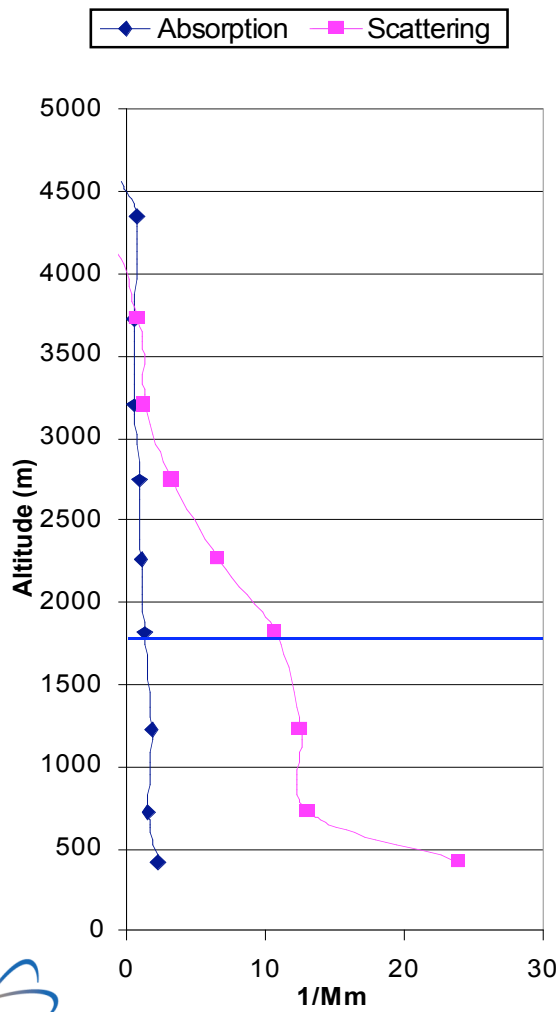
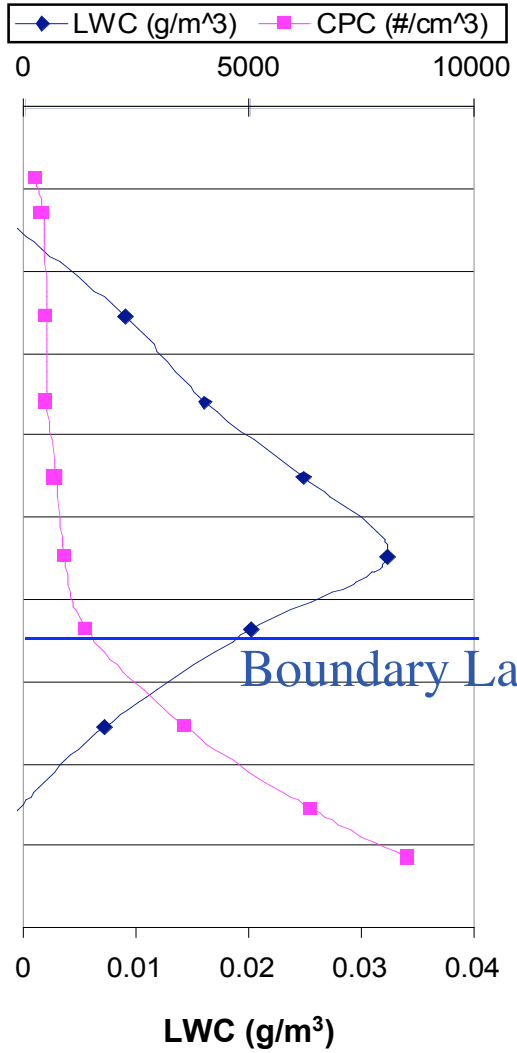
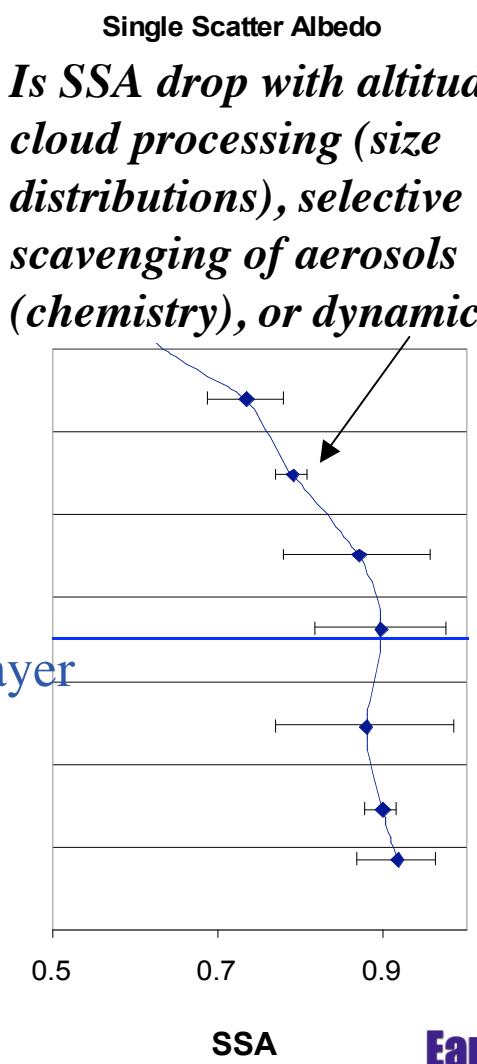
Vertical Statistics Abs, Scat, SSA, CPC, LWC







Is SSA drop with altitude from cloud processing (size distributions), selective scavenging of aerosols (chemistry), or dynamics?

Boundary Layer

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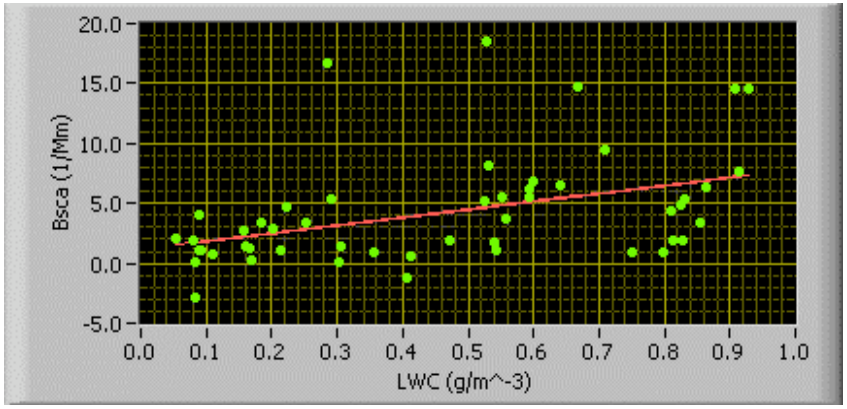
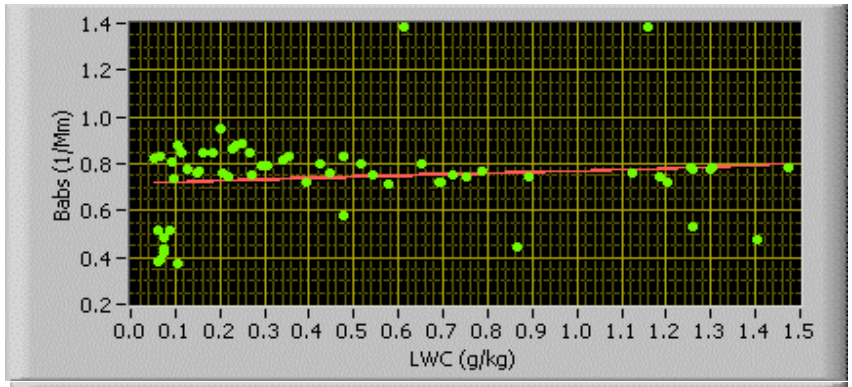
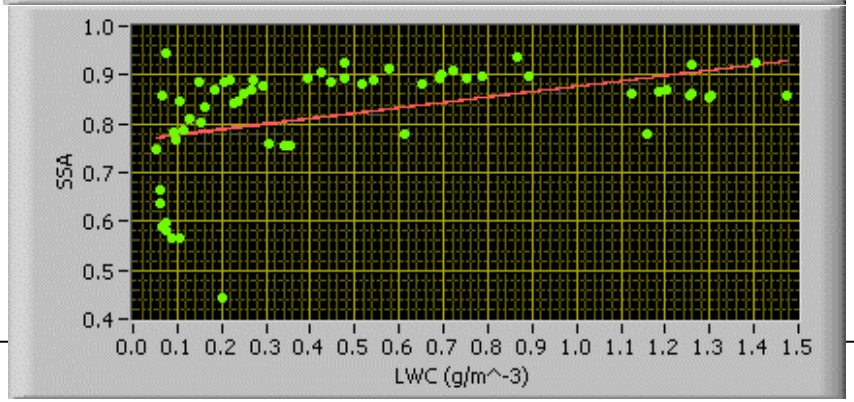
In Cloud Events: Scat, Abs, SSA vs LWC

GoMACCS
Houston
Aug-Sep 06



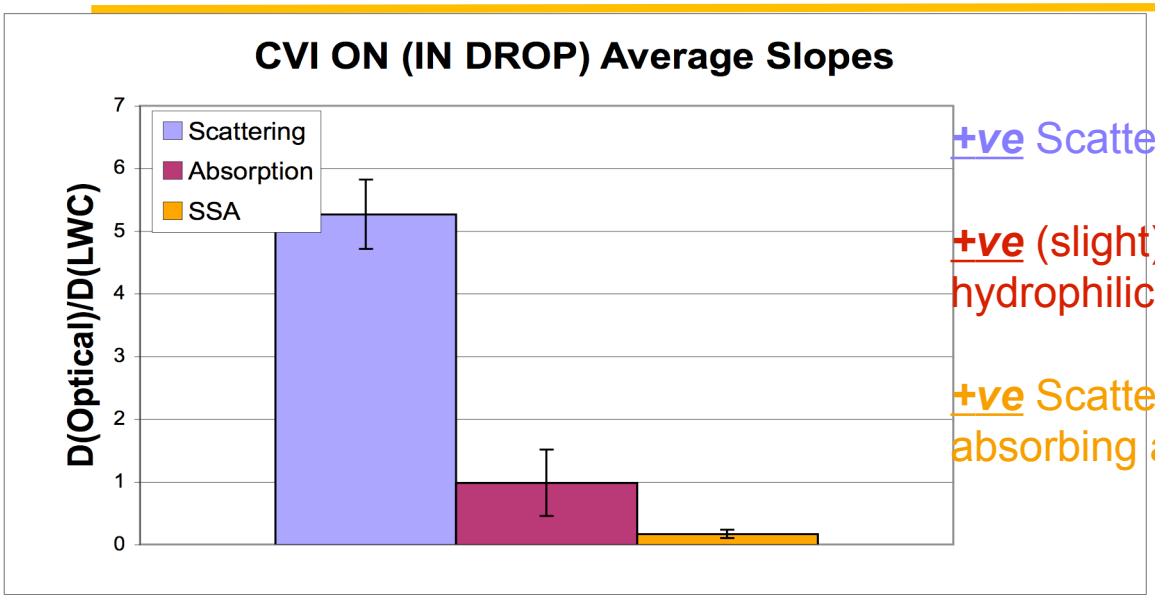




Either
CVI On (in drop)
100 minutes data
Or
CVI Off (interstitial)
60 minutes

N. Nussbaum

In Cloud Statistics of d(Opt. Prop.))/d(LWC)

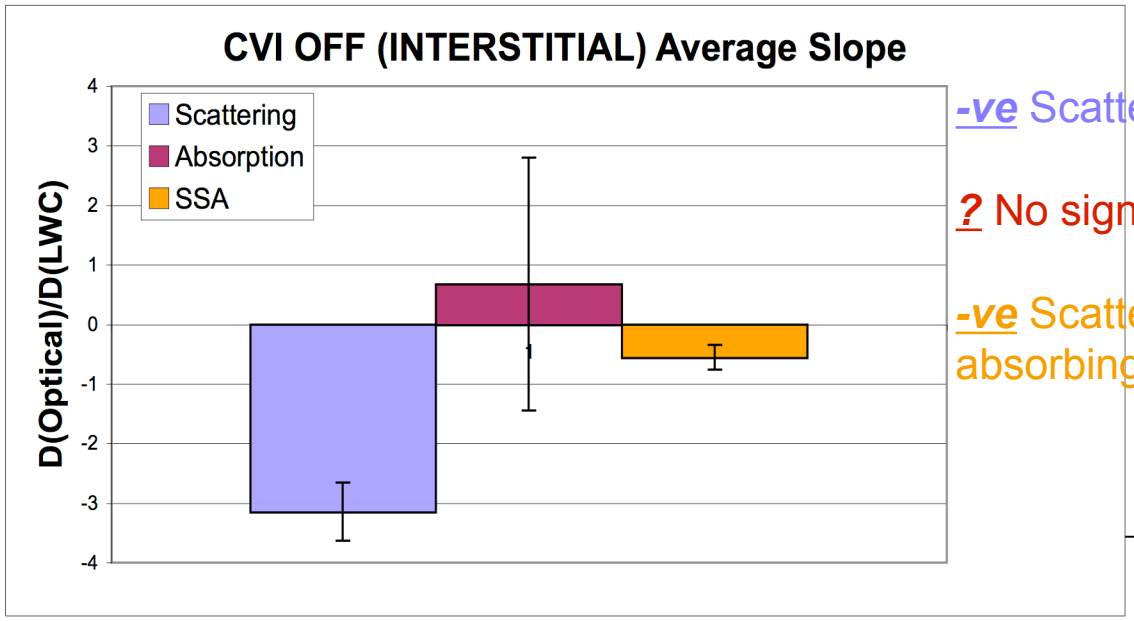


IN DROP

+ve Scattering aerosols are hydrophilic

+ve (slight) some Absorbing aerosols are hydrophilic (internal mix. with scat, aged soot)

+ve Scattering aerosols more hydrophilic than absorbing aerosols (some fresh soot)



INTERSTITIAL

-ve Scattering aerosols are hydrophilic

? No significant slope in absorption (weak stat)

-ve Scattering aerosols more hydrophilic than absorbing aerosols (some fresh soot)

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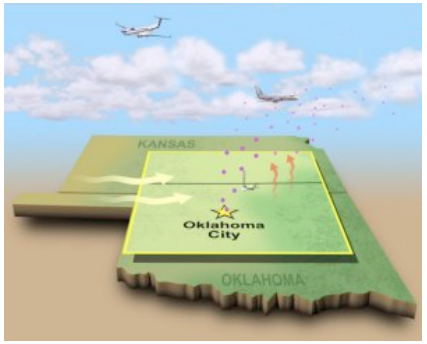
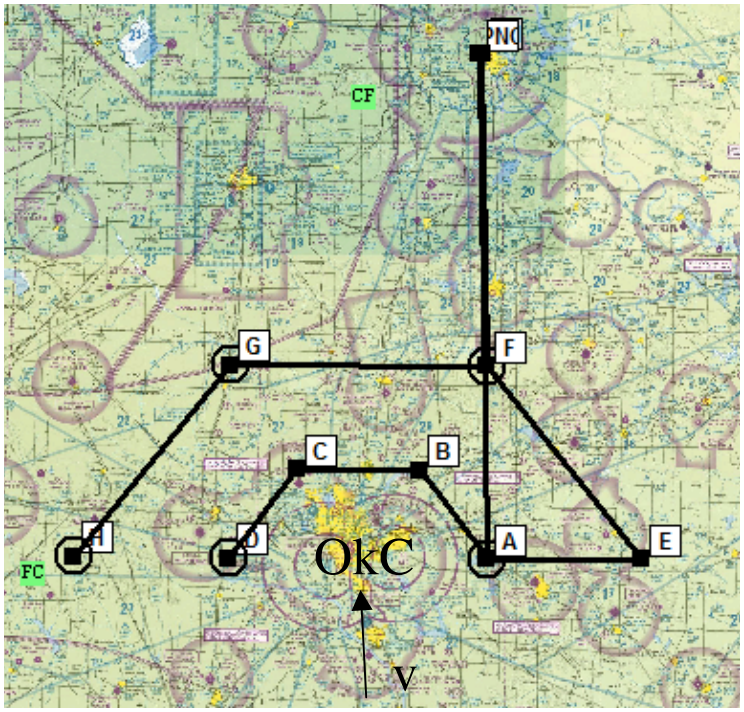
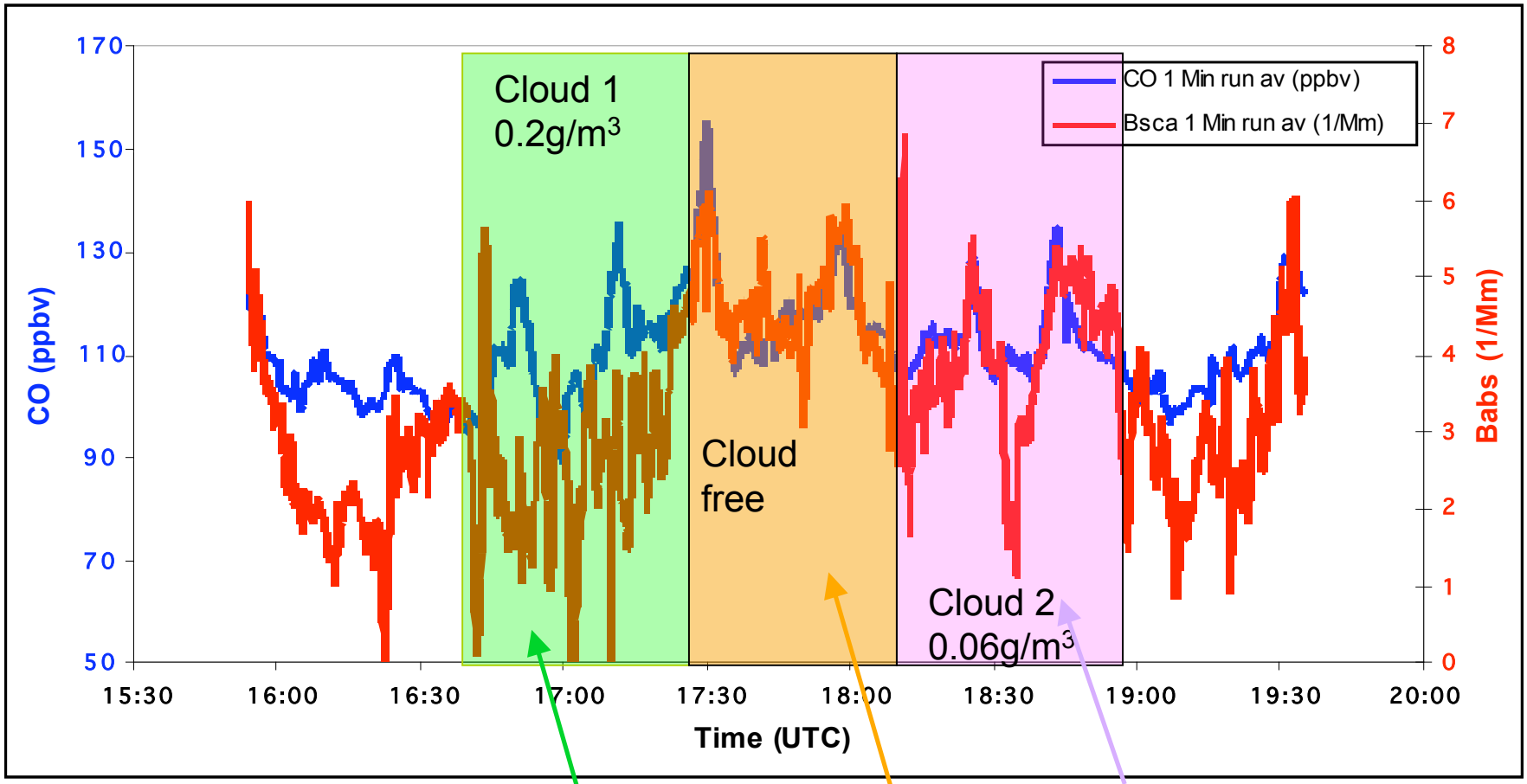
Cumulus Humilis Aerosol Plume Study (CHAPS)



6/25/2007

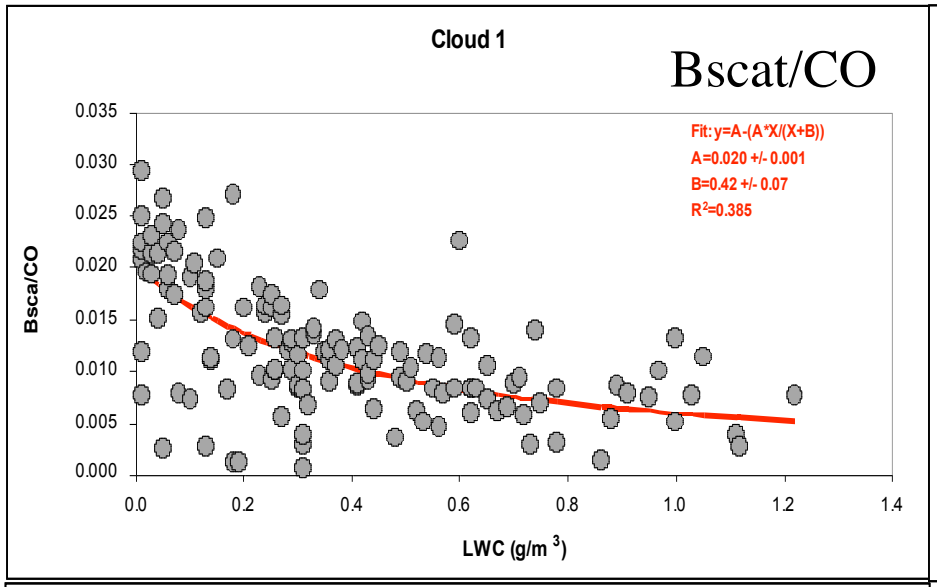


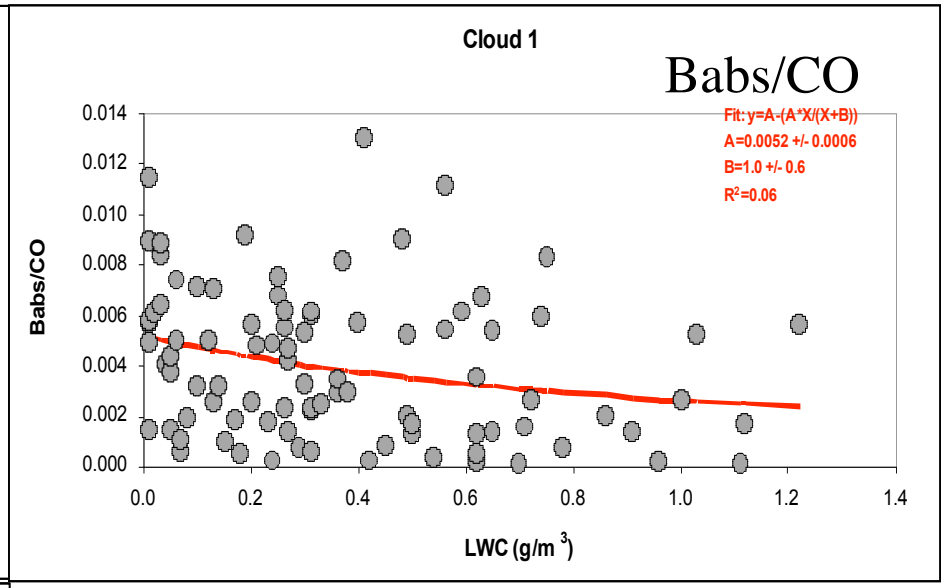
CHAPS 6/25/07 Oklahoma City Plume: CO & Bscat

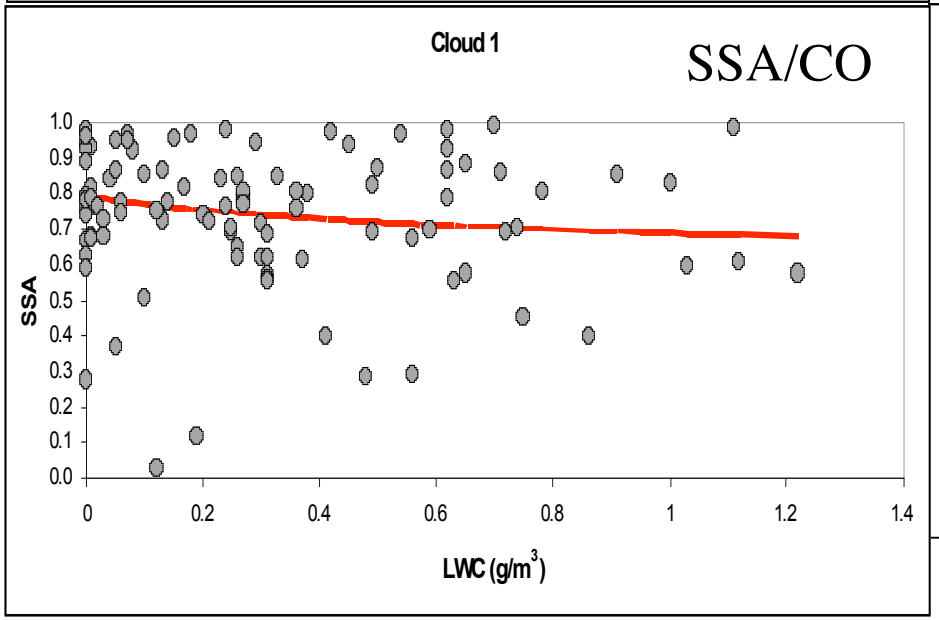


Positive CO/Scat correlation in cloud free and low water cloud which breaks down in high water cloud due to scavenging

Scavenging Kinetics: Optical Prop/CO vs LWC







- CO ratio normalizes dilution
- Scattering aerosols scavenged more effectively than absorbers
- Some external mixing (fresh soot)

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Conclusions

- In cloud soot-sulfate-salt aerosols are internally mixed (aged)
- Climatically significant cloud-albedo reduction only for extreme pollution, high τ_{opt} & low indirect effect (<0.05 W/m^2).
  - *Snow/ice-albedo sensitive to soot & significant ~0.2 W/m^2, needs observations*
- Cloud processing lowers aerosol SSA
- In fresh urban emissions scattering aerosols scavenged selectively over absorbing aerosols (some external mixture)