MIRAGE-Mex: Mexico City Pollution Outflow Experiment

SCIENCE OVERVIEW DOCUMENT

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NOTICE

This document is currently under consideration by the National Science Foundation (NSF). Contingent on NSF approval, the time-frame for submission of proposals by interested eligible participants is expected to be July-August 2004. Interested investigators should contact:

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http://www.nsf.gov/pubs/2004/nsf042/start.htm

ABSTRACT

An intensive observational mission is proposed to study the chemical and physical transformations of gaseous and aerosol pollutants in the outflow of the world's second largest metropolitan area, Mexico City. This mission, the Megacity Impacts on Regional and Global Environments – Mexico City case study (MIRAGE-Mex) is tentatively scheduled for 15 February – 15 March 2006, and will involve coordinated aircraft and ground-based measurements supported by extensive modeling and satellite observations. The overall scientific objectives of this study are:

(1) *Characterization of the extent, persistence, and potential impacts of the Mexico City plume in the surrounding regions.*

(2) *Increased understanding of the evolution of gas-phase reactivity over the urban-to-regional-to-global scale transition*.

(3) *Increased understanding of the physical and chemical evolution of aerosols and their radiative properties over the urban-to-regional-to-global transition.*

(4) *Increased understanding of the gas-aerosol interactions over the urban-to-regional-to-global transition*.

Participation by researchers from academia, government labs, and other institutions is essential for the success of this campaign. Significant opportunities for education and capacity-building are expected.

A. BACKGROUND

The export of air pollutants from urban to regional and global environments is a major concern because of wide-ranging potential consequences for human health and cultivated and natural ecosystems, visibility degradation, weather modification, changes in radiative forcing, and tropospheric oxidation (self-cleaning) capacity. Satellite, aircraft, and ground-based observations throughout the global atmosphere, as well as model simulations, indicate that air pollution can be transported over long distances, e.g., from eastern Asia to the western U.S., from North America to Europe, and from mid-latitudes to the Arctic[1-5]. Tropospheric oxidants, changes in precipitation chemistry, and reduced visibility are already significant environmental issues in much of the industrial Northern Hemisphere[6-9]. Globally, current levels of pollution-related tropospheric ozone and aerosols are significant contributors to the atmospheric "greenhouse" radiation budget [10-13]. Long-term changes in global OH concentrations, and therefore in the atmospheric residence times of many important gases, are a matter of great interest but remain highly uncertain[14,15].

Future population growth will undoubtedly exacerbate the large-scale impacts of urban air pollution. According to recent United Nations estimates[16] the world's population will increase by 36% from 2000 to 2030, led by a doubling of the number of urban dwellers in less developed regions. Many cities in developing countries already experience serious air quality problems and, for various socio-economic reasons, are likely to benefit more slowly from advances in emission-reducing technologies.

Clearly export of pollution is an enormously important issue, but also one for which scientific understanding is severely challenged by the complexity of physical and chemical processes that must be understood over a broad span of spatial and temporal scales. These include the primary emissions of a myriad of gases and particles; mixing

within and ventilation from the urban boundary layer; chemical transformations by thermal, photolytic, surface, and multi-phase reactions; microphysical processes such as nucleation, condensation, evaporation, and coagulation; perturbations of radiative heating and photolysis rates; convective and advective transport on local, regional, and global scales; mixing with reactive background air (e.g. biogenic emissions, lightning NO_x, biomass burning); removal of gases and aerosols from the atmosphere by wet or dry deposition; and eventual impacts on and feedbacks with the biosphere.

We believe that substantial progress in this understanding can be achieved through an intensive observational campaign that detects and follows the aging urban plume downwind of a large metropolitan area. Simultaneous measurements of a large suite of gas species and aerosol chemical/physical properties are now possible and offer unprecedented opportunities to identify and quantify the processes that control the evolution of the plume, its ultimate fate, and its potential large scale impacts. Previous field campaigns have studied pollutants in the remote troposphere, the outflow from extended source regions such as East Asia, the Indian subcontinent, and North America, and large scale transport (ACE-1[17], MLOPEX I&II[18,19], TOPSE[20], ACE-Asia[21], PEM-West A and B[22-24], PEM-Tropics A and B[24-26], TRACE-P[27-29], INDOEX[30], TARFOX[31], AEROCE[32], ACE-2[33], NARE I & II[34,35], ITCT 2K2[36]). Studies at the urban-regional transition scale have been limited mostly to the US and Europe (SOS[37-39], SCOS-97[40], TexAQS-2000[41], ESCOMPTE42, SLOPE[43], BERLIOZ[44], PIPAPO[45]). However, few if any previous field campaigns have focused on the polluted outflow from the major tropical megacities, and certainly not with the comprehensive chemical/physical instrumentation proposed here.

B. OVERVIEW OF THE PROPOSED CAMPAIGN AND ITS SCIENTIFIC OBJECTIVES

MIRAGE-Mex (Megacity Impacts on Regional and Global Environments – Mexico City case study) is an intensive observational mission to study the chemical and physical transformations of gaseous and aerosol pollutants in the outflow of the world's second largest metropolitan area, Mexico City (MC). This intensive field campaign is tentatively scheduled for 15 February – 15 March 2006, and will involve coordinated aircraft and ground-based measurements supported by extensive modeling activities and satellite observations. The NSF C-130 aircraft is requested for this campaign. The field campaign is an outgrowth of a workshop held in November 2002 at the National Center for Atmospheric Research in Boulder, Colorado. The workshop was attended by researchers from 28 universities (6 from outside the US) and various US and Mexican government agencies. Participants reached broad consensus on the need for such a campaign and contributed to the formulation of its scientific objectives. Continued participation by researchers from the community will be essential for the success of this campaign.

MIRAGE-Mex will address some of the major uncertainties in the atmospheric science of aging air pollution, specifically on the transport, physical and chemical transformations, and evolving radiative properties of gases and aerosols. The focus will be on understanding the life-cycle of these pollutants, from emission to ultimate removal from the atmosphere, and especially the "middle-age" transition from urban to regional and global scales. While this field campaign will benefit the MC region directly by

providing the first comprehensive characterization of its pollution problem, the mission is aimed more broadly at understanding the ***processing*** of pollutants in general urban outflows. This increased knowledge will result in significant improvement in model representations of these processes, and thus will be relevant to many other locations and especially to the growing number of tropical megacities. Such increased understanding, and the associated improvements in modeling capabilities, will be essential toward assessing the current and future impacts of global urbanization on climate, human health, and ecosystems. The overall objectives of MIRAGE-Mex fall into four interconnected categories, listed below.

(1) ***Characterization of the extent, persistence, and potential impacts of the Mexico City plume in the surrounding regions:*** Remarkably, although extensive air quality measurements have been made for many years inside MC, essentially no information is available on the composition of air in the surrounding areas, either at the surface or aloft. Satellite-based images (Fig. 1) and trace gas observations (Fig. 2), preliminary model results (Fig. 3), and anecdotal accounts all indicate that the impact of MC emissions should be observable for many hundreds of kilometers downwind. The environmental impacts (e.g. on regional air quality, vegetation, visibility) are currently unknown.

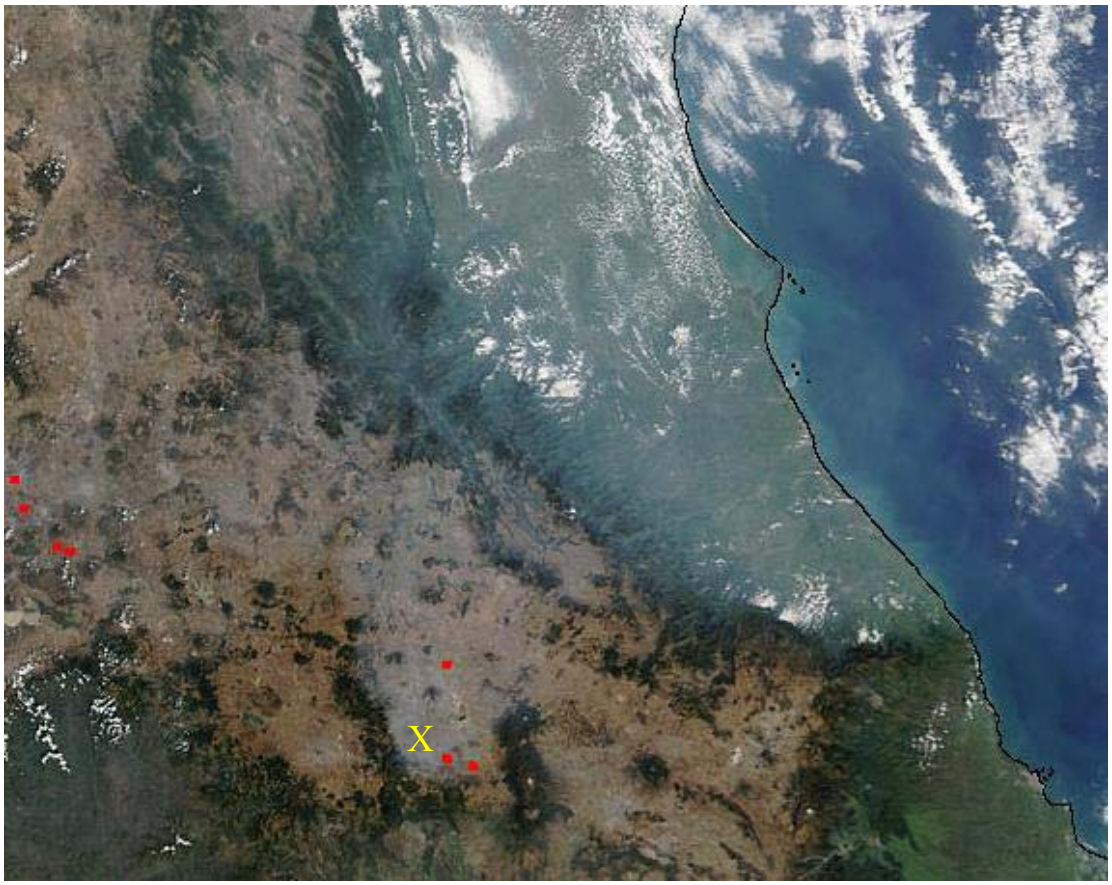

Figure 1*: Satellite image of central Mexico (4 Dec.'02). Fires are indicated by red dots, Mexico City by yellow X. Plume is visible as haze spreading from the plateau to the Gulf of Mexico. (Credit**:** Jeff Schmaltz, MODIS Rapid Response Team, NASA/GSFC)*

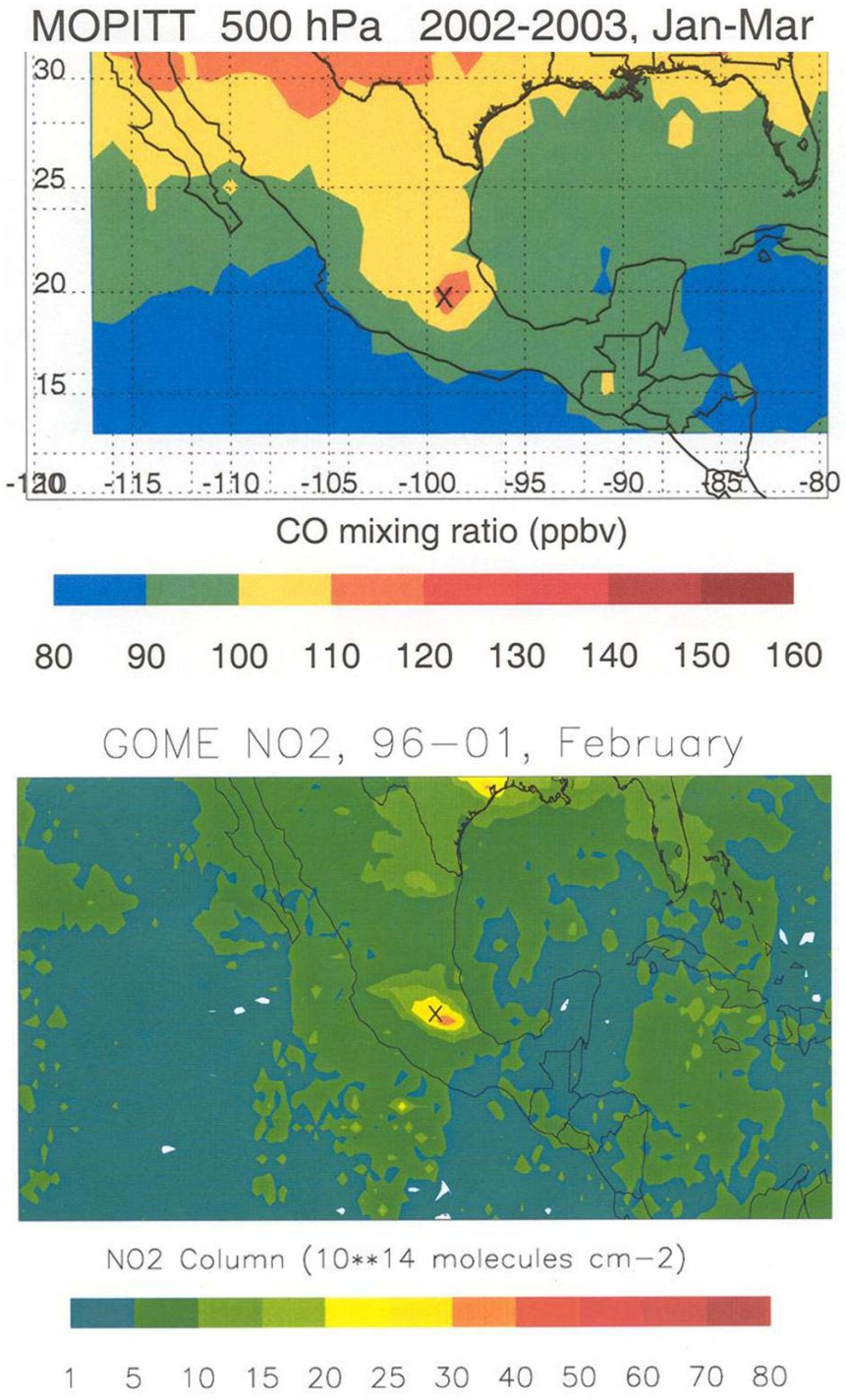

Figure 2: *Satellite-based observations of trace gases over Mexico. Top: Carbon monoxide (CO) concentration from MOPITT instrument aboard NASA's Terra satellite (MOPITT science team, NCAR). Bottom: Nitrogen dioxide (NO_2) tropospheric column from GOME instrument aboard ESA's ER2 satellite (courtesy John Burrows, U. Bremen).*

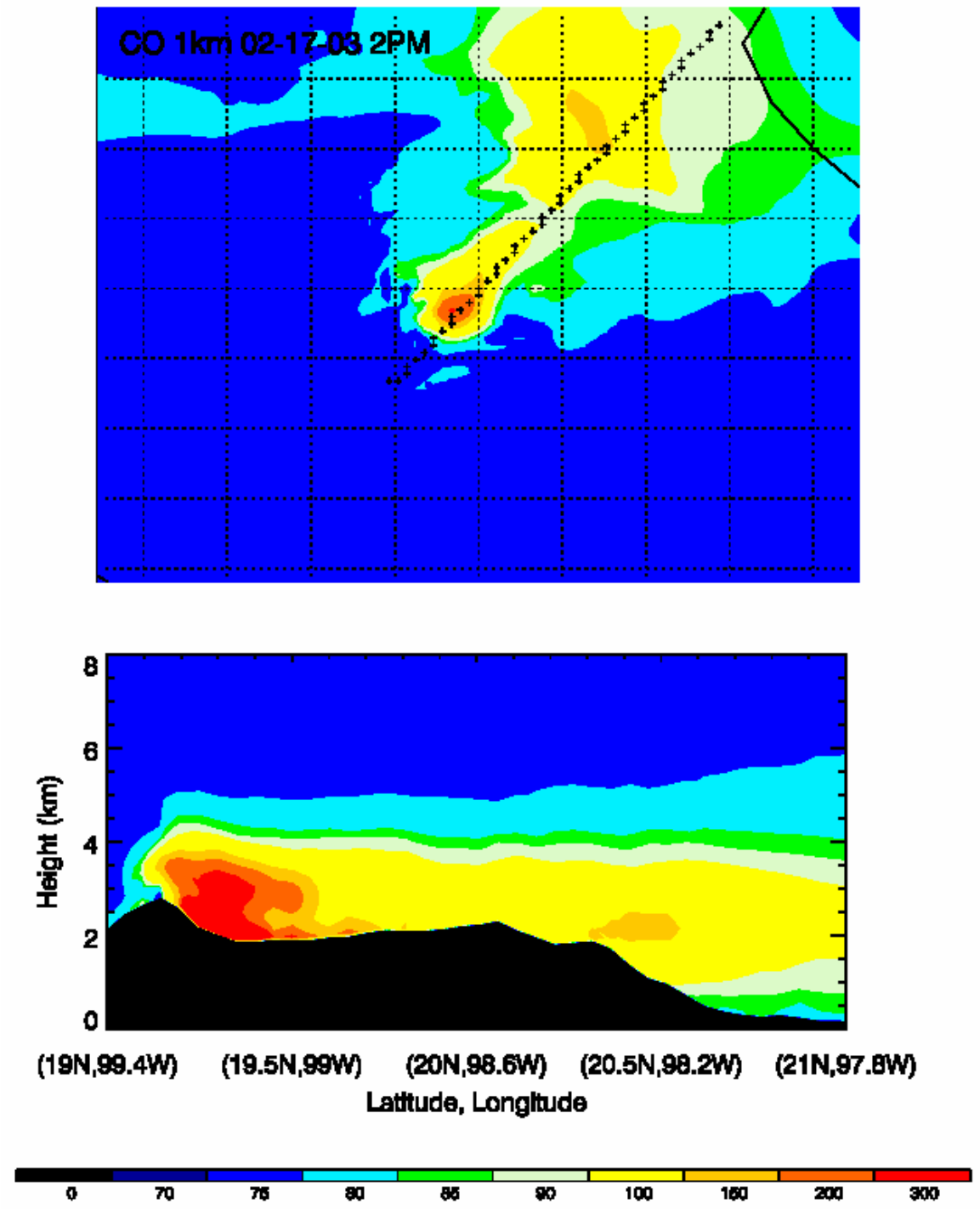

***Figure 3**: Carbon monoxide (CO) concentrations, ppbv, computed with a preliminary WRF-Chem 2-day simulation at 6 km resolution (600 x 600 km domain), 16-17 February 2003. Mexico City is located in the center of the top panel; the Gulf coast is visible in the upper right. Symbols in upper panel denote direction of cross section in lower panel. (courtesy X. Tie, NCAR)*

The region also experiences emission from vegetation and biomass-burning (depending on the time of the year), though no estimates are available on their importance relative to the urban plume. Interactions between the plume and regional emissions could fuel additional oxidant production, but no quantification exists at the current time. For MIRAGE-Mex, flight plans will be developed (and modified during the campaign according to next-day forecasts) to traverse the plume at different altitudes and several different distances from the city, in order to assess the plume extent and compare chemical concentrations in the plume and in the background air.

(2) ***Increased understanding of the evolution of gas-phase reactivity over the urban-to-regional-to-global scale transition*:** While urban reactivity is widely understood in terms of primary pollutants such as nitrogen oxides (NO_x = NO + NO_2) and a multitude of hydrocarbons (HCs), the reactivity of the aging plume is expected to be dominated by a much larger number of intermediates produced by the photo-oxidation of the initial compounds. This intermediate chemistry is arguably the most uncertain regime in our understanding of the atmospheric life cycle of anthropogenic emissions. The fully explicit chemistry, expected to involve thousands of intermediate species based on laboratory kinetic/mechanistic data, has so far been considered only in box (zero-dimensional) models such as the NCAR and Leeds Master Mechanisms. These detailed chemical models predict that elevated concentrations of intermediate volatile organic compounds (VOCs) persist in the atmosphere for many days (see Fig. 4) and can even dominate the gas phase reactivity (Fig. 5).

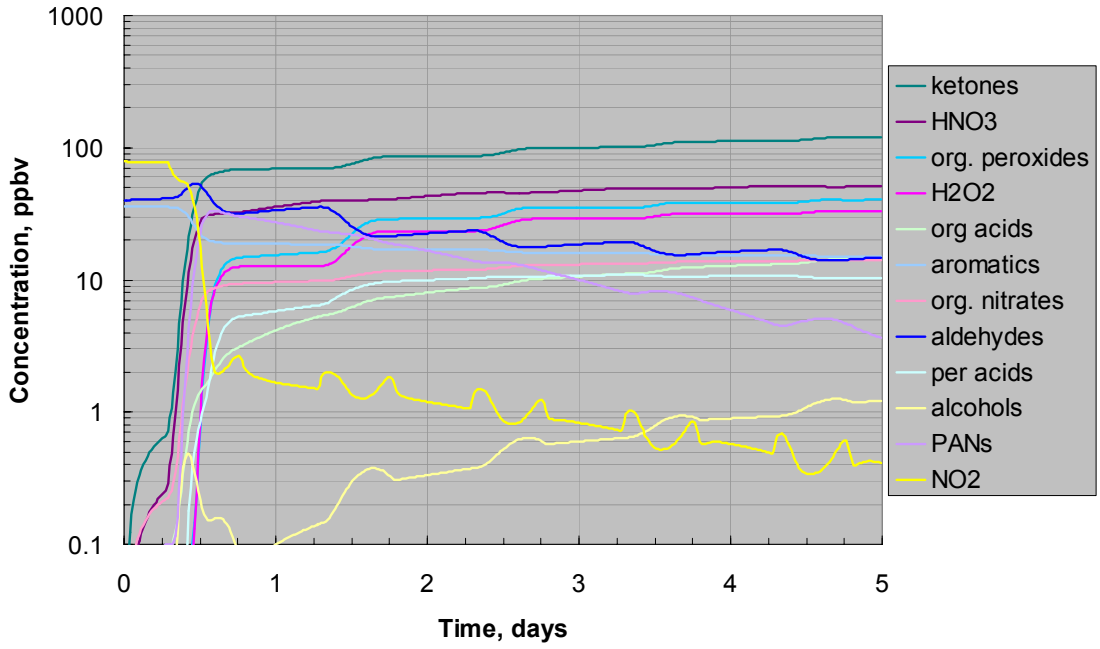

Figure 4: *Evolution of gas-phase species in an air parcel initialized with concentrations of NOx, CO, and hydrocarbons observed in Mexico City. Box model simulation with NCAR Master Mechanism, assuming no additional emissions, no dilution, and no heterogeneous processes.*

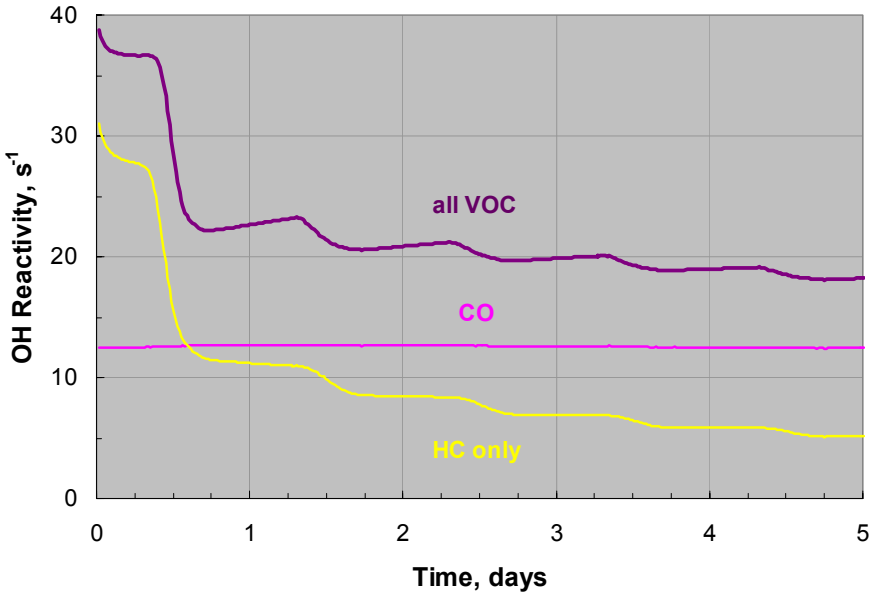

***Figure 5**: Evolution of OH reactivity for the air parcel of Fig.4. The reactivity is dominated by the volatile organic carbon (VOC) which includes hydrocarbons (HC) and intermediate HC oxidation products such as aldehydes, ketones, alcohols, and organic nitrates, peroxides, and acids.*

On the other hand, three-dimensional models typically use highly simplified chemical mechanisms in which surrogate or lumped species (a few hundred at most) are used to attempt to capture some chemical characteristics of the true mixture. It is currently unknown whether such simplified mechanisms are able to represent the chemistry downwind of large pollution sources such as MC. The MIRAGE-Mex campaign will provide a wealth of chemical measurements along the plume to evaluate (and improve) the models, including the concentrations of major oxidants (e.g. O_3, H_2O_2, organic peroxides, acids), radicals (primarily OH, HO_2, RO_2), longer-lived species that contribute to the global budgets of odd nitrogen (PANs, organic nitrates) and odd hydrogen (photo-labile organics such as ketones and aldehydes), aerosols (see below), as well as physical controlling variables such as temperature, pressure, relative humidity, and spectral actinic fluxes. These measurements will help establish whether the chemistry of the MC outflow air proceeds at a vigorous pace for several days, forms large amounts of NO_x and HO_x reservoir species, and is therefore likely to have substantial impacts on regional and global environments.

(3) ***Increased understanding of the physical and chemical evolution of aerosols and their radiative properties over the urban-to-regional-to-global transition:*** Megacities, including MC, are strong sources of both primary and secondary aerosols. While much aerosol evolution (e.g. emission, nucleation, coagulation, growth) is expected to occur rapidly within the city (see Fig. 6), an open question is how much additional processing

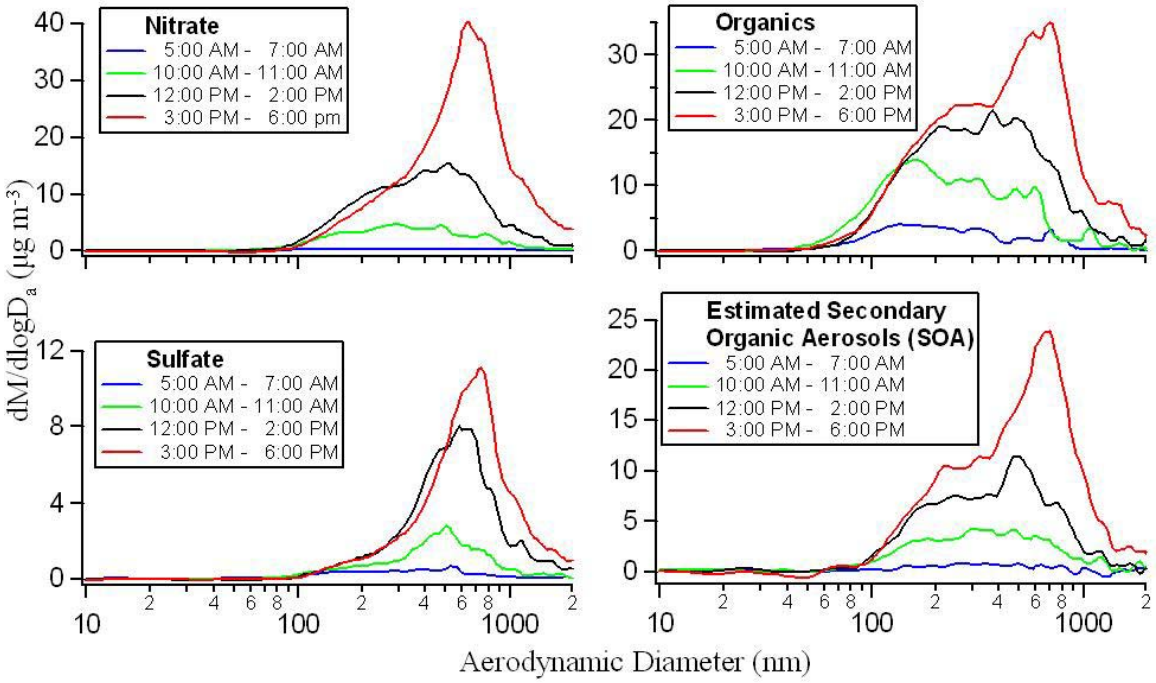

Figure 6: *Diurnal changes in non-refractory aerosol size distributions in México City measured during the MCMA2003 field campaign with the Aerosol Mass Spectrometer (AMS). Secondary organic concentrations, estimated from m/z=44, should be considered preliminary (courtesy Jose Jiménez, U. of Colorado).*

occurs downwind. Precursors of inorganic aerosol, such as SO_2, H_2SO_4, HNO_3, and NH_3 are expected to persist for some time in the plume. Furthermore, many photochemically-generated oxygenated organics are likely to contribute to aerosol evolution, through condensation, uptake and surface reaction. Aerosols can remove some species from the gas phase, transform others by surface reactions, and modify (by scattering and/or absorption) the ultraviolet radiation field that drives gas phase photochemistry. In addition, as particles age, they mix internally by coagulation and growth. These many processes ultimately determine the optical properties of aerosols (and therefore their direct impact on radiative budgets) and their microphysical characteristics (with potential impacts on clouds and precipitation). The MIRAGE-Mex campaign will provide measurements of aerosol chemical, microphysical, and optical properties along the plume, simultaneously with the gas phase measurements.

(4) ***Increased understanding of the gas-aerosol interactions over the urban-to-regional-to-global transition:*** Implicit in objectives (2) and (3) above, but worthy of further emphasis, is the important – but also highly uncertain – potential for gas-aerosol interactions in the outflow, for both inorganic and organic species. For example, many of the oxygenated organic intermediates generated by the photochemistry are expected to be multifunctional polar compounds with low vapor pressures. These compounds may nucleate new particles, adsorb on or absorb in existing particles, and/or react on the surface of aerosols. At the same time, their transfer to the condensed phase may have a profound influence on the residual gas-phase reactivity of the urban plume (e.g. suppressing the gas-phase reactivity of VOCs shown in Fig. 5). The state-of-the-science

aerosol and gas-phase measurements on the C-130 will provide insight into the nature of these interactions and their impacts on both the gas and aerosol phases. Furthermore, a suite of more detailed ground-based measurements (speciated aerosol composition, measurement of multi-functional gas-phase organic intermediates) is proposed.

C. THE MEXICO CITY CONTEXT

1. Rationale for Choosing Mexico City

No one megacity can fully represent all of the world's megacities, nor the thousands of smaller urban areas that contribute emissions to the regional and global troposphere. There are, however, several compelling reasons for selecting MC for the proposed pollution outflow study.

a) MC is, and will continue to be, one of the world's most populous cities, with a current population of ca. 18 million. It is situated in the tropics, as are most of the world's fastest growing megacities. Its emission characteristics are roughly intermediate between those of a city in emerging economies and those of a city in fully developed countries (see Table 1 for comparison with Los Angeles). The national O_3 standard (110 ppb/1hr) is routinely exceeded but some improvement occurred since the early 1990s when the Mexican government began instituting aggressive emission-reduction programs.

Table 1: *Comparisons between Los Angeles and Mexico City. From Ref. 47 except as noted.*

	Los Angeles Air Basin	Mexico City Metropolitan Area
population	15,000,000	18,000,000
total area (km^2)	27,800	5300
urbanized area (km^2)	17,500	1500
NO_x emissions ($Gg\ yr^{-1}$)	400	206
% NO_x from vehicles	80	80
VOC emissions ($Gg\ yr^{-1}$)	362	475
% VOC from vehicles	40	40
total VOC (ppmC)	0.36 (ref. 48)	4.1 (ref. 49)
aromatic VOC (ppmC)	0.05 (ref. 48)	0.25 (ref. 50)

b) Although regional air quality problems in the Mexico and Central America are widely acknowledged, this geographical area remains under-investigated.

c) MC is a very strong pollution source surrounded by a region that is only moderately polluted. The influence of urban emissions should be clearly discernible for at least several hundred kilometers down-wind, providing a unique opportunity to improve our understanding of the urban-to-regional scale transition.

d) Considerable knowledge on the air quality problem within the city has already been gained by local university, government, and industrial scientists, partly through collaborative studies with researchers from the U.S. and other countries. Inventories of anthropogenic emissions in MC have been developed and are being refined. Studies are underway to characterize biogenic emissions from the areas surrounding the city. A network of ground-based air quality measurements (HCs, NO_x, O_3) has been operating continuously for over a decade. Extensive data have also been gathered on aerosols

(chemical and physical properties), on surface radiation (visible, UV) and on boundary layer evolution and regional-scale diurnal circulations.

e) Compared with other tropical countries, Mexico offers good infrastructure and logistics for carrying out an intensive measurement campaign involving aircraft.

2. Meteorology of Mexico City and Surrounding Region

Mexico City is located at 19°N, 99°W in a basin roughly 2.2 km asl, surrounded by mountains on three sides. Two distinct seasons, wet (May – September) and dry (November – March) occur, with the other months being transitional. The preferred time for MIRAGE-Mex is the dry season, to avoid some of the additional complication associated with the more convectively dominated months and the less predictable transition months. Climatologic winds at 700 mb in February are shown in Fig. 7. Over central Mexico synoptic flow tends to transport pollution emitted from the city east-north-eastward into the Gulf of Mexico, although these winds are relatively weak and transport in other directions could occur. Forward 1-day trajectories (HYSPLIT-4[46], starting at 10, 1000, and 2000 m above ground) for February and March 2002 and 2003 were towards the NE on ~50% of the days, 25% to SW, 15% to NW, and 10% to SE.

The local circulation was studied extensively during the IMADA-AVER field campaign (February-March 1997, see below). Meteorological measurements at four main sites were equipped with radar wind profilers, sodars, and radiosondes that were released five times per day. Supplemental radiosondes were also launched at two sites outside MC. The measurements revealed circulations aloft that were previously unknown, and mesoscale modeling studies indicated that pollutants were vented out of MC on a daily basis with little multi-day accumulation. The simulations (Fig. 8) indicate that during

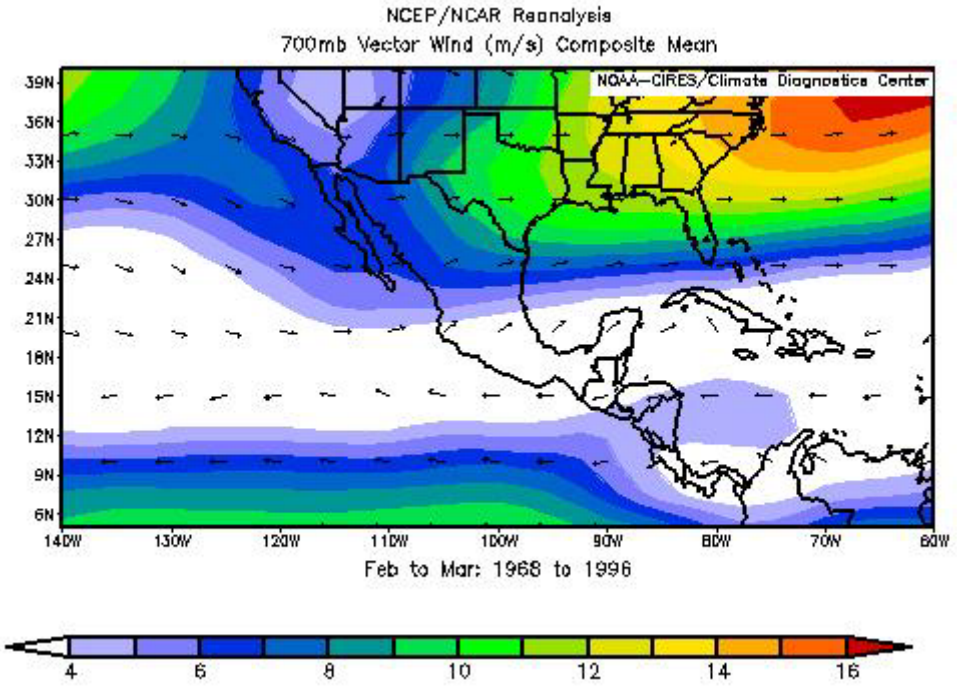

***Figure** 7: Wind climatology for February over the Mexico.*

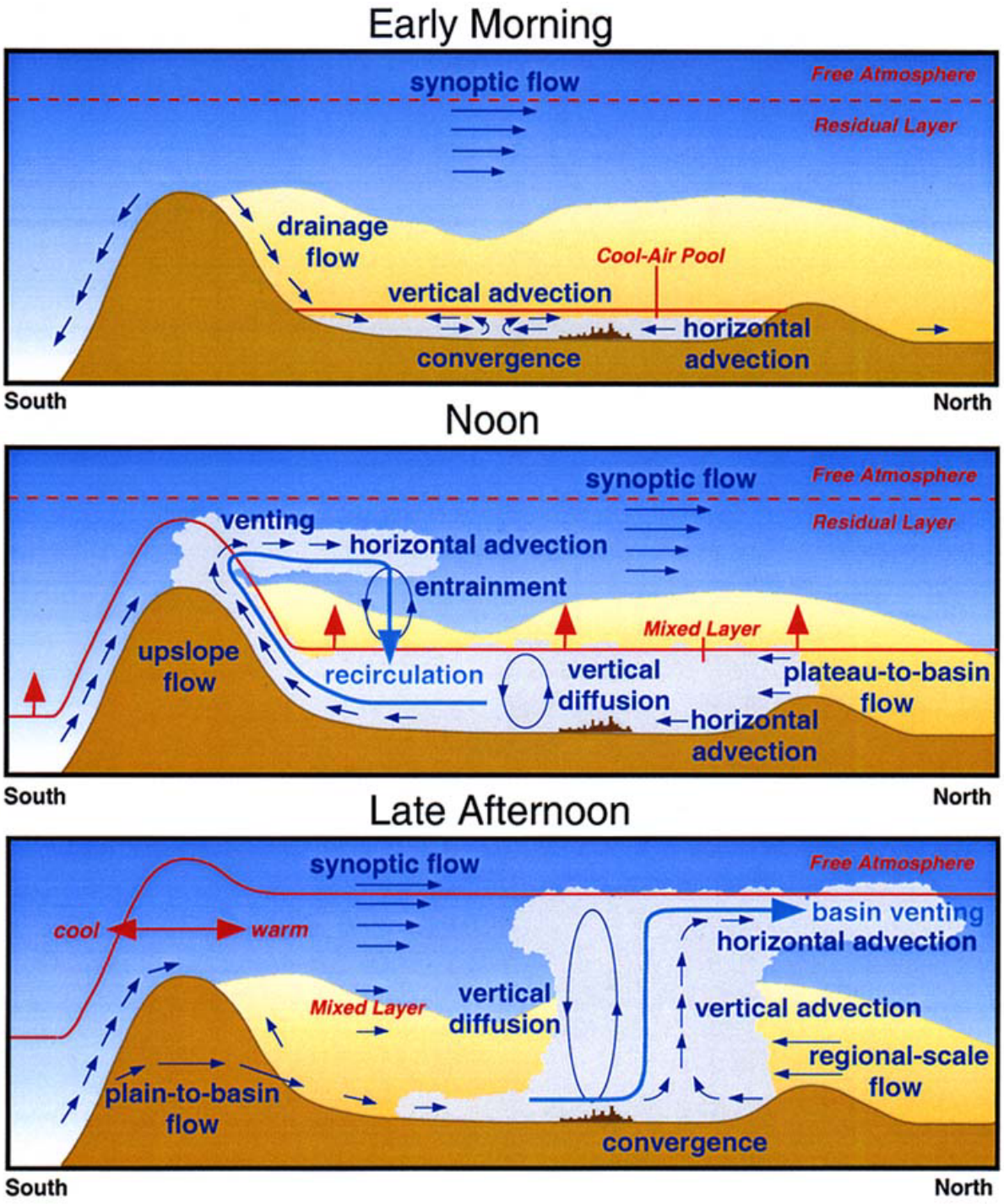

Figure 8: *Conceptual illustration of Mexico City's circulation and outflow patterns (courtesy Jerome Fast, PNNL).*

the dry season, pollutants may be vented out of the city at mid-tropospheric altitudes (4-5 km asl) due to the already high surface elevation, upslope flows on the surrounding mountains, and the diurnal growth of the PBL to depths of over 2 km above ground. A portion of the pollutant plume is likely to become decoupled from the surface as it is transported from the plateau over the lower terrain along the Gulf of Mexico.

A very different meteorological situation, common in February-March, is associated with the so-called "Norte" events. These events, caused by mid-latitude systems propagating into the tropics, are characterized by strong synoptic flow of cold air from the north, sometimes lasting several days. In the context of MIRAGE-Mex, the rapid and complete flushing of the basin towards the south during Nortes should make it easy to find and observe the outflow from the MC basin, and would additionally ensure a clean initial state for the MC atmosphere in the post-Norte days.

3. Previous Observations and Modeling of the Mexico City Region

The current state of knowledge about MC's air quality derives from numerous measurement and modeling programs, including long term monitoring and intensive observations by individual and multiple research groups. These studies have been reviewed extensively by Molina and Molina[47], and here we briefly summarize only some of the larger programs.

Continuous monitoring of air pollutants in MC began in 1986 with the establishment of several networks (RAMA, REDMA, REDMET)[51], now numbering 32 stations, to measure surface concentrations of O_3, NO_x, NO_2, CO, SO_2, TSP, and PM_{10}. Five additional stations are currently being integrated into the network, and additional measurement capability (formaldehyde, toluene, benzene, xylenes, $PM_{2.5}$) is being tested at some stations. Speciated hydrocarbon measurements are made every 6 months by the Instituto Mexicano del Petroleo (IMP). The RAMA measurements form the basis of the air quality index used for public information, and by the government to declare air pollution contingencies. Meteorological measurements, begun in 1948 by the Mexican National Meteorology Service, include hourly surface observations at 2 sites and two rawindsonde launches per day. These are complemented, since 1986, by surface observations at 10 RAMA sites and one UNAM site. RAMA has also been operating a sodar continuously at one location since 1999.

Several large intensive field campaigns have provided a wealth of additional meteorological and air composition data. The Mexico City Air Quality Initiative (MARI[52]), organized by IMP and Los Alamos National Laboratories, carried out four measurement intensives (Sep.'90, Feb.'91, Mar.'92, and Mar.'93). The first two intensives focused on vertical profiles of meteorological data, but also obtained some profiles of SO_2, O_3, and particulates (non-speciated). Interestingly, high O_3 values were found in the top part of the PBL, possibly due to strong vertical gradients of photolysis rates resulting from heavy aerosol loading[53]. The latter two campaigns included some sodar and lidar profiles, but primarily focused on *in-situ* hydrocarbon measurements at several sites, showing remarkably high VOC/NO_x ratios. The IMADA-AVER[49,54] activity was a large field campaign in the MC region from February 23 to March 22, 1997. In addition to extensive meteorological observations (Sect. C.2), surface air-quality measurements were made including PM_{10} and $PM_{2.5}$ and their chemical composition,

light scattering and absorption, nitric acid, ammonia, gas-phase hydrocarbons, polyaromatic hydrocarbons, and PAN.

The most comprehensive photochemical measurements within MC were made during the MCMA-2003 field campaign (March 31-May 4, 2003), led by L. and M. Molina of MIT in collaboration with researchers from many Mexican and US universities, government labs, and research institutions. Measurements at an urban "supersite" included, in addition to the more routine measurements, detailed speciated hydrocarbons and reactive nitrogen, size-resolved aerosol composition, LIDAR profiles of ozone and aerosols, as well as radical species. Additional measurements were made at a mountain site on the southern edge of the city, and throughout the city from a mobile laboratory developed by Aerodyne, Inc. This rich data set is currently being analyzed and prepared for publication, and will be of great utility in understanding the initial photochemical state of MC during the MIRAGE-Mex field campaign.

Emissions inventories for MC have been under development since 1986 and are periodically updated by the Mexican government. The most recent inventory, for the year 1998 by the Comision Ambiental Metropolitana[55], was constructed bottom-up from point, area, mobile, and natural sources, with totals of PM_{10} ~ 20, SO_2 ~ 22, CO ~ 1769, NO_x ~ 206, and HC ~ 475 Gg yr^{-1}. Substantial uncertainties exist, particularly concerning the HC emissions. An updated and refined inventory will be available in advance of the 2006 MIRAGE-Mex field campaign. A national emissions inventory is under development and is expected to be available in 2004 (P. Fields/ERG, personal communication, 2004).

Air quality modeling has been carried out in association with the major measurement programs (RAMA, MARI, IMADA, MCMA2003), as well as in individual studies summarized by Molina and Molina. Several of these studies suggest that current inventories are underestimating HC emissions, and the question of whether O_3 production is NO_x or HC limited remains open. More recent modeling studies have simulated wind-blown dust in the MC basin[56], particulate distributions and their effect on visibility[57] vertical recirculation patterns and their effect on pollutant concentrations[58] and the effect of a partial restoration of Lake Texcoco on local meteorology and ozone[59]. While these studies have increased our understanding of meteorological and chemical processes within the MC basin, there are no modeling studies to date that have examined the regional-scale evolution of trace gases and aerosols produced from MC emissions and how the MC plume mixes with regional and global background concentrations.

D. SPECIFIC SCIENCE OBJECTIVES

The MIRAGE-Mex field campaign will provide the first detailed characterization of tropospheric chemistry of the region surrounding MC. While this is an important goal in itself, we believe that the MC outflow offers unprecedented opportunities to study the complex array of chemical and physical processes occurring during urban plume aging. Many of these processes, such as the fate of organic oxidation intermediates, gas-aerosol interactions, and the evolving optical and microphysical properties of aerosols, are currently not well understood and are poorly represented in models. Major topics, related questions, and the proposed approaches are given below.

1. Geographical extent and temporal persistence of the urban plume: *How far from the city can its outflow be distinguished from background air? Can elevated regional levels of pollutants be correlated with urban outflow? Are there unique chemical signatures of the urban plume?*

Approach: The determination of the geographical extent and temporal persistence of the urban plume will primarily be established from the C-130 aircraft (see Sect. E.2). LIDAR measurements of O_3 and aerosols from onboard the C-130 will determine the vertical extent of the plume. Measurements of tracer species (e.g., aromatic hydrocarbons, acetylene/propane ratios, soot/PAH ratios, as MC outflow tracers; CH_3CN and K^+ in fine aerosol as biomass burning tracers) using canister sampling and/or on-line GC or PTR-MS techniques will further establish the extent of the plume and allow differentiation between the MC outflow and air masses influenced by biomass burning. Measurements of the by-products of MC outflow oxidative chemistry (e.g., O_3, nitric acid, peroxides, organic acids, unique products of aromatic hydrocarbon oxidation such as glyoxal and aromatic PAN derivatives) will also be used to distinguish the urban outflow from background air. These measurements will be used to test how well regional models (e.g. WRF-Chem) predict the spatial and temporal evolution of the plume.

The capabilities of satellites in monitoring key tropospheric indicators of air quality are currently expanding rapidly, and these data will be used to examine the magnitude and geographical extent of the MC plume as best as possible. Potentially useful remote sensing data include CO from MOPITT; NO_2 and CH_2O from SCIAMACHY; CO and O_3 from TES; and aerosol optical depth from MODIS and TOMS.

2. Regional oxidants production: *How much potential for O_3 production remains in the aging MC outflow, and how does it compare with the initial ozone production? How does net O_3 production (positive or negative) compare with that of background air? What are the concentrations of other oxidants (e.g., peroxides and acids) and how do they compare with background air? What is the role of secondary oxidation products in oxidant production? Are gas-phase oxidant precursors (radical and non-radical) lost on aerosols? Are actinic flux perturbations by aerosols altering substantially the oxidant photochemistry?*

Approach: The magnitude of the production of ozone and other oxidants in the MC outflow will be determined from aircraft-based measurements, inside and outside the plume, of the oxidants themselves, their radical and non-radical precursors, and related species (e.g., O_3, H_2O_2, organic peroxides, nitric and sulfuric acids, organic nitrates, aerosol nitrate and sulfate, organic acids, PANs, OH, HO_2 and organic peroxy radicals, NO_x and NO_y, and VOCs and their oxygenated by-products). In addition, LIDAR data will provide a measure of the total ozone burden integrated over the plume extent. Measurements of actinic flux and aerosol size distributions and composition will be made and used to determine the impact of the aerosols on photolysis and oxidant production rates.

Issues related to the uptake of multi-functional organics onto aerosols and the impact of these processes on oxidant production will be further investigated on the ground. Newly-developed instrumentation will be deployed at a "supersite" located outside MC

(see Sect. E.4) to determine gas-phase levels of oxygenated and multi-functional organics, as well as to speciate the organic content of the aerosols themselves.

3. Hydrocarbon oxidation products: *How does organic reactivity change with time downwind of MC (i.e., HCs versus oxygenated species)? What is the contribution of the oxygenates to the regional budgets of HO_x, O_3, and H_2O_2, and what is their impact on $HO/HO_2/RO_2$ ratios? To what extent are these secondary oxygenates taken up by aerosol or cloud water? What is the contribution of the MC outflow to the global budgets of some long-lived species (e.g. acetone, organic nitrates)?*

Approach: As with science objective #2 above, issues related to the evolution of organic reactivity in the MC outflow will be addressed via aircraft-based measurement of oxidants and the radical and non-radical species involved in their formation. Emphasis will be placed on the measurement of hydrocarbon oxidation products including aldehydes, ketones, alcohols, organic acids and nitrates and, to the extent possible, multi-functional species using CIMS and GC techniques. Note that, despite the complexity of the hydrocarbon oxidative chemistry, many of the oxygenated species predicted to be the most abundant (see Table 2) are routinely measurable. Concurrently, measurements of aerosol size distributions and composition will be made in the outflow to determine the extent to which gaseous organic intermediates partition to the condensed phase and to provide an assessment of the carbon budget in the plume. Comparisons of the gas- and condensed-phase measurements with detailed process models and regional chemistry/transport models will be made to evaluate model performance and guide improvements as needed. Comparisons between measured and modeled ratios of OH, HO_2, and RO_2 radicals will provide an estimate of the contribution of organic intermediates to radical budgets/partitioning and to oxidant production/loss.

Table 2: *Major non-methane hydrocarbons present in the MC (in approximate order of OH-reactivity), anticipated oxidation products and methods for their quantification.*

Parent Hydrocarbon	**Anticipated (measurable) Oxidation Products**	**Measurement Technique(s)**
butenes	acetaldehyde, propanal; formaldehyde; PAN, PPN	GC-MS, PTR-MS; TDL absorption; CIMS
butanes	MEK, acetone, acetaldehyde; PAN; butyl nitrates	GC-MS, PTR-MS; CIMS; whole air samples
ethene	formaldehyde	TDL absorption
xylenes	glyoxal, methylglyoxal; aromatic PANs	PTR-MS, GC-MS; CIMS
propane	acetone, propanal; isopropyl nitrate	GC-MS, PTR-MS; whole air samples
propene	formaldehyde; PAN acetaldehyde	TDL absorption; CIMS, GC-MS, PTR-MS
toluene	glyoxal, methylglyoxal; aromatic PANs	PTR-MS, GC-MS; CIMS
hexanes	C2-C6 aldehydes and ketones; alkyl nitrates; hydroxycarbonyls, hydroxynitrates	GC-MS, PTR-MS; whole air samples; GC-MS, PTR-MS

Cloud processing of the MC plume will not be a focus of the MIRAGE-Mex campaign. However, if and when the opportunity arises, measurements will be made on the C-130 of oxygenated intermediates in the gas- and condensed-phase before and after a cloud is encountered. Furthermore, analyses of rainwater collected on the ground will be conducted.

Detailed hydrocarbon oxidation chemistry and the gas/aerosol partitioning of organic intermediates will also be a focus of measurements made at the ground-based super-site. Direct measurements will be made of OH reactivity, and these data will be compared with reactivity calculated from measurements of individual species. Also, as already mentioned in objective #2, cutting-edge instrumentation will be deployed at the supersite to measure as large an array of multi-functional gas-phase organics as possible, and to speciate the organic aerosol to the extent possible.

4. Reactive nitrogen: *What is the amount and partitioning of NO_y species in the outflow, and what is the potential impact on the regional and global NO_x budgets? What is the role of reservoir species on ozone production far from the MC source? How is the NO_x budget of the remote atmosphere impacted by export of reactive nitrogen from MC? Is there a significant contribution to the NO_y budget from unusual or unexpected species, such as multifunctional $RONO_2$ or PANs, organic peroxynitrates, HNO_4, or HONO?*

Approach: The budget for reactive nitrogen species in the outflow will be assessed via the aircraft-based measurement of NO_y and its individual components (NO, NO_2, HNO_3, PAN and its analogs, organic nitrates, etc.). Possible contributions from (unmeasured) multi-functional nitrogen-containing species will be assessed from NO_y "deficit" calculations. These measurements, in conjunction with regional and global modeling activities, will indicate the contribution of nitrogen reservoirs (PANs and organic nitrates) on the NO_x budget in the outflow and on broader scales, and the impact of these NO_x sources on regional and global ozone production. Remotely-sensed NO_2 data (from SCIAMACHY) will be used to provide, to the extent possible, a global context for the aircraft-based NO_x/NO_y measurements.

In addition to the species being measured on the aircraft, more detailed measurements of reactive nitrogen species will be possible at the ground supersite, e.g. total and speciated alkyl nitrates, total peroxynitrates, and nitrophenols.

5. Gas-aerosol chemical processes: *What are the chemical and physical processes responsible for the composition and evolution of aerosols? What processes are involved in new particle formation, and how is this influenced by gas phase composition, aerosol surface area & meteorology? How does the ratio of secondary/primary organics change as a function of size and age? How do uptake and release of gas phase species by aerosols affect gas phase oxidant chemistry? How does the chemical composition of these aerosols affect their potential to modify clouds?*

Approach: The aircraft-based aerosol measurements will include size-resolved composition (volatile organic fraction, elemental carbon, and inorganic ions), CN and CCN number, and hygroscopicity. Differences between daytime and nighttime gas-aerosol processes will be investigated on some flights. Combined with simultaneous measurements of gaseous inorganic (NH_3, HNO_3, SO_2, H_2SO_4) and organic (multifunctional VOCs) precursors, these measurements should provide a rich data set for

examining gas-aerosol processes and evaluating models of aerosol evolution. Additionally, ground-based measurements will provide more comprehensive information, e.g. more complete organic aerosol and gas speciation, and analysis of collected rainwater.

6. Aerosol radiative properties: *How do aerosol optical properties (esp. optical depth and single scattering albedo - both as a function of wavelength) evolve during the outflow? How are these affected by aerosol size distributions, composition, and mixing state (internal vs. external)? How are aerosols distributed horizontally/vertically? How large are the aerosol impacts on spectral irradiances and actinic fluxes, and therefore on radiative budgets and photochemistry? What are the potential indirect effects of aerosols, e.g. in modifying clouds and precipitation?*

Approach: The simultaneous aircraft-based measurements of (i) aerosol vertical profiles by LIDAR, (ii) in-situ aerosol absorption and scattering coefficients at several wavelengths, and (iii) spectral actinic fluxes and spectral irradiances, will allow fairly stringent testing of radiative transfer models under these polluted conditions. Other in-situ measurements, including aerosol size distributions, inorganic ion content, size-resolved organic and elemental carbon, and possible aromatic and nitrate group absorbers, are aimed at understanding the optical properties in terms of aerosol composition and microphysics. Measurements of aerosol hygroscopicity, CN, and CCN number will address possible impacts on clouds and precipitation. All of these measurements will be made as a function of distance from MC, so that a clear picture of aerosol evolution can be developed and related to transformations (e.g. gas-aerosol interactions).

Ground-based measurements will include all or most of the aerosol and radiation measurements planned for the C-130, and additionally more detailed aerosol speciation, total (surface to space) aerosol spectral optical depths and diffuse/direct ratios. These measurements will be used to characterize the diurnal evolution of aerosol chemical and optical properties, their vertical structure (e.g. as related to PBL evolution), and their radiative impacts at the surface (e.g. heating rates, photolysis rates).

7. Regional surface-atmosphere interactions: *What is the magnitude of regional emissions, esp. from biomass and waste burning and from vegetation? How does it compare to urban emissions? How do regional emissions interact with the urban plume?*

Approach: The presence and extent of biomass burning plumes will be monitored from satellite data (e.g., fires from AVHRR, aerosols from MODIS, and CO from MOPITT or TES). Observations of biomass burning tracers (CH_3CN, HCN, fine-aerosol K^+, and HC ratios) from the C-130 will provide a measure of the extent of biomass burning impact on the air masses being sampled. Intersection of the MC outflow with biomass burning plumes is likely and could lead to significant changes in chemistry (e.g. O_3 production). Flight-paths will occasionally be chosen to maximize the likelihood of sampling plume interaction.

Biogenic emission algorithms (based on land-use) will be evaluated with measurements of concentrations and fluxes during flights upwind and downwind of MC city. Ground-based measurements at the supersite will include deposition fluxes of aerosol and trace gases (O_3, NO_x, NO_y, selected HCs and OVOCs).

E. IMPLEMENTATION PLAN

1. Timing of the field campaign

We propose to carry out a one-month long aircraft- and ground-based observational intensive during early 2006, nominally 15 February - 15 March. This time window is predicated by our wish to study the dry season (November - March,) and by consideration of local holidays from late December to early January. Uploading and downloading of instruments on the C-130 are estimated as 4 and 2 weeks, respectively. Assuming 7-8 hour flights on alternate days, this would require a total of ca. 110 hours of C-130 flight time. Transit and test flights will add an additional ~20 hours of flight time.

2. Aircraft-Based Measurements

The main observational platform for the outflow measurements will be the NSF EC-130Q Hercules aircraft (the C-130). The aircraft is operated by NCAR's Research Aviation Facility (RAF). The payload capacity (5900 kg with full fuel), range (3300 km at 1000ft, 5700 km at 20000ft) and maximum operating altitude (7.9 km) are well suited for the mission. The primary base of operation will likely be one of the following sea level international airports: Tampico (TAM, 340 km NE of MC) or Veracruz (VER, 320 km E of MC). Airports closer to MC (MEX, TLC, CVJ) were found to be less suitable due to limitations on takeoff weight at higher elevations. The C-130 will be deployed to study the evolution of the MC plume from the initial stages up to several days downwind. Flight paths (see Sect. E.3) are designed to sample air upwind of MC, followed by flights across the outflow at several distances down-wind and several altitudes.

The C-130 will be equipped with RAF-supplied and user-supplied instrumentation for measuring trace gases, aerosol properties, and radiation. In particular, the C-130 payload will include instruments for comprehensive characterization of the photochemical state of the atmosphere, including UV actinic flux, ozone (*in-situ* and LIDAR), reactive nitrogen species, carbon monoxide, hydrocarbons and selected oxygenated organics, SO_2, H_2SO_4, H_2O_2, CO_2, and aerosols size distribution, chemical composition, and optical properties. The measurements identified as highest priority for the aircraft to ensure the success of the mission are summarized in Appendix 1, together with the expected amount of rack space required in the aircraft. The instrument package will require 12 to 13 standard double aircraft racks plus the space required for the LIDAR system. During the TOPSE mission (flown February to May, 2000), 8 aircraft racks plus a large LIDAR system were fitted onto the C-130, with 5 crew and 14 investigators requiring 15 seats in the main cabin. For MIRAGE-Mex, more compact automated instruments will be desirable. Some of the instruments that required an operator during TOPSE have already been automated and some have been modified to require less space than in 2000, so that we are reasonably confident that a payload as outlined in Appendix 1 can be fitted onto the aircraft.

While the C-130 payload is designed as a stand-alone set of instruments, discussions are ongoing with potential collaborators concerning the use of additional aircraft, including DOE's G-1 and several smaller craft. Coordinated G-1 and C-130 flights would enhance the data base of outflow observations considerably, and the smaller aircraft

could be used to better characterize the composition of polluted air near its origin, i.e. to obtain cross sections and vertical profiles (e.g. of O_3, NO_x, total VOC, UV radiation, aerosols samples) over the MC basin. Additional aircraft will not only expand the number of measurements that can be made but more importantly, through carefully chosen overlap in some key measurements, will enable closer examination of the processes of air mass aging in the outflow of the city by simultaneous measurements at different distances downwind of the city. Some intercomparison flights will also be scheduled.

3. Nominal flight paths

Actual flight plans will vary depending on the meteorological conditions and the primary focus of each mission. Guidance for each flight will be provided by pre-flight meteorological and chemical model forecasts, and in-flight by the on-board LIDAR system to detect the plume. Nominal plans are shown in Fig. 9 for several 7-8 hour flights to examine the outflow at different stages of plume age and dilution with background air, based on typical meteorological situations expected for February. Four different types of mission are represented:

(1) An investigation of inflow air followed by a pass inside the MC valley (missed approach on MEX) followed by several passes immediately downwind of the city to investigate the fresh outflow. The second part of the flight examines the outflow from the previous day over the coastal area at two different altitudes. This flight would require a takeoff time around noon and 8 hours of flight time.

(2) An investigation of just the one-day old outflow. Here, several altitude profiles and passes through the plume at three different altitudes would be possible in 8 hours flight time. Takeoff time would be late morning. A very similar flight plan would apply to a nighttime takeoff (~2:00 a.m.) to investigate nighttime chemistry in the plume emitted in the afternoon on the day before. The flight would last well into the morning daylight hours to observe the onset of photochemistry in the plume.

(3) A survey flight plan to investigate the impact of the MC plume two to three days downwind over the Gulf of Mexico. Guidance from accurate meteorological forecasting and the on-board LIDAR will be critical for this type of mission.

(4) A Norte event. The flight plan shown contains an altitude profile north of the city followed by a missed approach to MEX followed by a pattern to investigate the plume, working away from the city. Takeoff for this flight could be at any time but a morning takeoff would allow the outflow to be sampled midday into the early afternoon when photochemistry is fully active. The return flight could include a leg in or above the plume back towards the city. Additionally, it would be interesting to fly a mission like the one described in (1) immediately after the end of a Norte event, so the city essentially starts with a zero-background, i.e. no recirculation of previous days' pollutants, and to contrast the results from this flight with a similar mission flown after a southeasterly flow has been established for a few days.

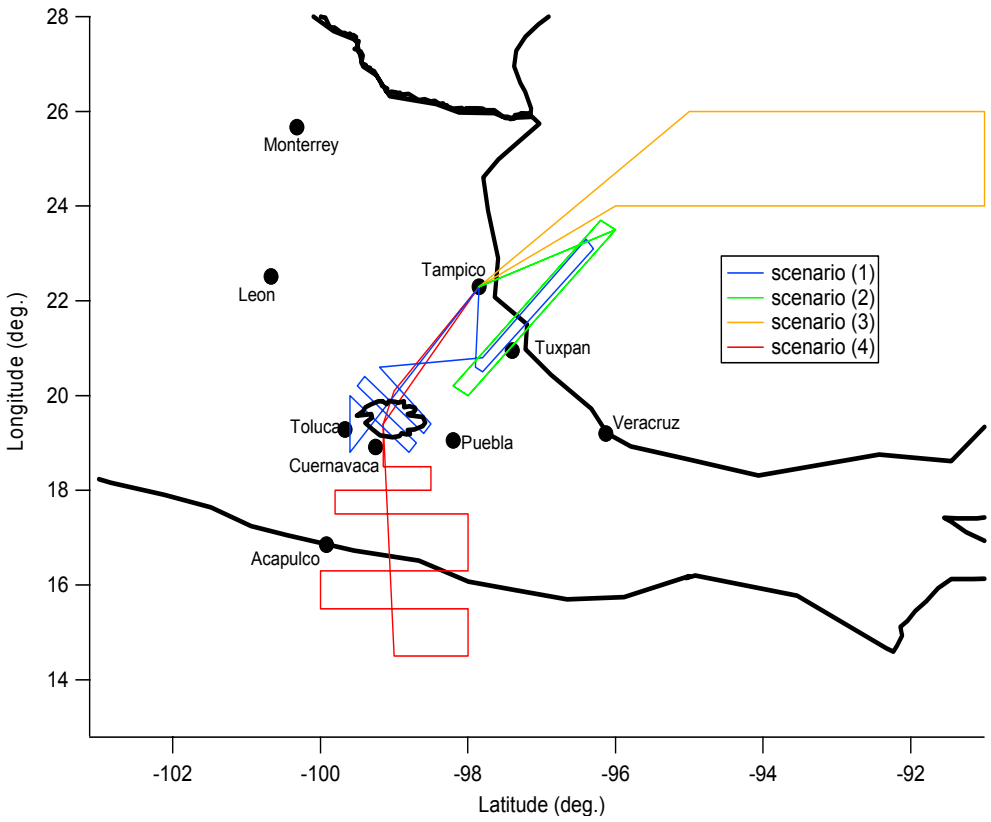

Figure 9*: Nominal flight paths (color coded) for the 4 possible scenarios described in the text. The black outline in the center is the approximate extent of the Mexico City metropolitan area. Tampico International Airport is the assumed base of operation.*

4. Ground-Based Measurements

The MIRAGE-Mex aircraft mission should be supported and enhanced by a comprehensive set of ground-based measurements (Appendix 2). These measurements can be categorized as (i) those that directly support the aircraft flights through acquisition of data needed for flight plans and model initialization (e.g. meteorological observations, PBL profiles), and (ii) those that widen chemical measurements beyond the capability of the C-130 payload, and thus provide a more detailed characterization of gas and aerosol processes. For logistic reasons and to facilitate data interpretation, most of these ground-based measurements will be co-located at a "supersite." The tentative location for this supersite is just outside of the city to the northeast (NE) in the expected prevailing direction of transport of the urban plume (exploratory ground-based campaigns are planned by Mexican researchers for March 2004 and November 2004 and knowledge gained in these campaigns will help determine the best location). Ground-based measurements will include:

i) Critical measurements to support the C-130 flights*:* Deployment of a radar wind profiler and the launching of GPS radiosondes several times per day from the supersite are required. The radar wind profiler will produce continuous wind speed and direction from a few hundred meters above the surface to 3-4 km above ground level, data that will be used to derive the initial air parcel trajectories as they exit MC. The radiosondes will

provide high-resolution vertical profiles of wind speed and direction, temperature, and humidity from the surface through the lower stratosphere. Vertical profiles (LIDAR) of ozone and aerosols will be used to further characterize the extent of vertical mixing and to determine the chemical characteristics of the plume aloft as it exits the city. A tethered balloon system for profiling other constituents (e.g. VOCs) is desirable.

Additionally, a radar wind profiler / radiosonde site is needed a few hundred kilometers downwind of MC, where the boundary layer structure and vertical wind profiles will probably be quite different from those close to MC. The location of this site is yet to be firmly established, but will likely be somewhere near the base of operation of the C-130 and will thus coincide with the majority of the C-130 flight paths.

ii) A "full" chemistry experiment: While the C-130 payload and flights are designed to probe the urban outflow at different distances from the source, much additional scientific understanding can be gained through simultaneous measurements of gas and aerosol chemistry at the supersite. The proposed measurements are similar to those made during MCMA2003 at an urban supersite, with the addition of several new instruments expected to be available by 2006. Because the supersite will be located just outside the city, these measurements - and their comparison to the MCMA2003 urban data - will give valuable information about the earliest stages of export. They will also provide a more accurate characterization of initial conditions for the regional scale measurements made with the C-130. Thus, the supersite measurements will facilitate “bridging” between urban and regional observations.

As already described in Sect. D, the supersite will allow us to address specific science objectives in more detailed ways with instruments that are not aircraft-ready. These include measurements of total and more fully speciated organic carbon, total OH reactivity, HONO and NO_3, and a comprehensive aerosol characterization package including size-resolved composition (esp. speciated organics), functional group analysis of aerosol filter samples, and collection/analysis of any rainwater. Measurements of deposition fluxes and spectral radiation will also help establish the near-field impacts of the plume on the surface environment.

The measurements at the supersite can be done in part through NSF sponsorship, but will also rely on contributions from other participants including a significant involvement by the Mexican research community. In fact, we anticipate that the supersite will provide numerous concrete opportunities for capacity building through collaborative studies, instrument intercomparisons, and exchange of technical and scientific know-how.

5. Expanded Measurement Activities

Unprecedented opportunities exist for simultaneous (or nearly so) studies that address important related scientific issues. These include (i) urban meteorology, (ii) urban pollution, and (iii) regional emissions and deposition. While none of these components is considered mission-critical, MIRAGE-Mex provides the foundation on which a project of larger scope can be developed. Further support for these enhancements of the campaign can be envisaged with the participation and support of agencies other than the NSF. External collaborations with other government agencies and universities will be encouraged to provide meteorological, trace gas, and aerosol measurements in the

vicinity of MC and perform urban-scale coupled meteorological-chemical-aerosol modeling.

(i) Urban meteorology: Previous studies have shown the meteorological and chemical processes in the vicinity of MC to be quite complex (Section C.2). It would be very useful to coordinate urban-scale measurements with those supported through MIRAGE-Mex. Additional meteorological instrumentation could include radar wind profiler / radiosonde / sodar sites within and around MC to describe local boundary layer structure and circulations where pollutants originate, surface meteorological stations located on the mountain slopes to characterized near-surface terrain-induced flows, and sonic anemometers to determine near-surface vertical fluxes. Some of the questions that could be addressed by other investigators include:

- *How much accumulation or recirculation of pollution occurs on a day-to-day basis?*
- *How much venting is produced by local thermally-driven circulations?*
- *What are the forecast errors associated with the evolution of the boundary layer? What refinement to physical parameterizations are needed to improve model forecasts?*
- *How well can we forecast the timing, direction, and altitude of outflows? How are the outflows affected by local, regional, and synoptic-scale circulations?*
- *How do changes in surface heating and aerosols produced by urbanization affect PBL stability, vertical mixing, and photolysis rates?*
- *How is the local meteorology affected by urbanization through changes in the surface heat, moisture, momentum, aerosol, and gas fluxes?*

Understanding these complex processes on the urban scale is not a primary objective of MIRAGE-Mex, but would be beneficial in understanding the earliest stages of outflow.

(ii) Urban pollution: Additional trace gas and aerosol measurements from ground stations, research aircraft, and remote instrumentation would be useful to describe the evolution of pollutants trace gases and aerosols for the several hours before they exit MC. Routine monitoring networks (PM_{10}, $PM_{2.5}$, CO, NO_x, O_3) are expected to continue operations during the MIRAGE-Mex field campaign and will provide valuable information for initialization of models. Supplementary measurements are desirable, such as canister sampling to provide more detailed speciation of the local VOC concentrations and their relation to emission estimates. Aerosol size distribution and chemical composition measurements would be also valuable, for comparisons with similar measurements made downwind. Surface radiation measurements (short-wave and UV irradiances, spectral actinic fluxes) are needed to characterize the aerosol optical properties and derive photolysis rates. Mobile measurement laboratories (e.g., Aerodyne and PNNL) could be deployed to provide further information on urban air quality and to follow the outflow. Mobile measurements throughout the basin of column CO and other pollutants were made during MCMA2003 (Chalmers U.) and would be useful during MIRAGE-Mex. Further information on vertical structure of the urban atmosphere could be obtained from over-city flights by smaller aircraft. Direct flux measurements, using systems mounted on towers or tall buildings, and mass balance studies (downwind minus upwind concentrations) would both be desirable.

(iii) Regional emissions and deposition: Several medium-sized urban areas are located within a few hundred km of MC; biomass burning activity is widespread; and emissions from vegetation are not negligible. Therefore, it will be useful to have some characterization (e.g. aerosols, CO, NO_x, VOCs, O_3) of these surrounding areas, as well as of the contribution of MC outflow to surface mixing ratios. An additional aircraft with flux measurement capabilities, if available, would be valuable for surveying aerosol and trace gases (O_3, NO_x, NO_y, selected HC and OVOCs) over a larger area than covered by the C-130. Measurements of deposition and emission fluxes of aerosols and selected trace gases would be desirable to characterize interactions between the MC plume and the biosphere downstream, and could be obtained from a mobile surface laboratory.

6. Satellite observations

Distributions of clouds, aerosols, and, depending on available satellite platforms, of CO, NO_2, CH_2O, and O_3 may be available to help in the planning and interpretation of the results. Table 3 lists satellites that are likely to be operating in 2006 and relevant data that may be available. The OMI experiment on the AURA platform (with an expected launch date of mid-summer 2004) will measure NO_2 columns at a horizontal spatial resolution finer than that of GOME and SCIAMACHY (currently in orbit). Monitoring of biomass burning (e.g. from AVHRR) will be critical during the campaign, as this information will be used in flight planning to either intersect or avoid plumes from such fires. Satellite observations during the field campaign will be compared to in-situ observations in order to validate the satellite capability. After the field campaign, analyses of satellite observations will be used in conjunction with regional model calculations to quantify the export of species, such as CO and NO_2, from Mexico City into the regional scale.

Table 3: *Satellite platforms and datasets relevant to Mirage-Mex that may be available in 2006.*

Satellite Platform	Relevant Data Products	Vertical Resolution
MOPITT	Tropospheric CO	approx. 4 km
MODIS	Aerosol optical depth, cloud droplet size and reflectivity	Trop. column
SCIAMACHY	O_3, NO_2, CH_2O	Trop. column
OMI	O_3, NO_2, aerosol	Trop. column
TES	O_3, CO	approx. 4 km

7. Modeling

A vigorous modeling activity will be required at all stages, from early planning, through field campaign execution, to analysis and interpretation of the results. Well in advance of the campaign, meteorological and chemistry-transport models will be used with representative episodic conditions to refine the overall flight plan strategy. During the campaign, forecast models will be used to assist with specific flight planning. Post-campaign, the observational data base will be used to improve and evaluate detailed models of processes (e.g. hydrocarbon oxidation, aerosol formation and evolution, gas-aerosol interactions), to evaluate high (or variable) resolution regional chemistry-transport models, and to assess large-scale impacts using global models.

It is anticipated that the Weather Research Forecast (WRF) model developed by NCAR's Mesoscale and Microscale Meteorology (MMM) division will be used extensively for the field campaign. Additionally, we are working with Georg Grell (NOAA/FSL) to adapt and improve an early version of WRF with on-line chemistry (WRF-Chem) to the Mexico region (preliminary results were shown in Fig. 3). Meteorological and chemical modeling efforts focused on Mexico City are also ongoing at other institutions (e.g. UNAM and MIT among others). We fully expect that these as well as other models, both regional and global, will benefit from the extensive observational data base that will be obtained during MIRAGE-Mex. Modeling collaborations and intercomparisons will be encouraged.

F. NATIONAL AND INTERNATIONAL COLLABORATIONS

We have identified, through their attendance at a planning workshop or by direct contact, potential participants from 28 universities (22 from the US, 2 from Mexico, one each from Argentina, Germany, Switzerland, and the UK), from government agencies in the U.S. (DOE, NOAA, NASA) and Mexico (SMA-GDF, SEMARNAT, CONACyT), and 4 from the private sector (Aerodyne Inc., IMP, FZK/IFU, and Sonoma Technology, Inc.). Some of these (e.g. DOE) could potentially contribute several research teams and provide additional aircraft. We will work closely to coordinate these teams.

We will seek formal recognition of MIRAGE-Mex by international scientific coordinating organizations, specifically NARSTO and IGBP, to encourage international collaborations, to facilitate wide-spread utilization of the collected data (see section G), and to ensure coordination with other related upcoming activities such as INTEX, Houston-2006, and the Asian Brown Cloud experiments.

G. DATA MANAGEMENT

Data archives will be maintained by the Atmospheric Chemistry Division of NCAR. After a period of 1 year (to allow for quality control and initial interpretation by the participating researchers) the data will be made available to the community, in a standard format (NARSTO) so that they may be easily accessed and used by other researchers. Results of model simulations will also be put in the public archive. The scientific results of the MIRAGE-Mex field campaign will be submitted for publication in the peer-reviewed literature.

H. BROADER IMPACTS

Significant opportunities for the integration of research and education are expected from the MIRAGE-Mex activities. Through university participation, graduate students and post-doctoral fellows will be involved in all aspects of the field campaign, including instrument development, measurements, modeling, data analysis and interpretation, and participation in workshops and preparation of publications. In particular, participation by underrepresented groups is expected and will be encouraged, and has already begun at NCAR through the recent involvement of SOARS (Significant Opportunities in

Atmospheric Research and Science) students in analyzing and interpreting Mexico City's pollution data.

A strong partnership with our Mexican colleagues in the academic, government and private sectors is essential to the success of MIRAGE-Mex, and is expected to provide significant enhancements in infrastructure for research and education. In particular, measurement capabilities and logistical support afforded by Mexican scientists will provide the foundation upon which the ground supersite will be built. At the same time, the participation of U.S. and international scientists increases the scope of scientific questions that can be addressed, presents opportunity for informal instrument intercomparison and technology transfer, and provides a greater range of experiences for students and early-career scientists in Mexico than would otherwise be possible. Formal collaborations will continue from the earliest stages of planning through final preparation of manuscripts, and longer-term collaborations are likely to evolve.

The need for an integrated multidisciplinary approach to understand the impacts of megacities on regional and global air quality was recognized at NCAR through the recent establishment of a MIRAGE Strategic Initiative. In addition to coordinating the MIRAGE-Mex field campaign, the MIRAGE Strategic Initiative office will sponsor workshops, international visitors, and modeling efforts related to the topic of export of pollution from megacities. These activities will bring together atmospheric and social scientists, economists, and urban planners to assess how well we can predict future emissions, and what improvements in urban design could ameliorate the problems. The MIRAGE office will maintain a web site related to the MIRAGE-Mex field campaign (including data archives) as well as a more general web site to enhance public awareness and education. MIRAGE has also established a partnership with the Integrated Program on Urban, Regional and Global Air Pollution, a related research and education program initiated at MIT.

By providing the first characterization of air quality outside Mexico City, MIRAGE-Mex will be the first step toward assessing the environmental and economic impacts of pollution emanating from this megacity - information that is necessary for the development of future environmental policy in this region. More broadly, MIRAGE-Mex will contribute to the understanding of how air pollution of urban origin affects regional and global scale atmospheric chemistry, weather, climate, and ultimately ecosystems. This understanding is essential for the development of scientifically informed and effective strategies to mitigate the consequences of globally increasing urbanization.

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APPENDIX 1: Proposed C-130 User-Supplied Measurements and their Relation to Science Objectives.	**Plume extent**	**Regional oxidant production**	**HC oxidation products**	**Reactive nitrogen**	**Gas-aerosol chemical processes**	**Aerosol radiative properties and impacts**	**Regional surface-atmosphere interactions**	**Available instrument / method (a)**	**Rack space (b)**
In-situ Gas Phase Measurements									
O_3	x	x	x		x		x	CLD*	1
NO	x	x	x	x			x		
NO_2	x	x	x	x			x		
NO_y	x	x	x	x	x		x		
H_2O		x	x		x	x	x	Lyman-α* or TDL*	
CO_2	x		x				x	TDL* or Licor*	0.5
CO	x	x	x				x		
$OH/HO_2/RO_2$	x	x	x		x			Four-channel CIMS*	1+
HNO_3	x	x		x	x		x		
H_2SO_4	x				x		x		
NH_3					x		x		
PANs	x	x	x	x			x	CIMS*	0.5
H_2O_2	x	x	x		x		x	TDL* or enzyme	
CH_3OOH		x	x		x		x		0.5
ROOH		x	x				x		
HCHO	x	x	x					TDL* or enzyme	1
SO_2	x				x		x	CIMS or TE	0.5
speciated HCs	x	x	x		x		x	WAS	1
organic nitrates	x	x	x	x					
halocarbons	x						x		
OVOC	x	x	x		x		x	GC-MS*	1
organic acids	x	x	x		x		x	PTR-MS*	0.5
HCN/RCN	x						x		

Aerosol Measurements									
number size distribution	x				x	x		PMS*	w
total number CN						x		CNC*	~0
size-resolved composition (non-refractory)					x	x		AMS	1
soluble organics & inorganics					x	x		PILS	0.5
soot size distribution						x		SP2	1
multi-wavelength optical properties (absorption and scattering coefficients)					x	x		nephelometer, aethelometer, PSAP	.25
size-resolved hygroscopicity					x			HTDMA or TDMA	.75
size-resolved volatile content			x		x				
CCN	x					x		CCN counter	~0
Radiative Measurements									
O_3 (nadir/zenith)	x	x						LIDAR	2
aerosol (nadir/zenith)	x					x			
spectral actinic flux		x	x	x				SAFS*	.25

(a) Specific instruments are listed to indicate existing measurement capability. In some cases, alternative instruments may be available. See Appendix 3 for instrument abbreviations. Instruments currently available at NCAR are denoted by *.

(b) w = wingpod or wingprobe-mounted instrumentation (no rack space required).

APPENDIX 2: Proposed User-Supplied Ground-based Measurements and their Relation to Science Objectives.	**Vertical Plume Extent**	**Regional oxidant production**	**HC oxidation products**	**Reactive nitrogen**	**Gas-aerosol chemical processes**	**Aerosol radiative properties and impacts**	**Surface-atmosphere interactions**	**Available Instrument / Method (a)**
Gas Phase Measurements								
O_3		x	x		x		x	UV abs.
NO		x	x	x				CLD
NO_2		x	x	x				
NOy		x	x	x			x	
HNO_3		x	x	x	x		x	FT-IR
O_3 remote	x	x						LIDAR
H_2O remote	x	x						LIDAR
CO		x	x				x	canister
CO_2			x				x	Licor
CO column	x	x	x					FTIR*
OH (HO_2/RO_2 ??)		x	x	x	x	x	x	LIF
OH reactivity		x	x	x	x	x	x	
NH_3		x		x	x	x	x	FT-IR or TDL
PANs		x	x	x			x	GC* or CIMS
H_2O_2		x	x		x		x	TDL or FT-IR or LC-enzyme
CH_3OOH		x	x					LC-enzyme
ROOH		x	x					LC-enzyme
HCHO / RCHO		x	x		x		x	TDL or cartridges
SO_2		x			x		x	CIMS or TE
speciated HCs		x	x		x		x	canister samples or GC-MS*
organic nitrates		x	x	x	x		x	
Halocarbons							x	
HCN/RCN							x	
OVOC		x	x	x	x		x	
total peroxy nitrates		x	x	x			x	TD-LIF
speciated organic nitrates		x	x	x			x	
multifunctional OVOC		x	x		x		x	GC-MS or LC-MS
HONO		x	x	x	x		x	DOAS
NO_3		x	x	x	x		x	
HCHO		x	x				x	
glyoxal		x	x				x	
RCHO		x	x		x		x	GC-MS or cartridges

Tethered Balloon Measurements								
NO_x	x	x		x			x	CLD
O_3	x	x					x	UV
HC	x	x	x				x	canisters-GC*
met. params.							x	standard
Aerosol Measurements								
number size distribution					x	x		PMS
total number CN						x		CNC
size-resolved composition (non-refractory)		x	x		x	x		AMS
soluble organics & inorganics		x	x		x	x		PILS
soot size distribution						x		SP2
multi-wavelength optical properties (absorption and scattering coefficients)		x			x	x		Nephelometer and (Aethelometer or PSAP)
size-resolved hygroscopicity					x	x		HTDMA and TDMA
size-resolved volatile content			x		x	x		
speciated organic aerosol content			x		x	x		GC-MS-MS
rainwater chemistry			x		x	x	x	GC-MS, LC-MS, ion-GC, etc.
single particle composition (ultrafine)			x		x	x		TDCIMS*
aerosol organic content - functional group analysis			x		x	x	x	Filter and FT-IR
PM_{10}, $PM_{2.5}$					x	x		aerosol probes
CCN					x			CCN counter
Deposition and Flux Measurements								
O_3		x			x		x	CLD
NO_x		x		x	x		x	CLD
HNO_3/NO_y		x		x	x		x	CIMS/CLD
PANs		x		x	x		x	CIMS
VOC		x	x		x		x	PTR-MS
aerosols					x	x	x	AMS
Meteorological Measurements								
surface std. meteorology	x							various*
wind profiles	x							radar wind profiler*
vertical profiles of winds and temperature/humidity	x							radiosondes*

Radiative Measurements								
spectral irradiance		x	x	x		x		spectroradiometer*
spectral actinic flux		x	x	x		x		SAFS*
diffuse/direct radiation		x				x		MFRSR
aerosol optical thickness	x	x				x		DIAS*, filter photometers

(a) Specific instruments are listed to indicate existing measurement capability. In some cases, alternative instruments may be available. See Appendix 3 for instrument abbreviations. Instruments currently available at NCAR are denoted by *, excluding those for use on C-130.

APPENDIX 3: Abbreviations

Instruments:

AMS	Aerosol Mass Spectrometer
CIMS	Chemical Ionization Mass Spectrometer
CLD	ChemiLuminescence Detector
CNC	Condensation Nuclei Counter
DIAS	Direct irradiance atmospheric spectrometer
DOAS	Differential Optical Absorption Spectrometer
FT-IR	Fourier Transform InfraRed spectrometer
GC	Gas Chromatography
HTDMA	Hygroscopicity Tandem Differential Mobility Analyzer
LC	Liquid Chromatography
LIDAR	LIght Detection And Ranging
LIF	Laser-Induced Fluorescence
MFRSR	Multi-Filter Rotating Shadowband Radiometer
MS	Mass Spectrometer
PILS	Particle-Into-Liquid-Sampler
PMS	Particle Measuring Systems
PSAP	Particle/Soot Absorption Photometer
SAFS	Scanning Actinic Flux Spectroradiometer
SP2	Soot Photometer
TDCIMS	Thermal Desorption CIMS
TDL	Tunable Diode Laser
TDMA	Tandem DMA
TE	ThermoElectron
UV	UltraViolet absorption
WAS	Whole Air Sampler

Organizations:

CONACyT	Consejo Nacional de Ciencia y Tecnologia
DOE	Department of Energy
FZK/IFU	Forschungszentrum Karlsruhe
IGBP	International Geosphere-Biosphere Programme
IMP	Instituto Mexicano del Petroleo
MIT	Massachusetts Institute of Technology
NARSTO	North American Research Strategy for Tropospheric Ozone
NASA	National Aeronautic and Space Administration
NCAR	National Center for Atmospheric Research
NOAA	National Oceanic and Atmospheric Administration
NSF	National Science Foundation
RAF	NCAR's Research Aviation Facility.
RAMA	Red Automatica de Monitoreo Atmosferico
REDMA	Red Manual de Monitoreo Atmospherico
REDMET	Red de Parametros Meteorologicos
SMA-GDF	Secretaría de Medio Ambiente (Mexico City)
SEMARNAT	Secretaría de Medio Ambiente y Recursos Naturales (Mexico, federal)
UNAM	Universidad Nacional Autonoma de Mexico