OBSERVATIONS OF PROCESSED ASIAN POLLUTION WITH A HIGH RESOLUTION AERODYNE AEROSOL MASS SPECTROMETER (HR-ToF-AMS)FROM THE C-130 AIRCRAFT DURING THE INTEX-B FIELD CAMPAIGN

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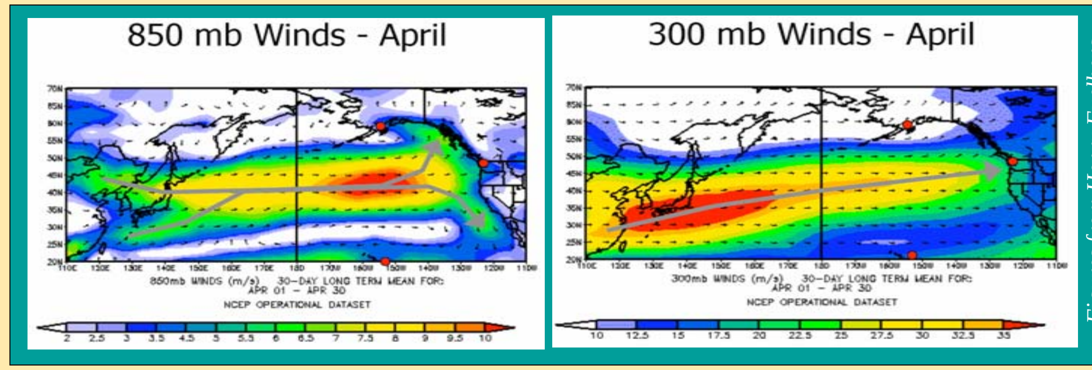
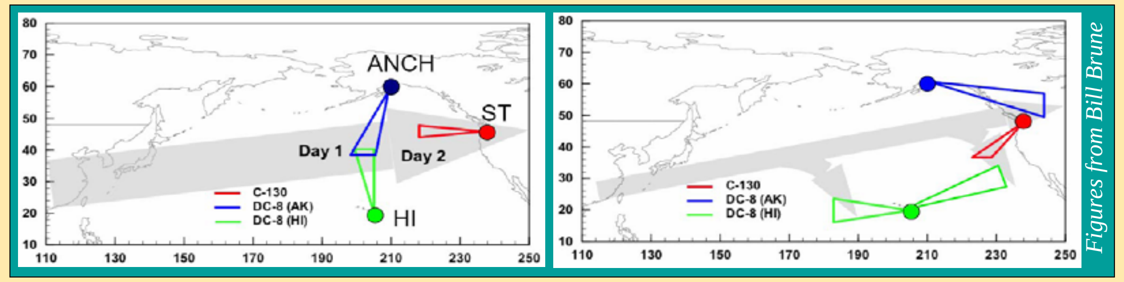
Goals of INTEX-B Campaign

Intercept Asian Pollution During Transport Across Northern Pacific Ocean

- Goal = Quantify amount of Asian pollution transported to North America
  - Asian pollution estimated to enhance N. American surface O_3 by several ppb, aerosol mass loading enhanced by 0.1 μg m^{-3}
  - Improve understanding of chemical transformation of polluted air masses, both aerosol and gas phase, on days to weeks time scale
  - Validation of chemical and transport models & satellite measurements
- INTEX-B campaign during spring 2006
  - Season for most efficient transport across Pacific (see figures below)
  - C-130 based in Seattle; DC-8 based in Hawaii and Alaska
  - Lagrangian experiments coordinated with DC-8 aircraft (see below)
  - Specifically attempted to observe descent of Asian pollution into boundary layer of North America



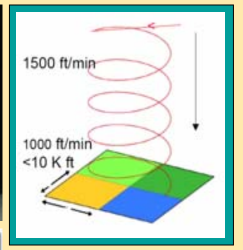
Figures from Henry Fuelberg



Figures from Bill Brune

Intercomparisons

- Satellite
- Ground Sites
  - Mt. Bachelor
  - Mt. Whistler
- Other Aircraft
  - DC-8
  - Duchess (WSU)
  - Canadian Cessna



Theoretical spiral underneath satellite overpass (*Figure from Hanwat Singh*)

C-130 fly-by Mt. Bachelor

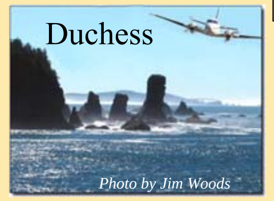


Photo by Jim Woods

Photo by Glenn Wolfe

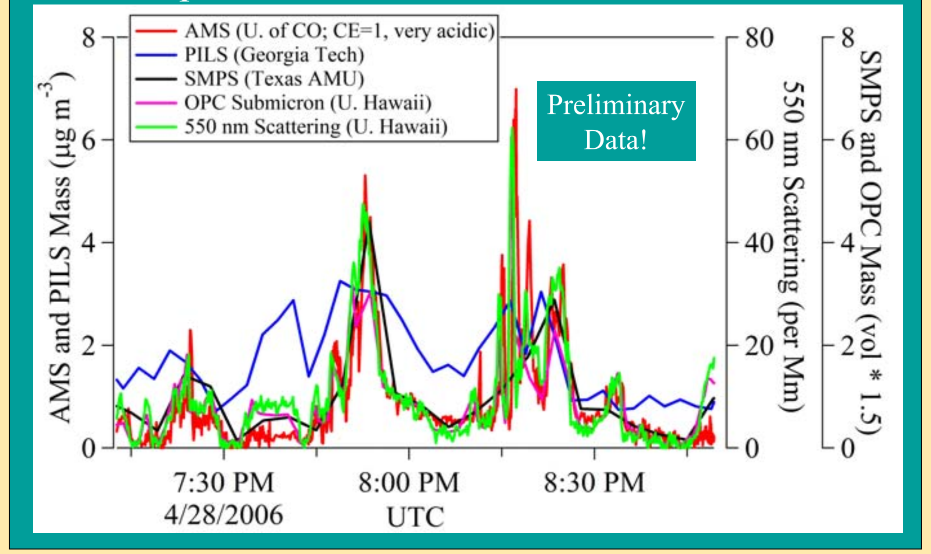
HR-ToF-AMS Installation and Performance

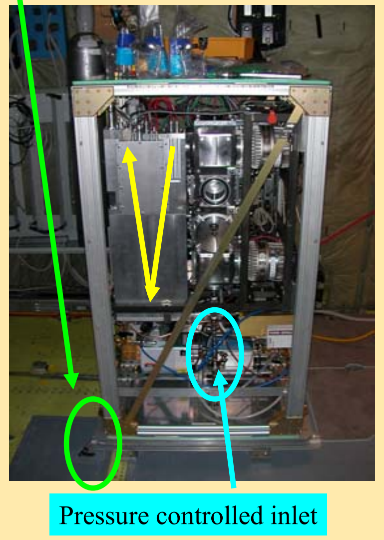
AMS Performance During INTEX-B Campaign

- First deployment of HR-ToF-AMS on airplane during MIRAGE/INTEX
  - For description of HR-ToF-AMS see DeCarlo talk 8D4, Wed. 12:20pm
- INTEX = 12 Flights (> 90 hrs in air)
- **No instrument down time**

For description of MIRAGE campaign, AMS schematic, and further comparison figures, see DeCarlo et al., paper #14H49

Comparison with Other Aerosol Instruments



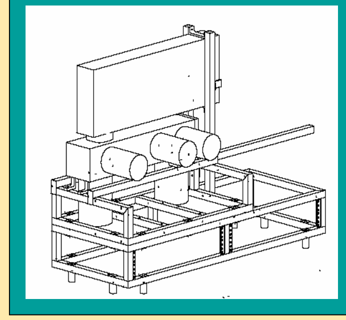


Photos courtesy of P. DeCarlo

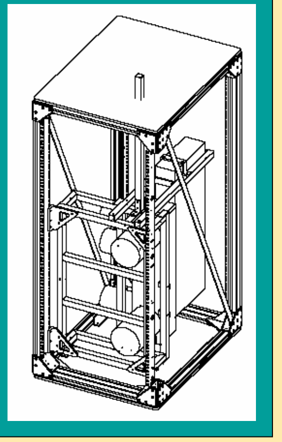
AMS Mounted in HIAPER Instrument Racks

- Custom built internal rack
- Finite element analysis ensured specifications met for HIAPER, C-130 and P-3 aircraft
- Allows mounting in HIAPER racks AND in custom P-3 rack

Instrument Mounting Schematics



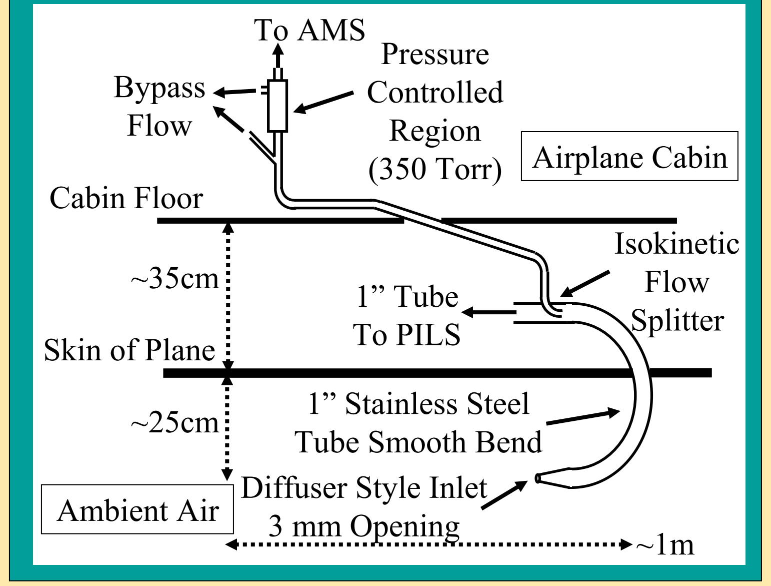
P-3 Installation



C-130 Installation

AMS Inlet System

- 1st deployment of custom designed inlet system
- Belly inlet ~ 50 ft from nose of plane
- Near isokinetic sampling at inlet tip
- Expansion to larger tube at low angle (9.9º)
- 37 lpm total flow through inlet (30 lpm to PILS; 7 lpm to AMS + bypass)
- Cabin and ram heating means dry aerosol sampled

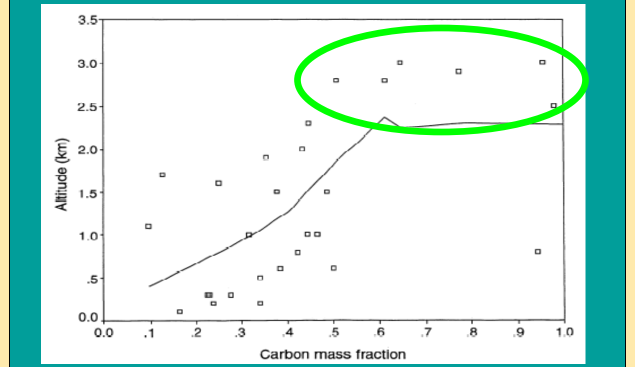


Previous Observations

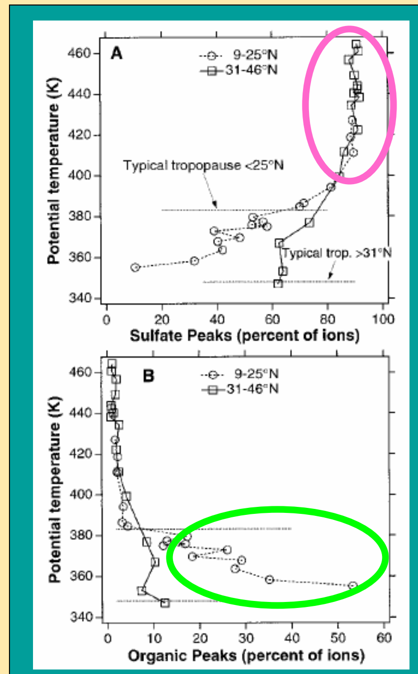
General Observations

- Chemical composition of atmospheric submicron aerosols generally dominated by organics and sulfate
- Organics to sulfate ratio high in free troposphere, low in stratosphere
- Prospero et al., 2003 observe increasing SO_4 at Midway Island → Asian pollution
- Quinn et al., 2000 found large fraction of sub-micron marine aerosol was organic

High Organic Loading in the Free Troposphere



Airborne measurements with nephelometer & grab samples off east coast of U.S.; Novakov et al., 1997

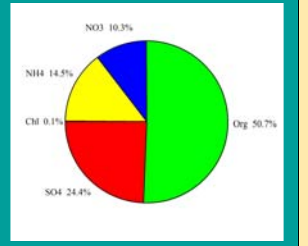


Almost Entirely SO_4 and Water Particles in the Stratosphere

Airborne Measurements with PALMS; From Murphy et al., 1998

High Organic Loading in Tropical Upper Troposphere

AMS measurements of free tropospheric aerosol from Storm Peak mountain-top observatory during INSPECT-II campaign, 2004



From Dunlea et al., in prep.

Asian Transport

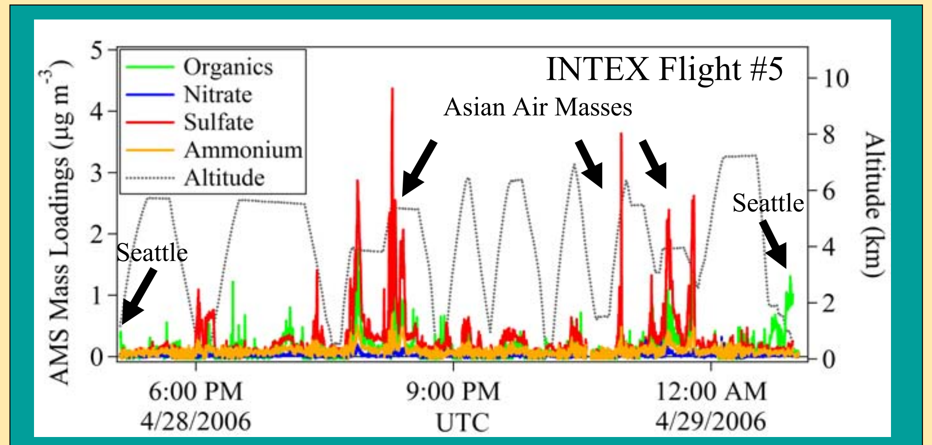
- Brock et al., 2004, ITCT-2k2, observe relative depletion of organics during lifting near Asia; Followed by SO_2 → SO_4 conversion over Pacific
- VanCuren, 2003 measured organics associated with Asian dust in filter samples
- Jaffe et al., 2005 observe organics in Asian outflow over North America
- Heald et al., 2006 model transport of Asian sulfate aerosol with little organics

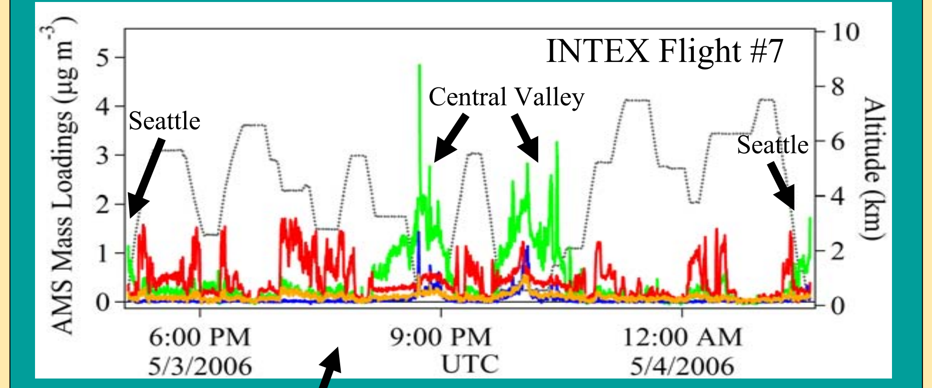
References for Map Below

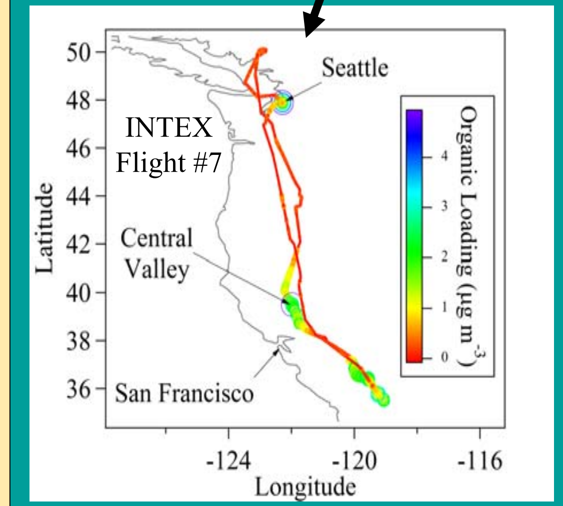
1) Bahreini et al. (ACE-Asia, 2001-Apr), JGR, 108, D23, 8645, 2003
2) Kaneyasu et al., (Sapporo, Japan, 1987 to 1988 all year), Atm. Env., 29(13), 1559, 1995
3) Heubert et al. (ACE-Asia, 2001-Apr), JGR, 109, D19S11, 2004
4) Dunlea et al. (Storm Peak, 2004-Apr), in prep.
5) Jaffe et al., (Crater Lake, 2001 to 2002 all year), Atm. Env., 39, 297, 2005
6) Salcedo et al. (Mexico City, 2003-Apr), ACP, 6, 925, 2006
7) Brock et al. (ITCT-2k2, 2002-May), JGR, 109, D23S26, 2004
8) This Study (INTEX, 2006-Apr), Flight #3
9) This Study (INTEX, 2006-Apr), Flight #4
10)This Study (INTEX,2006-Apr), Flight #5
11)This Study (INTEX,2006-May) Flight #7

Processing of Asian Aerosol During Trans-Pacific Transport

Observations of Organics and Sulfate





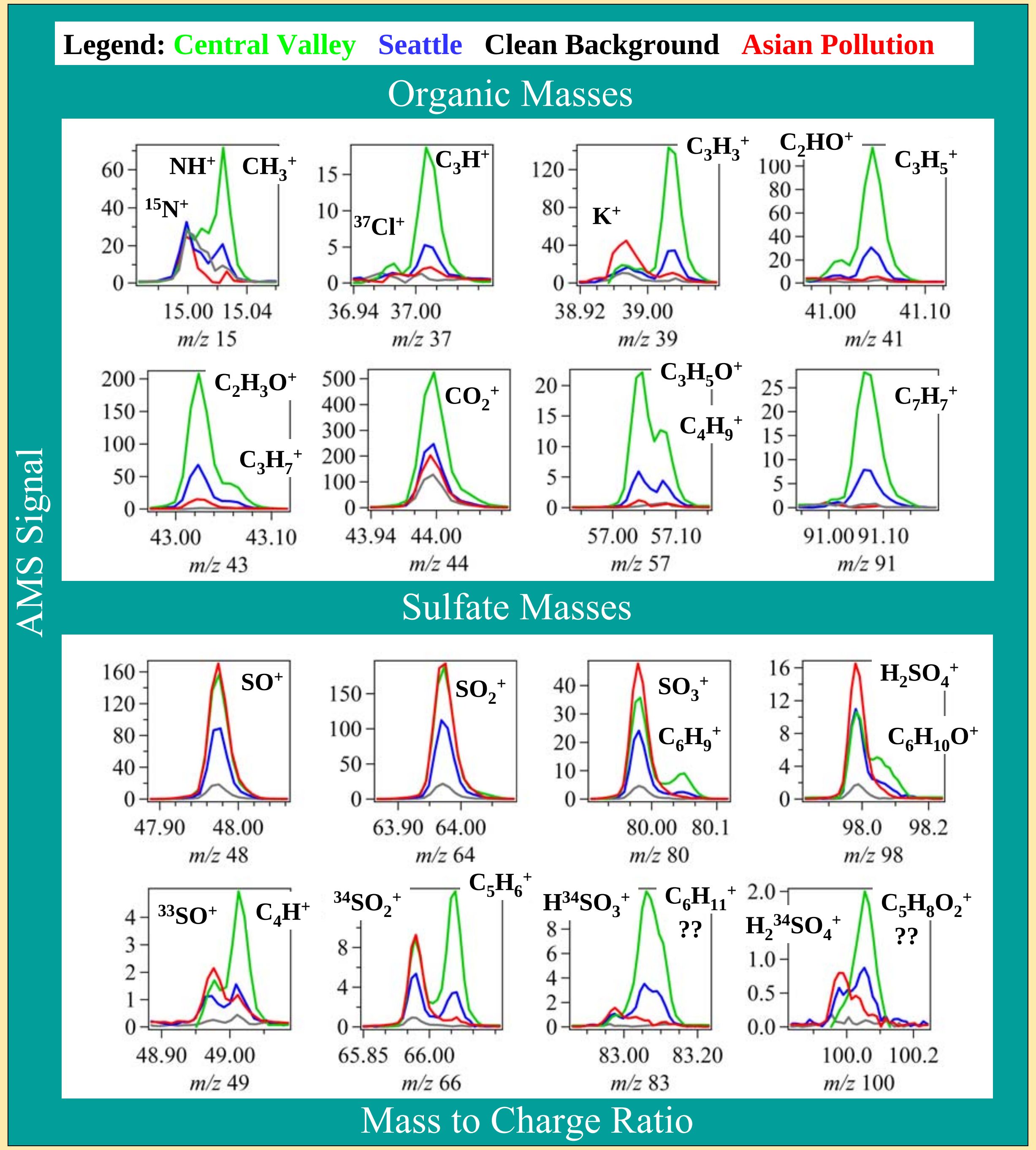


- Asian pollution layers intercepted over ocean (top graph) show clear enhancements in sulfate
- Central Valley and Seattle pollution shows clear enhancements in organics

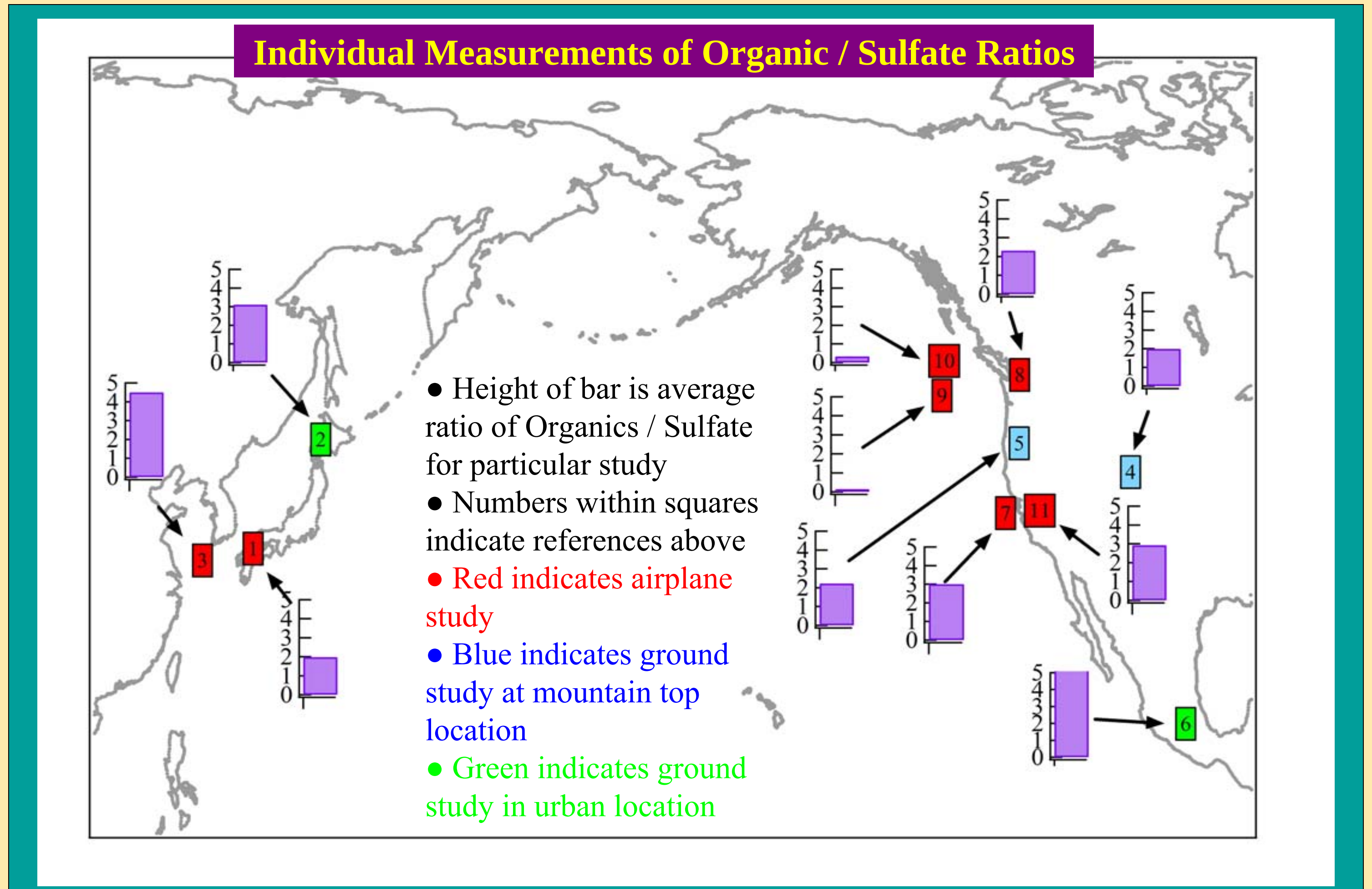
Urban Organic Aerosol and Other Observations

- Flights shown here from:
  - Central Valley (see graph on left)
  - Over Seattle area
  - Clean period over Pacific ocean
  - Asian pollution events (see left)
    - Also identified by CO, NO_y and other tracers during flight
- Central Valley and Seattle are similar in chemical composition
  - Central Valley had higher loadings for these particular flight legs
  - Similar "slightly aged urban" pollution (hours to days downwind)
- Aged urban aerosol characterized by mixture of oxygenated ($C_xH_yO_z$) and non-oxygenated fragments (C_xH_y)
  - See graphs to right for organics
  - $C_xH_yO_z$ slightly higher percentage
- Asian pollution shows enhanced potassium fragment (m/z 39)
  - Known marker for biomass burning, possible evidence for this
- Asian pollution shows complete lack of fresh aromatic fragment (*m/z* 91)
- No obvious organo-nitrate or organo-sulfate peaks found in these flights

High Resolution Mass Spectra



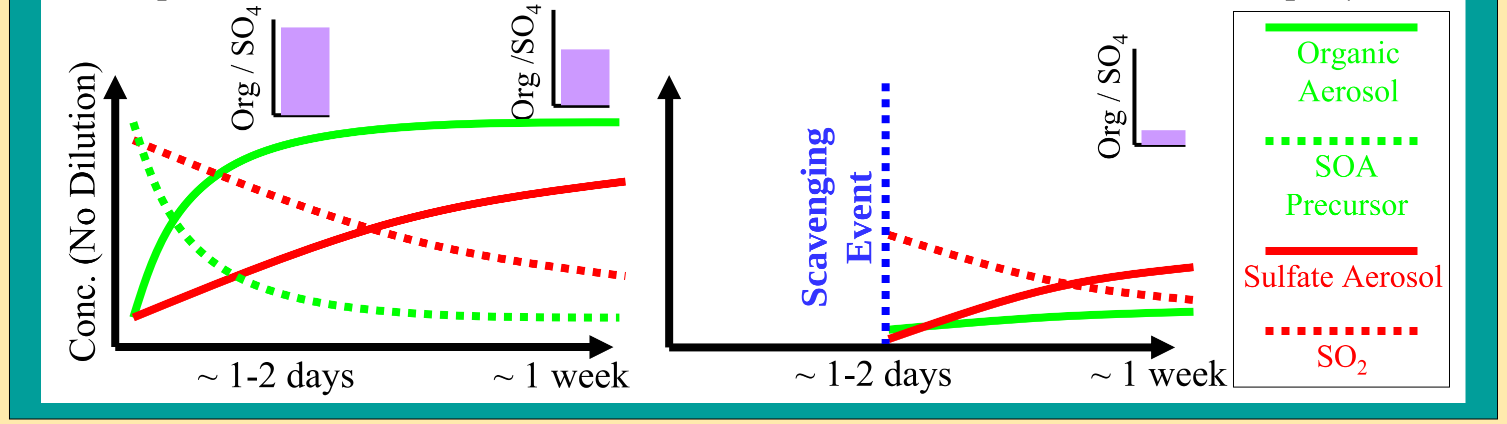
Ratio of Organics to Sulfate Across the Pacific



High Org / SO_4 Ratio Observed Near Asian Sources	Low Org / SO_4 Ratio Observed Over Pacific	High Org / SO_4 Ratio Observed Over North America

Large Asian Emissions of Organic and SO_4 Aerosol and Their Precursors	Scavenging Preferentially Removes Organic Aerosol and/or Organic Precursors Relative to SO_2	New Aerosol Formation over Pacific – Primarily SO_4	Further POA and SOA from North American Emissions

- Idea originally proposed by Brock et al., 2004; our observations expound upon this story
- Organic aerosol formed relatively quickly (precursors consumed) compared to sulfate
- Scavenging event, via lifting in storm system, removes aerosol mass (& organic precursors?)
- SO_2 available to begin aerosol formation after scavenging event → acidic aerosol (H_2SO_4)



Acknowledgments

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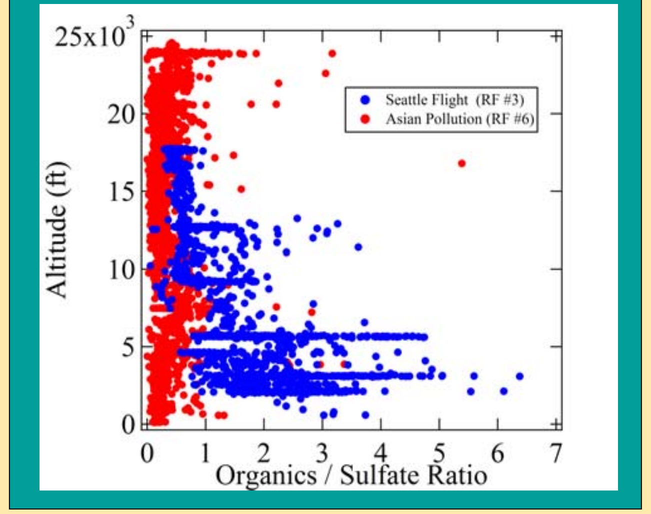

Acidic Aerosol Observed in Asian Pollution

- Low altitude Seattle pollution shows high Org/SO_4 ratio
  - Urban pollution aged ~ 1 day (see graph on left)
- Asian pollution shows low Org/SO_4 ratio
- Asian pollution has less measured ammonium than predicted from inorganic ions (see graph on right)
  - NH_4 predicted = $(36/98)*SO_4 + (18/63)*NO_3 + (18/35)*Cl$
- Asian pollution has enhanced H_2SO_4 and fragments in high resolution mass spectra compared to urban pollution
  - See graphs above (*m/z* 98 and 100 in particular)
- All indicative of <u>acidic</u> aerosol during Asian pollution events