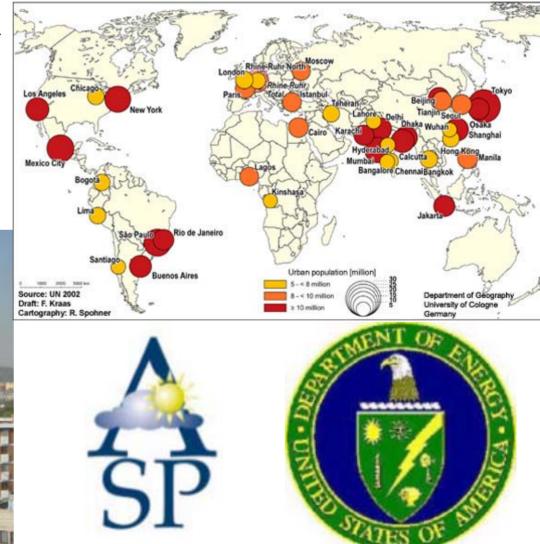
### **Megacities: Mexico City and Houston as Potential Field Sites**

**Nancy Marley and Jeff Gaffney** 

**Argonne National Laboratory** 





#### **Aerosol Radiative Forcing – You Need to Determine:**

- Optical Properties -(Size, Chemical Composition as Function of Size, Optical Constants, Etc.)
- Sources (Natural and Anthropogenic)
- Lifetimes (Size and Hygroscopicity)
- Position in Atmosphere (Vertical & Horizontal Distributions)

#### NEED TO MINIMIZE MEASUREMENT UNCERTAINTIES

- NEED REASONABLE AEROSOL LOADINGS
- SENSITIVE and SELECTIVE INSTRUMENTATION

#### **URBAN AREAS – HIGH AEROSOL LOADINGS**

#### MAJOR SOURCES OF BOTH PRIMARY AND SECONDARY AEROSOLS to REGIONAL & GLOBAL SCALES

#### ASP HAS CONSIDERABLE HISTORY IN URBAN FIELD WORK

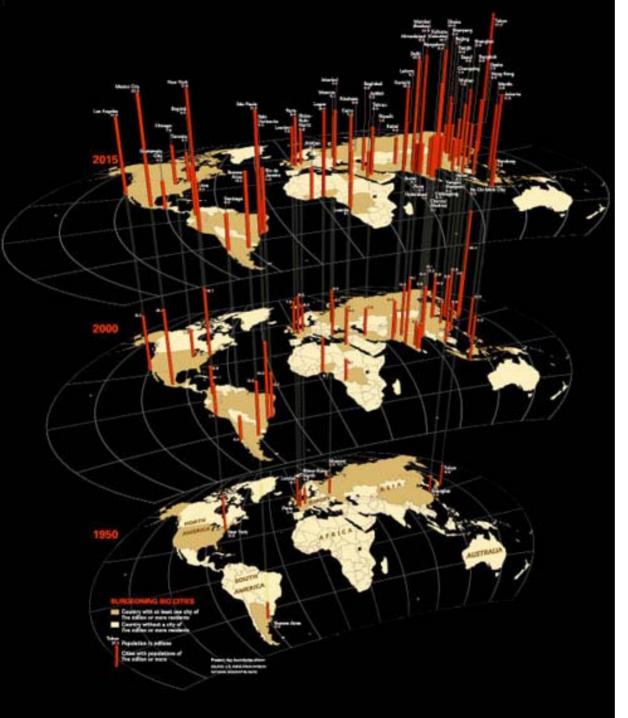
Mexico City – Aerosol Study – 1997

Northeast Oxidant and Particle Study (NEOPS), Philadelphia-1999

Phoenix, AZ 1998, 2001

Texas Air Quality Study, TexAQs 2000 (**Houston**) Mexico City Megacity 2003 – MCMA 2003 (MIT)





**MEGACITIES** ► 10 Million 1950 – 1 (NYC) 1995 - 142015 - 21**Mini – MEGACITIES** 5 Million – 10 Million 1995 - 72015 - 37ASIA – AFRICA 2/3 rural to  $\frac{1}{2}$  urban by 2025

#### MEGACITIES AND MINI-MEGACITIES MAJOR SOURCES OF AEROSOLS AND GREENHOUSE GASES

THESE SOURCES WILL BE CHANGING OVER TIME AS THE CITIES DEVELOP AND THE TECHNOLOGIES EVOLVE

CARBONACEOUS AEROSOLS (ORGANIC & BLACK CARBON) SULFATE, NITRATE – FOSSIL FUEL COMBUSTION BLACK CARBON – DIESEL AND TWO-CYCLE ENGINES, ETC. SECONDARY ORGANIC AEROSOLS – FOSSIL AND BIOGENIC

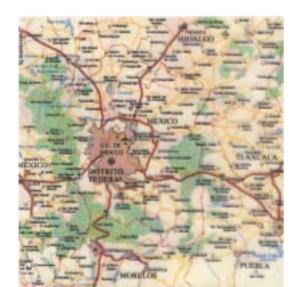
NEED TO BETTER CHARACTERIZE THE EMISSIONS AND THEIR PROPERTIES (SIZE, ETC.)

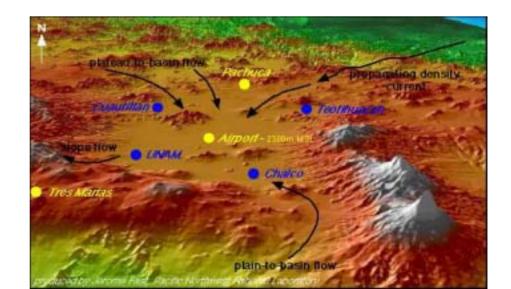
#### **MEGACITIES** - TOP TEN

	Population	
Megacity	1991	2000
Tokyo, Japan	27,245,000	29,971,000
Mexico City, Mexico	20,899,000	27,872,000
Sao Paulo, Brazil	18,701,000	25,354,000
Seoul, South Korea	16,792,000	21,976,000
New York, USA	14,625,000	14,648,000
Osaka, Japan	13,872,000	14,287,000
Bombay, India	12,101,000	15,357,000
Calcutta, India	11,898,000	14,088,000
Rio de Janeiro, Brazil	11,688,000	14,169,000
Buenos Aires, Argentina	11,657,000	12,911,000

#### TARGET OF OPPORTUNITY - MEXICO CITY

- 2<sup>ND</sup> 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





# Mexico City 1997

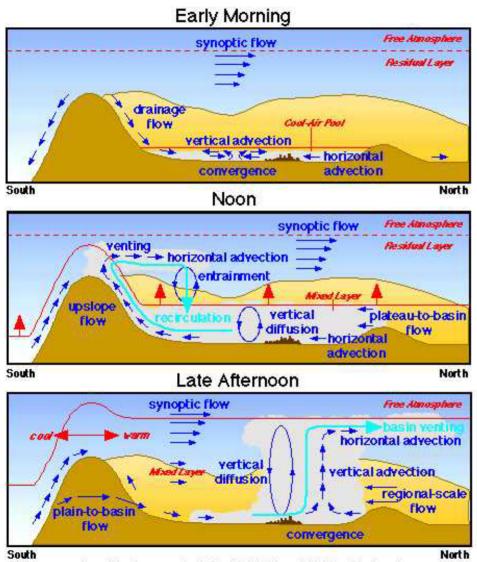
#### LOTS OF AEROSOLS – ON A DAILY BASIS!

- $> 50 \ \mu g/m^3 PM-2.5$
- 50% Organic and Black Carbon (Soot)
- Fast NO to NO<sub>2</sub> Conversion & NH<sub>4</sub>NO<sub>3</sub> Production
- **NH<sub>3</sub> Important!**
- **NH<sub>3</sub> Sources?**

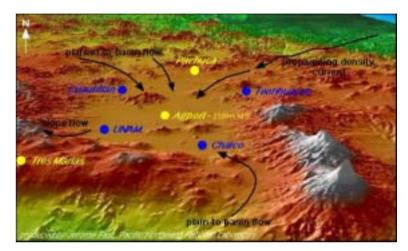


Mexico City 7:50 am 2/21/97

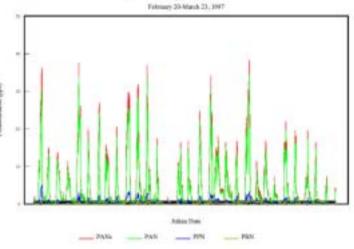
#### ASP METEOROLOGY – SHOWED STRONG DIURNAL TRANSPORT IN MEXICO CITY BASIN! TYPICAL?



produced by Jerome Fast, Pacific Northwest National Laboratory



Measurements of PANs - IMP, Mexico City



#### **MEXICO CITY MEGACITY 2003 – APRIL**

Collaborative Effort with MIT – Luisa and Mario Molina

Mexico City Metropolitan Area 2003 (MCMA) Study

**NARSTO Effort** 









#### **Preliminary Findings of Note:**

- High Levels of Black Carbon Not Washed Out in Rain Event!
- Important Regional Climate Implications
- Obtained Radiation Data as well as Comprehensive Aerosol and Gas Phase Data Sets

Aerosol Mass Spectrometers (Aerodyne)

DOAS, LIDAR, TDLAS, MFRSR instrumentation

High Levels of Ammonia – Anti-correlated with NH<sub>4</sub>NO<sub>3</sub>



New Cars – NH<sub>3</sub> Sources!

#### April 2003 Intensive Field Measurement Campaign

#### Supersite at CENICA (Ixtapalapa)

- □ Fixed Site Aerosol Mass Spectrometer (Aerodyne)
- □ Tall flux tower (Washington State University)
- **UV-VIS DOAS (University of Heidelberg/MIT)**
- □ LIDAR (University of Berlin/MIT)
- **Tethered balloon (CENICA)**
- □ Vertical atmospheric radiosondes (IMP/MIT)
- **Fast GC with Luminol detection method PAN, NOx**
- □ Aethalometer Black Carbon
- **Tunable-diode laser system for NH**<sub>3</sub>
- **VOC Canister sampling**
- □ Fast GC with OLEFIN Detector- isobutene
- **Nitroarenes** (Arey, Atkinson, UCRiverside)
- **Organic Carbon/Elemental Carbon (LBNL)**
- □ MFRSR & Aerosol Characterization (PNNL)
- **PTRMS/Aerosol Mass Spectrometer– VOCs (PNNL)**



POOLING RESOURCES THROUGH COLLABORATIVE FIELD WORK!

#### MCMA 2003 – MIT/AERODYNE MOBILE LAB

#### **SPATIAL INFORMATION**

- •NH<sub>3</sub>/HNO<sub>3</sub>/HONO/HCHO QC-TILDAS (Aerodyne)
- AEROSOL MS
- •Chemiluminescent NOy instrument (MIT)
- PTR-MS (MSU)
- •Real-time Canister/Cartridge Autosampler (WSU)
- PAN/NO<sub>2</sub>
- AETHALOMETER (LBNL)





#### **BLACK CARBON – PRIMARY AEROSOL**

Strong Absorber of Short Wave And Long Wave Radiation.

#### DIESELS

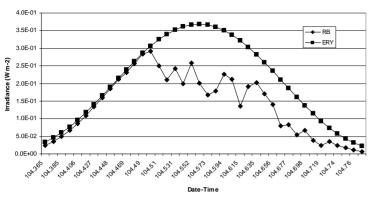




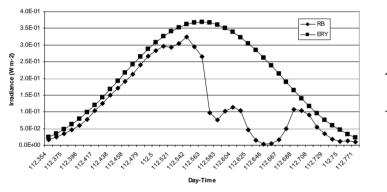
#### BIOMASS BURNING



Day 104: RB Measured and Clear-Sky Erythemal Calculation



Day 112: RB Measured and Clear-Sky Erythemal Calculation



#### **PHOTOCHEMISTRY EFFECTS**

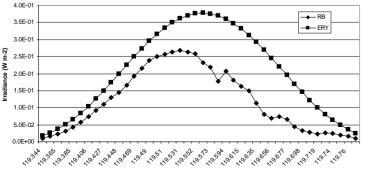
#### **BC** Absorbs Actinic Radiation

Modeled UVB vs. Robertson-Berger Meter

Cloud Effects (Added Bonus!)

April 22 – Julian Date 112 – RAIN EVENT UV-B at zero!



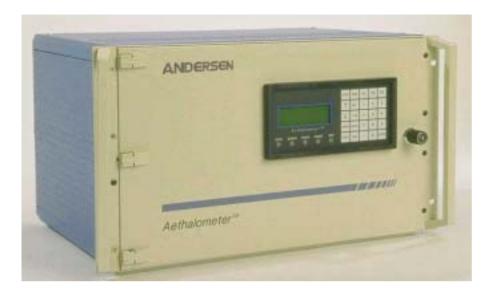


Early morning – Black Carbon  $CH_2O$ ,  $NO_2$ , etc.  $\rightarrow$  Ozone



#### **IMPORTANCE OF PHOTOCHEMICAL FEEDBACKS**

- Less UV from BC absorption
- **Reduced Secondary Greenhouse Gas Formation (e.g. O<sub>3</sub>)**
- **Increased Lifetimes of Other Greenhouse Species (e.g. RH, CO)**
- **Reduced Rates of Formation of Sulfates, Nitrates, Secondary Organic Aerosols**
- Biota Effects as Well  $CO_2$  Uptake Evapotranspiration, etc.
- **Others?**



#### AETHALOMETER

Optical Method Developed by DOE ASP Supported Research



Commercial Instrument Seven Wavelengths 1-2 Minute Time Resolution Semi-Continuous Operation BC Data – 30 days – Aethalometer – Mexico City April 2003
Daily Average - 5000 ng m<sup>-3</sup> (Std dev. 1500 ng m<sup>-3</sup>)
Daily Maximum Ave 14300 ng m<sup>-3</sup> (Std dev. 6100 ng m<sup>-3</sup>)
Daily Minimum Ave 1800 ng m<sup>-3</sup> (Std dev. 850 ng m<sup>-3</sup>)

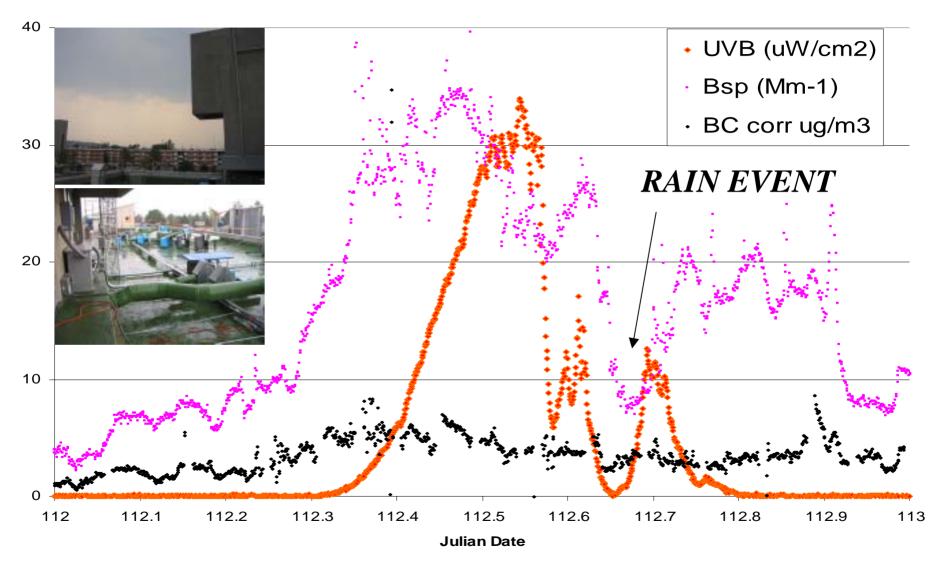
#### PREVIOUS WORK in 1997 Ave PM-2.5 was 50 µg m<sup>-3</sup>

10 % BC estimated from thermal analyses compares very well with this work.

#### DATA TAKEN DURING HOLY WEEK – SHOWS BC REDUCED

#### April 22, 2003

Int UVB, Bscat, Black Carbon-ANL-MCMA 2003 CENICA



## Most Models – BC Treated Same as Hygroscopic Aerosols Lifetimes from 2-7 days

This work and other data indicates BC has longer lifetime.

Models Typically Underestimate Black Carbon and Organic Carbon (Secondary Organic Aerosols) by Factors of 3-4 (or more) (Chung & Seinfeld 2002).

NEED TO ADDRESS AGING OF BC

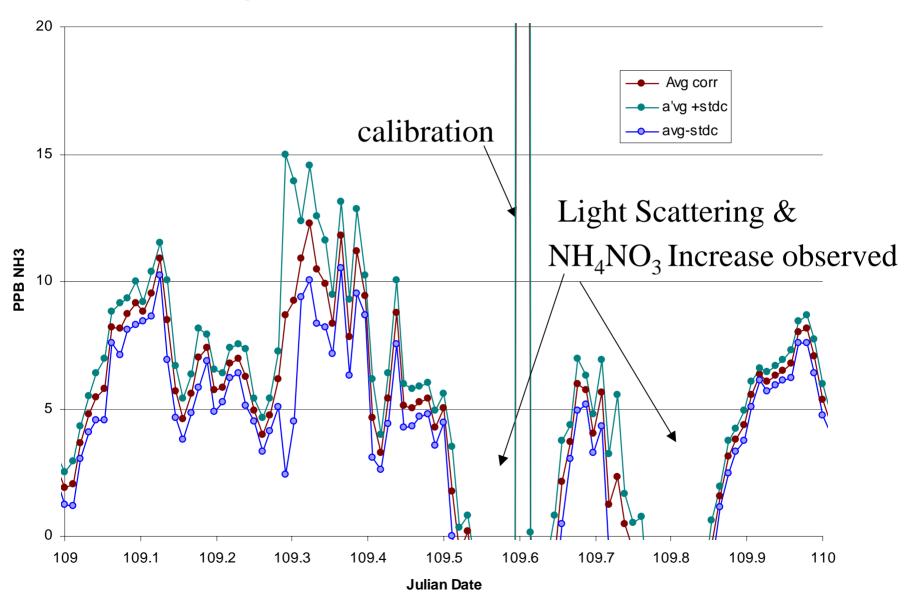


Mexico City Plume

#### AMMONIA – IMPORTANT IN FORMATION OF AEROSOLS TUNABLE DIODE SYSTEM – NEAR-IR LINE – 0.8 PPB DETECTION LIMIT TELESCOPE/REFLECTOR SYSTEM – 244 M PATH



#### NH3 Open Path LASIR - CENICA- MCMA 2003



#### Preliminary Observations from Mexico City Megacity 2003 – MCMA 2003

#### Importance for Regional and Global Climate

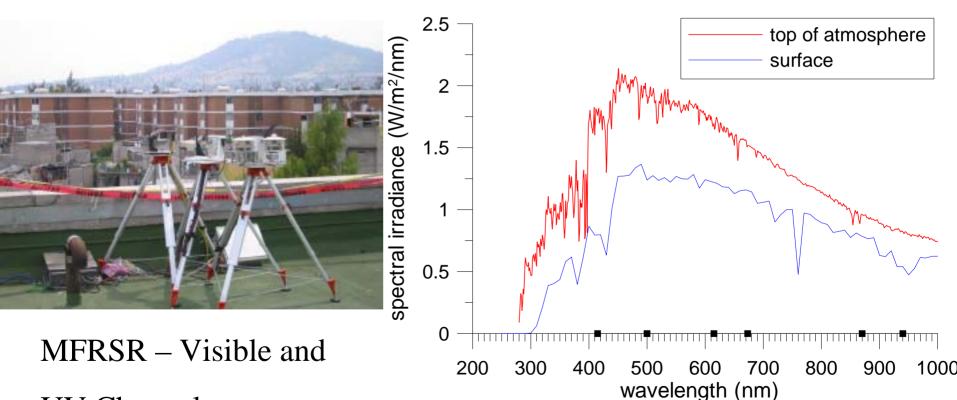
#### **MEXICO CITY MAJOR SOURCE OF BC and NH<sub>4</sub>NO<sub>3</sub>**

BC Can Heat as Well as Scatter – So BC Acts as Greenhouse Species (Hansen et al, 2000; Jacobsen, 2000, etc.)

BC Direct Effect – Semi-Direct Effect (Change in Relative Humidity)

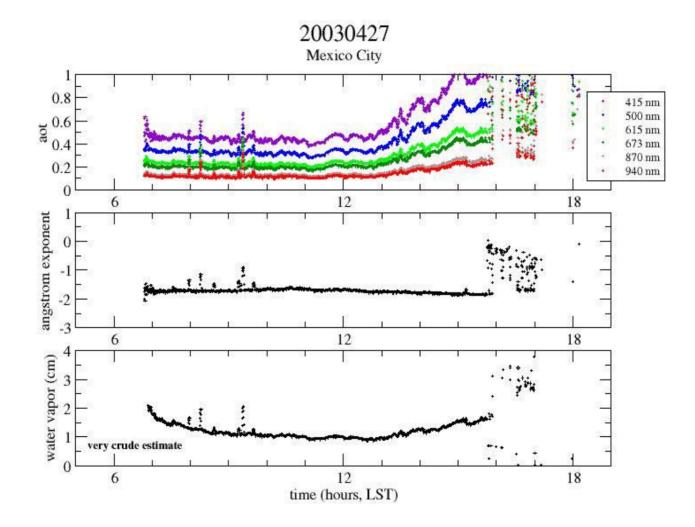
NH<sub>4</sub>NO<sub>3</sub> – Direct and Indirect Effects – Scattering

## MFRSR Solar Radiation

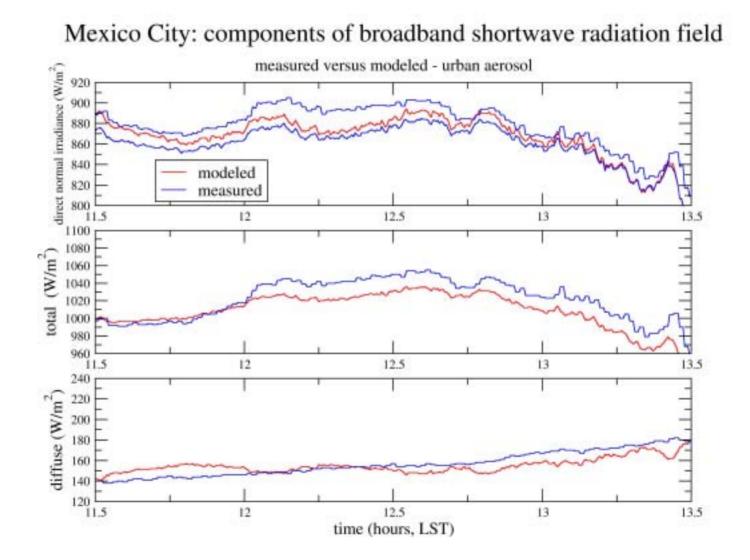


UV Channels

#### Aerosol Optical Thickness



# Run Measured vs Modeled experiment (narrowband to broadband)



## We get excellent agreement for $\varpi_o = 0.80$ for diffuse radiation

## Broadband Fluxes (W/m<sup>2</sup>)

	Modeled	Measured	Error
direct	867	878	1.3%
diffuse	155	157	1.3%
total	1009	1022	1.3%

#### SINGLE SCATTERING ALBEDO REFLECTS BC IMPACT!

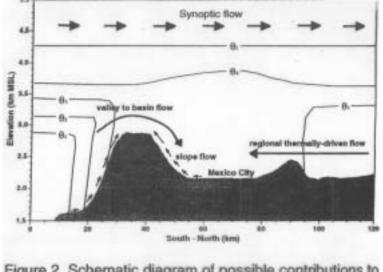


Figure 2. Schematic diagram of possible contributions to local flow patterns in the Mexico Citv area.

ASP Meteorologists

Showed Regional

"Flushing" of Basin

In Mexico City 1997 Study .

LIDAR AND AEROSOL DATA – BC, SULFATE, NITRATE, NH<sub>3</sub> Single Particle Mass Spectrometers, DOAS, etc.

→Daily Flux Estimates of the Mexico City Plume!

→Comparison to Modeled Emissions

→SPATIALLY CORRECT?

#### **MEXICO CITY MEGACITY 2006**

#### PLANNING INITIATED FOR JOINT AIRCRAFT & GROUND BASED STUDY COLLABORATION WITH MCMA 2006 – MIT MIRAGE 2006 - NSF









#### SOME FOCUS AREAS

#### • DETAILED CHARACTERIZATION OF A MEGACITY PLUME

INTEGRATED AEROSOL SOURCE STRENGTHS FROM A MEGACITY – FOR MODEL COMPARISON

NSF and DOE Aircraft

ASP Meteorology (RASS, SODARS, LIDARS)

Ground Based Measurements

SECONDARY AEROSOL FORMATION & AGING – DOWNWIND PLUME MEASUREMENTS – BLACK CARBON, ETC.

NIGHTTIME RADIATIVE FORCING – "SMUDGE POT" EFFECTS (Jacobsen 2002) – GROUND BASED MEASUREMENTS.