Sources of Primary and Secondary Particles in the Houston Area in 2006

Charles Brock, Adam Wollny, Rebecca Washenfelder, Ann Middlebrook, Roya Bahreini, Ryan Spackman, Shuka Schwarz, John Holloway, Jeff Peischl, Tom Ryerson, Andy Neuman, John Nowak, Joost de Gouw, Carsten Warneke, Michael Trainer, Andreas Stohl, Tahllee Baynard, and others. . ..

Objectives

- **Examine relative importance of primary and secondary aerosol sources**
- **Evaluate contribution from Parish power plant and industrial emissions**
- **Compare with TexAQS 2000 data**

Aerosol Measurements on the NOAA P-3

Particle size distributions, .005-8 µm, 1 sec.

- **Calculate integrated number and volume (proportional to mass), uncertainties in integrated volume ~25%**

Aerosol mass spectrometer, submicron sulfate, nitrate, ammonium, organics, 10 s

- **mission absolute uncertainties ~50% (single-flight variability ~25%), good sensitivity**
- **Middlebrook talk, Bahreini poster**

- **Particle-in-liquid sampler, submicron water-soluble organic (Hecobian poster)**
- **Single particle soot photometer (Spackman poster)**
- **Cavity ringdown extinction, f(rh), absorption (Baynard poster; Lack talk, Massoli poster for ship instruments)**
- **Actinic flux spectral radiometer (Stark poster)**

Aerosol Measurements on the NOAA P-3

Particle size distributions, .005-8 µm, 1 sec.

- **Calculate integrated number and volume (proportional to mass), uncertainties in integrated volume ~25%**

Aerosol mass spectrometer, submicron sulfate, nitrate, ammonium, organics, 10 s

- **mission absolute uncertainties ~50% (single-flight variability ~25%), good sensitivity**
- **Middlebrook talk, Bahreini poster**

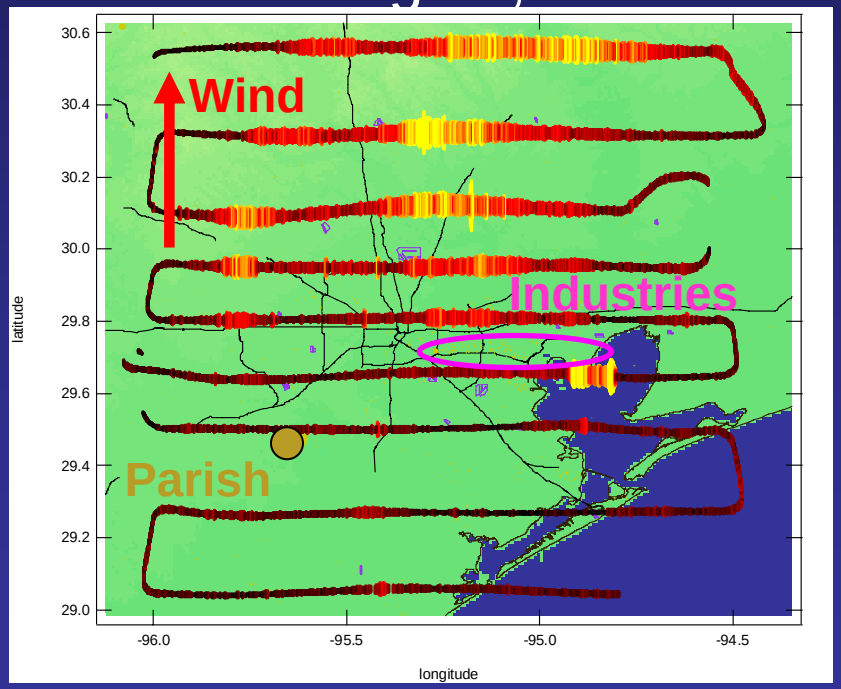
- **Particle-in-liquid sampler, submicron water-soluble organic (Hecobian poster)**
- **Single particle soot photometer (Spackman poster)**
- **Cavity ringdown extinction, f(rh), absorption (Baynard poster; Lack talk, Massoli poster for ship instruments)**
- **Actinic flux spectral radiometer (Stark poster)**

Not available in 2000 campaign

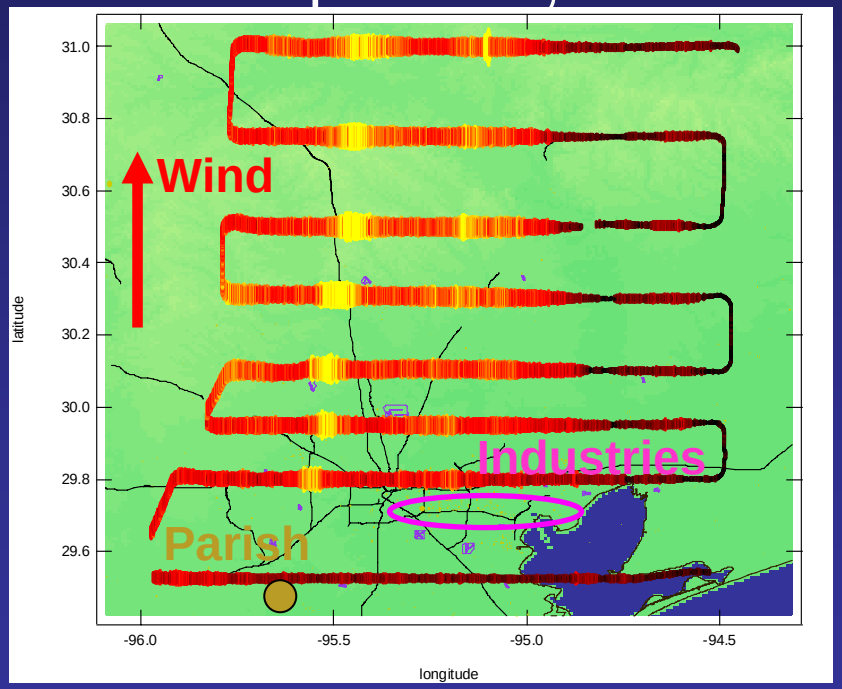
Focus on 27 September 2006:

- *High O_3 day*
- *Wind ~constant in direction, ~2x variable in speed*
- *Many downwind transects*
- *Source plumes ~separated*
- *Qualitatively similar to days examined in 2000*

28 August, 2000



27 September, 2006

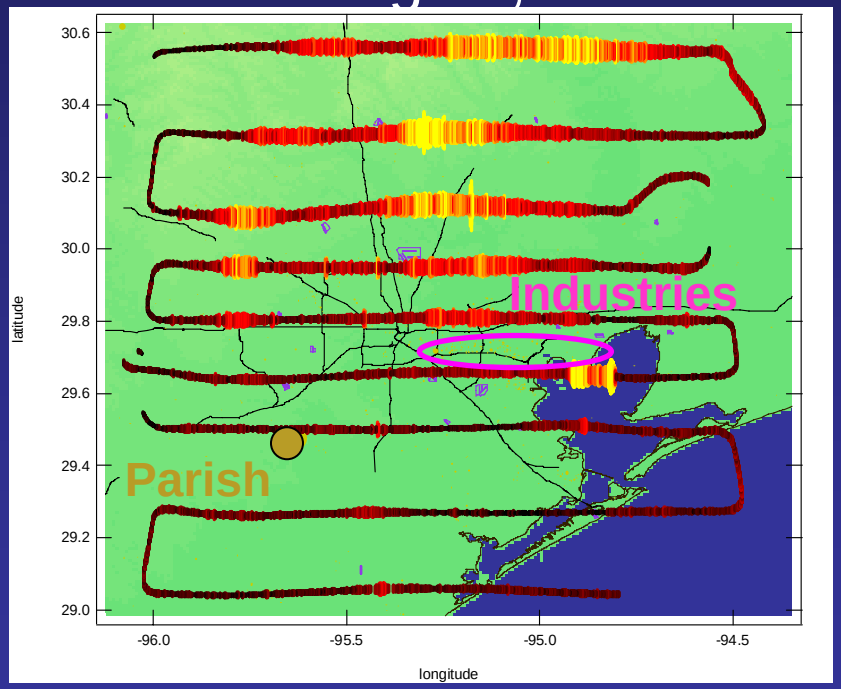


Color & size proportional to particle volume. Scales differ.

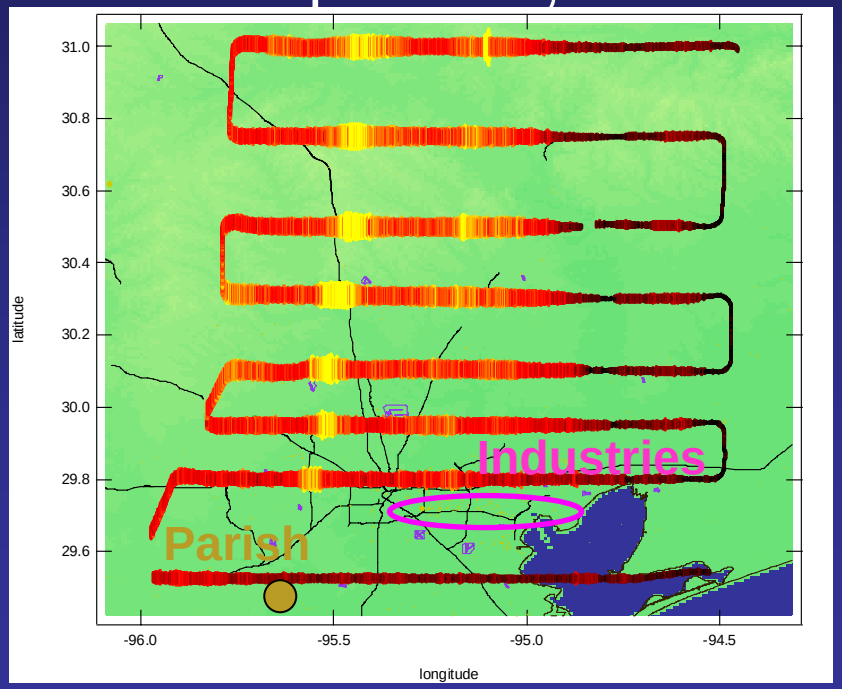
But relative to August 2000. . .

- *Temperature cooler*
- *Mixing less vigorous ↕ and ↔*
- *Fewer photons*
- *Background more variable, so uncertainties in Δ larger*

28 August, 2000

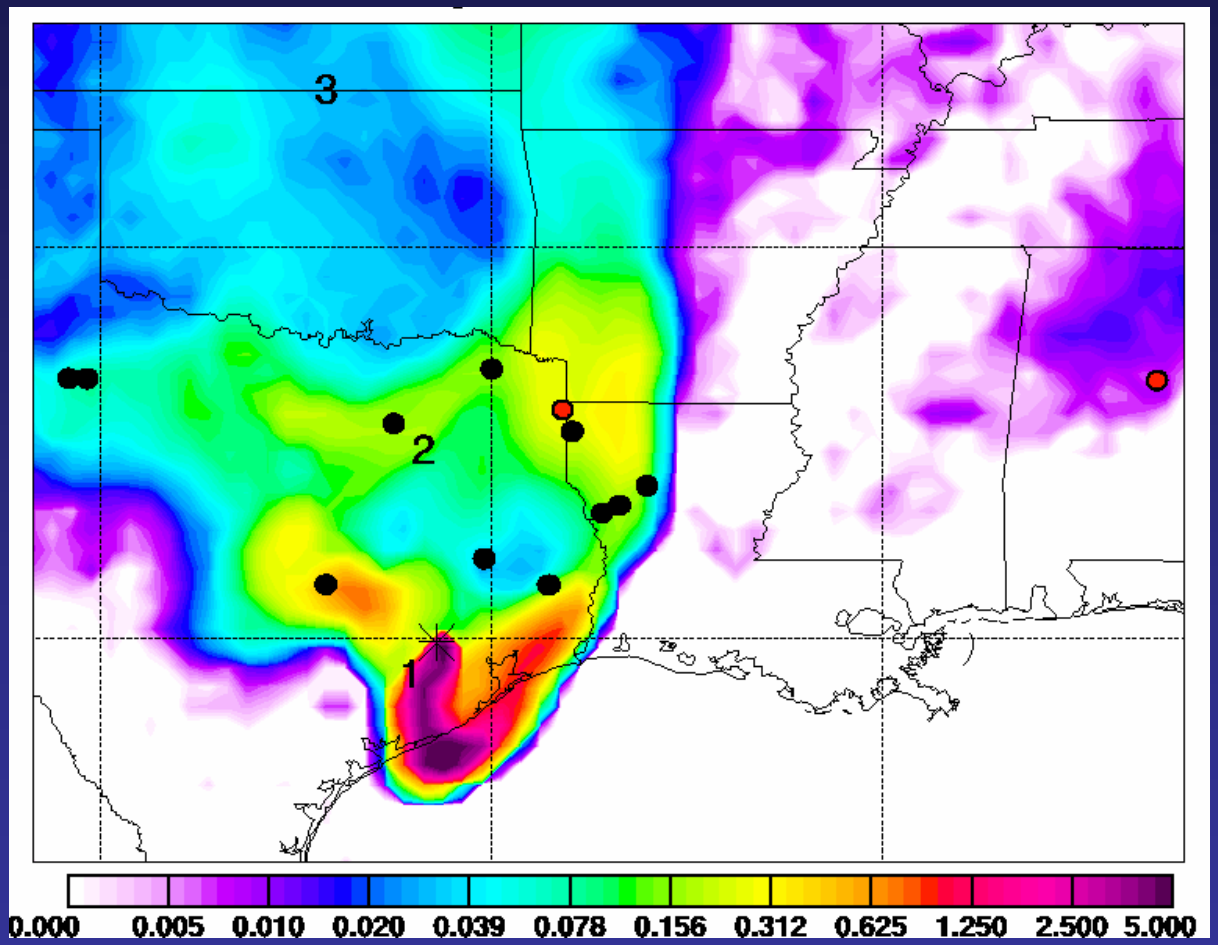


27 September, 2006



Color & size proportional to particle volume. Scales differ.

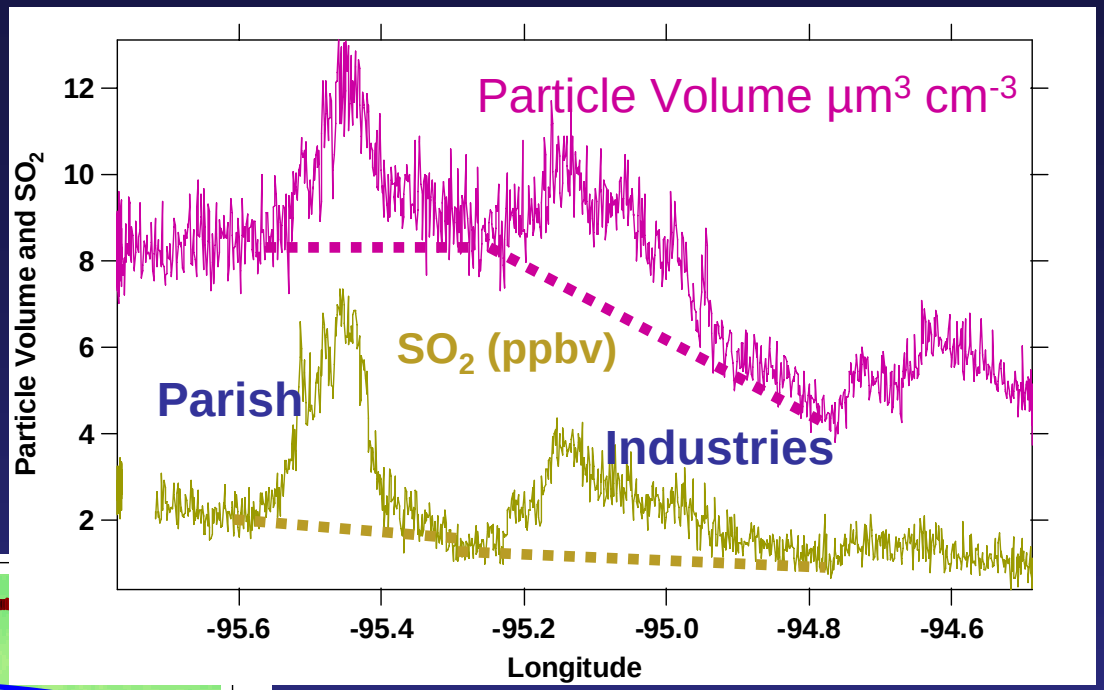
Background variability caused by recirculation of Houston/Beaumont area plume from previous day.

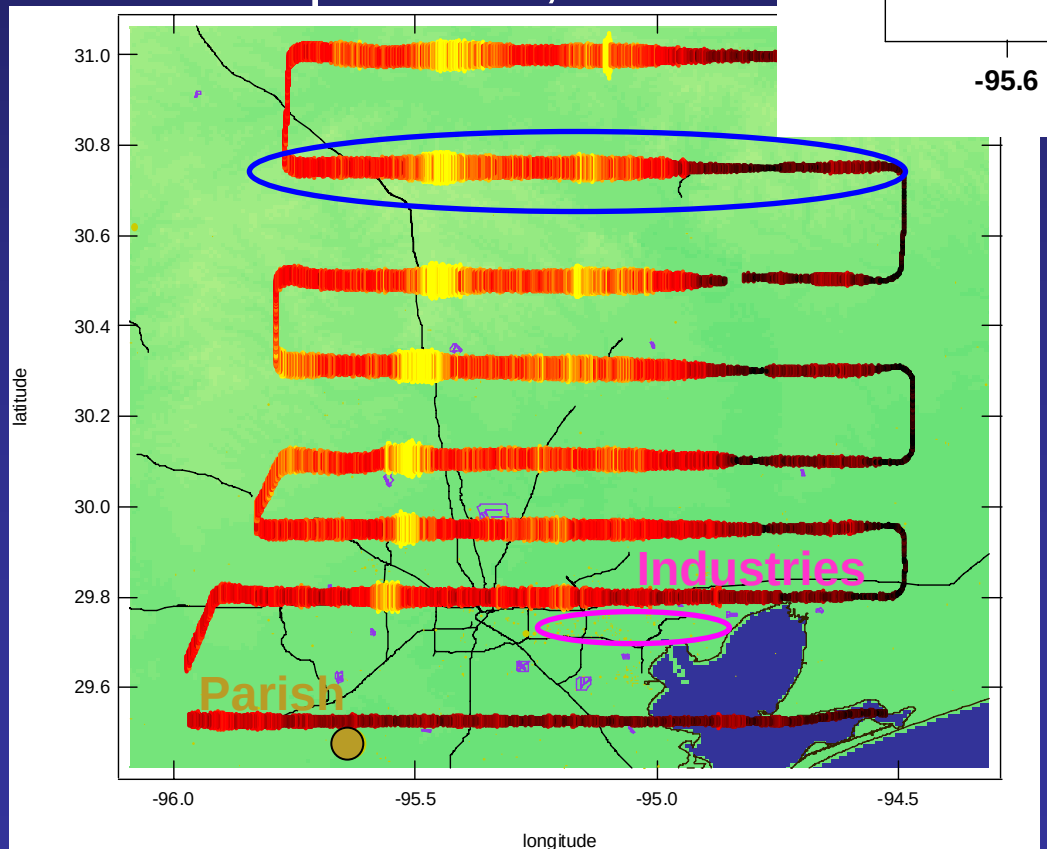


FLEXPART model transport footprint from 40,000 backtrajectories at aircraft location. Color proportional to backtrajectory residence time in lowest 100 meters. Dots are MODIS fire hotspots.

Subtracting the background to isolate Parish and ship channel industrial plumes:

27 September, 2006

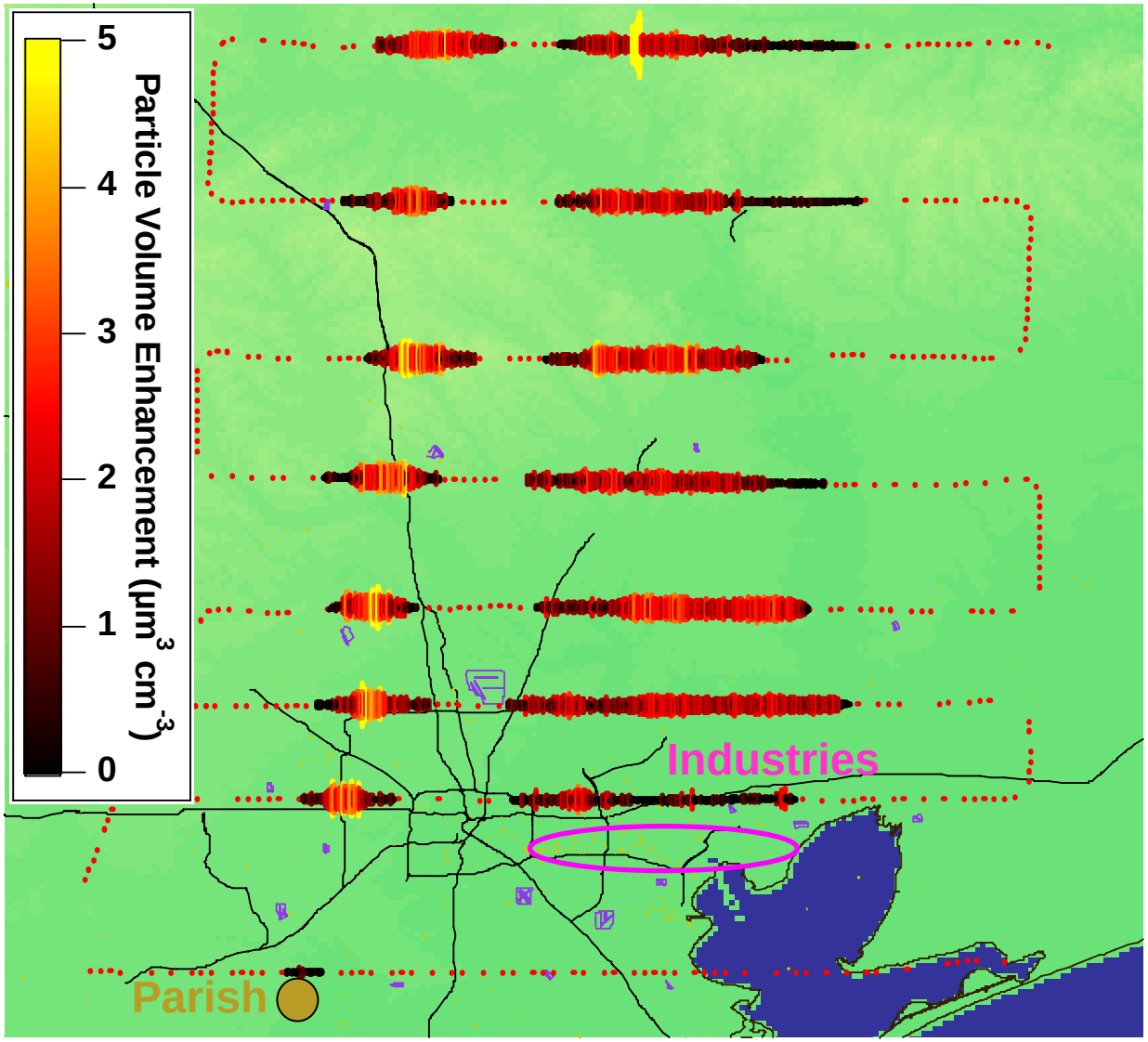




Subjective and prone to error-- >30% in volume?

Particle volume enhancements above background

27 September, 2006

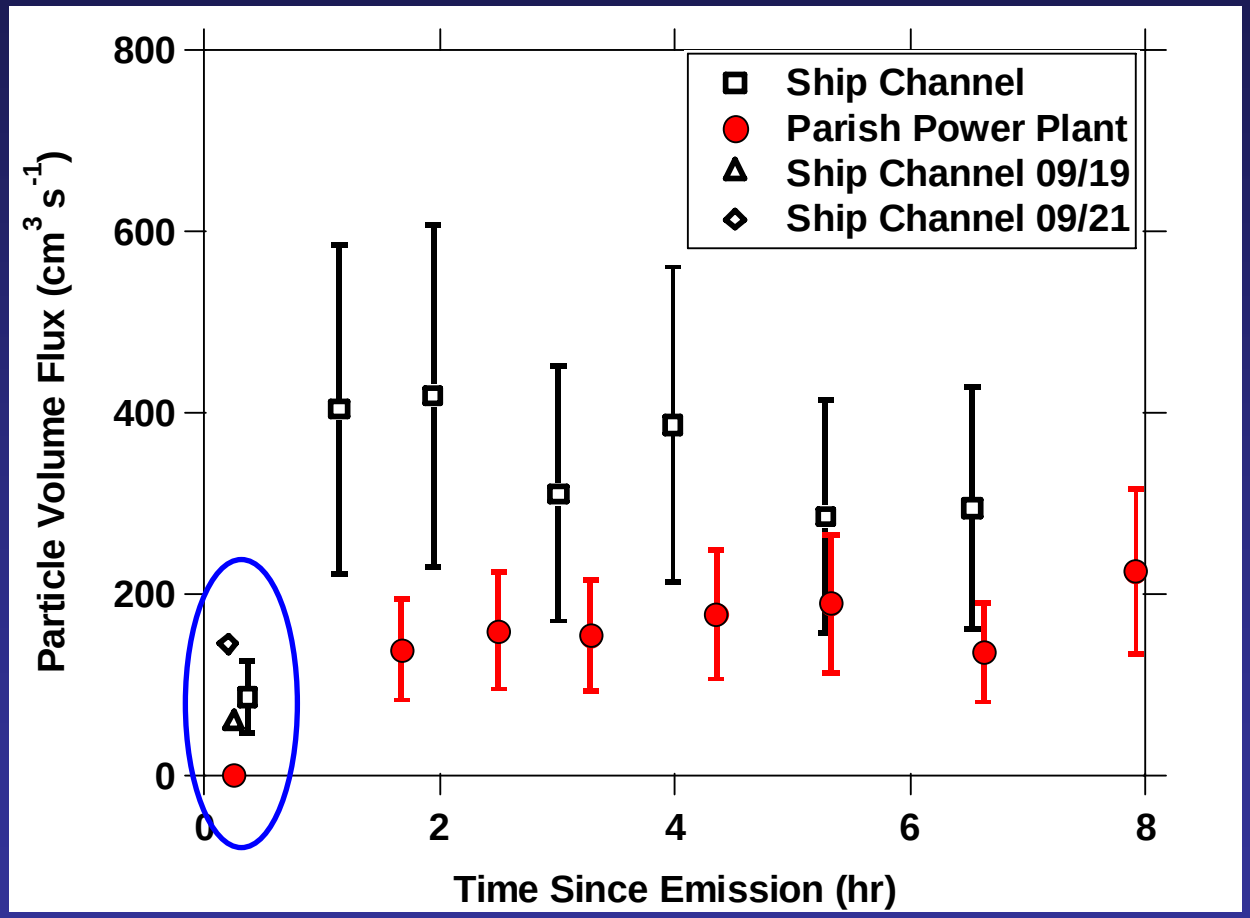


Enhancements are <2 μm^{3}cm^{-3} on average

Growth does not continue far downwind

Very little volume enhancement immediately downwind of sources

Estimate flux of particle volume (mass) in ship channel and Parish power plant plumes

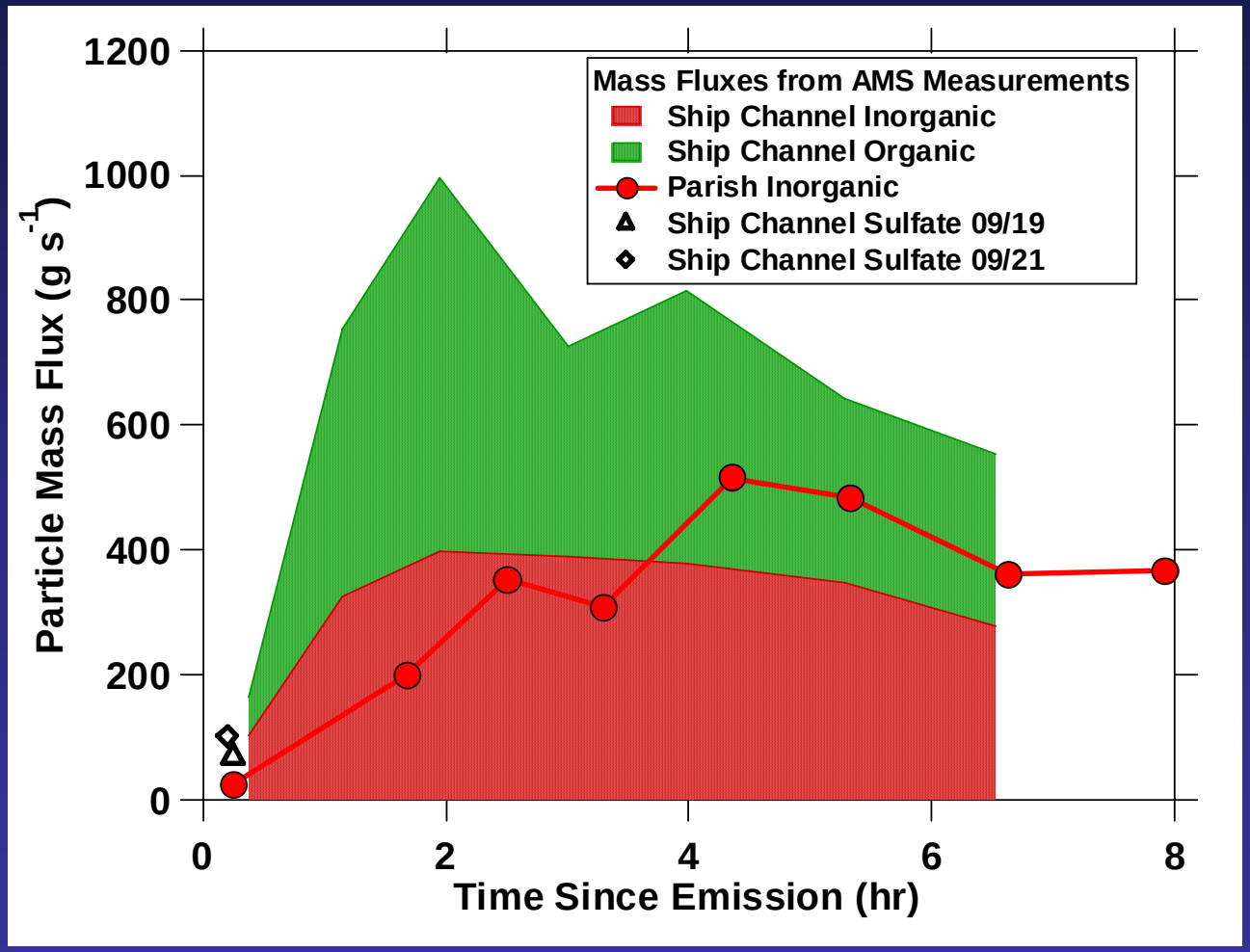


Uncertainties large due to background subtraction, wind speed at time of emission, PBL height (transport time uncertainties not shown)

Ship channel flux ~2x Parish flux

"Primary" mass emissions are small compared with secondary formation, but not negligible in ship channel area

Composition of particles from ship channel and Parish power plant

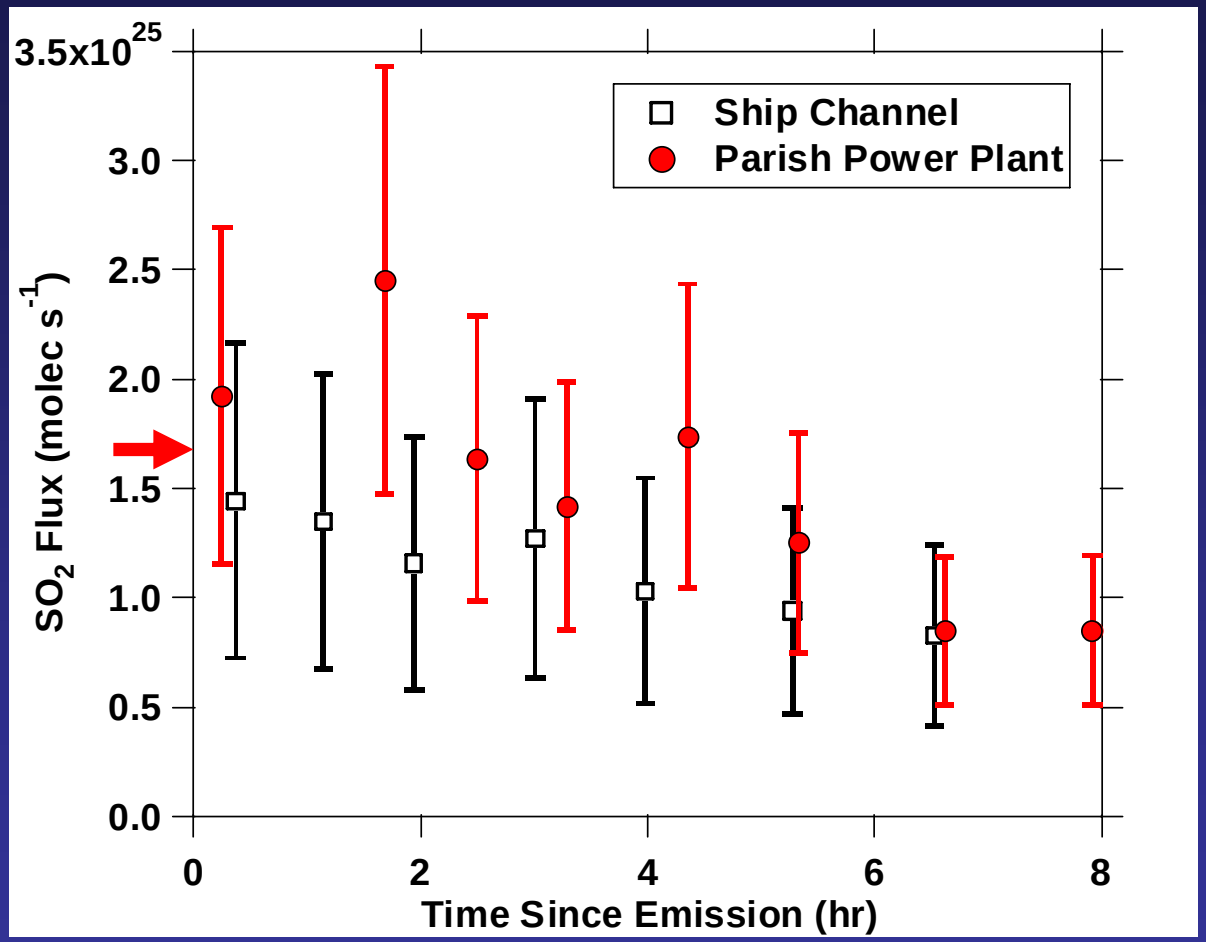


Ship channel particle mass is ~50% organic

Parish plume has no significant organic component

Black carbon flux from ship channel ~10 g s^{-1} (not shown), small but not negligible

Fluxes of SO_2 from Ship Channel and Parish are comparable



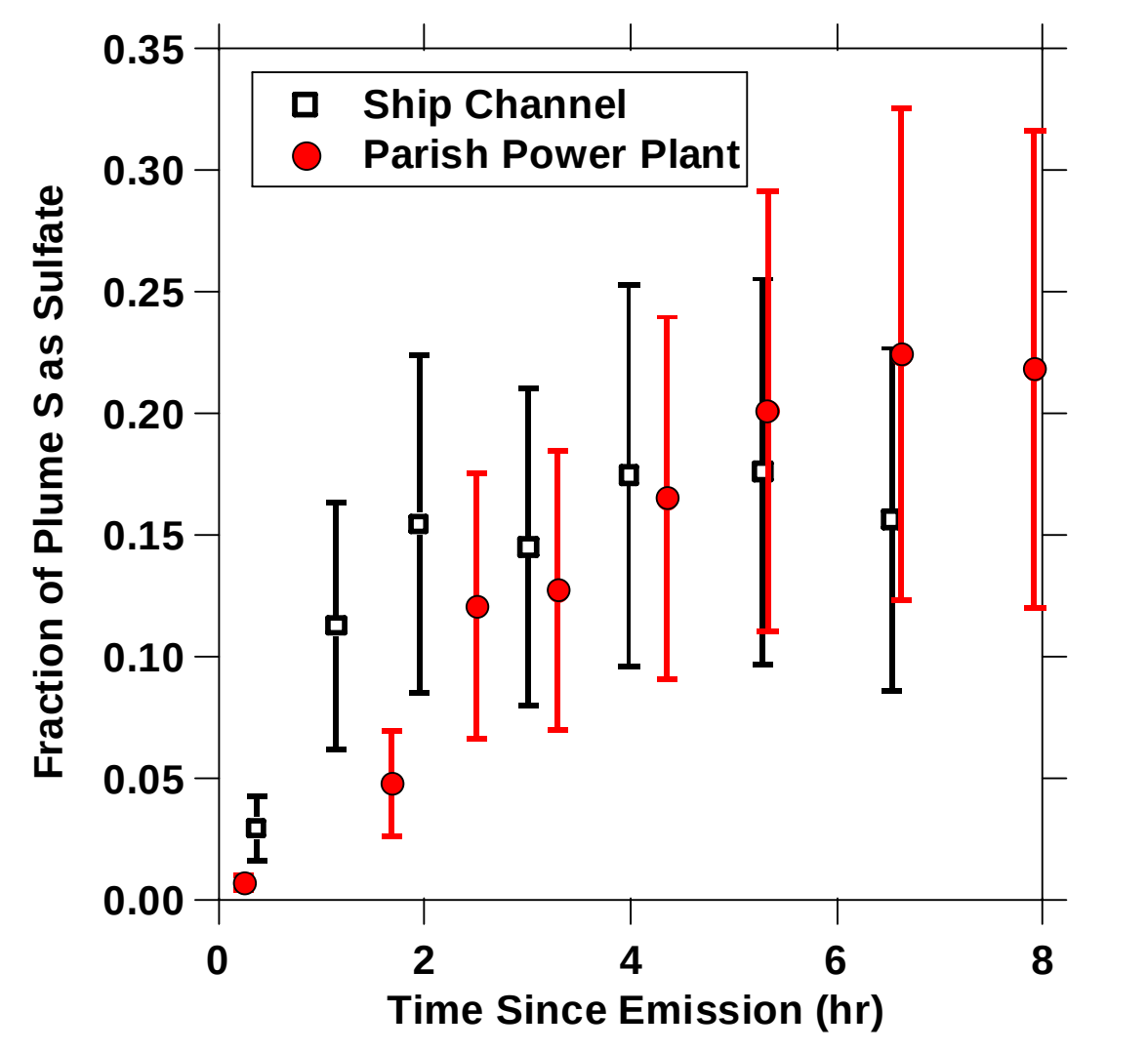
First Parish transect is in agreement with CEMS

Decline in flux with time may be caused by:

- lower fluxes at time of emission (<10% var.)
- dry deposition
- g-p conversion (~20%)
- background subtraction and other analysis biases

See Washenfelder et al. poster

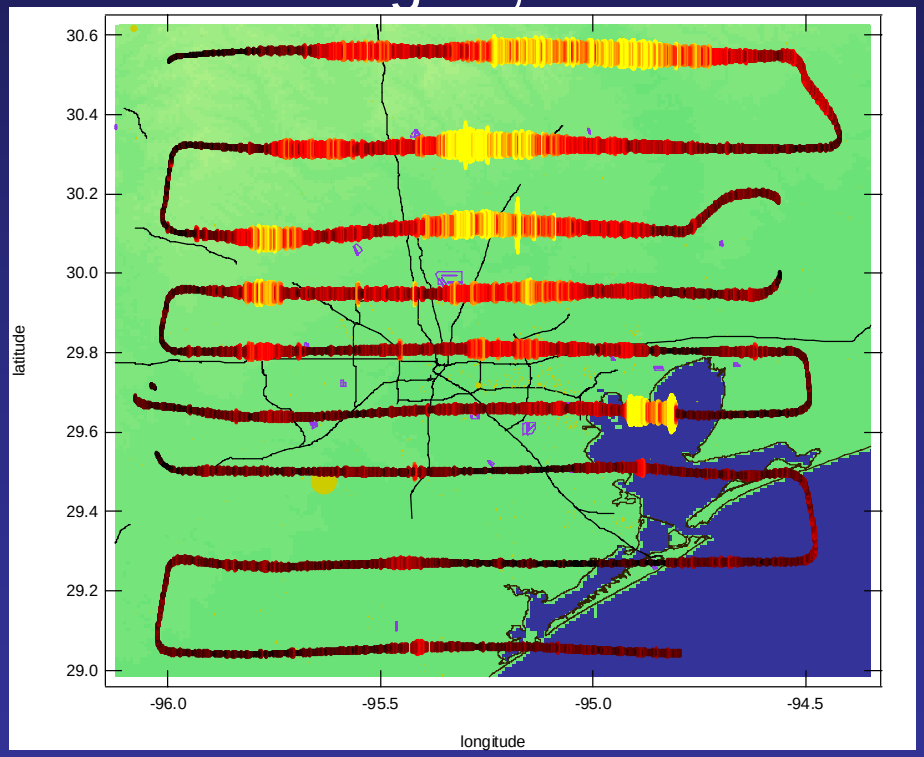
Oxidation of SO_2 to form sulfate is comparable in Ship Channel and Parish plumes



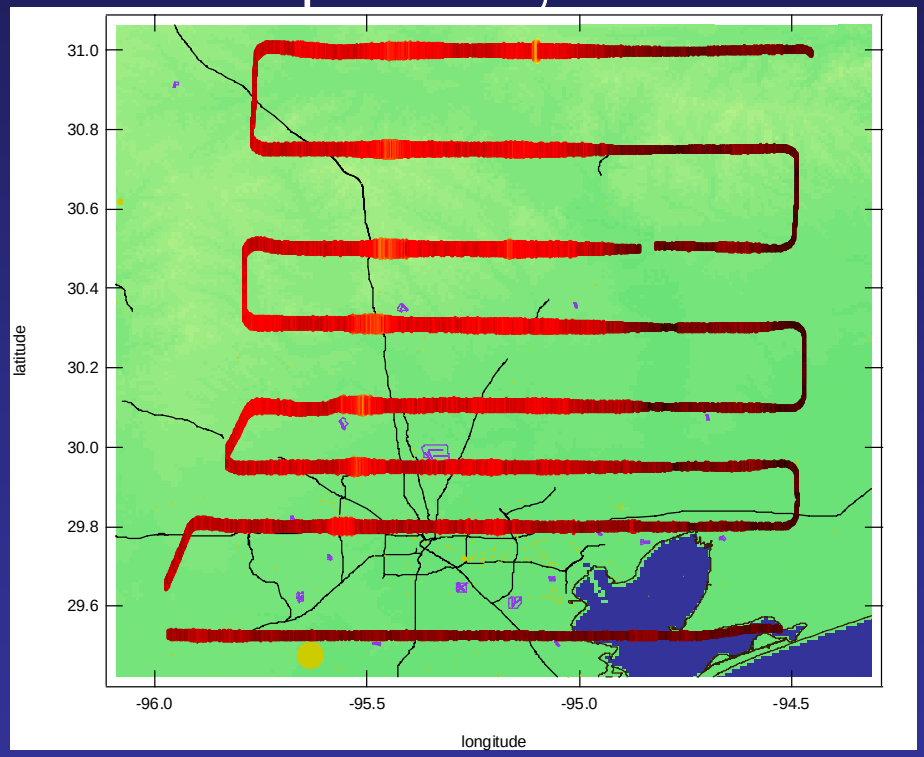
Compare 2006 volume fluxes with 2000

- *Fluxes appear to be 5-10x lower in ship channel plume in 2006*
- *Fluxes 2-4x lower even in Parish plume in 2006*

28 August, 2000



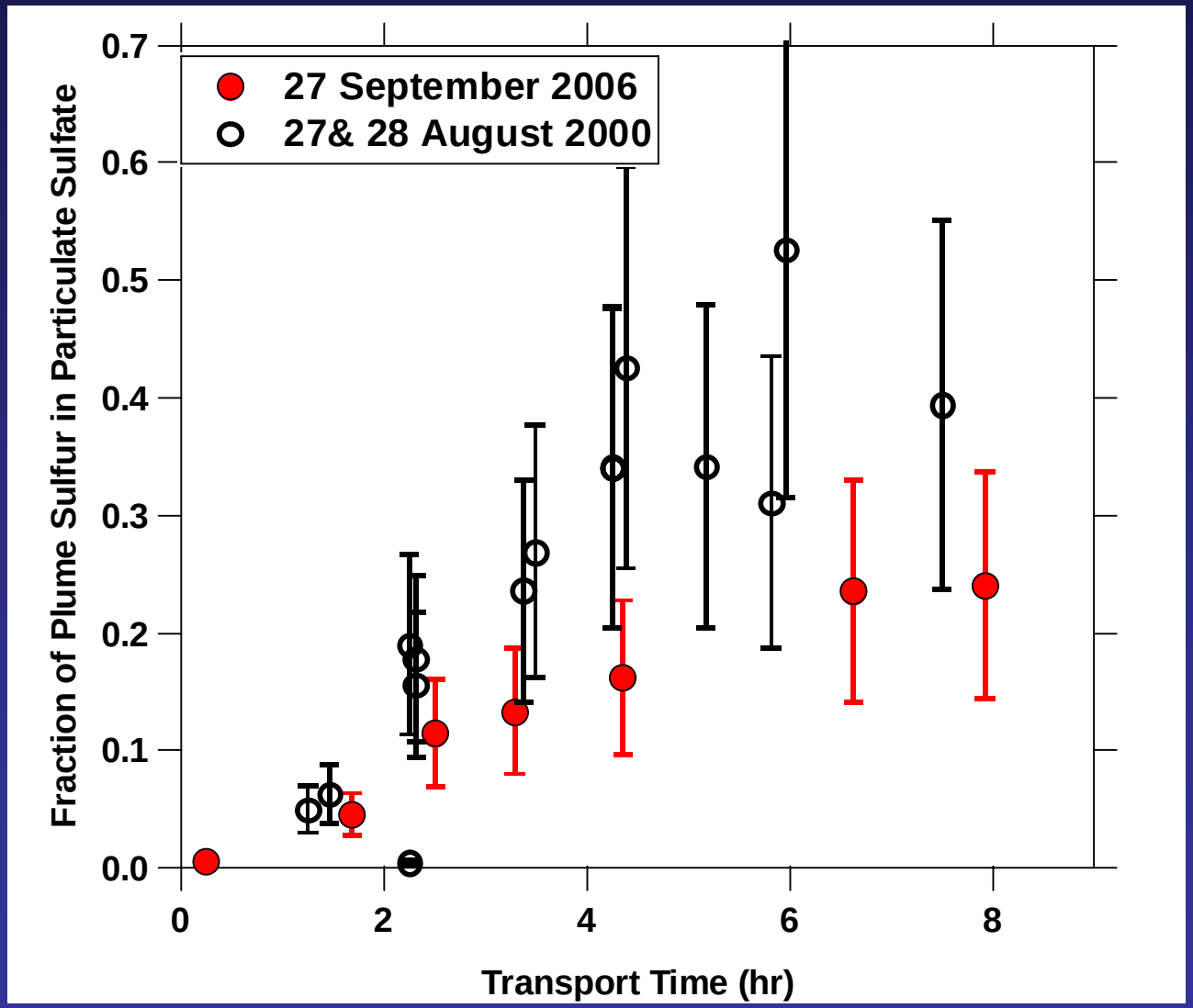
27 September, 2006



Same Scale!

Color & size proportional to particle volume, same dynamic range.

Compare sulfur oxidation in Parish plume 2006 vs 2000



Oxidation is ~2x lower in Parish plume in 2006 than estimated in 2000

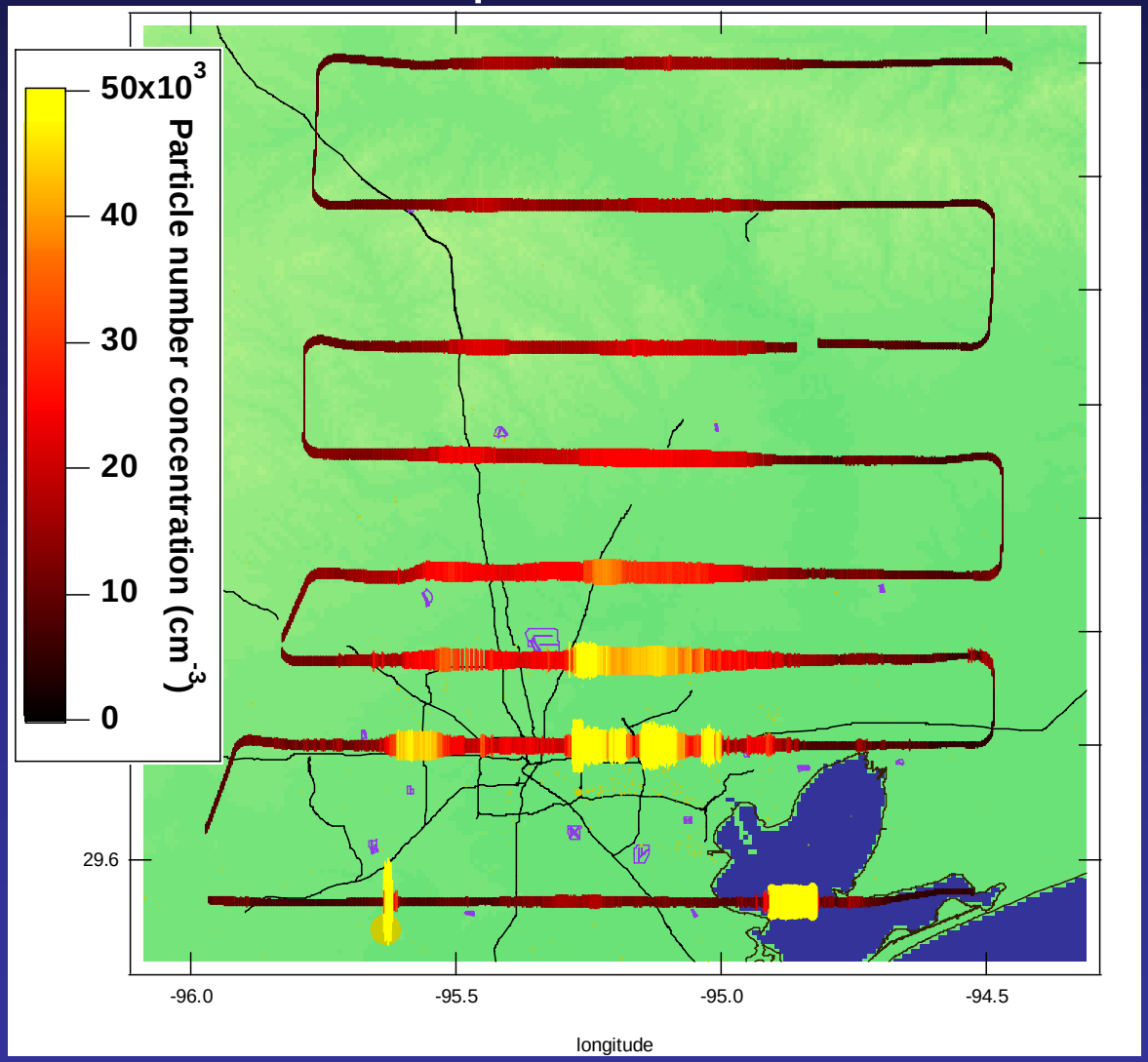
Remember SO_2 flux decreases of ~2x also due to dry deposition or analysis biases.

Many possible causes: need careful evaluation of oxidative environment in 2006 and 2000

Not in time for RSS!

Small particles are rapidly formed downwind of sources, especially of SO_2.

27 September, 2006



Number may be correlated with health effects

As in 2000 data, number maximum occurs immediately downwind of SO_2 sources

Coagulation rapidly depletes number

Map color-coded by particle number concentration

Sources of Primary and Secondary Particles in the Houston Area in 2006

Conclusions

- Secondary sources of particle mass dominate primary sources. "Primary" emissions (<20 minutes old) are *not* negligible in ship channel
- On this day, the ship channel region produced fluxes of particle mass ~2x that of the Parish power plant
- On this day, secondary mass formation ~2-10x lower than on comparable day during TexAQS 2000. Variability driven by meteorology, emissions, photochemistry, analysis biases? Need much more analysis for concrete results