Max Mex: Regional Climate Effects of Mexico City

Jeffrey S. Gaffney
University of Arkansas at Little Rock

NARSTO Meeting
Washington, D.C. March 27, 2007
Radiative Forcing by Tropospheric Aerosols

Partial Reflection and Absorption of Incoming Solar Radiation

Aerosol Haze → Clouds

- Organics
- Dust
- SO₂
- Soot
- Sea salt

Land Use Changes → Industrial Emissions → Biomass Burning → Ocean
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

condensation

evaporation

surface chemistry

coagulation

water uptake

oxidation

precursor emissions

activation

scavenging

diffusion

subcloud scavenging

evaporation

new particle formation

aqueous chemistry

primary emissions

dry deposition

Ghan and Schwartz 2007 in press BAMS
Types of Aerosols

**Primary Aerosols**
- Combustion
- Diesel Soots
- Biomass Burning

**Secondary Inorganic Aerosols**
- $\text{NH}_4\text{NO}_3$, $(\text{NH}_4)_2\text{SO}_4$, 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

*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

*National Geographic*
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 $\text{SO}_2$, $\text{NO}_x$.

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 HO$_2$ concentrations – High UV
- 1997 and 2003 – strong diurnal variation – indicating basin clearing on daily basis
2003 – Mexico City Dependence of aerosol absorption on time of day

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

Strong 370 nm absorption by secondary organic aerosols in afternoon – and impact by biomass burning – supported by \(^{14}\text{C}\) and other tracers.

1/\(\lambda\) dependence for broadband absorbers

Marley Gaffney, UALR
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

J. Fast, PNNL
Pre Field Campaign Activities

Ground Sites (DOE/NSF)

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

Aircraft Operations - Veracruz

DOE G-1

NASA King-Air
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 Schmoltner

**INTEX-B** – *Intercontinental Chemical Transport Experiment (NASA, NSF)*
NASA: Lead Scientist, Hanwant Singh
Program Manager, Bruce Doddridge

---

**MCE²**
Geographic Relation of Projects

G-1 (DOE)  C-130 (NCAR)  King-Air (NASA)  Twin Otter (U Montana)  J-31 (NASA)  DC-8 (NASA)
<table>
<thead>
<tr>
<th>Task</th>
<th>Details</th>
</tr>
</thead>
</table>
| 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                                                                                               |
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
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, NOₓ, 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)
### MAX-Mex Participants and Instrumentation

<table>
<thead>
<tr>
<th>Institution</th>
<th>Instrument/Device</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>ANL</td>
<td>Aethalometer (7 channel)</td>
<td>BC Aerosol absorption</td>
</tr>
<tr>
<td>ANL</td>
<td>Multi-angle Absorption</td>
<td>BC Aerosol absorption</td>
</tr>
<tr>
<td>ANL</td>
<td>Nephelometer 3 wavelength</td>
<td>particle scattering</td>
</tr>
<tr>
<td>ANL</td>
<td>Nephelometer 1 wavelength dry</td>
<td>dry particle scattering</td>
</tr>
<tr>
<td>ANL</td>
<td>Nephelometer 1 wavelength wet</td>
<td>wet particle scattering</td>
</tr>
<tr>
<td>ANL</td>
<td>Filter Sampler</td>
<td>OC/EC, humic like substances</td>
</tr>
<tr>
<td>ANL</td>
<td>Open path NIR TDLAS</td>
<td>NH3</td>
</tr>
<tr>
<td>ANL</td>
<td>Filter Sampler</td>
<td>14C, 40K, 210Pb, 7Be, 210Po, 210Bi,</td>
</tr>
<tr>
<td>ANL</td>
<td>RB Meter</td>
<td>UVB</td>
</tr>
<tr>
<td>ANL</td>
<td>Weather Station</td>
<td>wind speed/dir., rain, temp, press, RH</td>
</tr>
<tr>
<td>BNL</td>
<td>CCN Counter</td>
<td>cloud condensation nuclei</td>
</tr>
<tr>
<td>BNL</td>
<td>Scanning Mobility Particle Sizer</td>
<td>aerosol size distributions</td>
</tr>
<tr>
<td>DRI, U of Nev, Reno</td>
<td>Photoacoustic Spect.</td>
<td>aerosol absorption</td>
</tr>
<tr>
<td>PNNL</td>
<td>MFRSR</td>
<td>radiation, aerosol optical depth</td>
</tr>
<tr>
<td>PNNL</td>
<td>Solar Tracker</td>
<td>broad band radiometer</td>
</tr>
<tr>
<td>PNNL/EMSL</td>
<td>DRUM Aerosol Sampler</td>
<td>sampling for PIXE/PESA/STEM</td>
</tr>
<tr>
<td>PNNL/EMSL</td>
<td>TRAC Aerosol Sampler</td>
<td>sampling for TEM, SEM/EDX analysis</td>
</tr>
<tr>
<td>Aerodyne Res. Inc.</td>
<td>ARI H.R. TOF-AMS/soft ions</td>
<td>non-refractory fine PM size &amp; comp.</td>
</tr>
<tr>
<td>University of Colorado</td>
<td>High Res. TOF-AMS</td>
<td>aerosol size and composition</td>
</tr>
<tr>
<td>University of Colorado</td>
<td>Thermal Denuder</td>
<td>aerosol volatility before AMS</td>
</tr>
<tr>
<td>University of Colorado</td>
<td>Aerosol Concentrator</td>
<td>aerosol concentration before AMS</td>
</tr>
<tr>
<td>University of Colorado</td>
<td>Optical Particle Counter</td>
<td>aerosol size, number (Grimm 1.109)</td>
</tr>
<tr>
<td>University of Colorado</td>
<td>TSI SMPS</td>
<td>particle size distribution</td>
</tr>
<tr>
<td>University of Colorado</td>
<td>TSI nano-SMPS</td>
<td>nanoparticle size distribution</td>
</tr>
<tr>
<td>University of Colorado</td>
<td>TSI DustTrak</td>
<td>aerosol concentration</td>
</tr>
</tbody>
</table>
Field Campaign Period

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

*Example Briefing Material Available at [http://catalog.eol.ucar.edu/cgi-bin/milagro/report/index](http://catalog.eol.ucar.edu/cgi-bin/milagro/report/index)*

*Veracruz Forecasting Team*
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.

J. Fast, PNNL
G-1 Flights

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

S. Springston, L. Kleinman, BNL
Aircraft instrument intercomparisons provide high confidence in the measurements.

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

Aerosol Mass Spectrometer Measurements from G-1

Eastern U.S. Regional Pollution

- Sulfate
- Organics
- NH$_4^+$

Mexico City Urban

- Sulfate
- Organic
- Cl$^-$

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

L. Kleinman, BNL
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

S. Springston, L. Kleinman, BNL
Layering of Aerosols above T1

Aerosols lofted from Boundary Layer into Lower Troposphere

Wind Profiler

Ground-Based Micropulse Lidar
(Lidar return amplitude)

R. Coulter, ANL
Spatial Distribution of Aerosol Properties

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

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

complex structure over Mexico City
MC emissions transported southwest of city

C. Hostetler, R. Ferrare, NASA
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.

LaRC Airborne HSRL Measurements, Mexico City, March 13, 2006
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

Hostetler, Ferrare NASA; J. Fast, PNNL
Lidar Data Provide Vertical Context for In-situ Measurements

March 9, 2006

- HSRL and G-1 measurements show changes associated with MC pollution
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.
Aerosol Absorption – Changes due to Secondary Organic Aerosols and Biomass Burning

2003 – Mexico City
Departure from $1/\lambda$ behavior of 370 nm / 880 nm ratios for broadband absorbers. Note changes in aerosol absorption ratio vs time of day: AM (2400 – 1200) and PM (1200 – 2400)

MODIS satellite image showing wildfires April 18, 2003

- Satellite data MODIS indicated that plumes from Yucatan fires impacted the Valley of Mexico during this time in April 2003.
- $^{14}$C is consistent with transport from Yucatan to Mexico City during same time period.
- $^7$Be is produced in upper troposphere. Low levels are consistent with little upper air transport and mixing during this period
- $^{14}$C contributions could also be from SOA from natural hydrocarbons and from “biomass trash burning”

$^{14}$C results show 70% biogenic carbon in aerosols in Mexico City samples during end of April 2003. 4/21-4/27

Marley/Gaffney UALR
CaO inclusions and S-rich inclusions in soot particles

Y. Desyaterik, R. Hopkins, M. Gilles and A. Laskin; PNNL, LBNL
New particle formation was frequently observed.

New particles exhibited variable hygroscopicity indicating external mixture.
New Particle Formation

Organics and sulfur compounds appear to be important in the initial stages of particle growth.

TDCIMS “mode-following experiment” analyzes particles at the peak of the growth mode in order to identify the species responsible for aerosol growth.

TDCIMS data show initially higher organics and nitrate (closed symbols) during start of event with constant levels of sulfur species (open symbols).

J. Smith, NCAR
Smaller particles activate in later AM and PM (observed during many days).

Implications on cloud albedo and aerosol scavenging by precipitation.

Relation to increases in soluble components (sulfate, nitrate, and oxidized organics) being investigated.

---

J. Wang, BNL
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.

C. Doran, J. Barnard, W. Shaw, PNNL
Publications – Atmospheric Chemistry and Physics (ACPD and ACP) Special Issues

The T1-T2 study: evolution of aerosol properties downwind of Mexico City

A meteorological overview of the MILAGRO field campaigns
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: Final Data Sets – March 2007
MILAGRO Science Meeting – October 2006
Publications – Atmospheric Chemistry and Physics (ACPD and ACP)
Special Issues
EGU Symposium – Vienna, Austria (April 16-20, 2007)
MILAGRO Science Team Meeting (Mexico City – May 15-20, 2007)
AGU Symposium – Acapulco, Mexico (May 22-25, 2007)

Acknowledgments
ASP MAX-MEX Science Team
Pilots and ASP Science Support personnel
MILAGRO Participants
Our Mexican Hosts – INE, CENICA, SENEAM, IMP, UT Tecamac

THE PROJECT WAS CARRIED OUT SAFELY WITH NO INCIDENTS.