

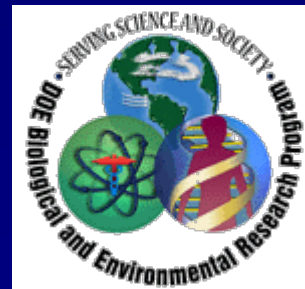
Reconfigured Atmospheric Science Program and 2006 Mexico City Campaign

Jeffrey S. Gaffney

University of Arkansas at Little Rock – Argonne National Laboratory

Department of Energy

Biological and Environmental Research Advisory Committee Washington, D.C. July 10, 2006



ASP Management

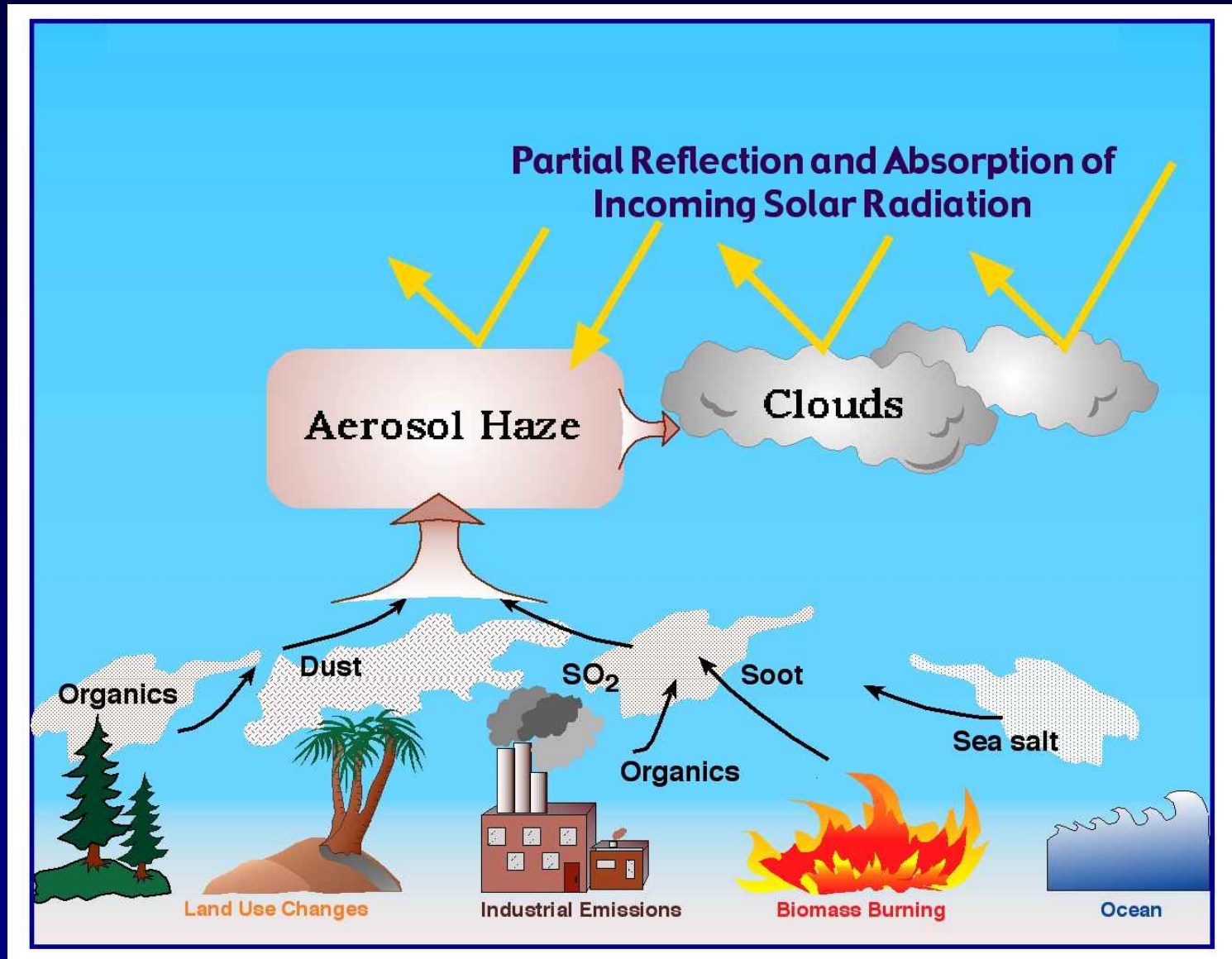
Atmospheric Science Program (ASP)

- **Program Managers**
- Ashley Williamson (DOE-CCRD)
- Rickey Petty (DOE-CCRD)
- **Chief Scientist**
Stephen Schwartz – BNL

ASP Major Milestones

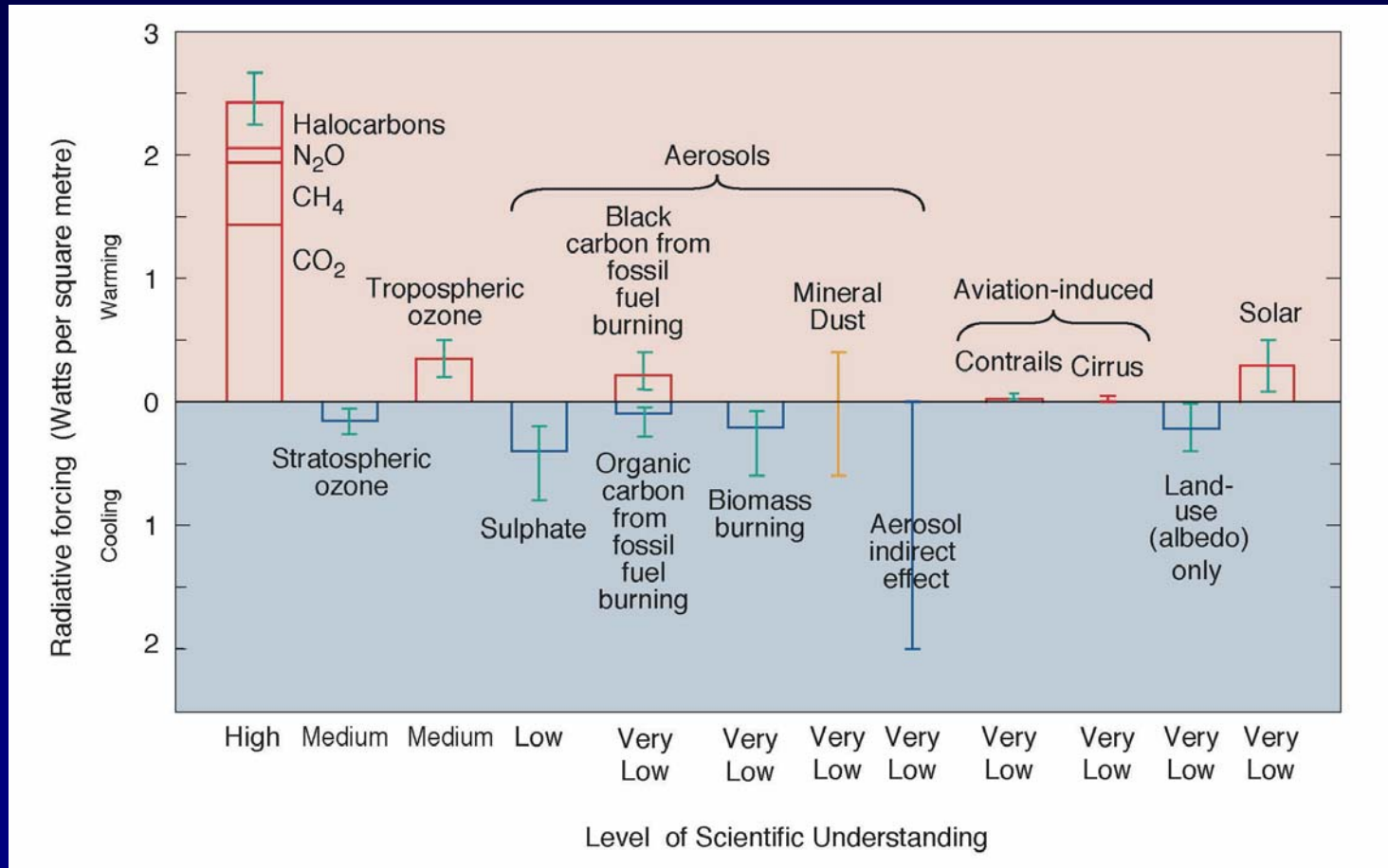
- 2004: March 2 - Chief Scientist selected
April 29 - BERAC approval of ASP reconfiguration
May 4 - Federal Register announcement
June 21 - Proposals due; ~154 proposals received
September 21 - Proposal reviews completed
- 2005: January 25 - Science Team meeting (FY 2005)
May - Black carbon measurement intercomparison
July - MASE - MArine Stratus Experiment
October 31 - Science Team meeting (FY 2006)
- 2006: **March - MAX-MEX Megacity Aerosol eXperiment - MEXico City**
August - MAX-TEX Megacity Aerosol eXperiment - TEXas
October - MILAGRO Data workshop
October - Science Team meeting (FY 2007)
- 2007: Solicitation for FY-08 ASP science team starts
June - CHAPS Cumulus Humilis Aerosol Processing Study

Radiative Forcing by Tropospheric Aerosols



Uncertainties in Global Climate Predictions

Global Mean Radiative Forcing of the Climate System for the Year 2000, Relative to 1750



Largest uncertainties associated with aerosols and clouds (IPCC 2001)

Objectives of ASP

<http://www.asp.bnl.gov>

Aerosol radiative forcing of climate:

- Enhance scientific knowledge needed to represent radiative forcing and other climatic influences of aerosols in climate models

Characterize, understand, and develop model representation of:

- Sources of particles and gaseous precursors
- Aerosol transformations
- Local and regional transport of particles and gaseous precursors
- Concentrations of gas-phase aerosol precursors
- Chemical, optical, microphysical, and cloud nucleating properties of aerosols
- Aerosol influences on atmospheric radiation

Aerosol Influences on Radiation and Climate

Direct Shortwave Radiative Effects (Clear air)

Light scattering – Cooling influence

Light absorption – Warming influence, depending on surface

Indirect Shortwave Radiative Effects – Aerosols influence cloud properties

More droplets – Brighter clouds

More droplets – Enhanced cloud lifetime

More droplets – Broadening of drop distribution – warming

Semi-Direct Shortwave Radiative Effect

Absorbing aerosol heats air and evaporates clouds

Longwave Radiative Effect (Clear sky)

Greenhouse effect of aerosol particles

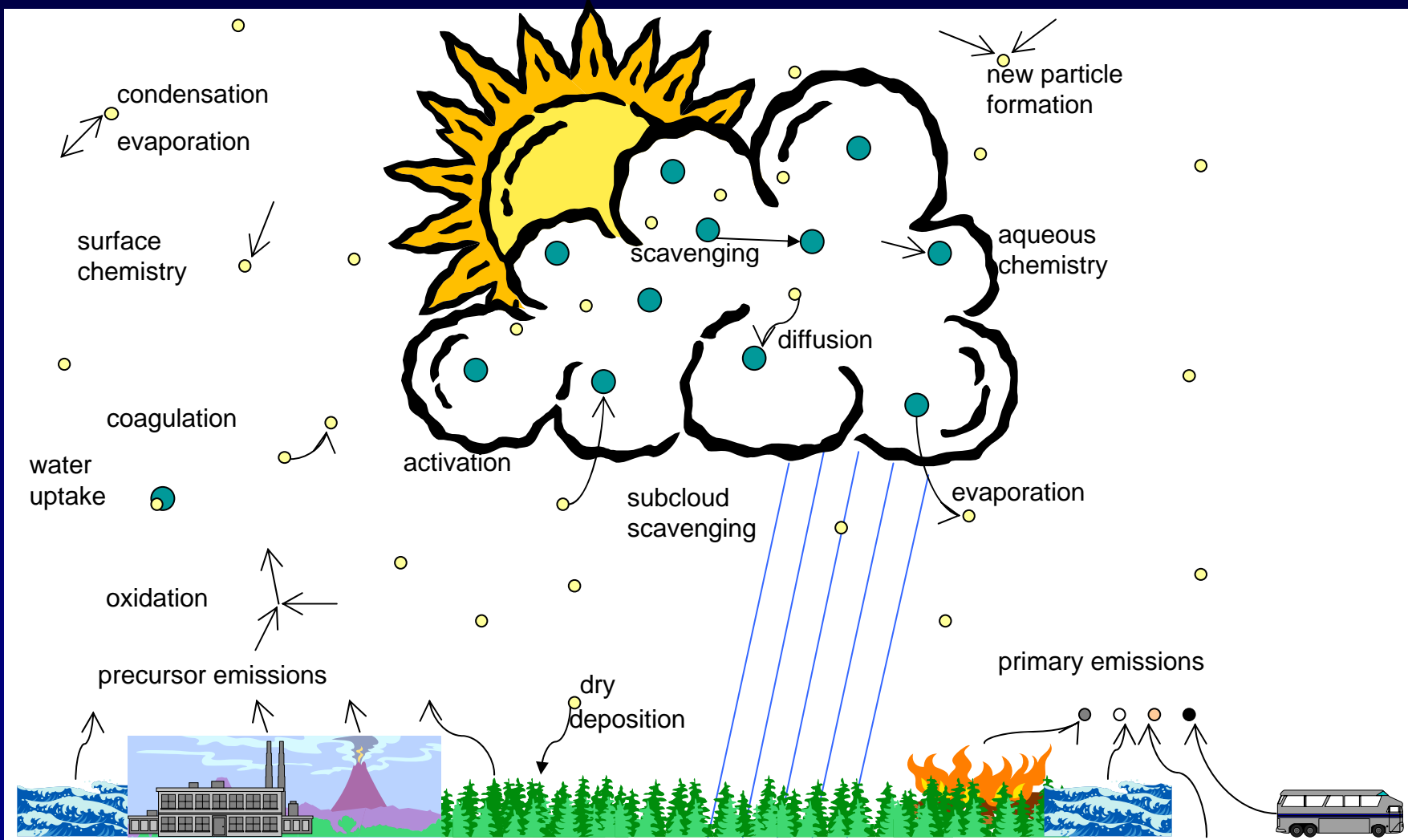
Hydrological Effects

Suppressed surface evaporation - spinning down the water cycle

Displaced precipitation - clouds last longer or evaporate

Aerosol Processes

Processes that Need to be Understood and Represented in Models



Types of Aerosols

Primary Aerosols

Combustion
Diesel Soots
Biomass Burning



Secondary Inorganic Aerosols

NH_4NO_3 , $(\text{NH}_4)_2\text{SO}_4$, etc.

Secondary Organic Aerosol

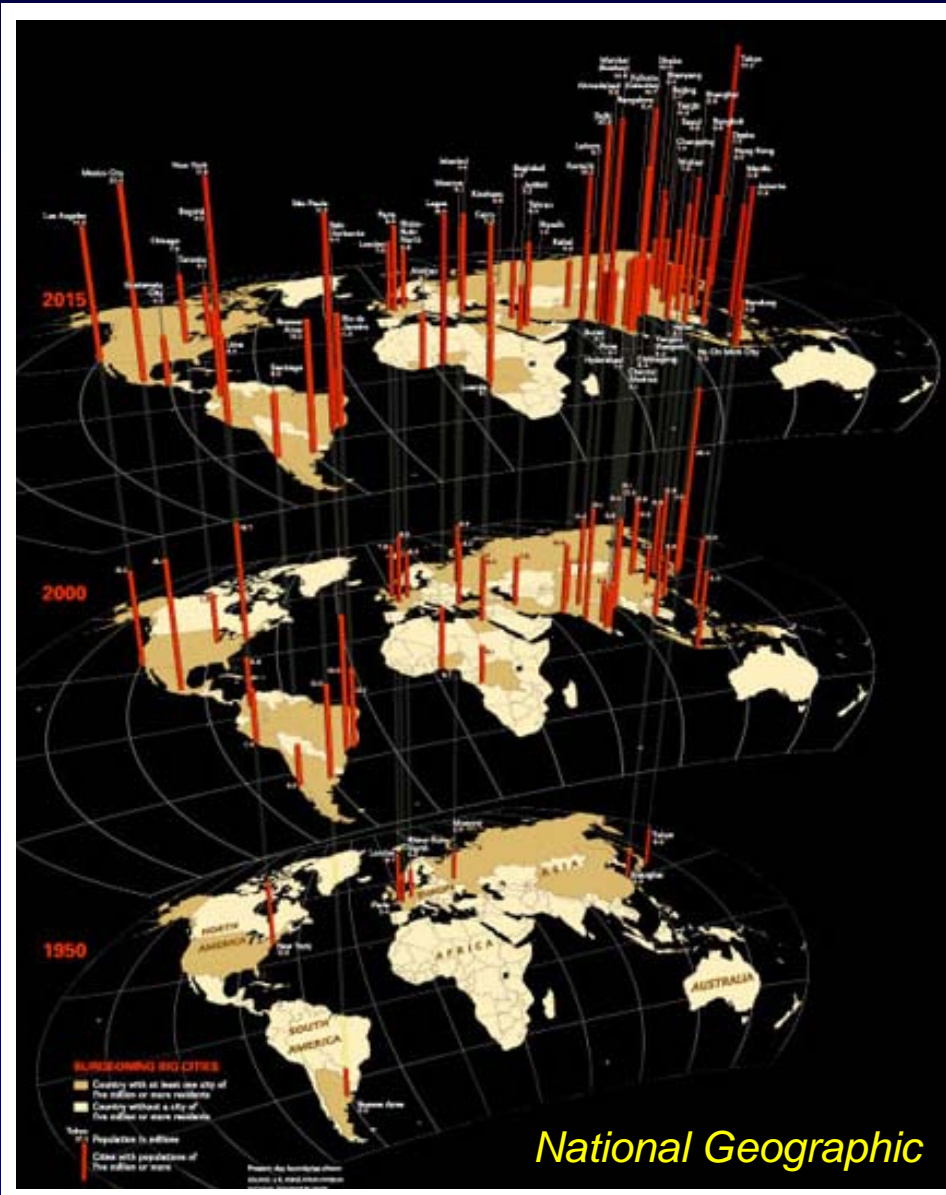
Humic Like Substances (HULIS)
Oxidized Organic Compounds

Ideal Field Study Situation

Attributes

- High aerosol concentrations in representative area - large signal
- High concentrations of primary black carbon (BC)
- High photochemical activity to maximize chemistry changes
- Abundant gas-phase organics to look at secondary organic aerosol (SOA)
- Good meteorology support and infrastructure
- Infrastructure conducive to safe operations
- Ground and aircraft operations - downwind sites

Megacities



Megacities

Population >10 Million

1950 – 1 (NYC)

1995 – 14

2015 – 21

Mini – Megacities

5 Million – 10 Million

1995 – 7

2015 – 37

Asia – Africa

2/3 rural to 1/2 urban by 2025

Sources of Aerosols and Precursor Gases

Megacities and mini-megacities are major sources

Need to better characterize aerosol properties and evolution for models.

Size, composition, size dependent composition, optical properties, cloud-nucleating properties.

High concentrations of aerosols and precursors

Carbonaceous aerosols: organic and black carbon; primary and secondary; fossil, biofuel, and biogenic; fossil and biogenic precursors

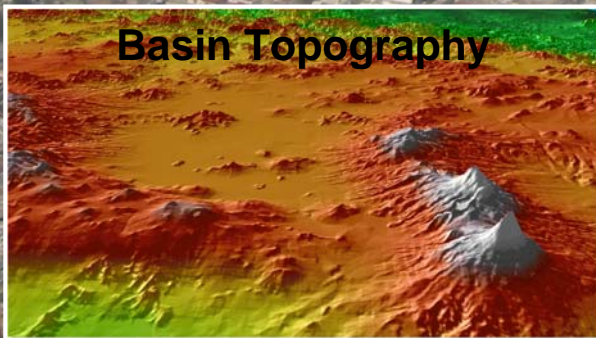
Inorganics: sulfate, nitrate and precursors SO_2 , NO_x .

These sources will be changing over time as cities develop and technologies evolve.

Mexico City

Ideal Field Study Opportunity

- **World's 2nd largest megacity**
- **Largest megacity in North America**
- **Basin meteorology - complex terrain**
- **Infrastructure connections!**
- **Size reasonable for aircraft and ground study**
- **Preliminary ground field studies - 1997 & 2003**



April 2003 Field Campaign

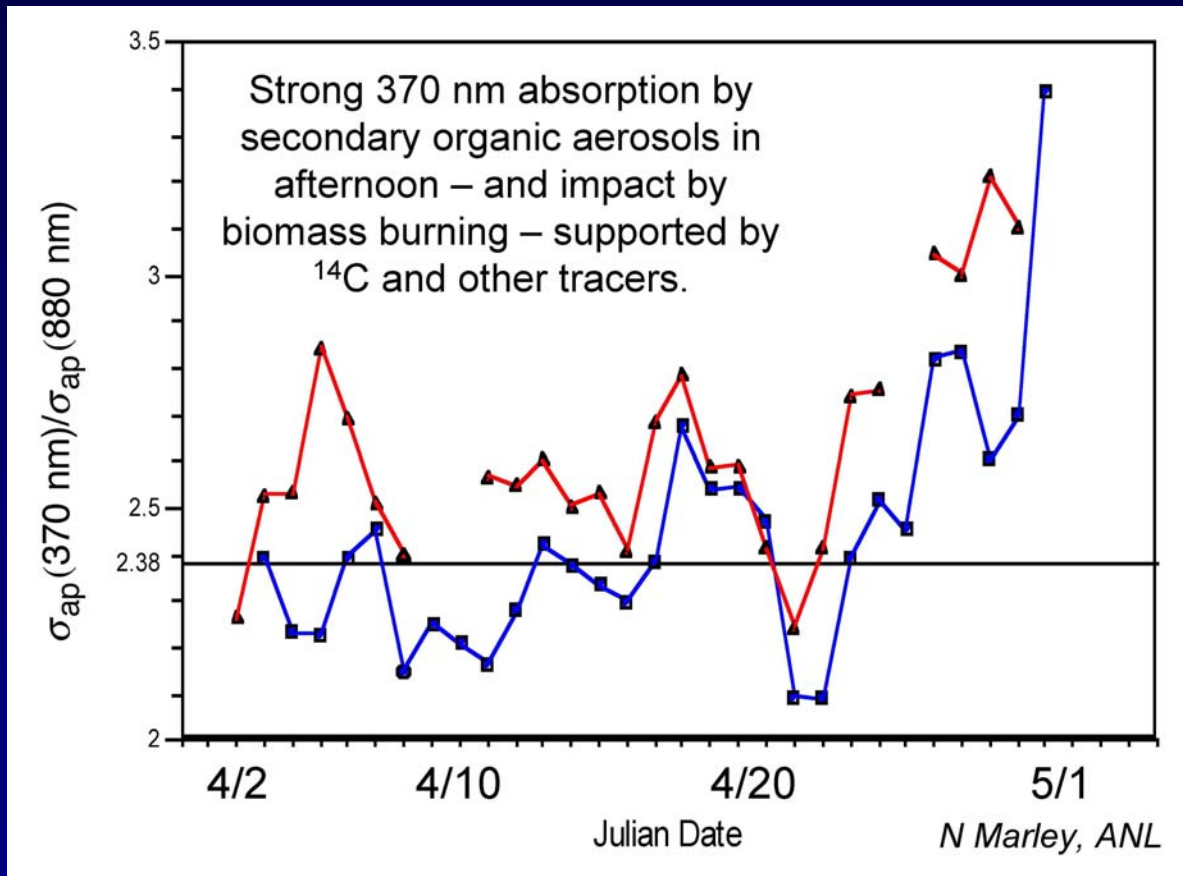
Mexico City Metropolitan Area (MCMA) 2003
DOE Mexico City Megacity 2003

Aerosol Related Findings:

- High BC – High 0.1 - 1.0 μm aerosol concentrations
- Evidence for significant secondary organic aerosol
- Evidence for SOA: 300 - 440 nm absorption
- Evidence for biomass burning – aerosol transport
- High precursor gas concentrations (ammonia and organics)
- Evidence for fresh black carbon resistance to washout
- High OH and HO₂ concentrations – High UV
- 1997 and 2003 – strong diurnal variation – indicating **basin clearing on daily basis**

Aerosol Absorption

2003 – Mexico City Dependence of aerosol absorption on time of day



$1/\lambda$ dependence
for broadband
absorbers

AM (2400 – 1200) and PM (1200 – 2400) average ratios of aerosol absorption
370 nm / 880 nm in Mexico City

MAX-Mex

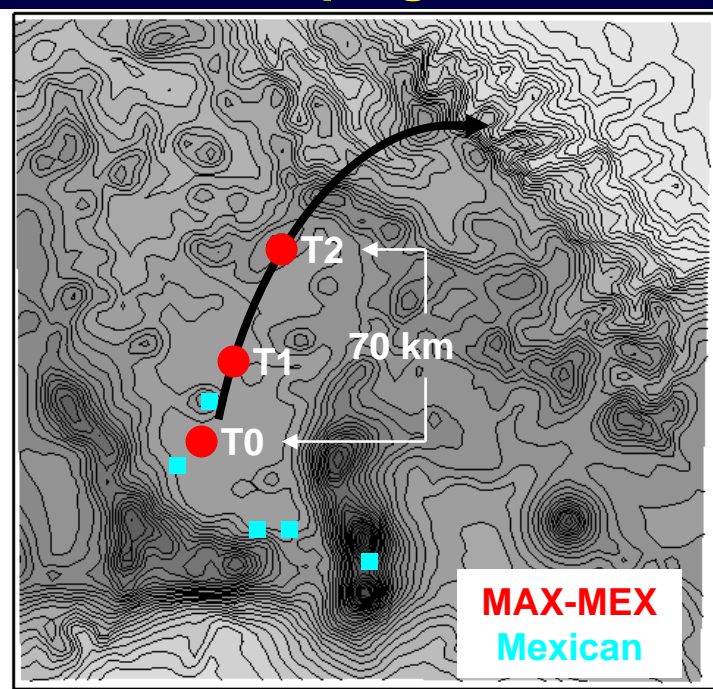
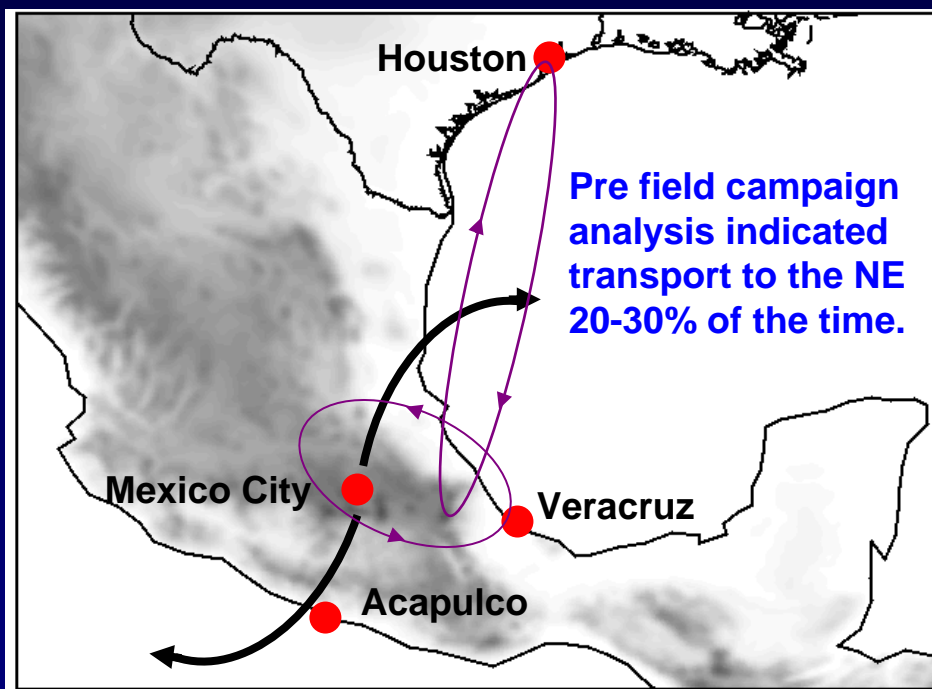
Megacity Aerosol Experiment - Mexico City (MAX-Mex)

Planning Began January 2005

- Jeff Gaffney (ANL) –
Lead Scientist
- Larry Kleinman (BNL), John Hubbe (PNNL) –
Research Aircraft Operations and Flight Plans
- Jerome Fast (PNNL) –
Modeling, Forecasts, Planning for Ground and G-1
- Christopher Doran (PNNL), Will Shaw (PNNL), Rich Coulter (ANL) –
Ground Site Identification, Meteorological Infrastructure Deployment

Pre Field Campaign Activities

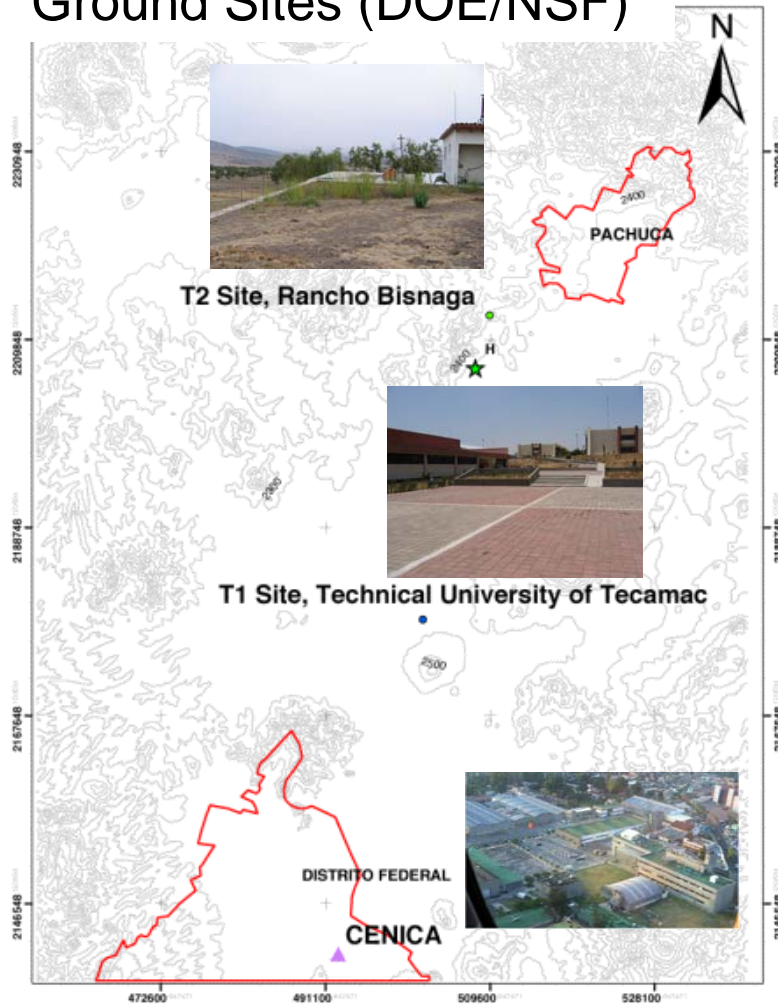
Modeling Used to Target Meteorology, Chemistry, and Aerosol Instrumentation Sites for 2006 Field Campaigns



- Operational rawinsondes (NSF): supplemented to 4 per day at Veracruz and Mexico City and 2 per day at Acapulco
- Aircraft operations: Veracruz – NASA at Houston
- Radar wind profilers: T0, T1, T2, Veracruz
- Microwave radiometer: T0
- GPS radiosondes: T1, T2
- Tethersonde: T1
- Micropulse Lidar: T1

Pre Field Campaign Activities

Ground Sites (DOE/NSF)



Ground Sites Identified in April 2005 – Veracruz Selected for G-1 Operation

Aircraft Operations - Veracruz



MILAGRO - March 2006

Megacity Initiative - Local and Global Research Observations

MCMA-2006 – *Mexico City Metropolitan Area – 2006*

Lead Scientist – Luisa Molina (Molina Center for Energy and Environment, MIT)

Adrian Fernandez – Instituto Nacional de Ecologia

MAX-Mex – *Megacity Aerosol Experiment – Mexico City*

DOE: Lead Scientist, Jeff Gaffney (ANL, UALR)

Program Manager, Rickey Petty

MIRAGE-Mex – *Megacity Impacts on Regional and Global Environments – Mexico City*

NSF: Lead Scientist, Sasha Madronich (NCAR)

Program Manager, Anne-Marie Schmoltnner

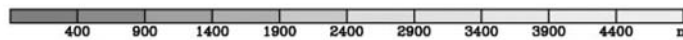
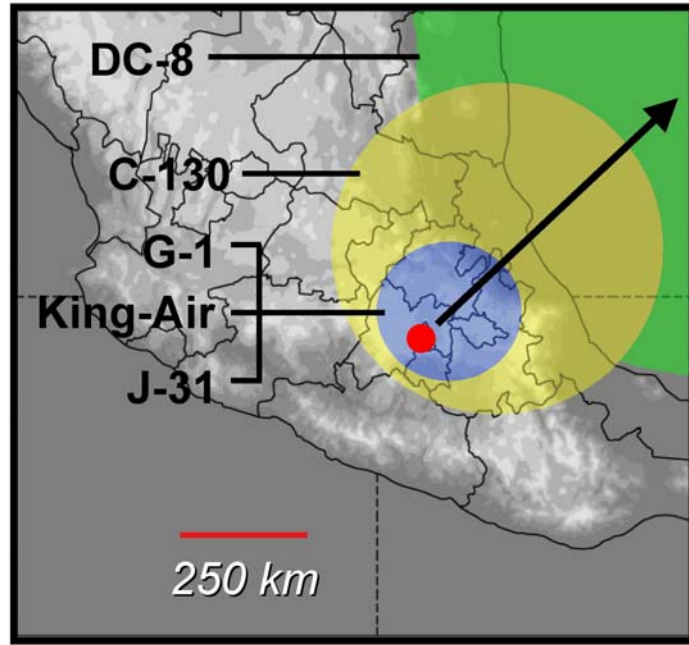
INTEX-B – *Intercontinental Chemical Transport Experiment (NASA, NSF)*

NASA: Lead Scientist, Hanwant Singh

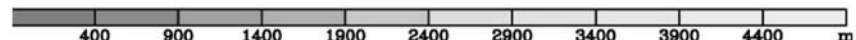
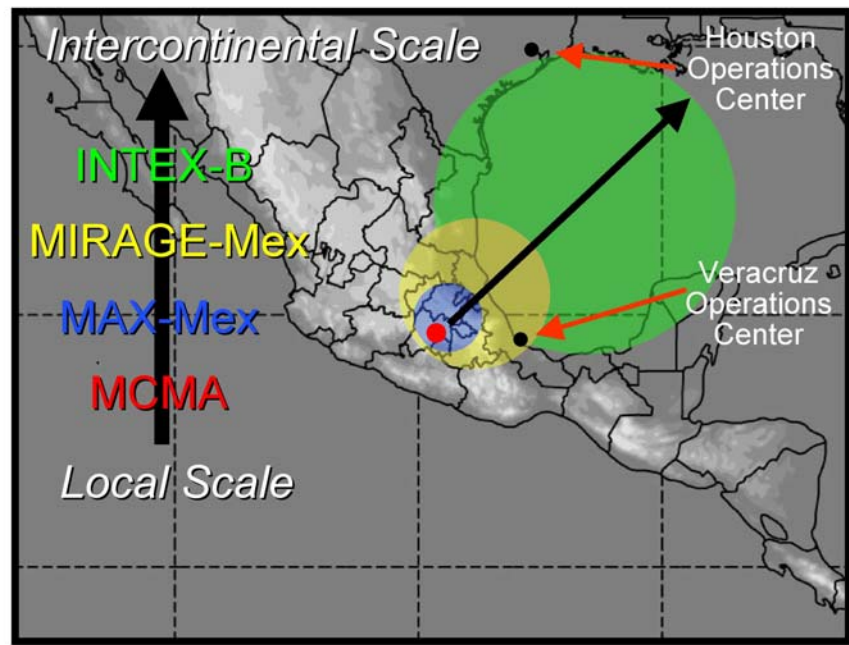
Program Manager, Bruce Doddridge



Geographic Relation of Projects



terrain height (meters)



terrain height (meters)



G-1 (DOE)



C-130 (NCAR)



King-Air (NASA)



Twin Otter (U Montana)



J-31 (NASA)



DC-8 (NASA)

MAX-Mex Science Tasks, Issues, Questions

Characterize aerosol size-dependent composition

Internal mixture vs external mixture

Water uptake dependence on relative humidity

Characterize aerosol optical properties and dependence on controlling variables

Composition, size dependence, size-dependent composition, humidity

Effects of chemical processing/aging

Contribution of BC and species other than BC to absorption

Characterize aerosol cloud nucleating properties and dependence on controlling variables

Composition, size dependence, size-dependent composition, humidity

Effects of chemical processing/aging

MAX-Mex Science Tasks, Issues, Questions, Cont.

Characterize and quantify secondary aerosol formation and aerosol evolution

New particle formation vs condensational growth

Role of coagulation in modifying size and composition distribution

Mechanism(s) of new particle formation and responsible species

Dependence on gas-phase precursors

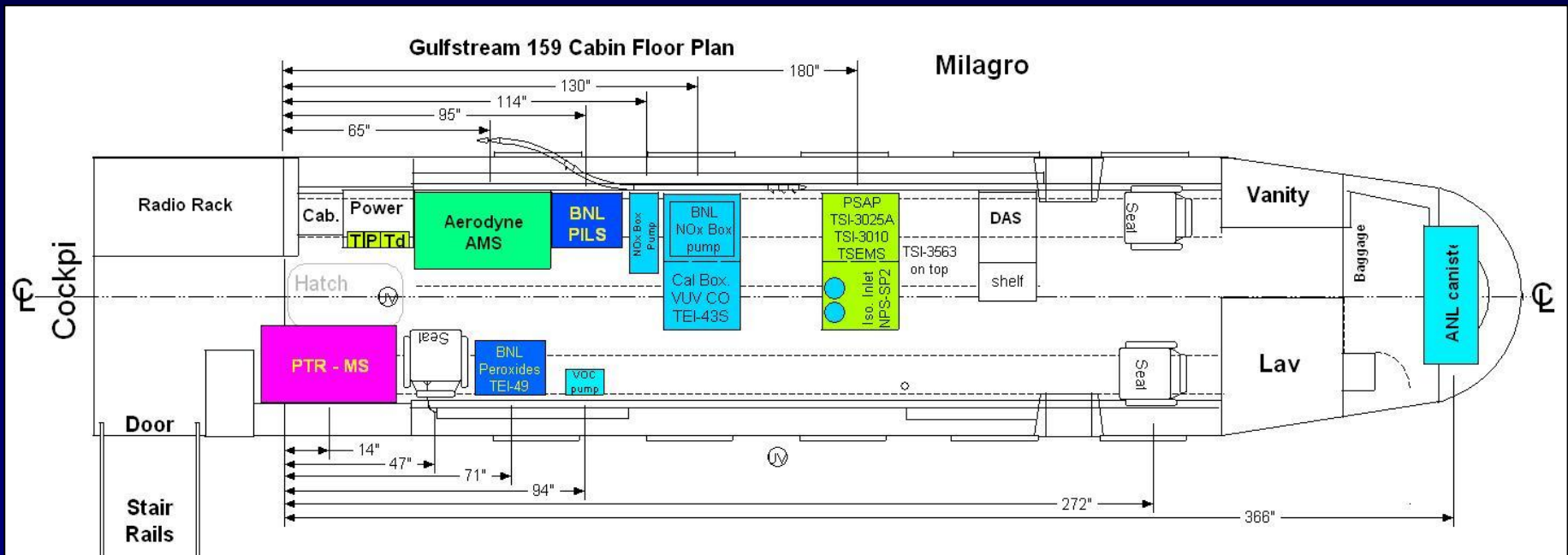
Urban vs regional vs global impacts – Effects of transport and scale for aerosol forcing

Spatial Impacts – Horizontal and Vertical – Temporal

DOE MAX-Mex Participants

- 63 Scientists
 - 3 Field sites
 - 2 Aircraft
 - 1 Mobile van
-
- MILAGRO total: 300 plus scientists
 - 6 aircraft
 - Multiple field sites
 - Multiple mobile vans
-
- Data to be shared among all participants of MILAGRO

ASP G-1 Research Aircraft Facility Layout

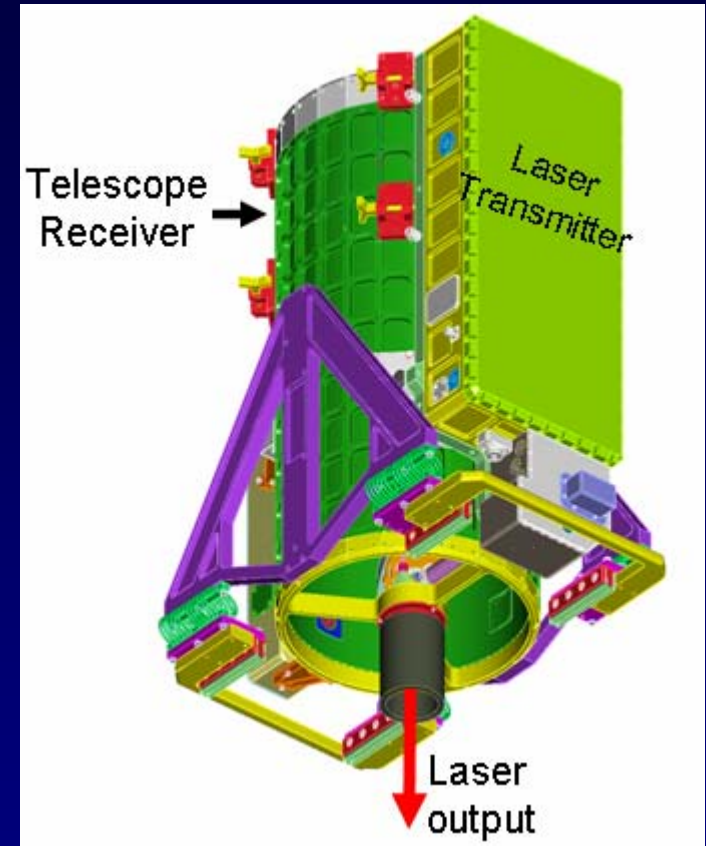


PCASP, CAPS – PNNL, BNL: Senum, Hubbe
 State – PNNL: Hubbe
 PTRMS - EMSL: Alexander, Ortega
 AMS - Aerodyne, EMSL: Alexander, Jayne
 Peroxides - SUNY, BNL: Lloyd, Bowerman
 VOCs – York: Hubbe, Rudolf
 PILS – BNL: Lee

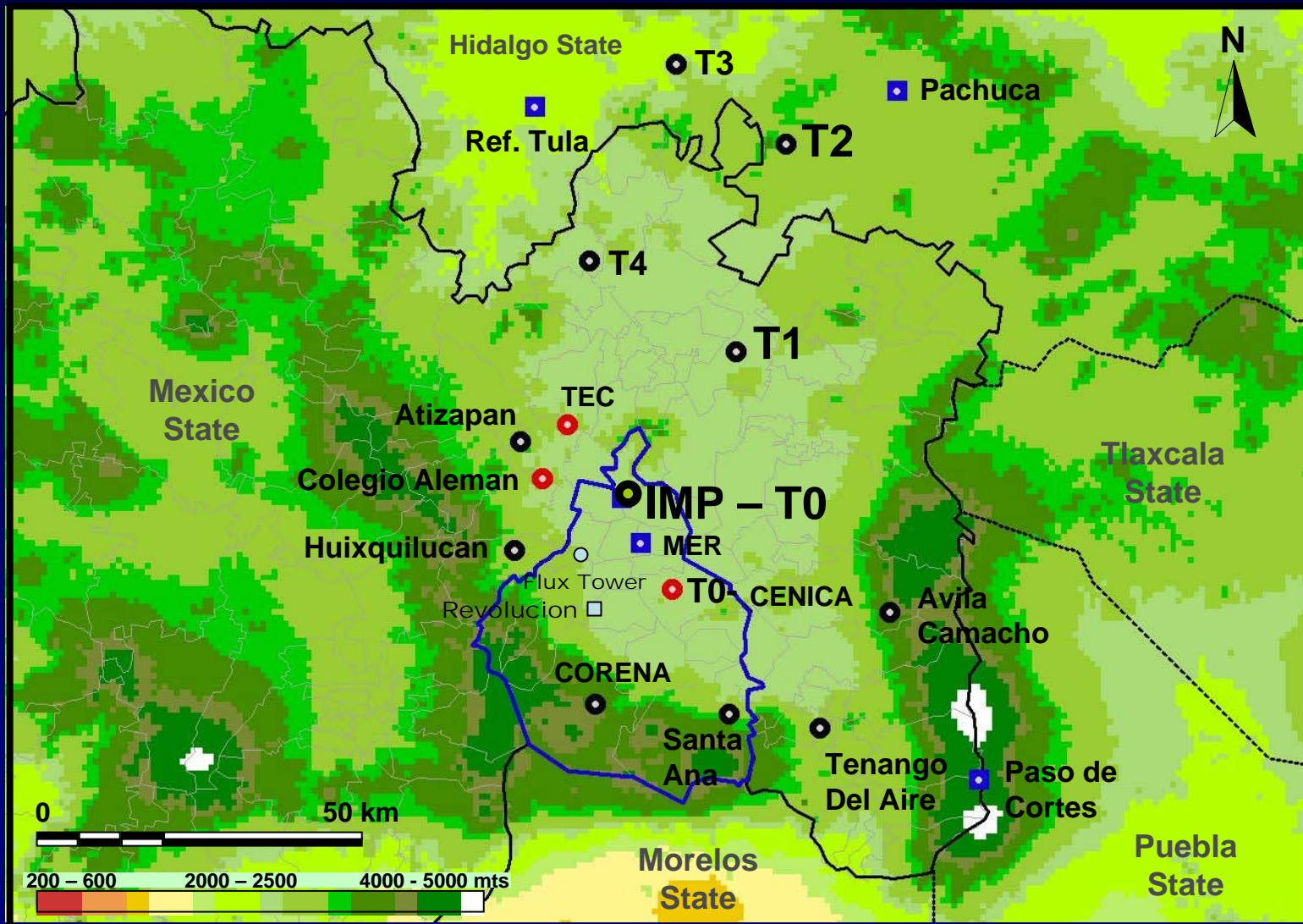
CO, NO, NO₂, NO_y, O₃, SO₂ – BNL: Springston, Senum
 PSAP, Neph, CNCs – PNNL: Group
 TSEMs – BNL: Wang
 MFRs – PNNL: Barnard
 SPSP – DMT, CIRPAS: Kok, Jonsson, Senum
 Balloons – PNNL: Zaveri, Hubbe
 Data – PNNL, BNL: Hubbe, Springston, Senum

NASA King Air

Base: Veracruz
Airborne High Spectral
Resolution Lidar (HSRL)



MILAGRO Measurement Sites



- Mobile site
- Fixed site
- Other measurements

MAX-Mex Participants and Instrumentation

T0 Supersite Mexico City IMP



MET RADIATION AEROSOLS

ANL	Aethalometer (7 channel)	BC Aerosol absorption
ANL	Multi-angle Absorption	BC Aerosol absorption
ANL	Nephelometer 3 wavelength	particle scattering
ANL	Nephelometer 1 wavelength dry	dry particle scattering
ANL	Nephelometer 1 wavelength wet	wet particle scattering
ANL	Filter Sampler	OC/EC, humic like substances
ANL	Open path NIR TDLAS	NH ₃
ANL	Filter Sampler	¹⁴ C, ⁴⁰ K, ²¹⁰ Pb, ⁷ Be, ²¹⁰ Po, ²¹⁰ Bi,
ANL	RB Meter	UVB
ANL	Weather Station	wind speed/dir., rain, temp, press, RH
BNL	CCN Counter	cloud condensation nuclei
BNL	Scanning Mobility Particle Sizer	aerosol size distributions
DRI, U of Nev, Reno	Photoacoustic Spect.	aerosol absorption
PNNL	MFRSR	radiation, aerosol optical depth
PNNL	Solar Tracker	broad band radiometer
PNNL/EMSL	DRUM Aerosol Sampler	sampling for PIXE/PESA/STEM
PNNL/EMSL	TRAC Aerosol Sampler	sampling for TEM, SEM/EDX analysis
Aerodyne Res. Inc.	ARI H.R. TOF-AMS/soft ions	non-refractory fine PM size & comp.
University of Colorado	High Res. TOF-AMS	aerosol size and composition
University of Colorado	Thermal Denuder	aerosol volatility before AMS
University of Colorado	Aerosol Concentrator	aerosol concentration before AMS
University of Colorado	Optical Particle Counter	aerosol size, number (Grimm 1.109)
University of Colorado	TSI SMPS	particle size distribution
University of Colorado	TSI nano-SMPS	nanoparticle size distribution
University of Colorado	TSI DustTrak	aerosol concentration

Field Campaign Period

**MAX-MEX/MILAGRO Began March 1, 2006
with Data Taken to March 29, 2006**

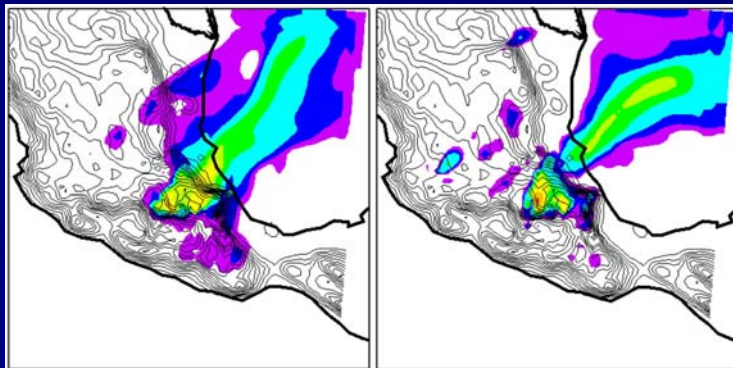


Forecast Support for Aircraft Operations

DOE leadership in forecast support

- Pre-field campaign activities to help identify surface sampling sites and dominant pollutant transport pathways
- Organized set of models (local, regional, and global scale) used during MILAGRO
- Lead forecasting team to develop daily briefings to support principal investigators in planning research aircraft flights
- Daily briefings disseminated to remote scientists via NCAR field catalog

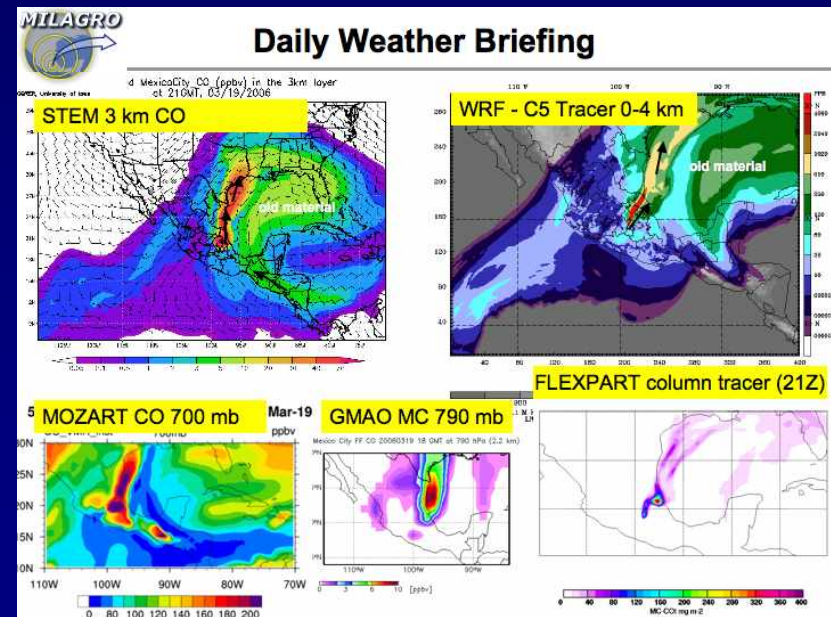
DOE / ASP pre-field campaign modeling of aerosol transport patterns



Veracruz Forecasting Team

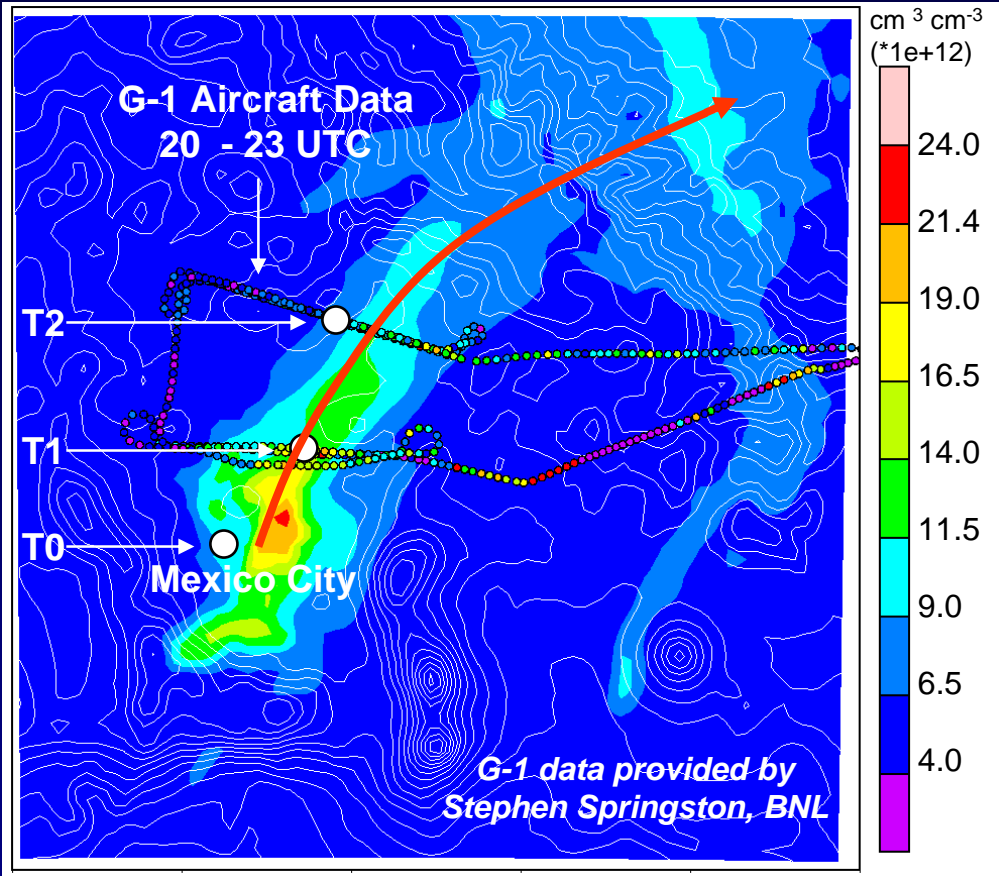


Example Briefing Material Available at <http://catalog.eol.ucar.edu/cgi-bin/milagro/report/index>

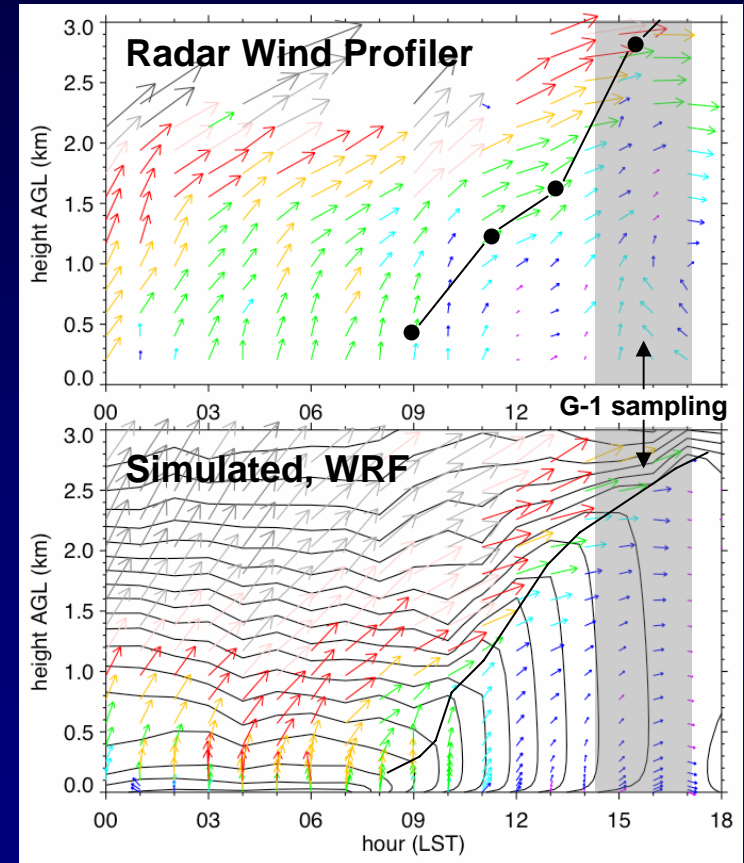


Sampling Strategy a Success

WRF-chem Particulate Volume Prediction 21 UTC 20 March, ~990 m AGL



Wind Profiles and PBL at T1, 20 March



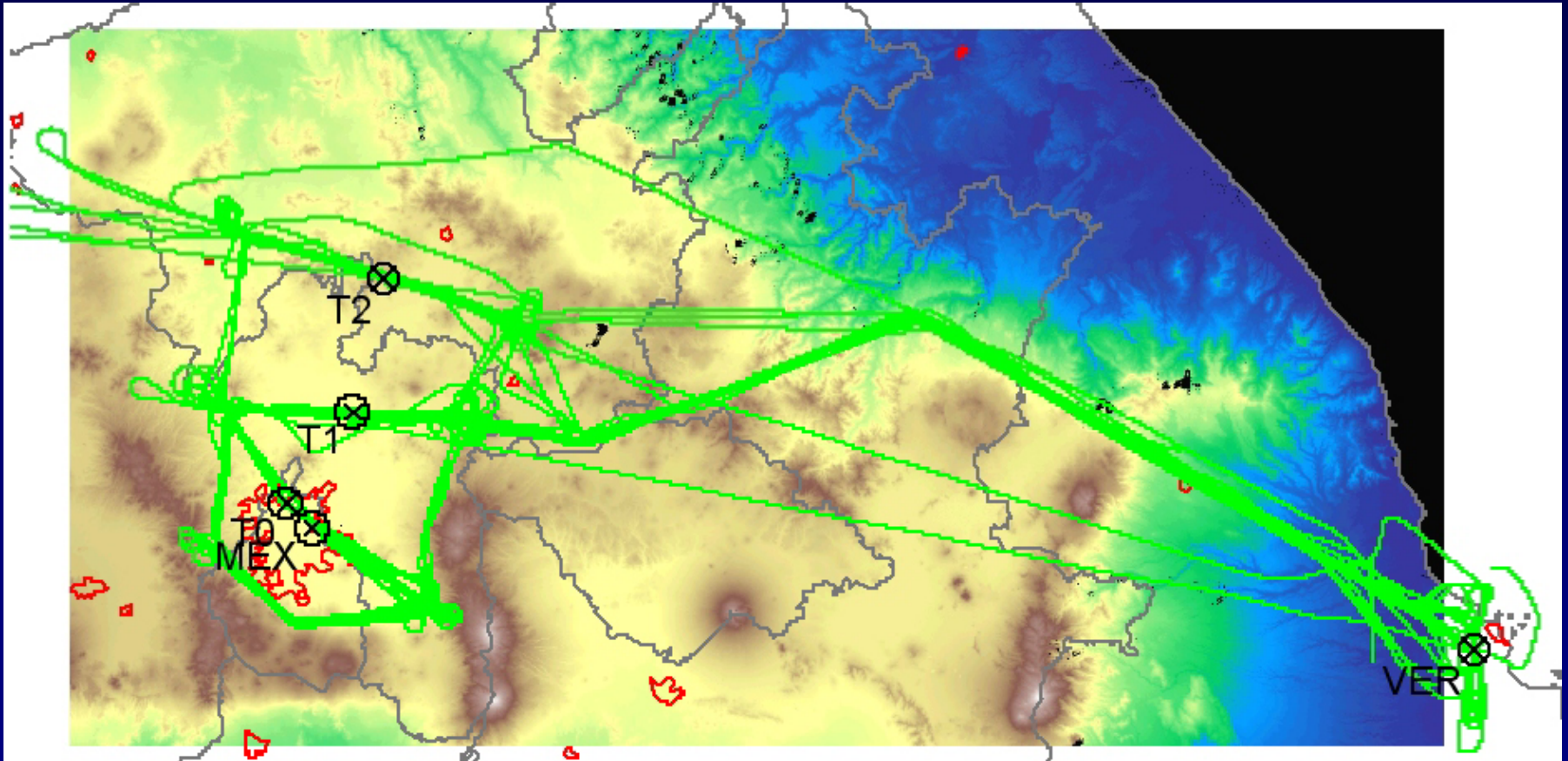
- T1/T2 transport scenario occurred on 4-5 days during March 2006.
- Simulated downwind plume in good agreement with observations.

- Strong ambient SW flow aloft decreases during day.

G-1 Flights

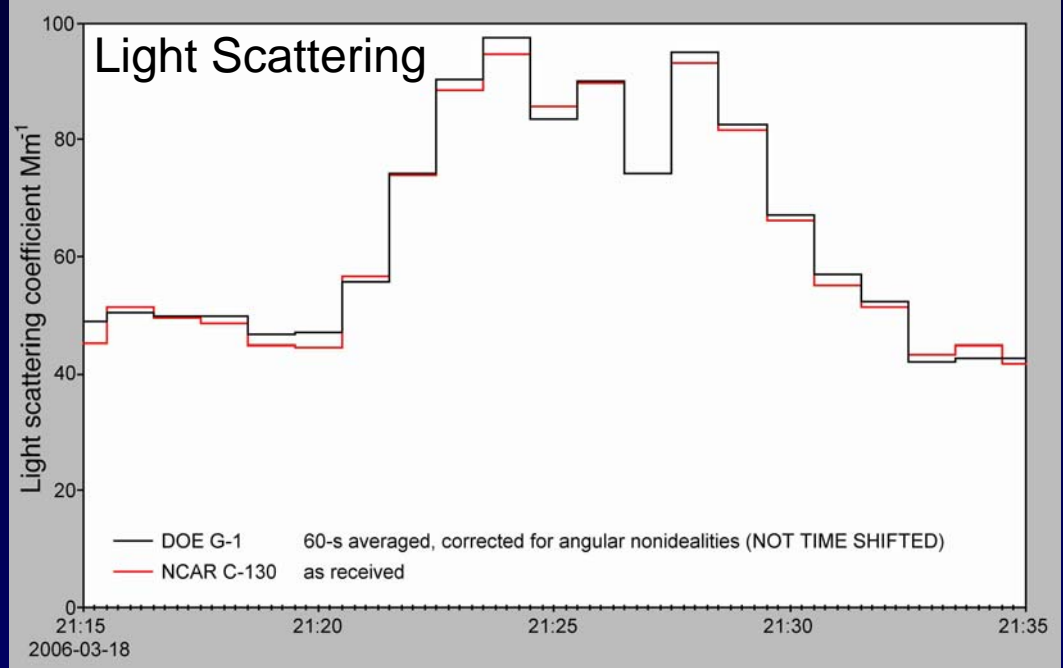


15 Flights, 48 hours Flight Time
Over-flights of T0 (12), T1(37), T2 (23)

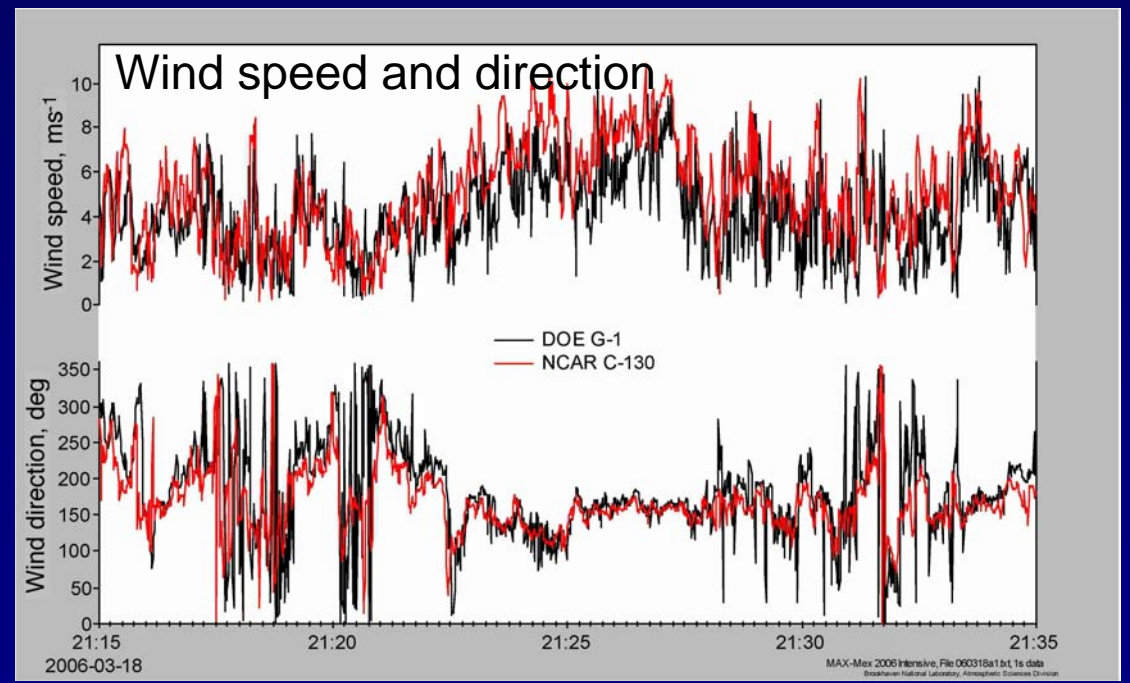


Aircraft Intercomparisons

Aircraft instrument intercomparisons provide high confidence in the measurements.



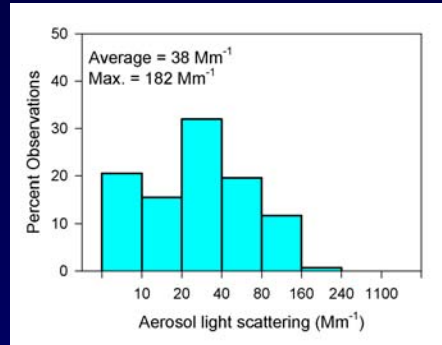
DOE G-1 and NSF C-130 wing-to-wing intercomparisons show excellent agreement.



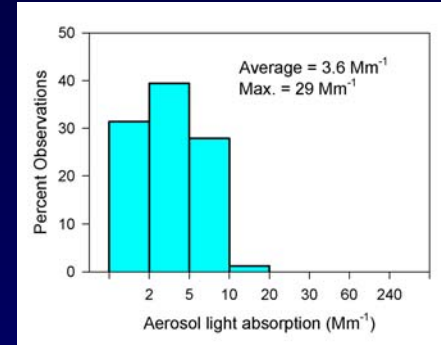
Aerosol Optical Properties

Eastern U.S. (2002, NEAQS)

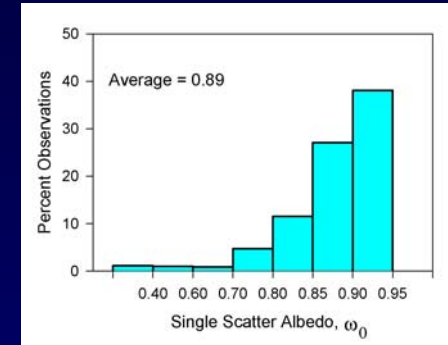
Scattering



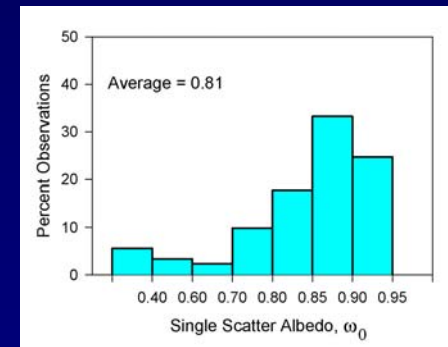
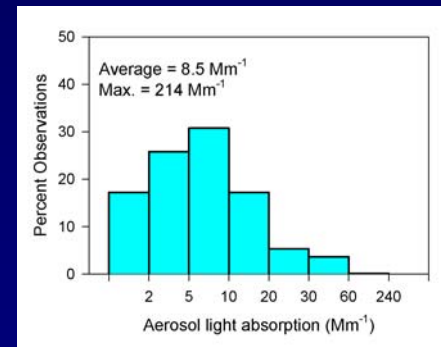
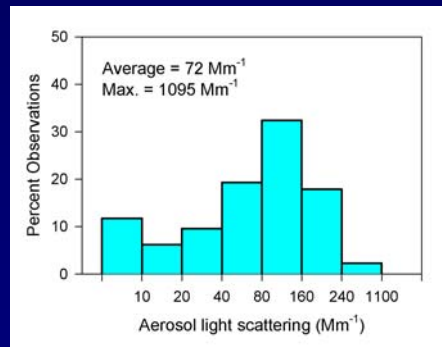
Absorption



Single Scatter Albedo



Mexico City Basin

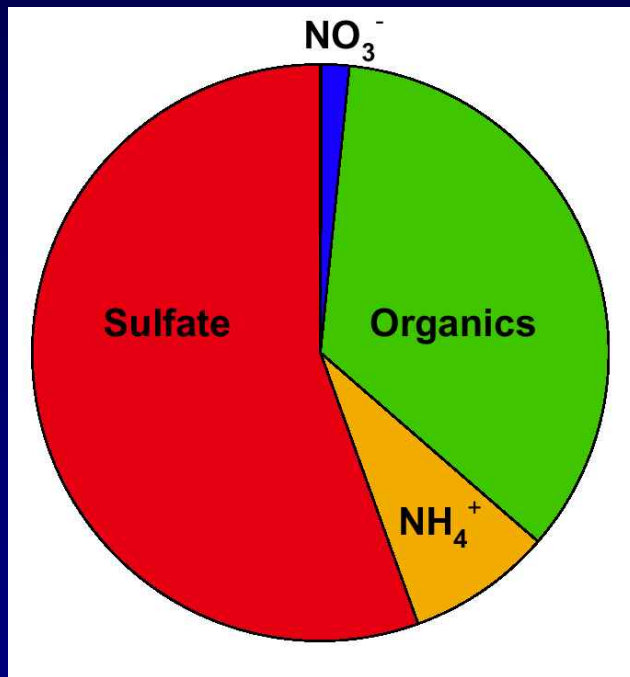


- Light scattering, absorption were 2 times Eastern U.S.
- Urban area routinely surpassed peak U.S. values
- Higher proportion black carbon in Mexico aerosol
- Mexico aerosol is warming ($\omega_0 < 0.85$), U.S. cooling

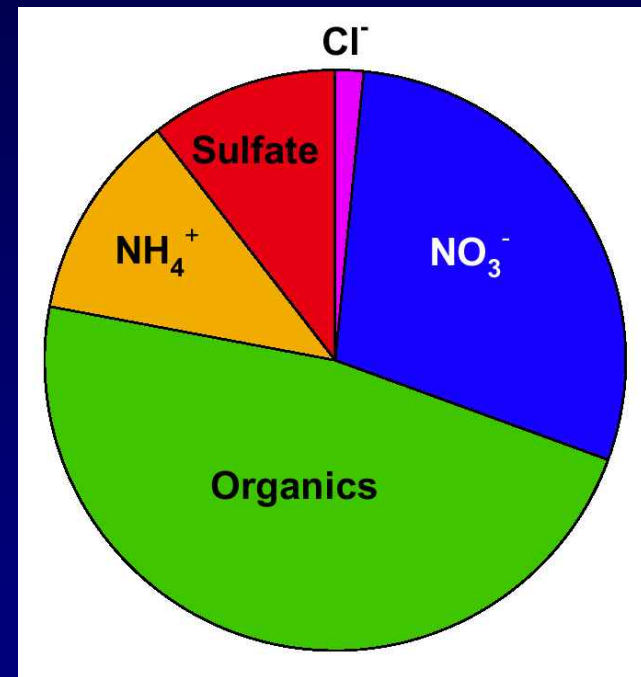
Aerosol Composition Comparison

Aerosol Mass Spectrometer Measurements from G-1

Eastern U.S. Regional Pollution



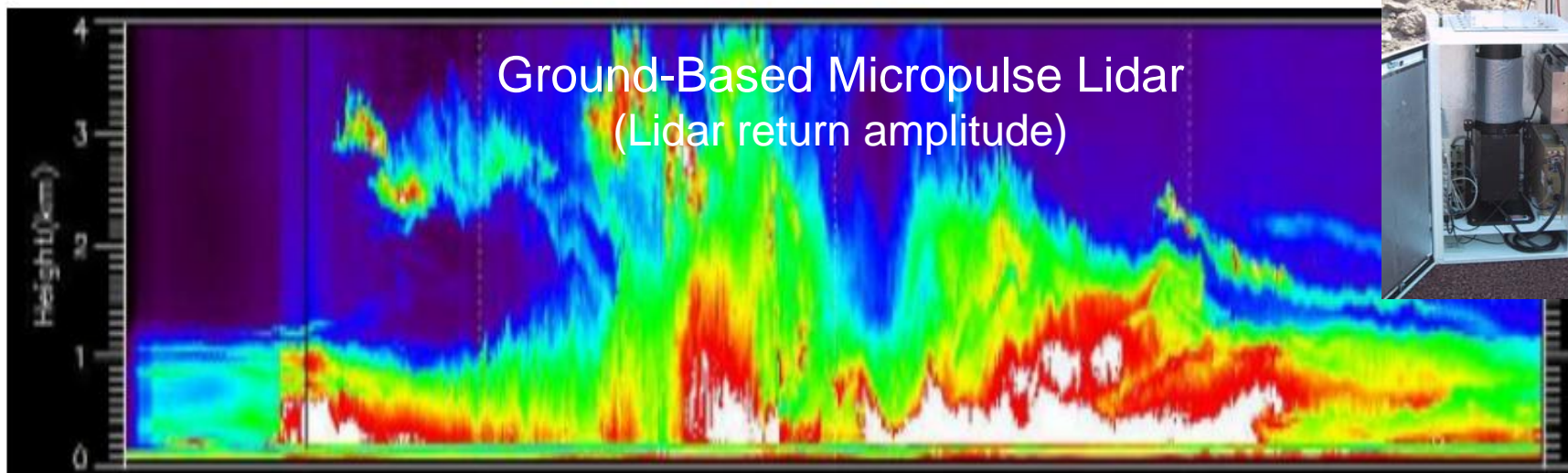
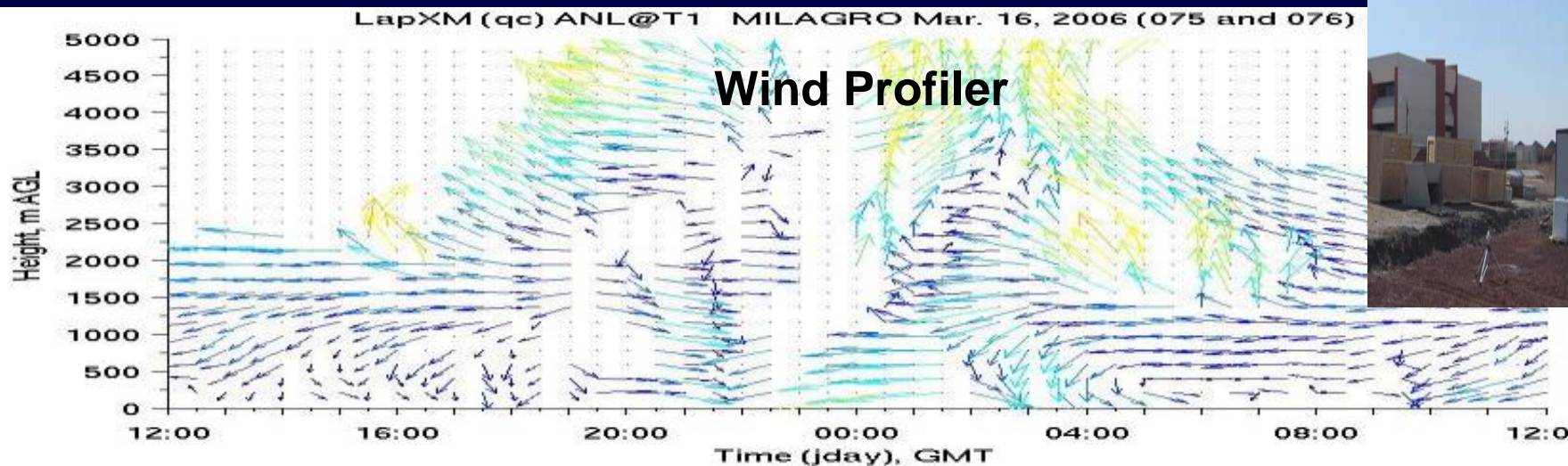
Mexico City Urban



- Eastern U.S. episodes - sulfate dominated
- Mexico urban aerosol - organic dominated, with nitrate

Layering of Aerosols above T1

Aerosols lofted from Boundary Layer into Lower Troposphere

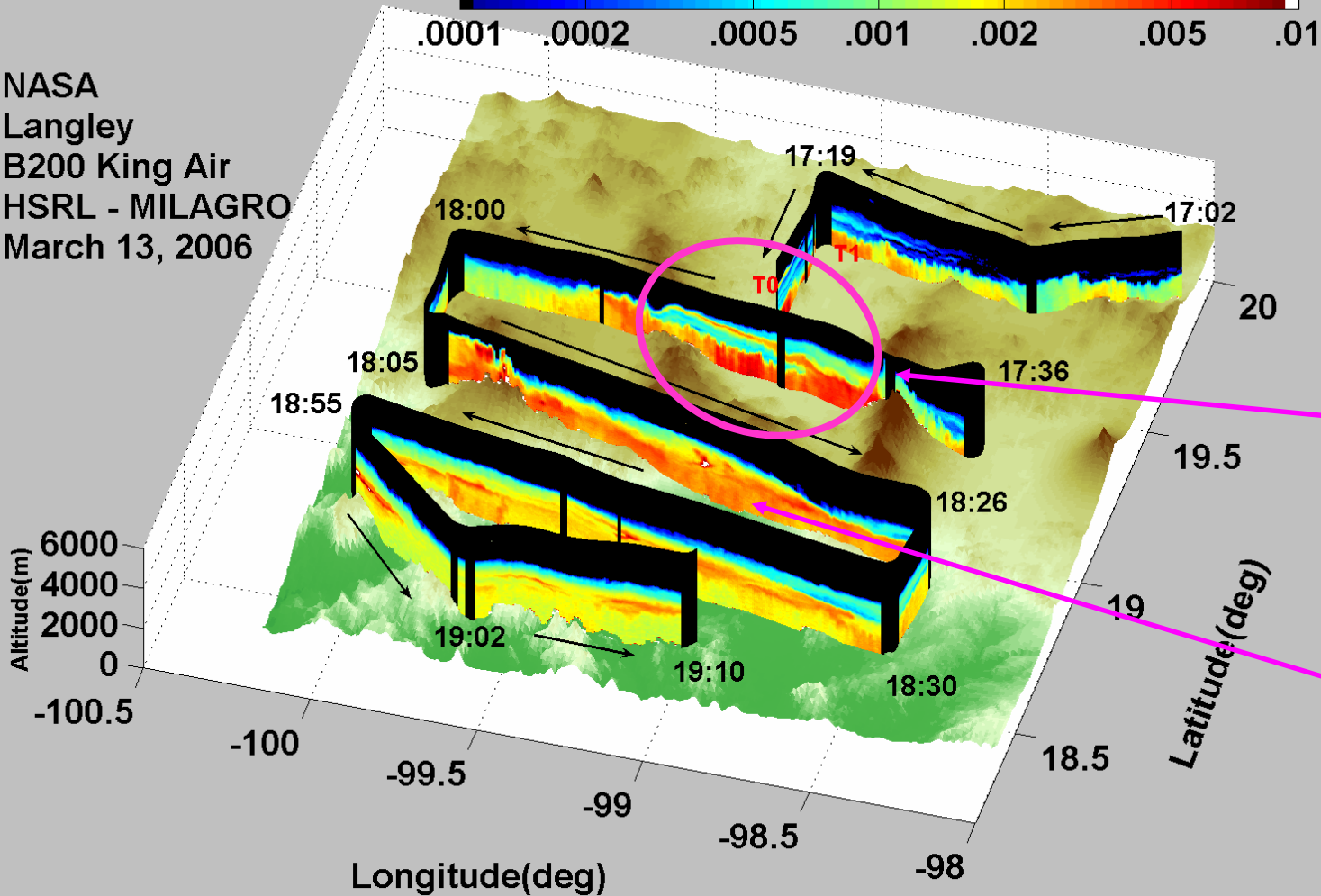


Spatial Distribution of Aerosol Properties

Aerosol Backscatter Coefficient (532 nm) (km-sr)**-1



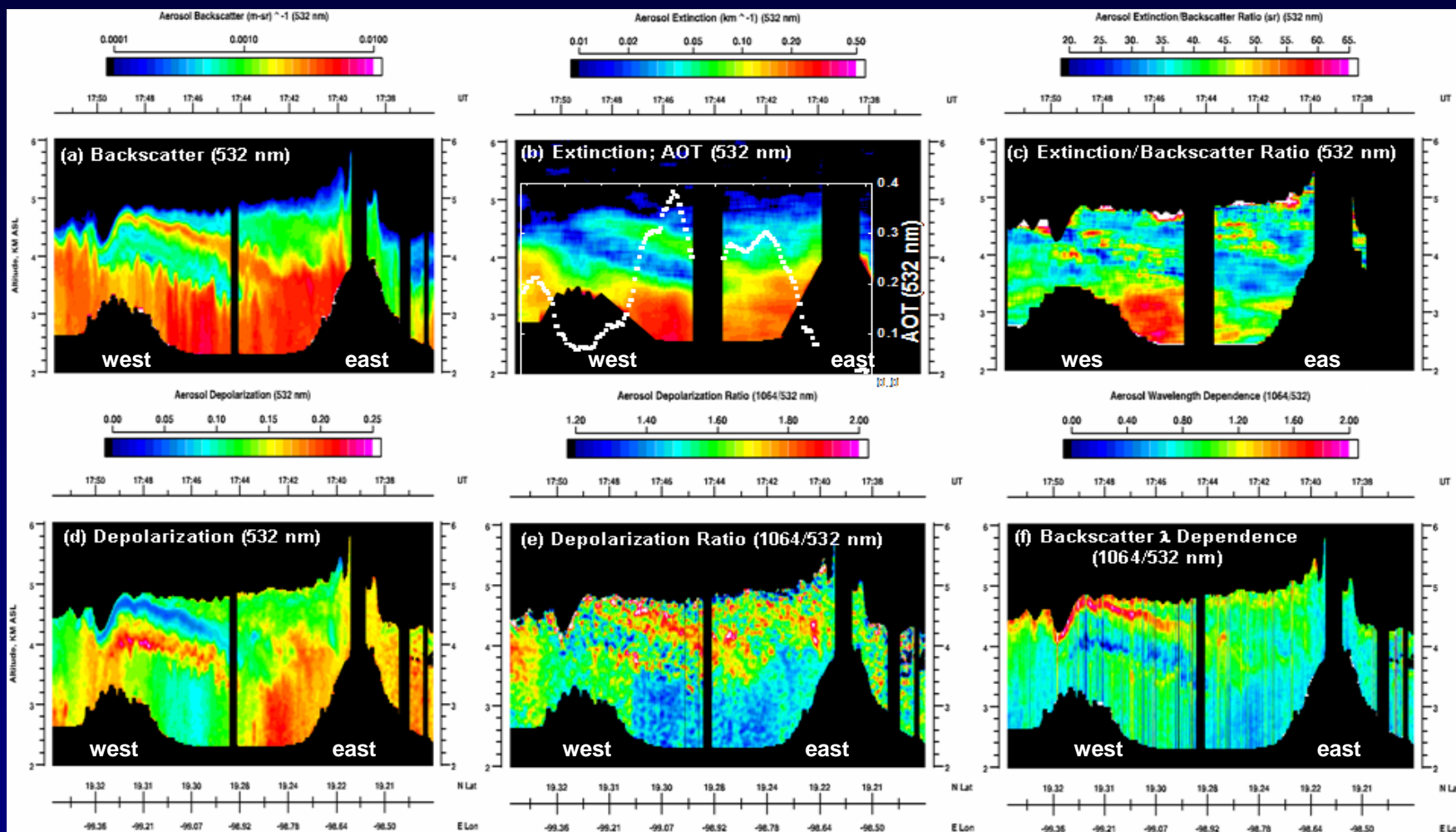
NASA
Langley
B200 King Air
HSRL - MILAGRO
March 13, 2006



complex structure
over Mexico City

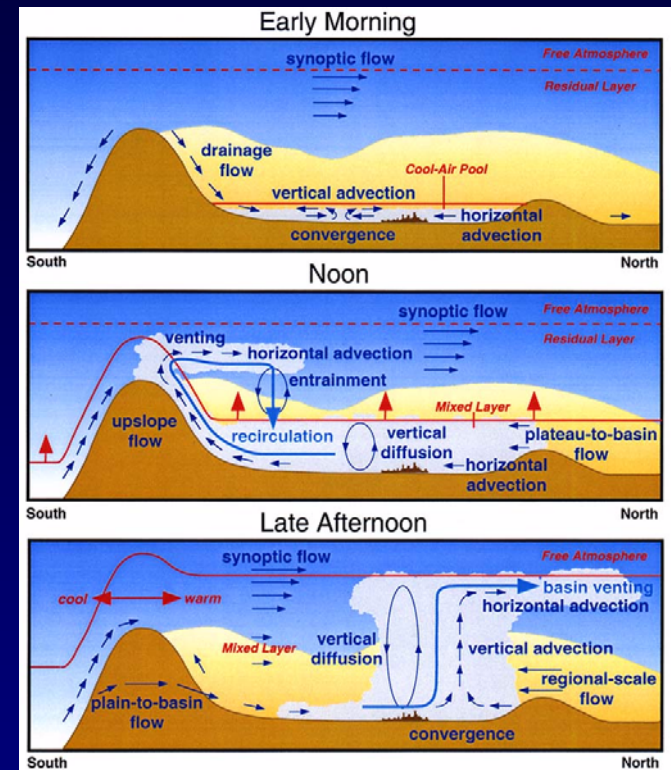
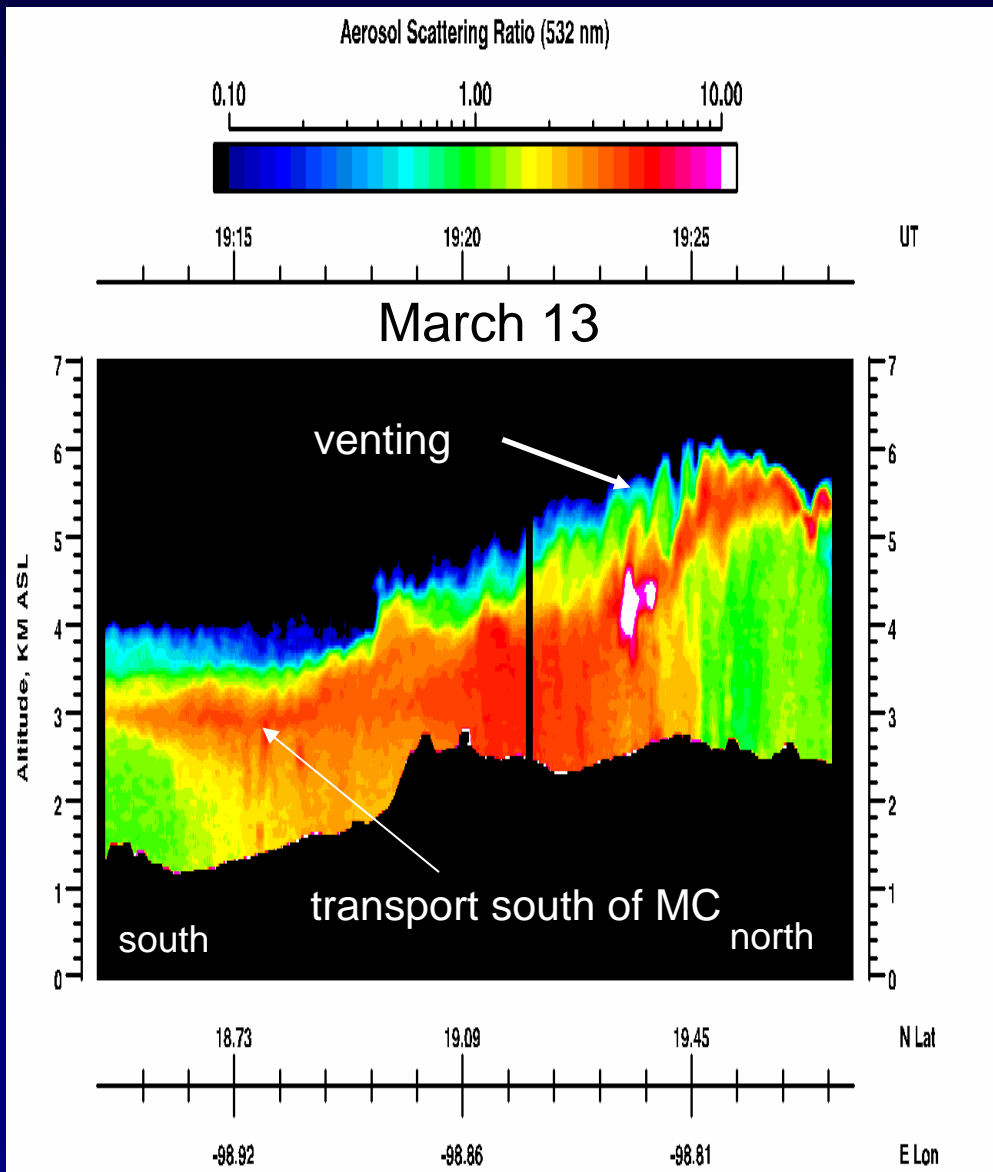
MC emissions
transported
southwest of city

Horizontal Distribution of Aerosol Types



Western part of city- high extinction/backscatter ratio, high wavelength dependence, low depolarization – urban aerosol.
 Eastern part of city - low extinction/backscatter, low wavelength dependence, high depolarization – dust.

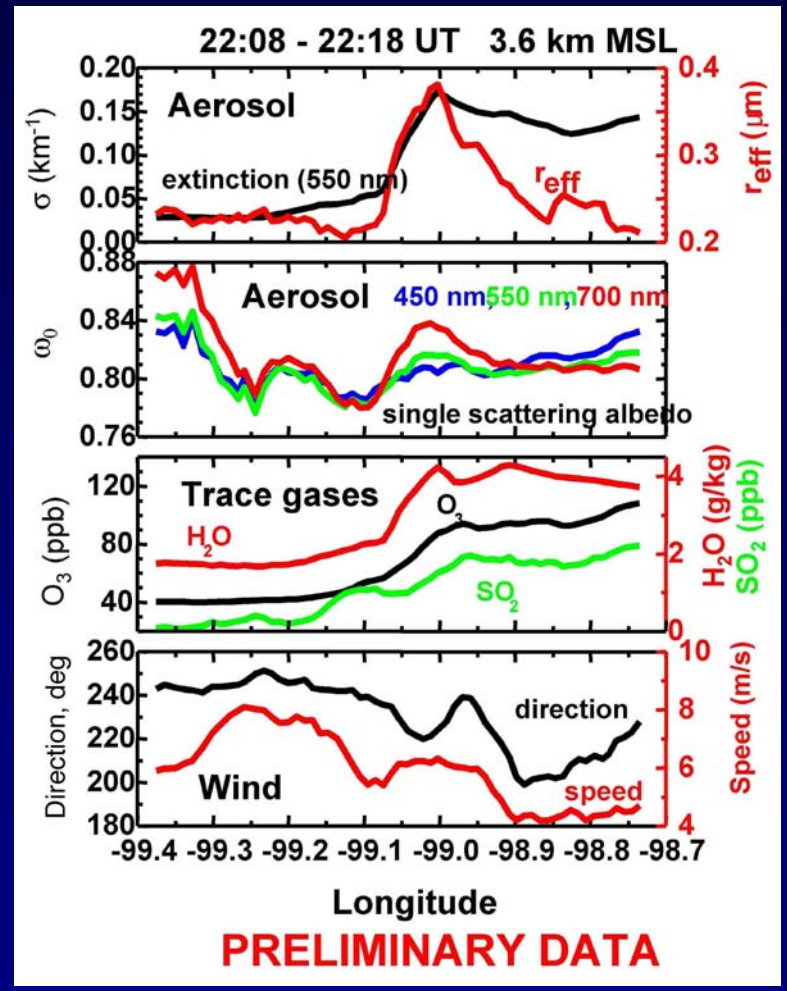
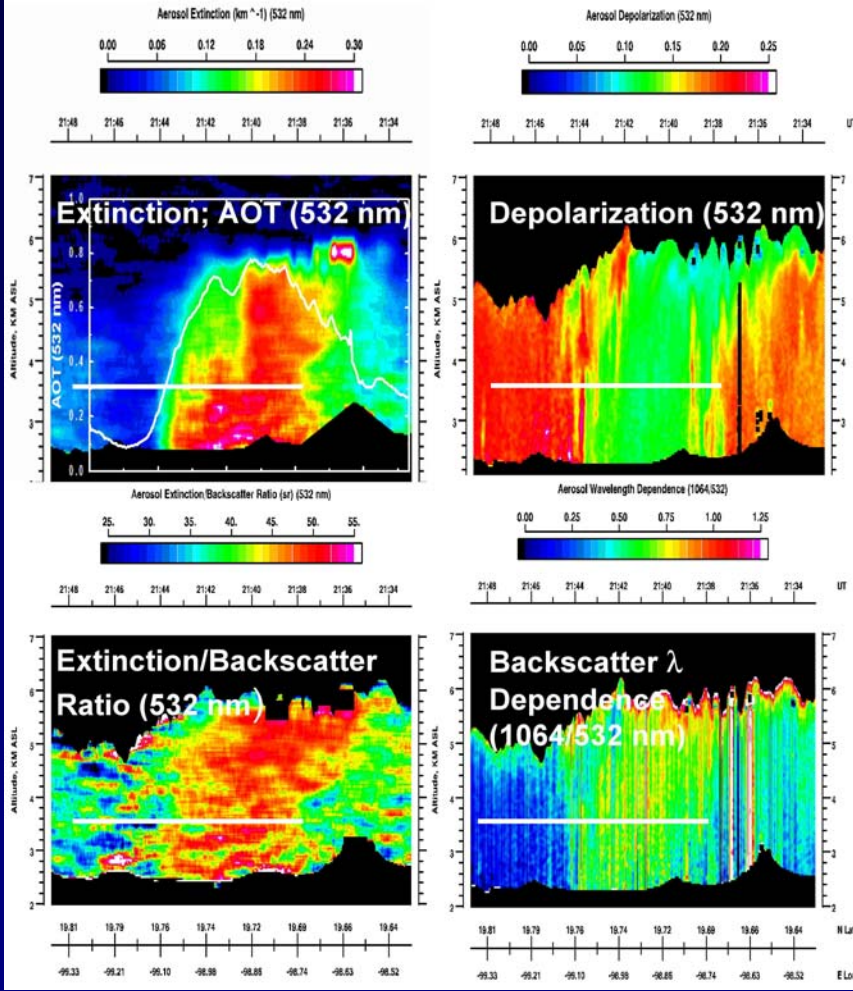
Aerosol Venting into Free Troposphere



- Lidar measurements confirm earlier DOE / ASP modeling studies
- Models are being used to determine the effect of venting on aerosol evolution and radiative forcing

Lidar Data Provide Vertical Context for In-situ Measurements

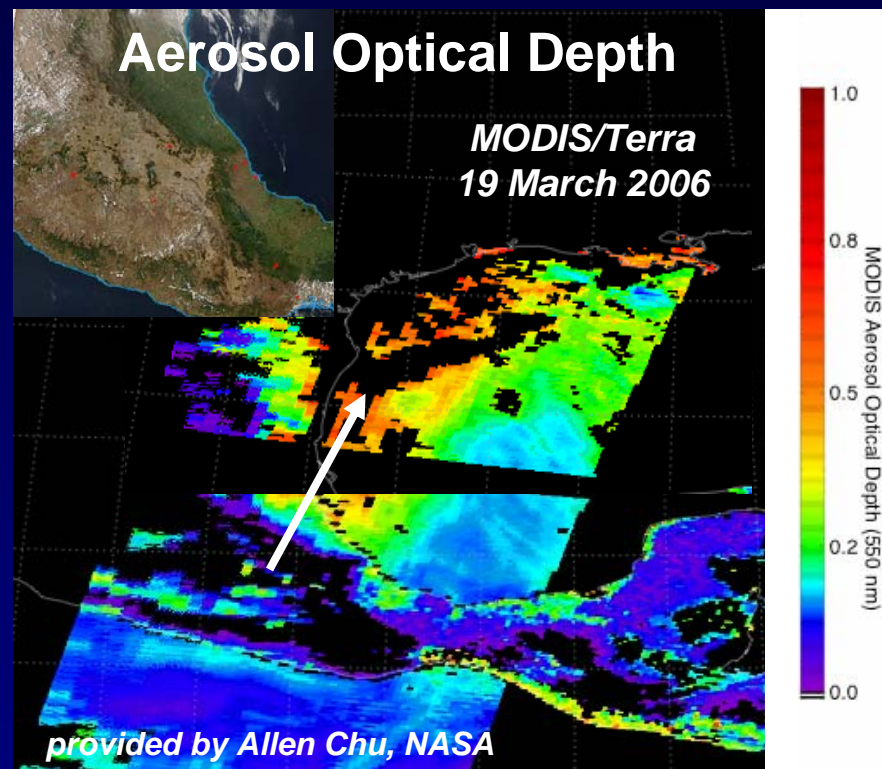
March 9, 2006



- HSRL and G-1 measurements show changes associated with MC pollution

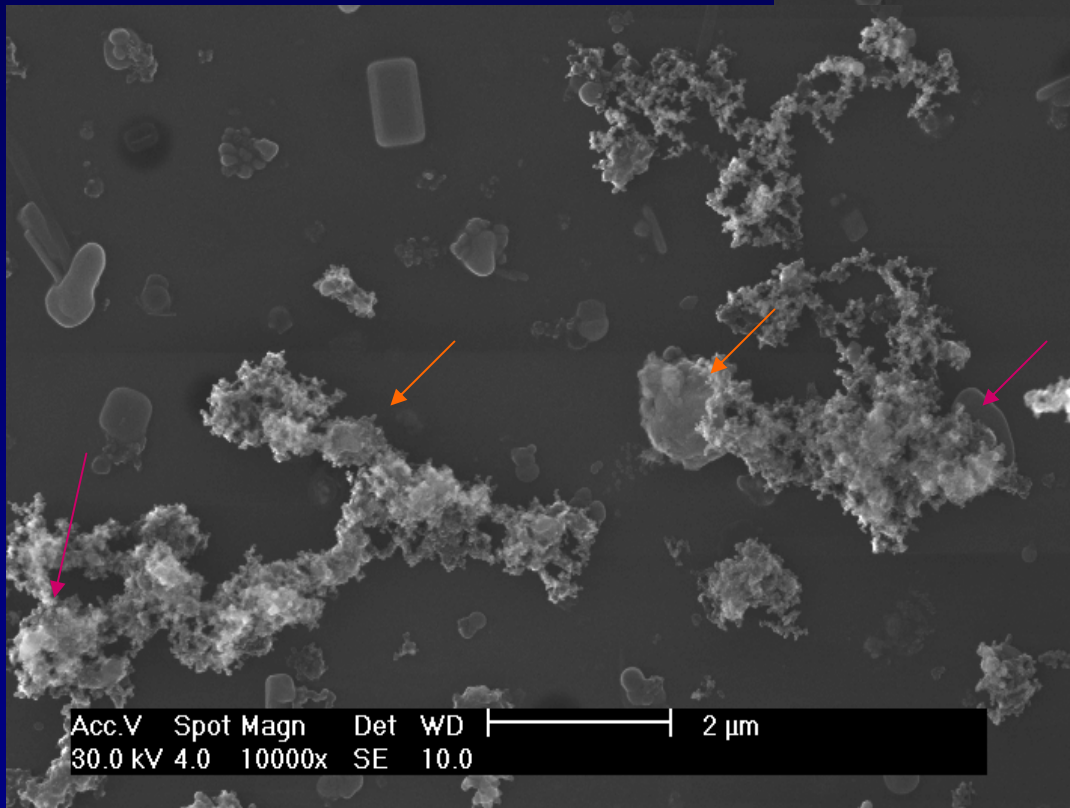
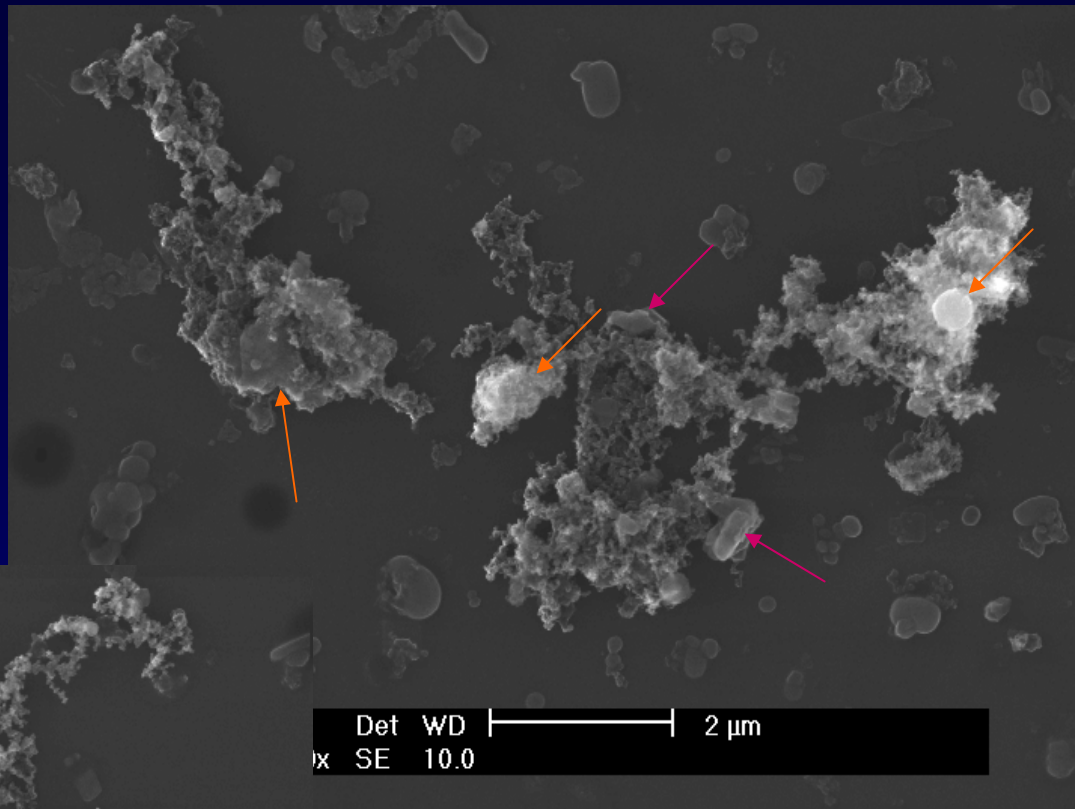
Use of Satellite Measurements

Satellite and Aircraft Intercomparisons for Biomass Burning and Dust Events



- Very dry conditions contributed to higher-than-normal biomass burning events.
- Mixing of anthropogenic and biomass burning particles.
- Aircraft measurements permit determination of the height of particles downwind of Mexico City and the relative contribution of anthropogenic and biomass burning sources to aerosol optical depth.

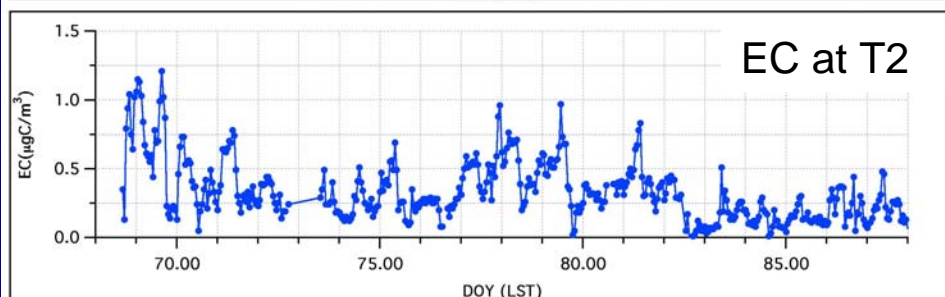
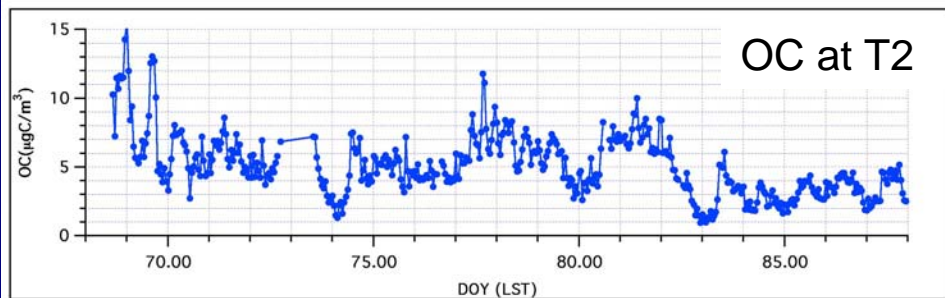
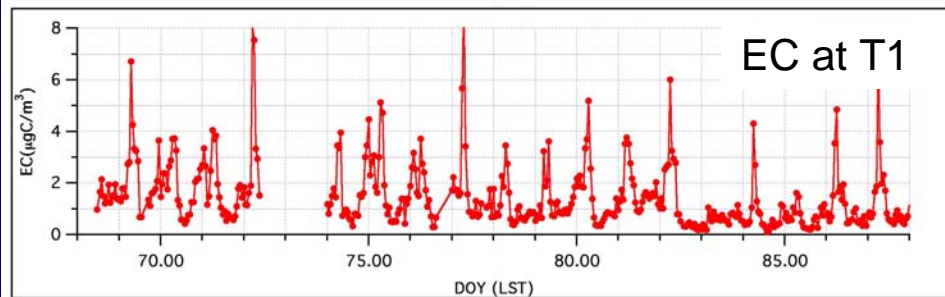
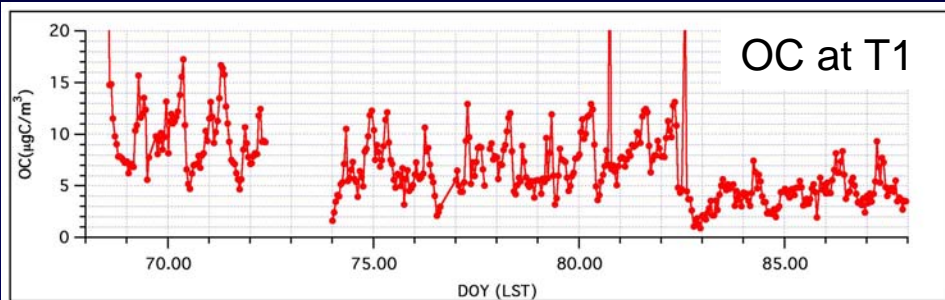
Scanning Electron Microscopy Images of Particles at T0



**CaO inclusions and
S-rich inclusions
in soot particles**

Y. Desyaterik, R. Hopkins,
M. Gilles and A. Laskin;
PNNL, LBNL

EC and OC Measurements at T1 and T2



Organic Carbon / Elemental Carbon
Measurements at T1 and T2

EC - Tracer of Transport

OC - Tracer of Secondary Organic
Aerosols

Higher concentrations
and stronger diurnal
variations at T1 than T2
as expected.

Data will be compared
to optical data and
aerosol composition.

Anticipated Further Analyses and Results

- Examination of size-dependent aerosol composition as function of “age” subsequent to emission and chemical processing.
- Attribution of changes in size-dependent composition to specific processes.
- Quantification of secondary organic aerosol production.
- Comparison of properties of biomass burn and urban soot aerosols.
- Examination of dust events and dust interactions with urban aerosol.
- Examination of hygroscopic growth, CCN properties, and precipitation scavenging in relation to aerosol properties.
- Quantitative description of aerosol transport.
- Examination of evolution of composition and optical properties of black carbon and secondary organic aerosol.
- Evaluation of performance of current models.
- Development of new and/or improved treatments of aerosol processes.

VERY RICH DATA SET!

MAX-Mex and MILAGRO Continue

Data:

Preliminary Data - September 2006

Final Data Sets – March 2007



MILAGRO Science Meeting – October 2006:

Preliminary Analyses

Begin Data Assessments for Publications

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ASP MAX-MEX Science Team

Pilots and ASP Science Support personnel

MILAGRO Participants

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THE PROJECT WAS CARRIED OUT SAFELY WITH NO INCIDENTS.