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Assessing the regional impacts of Mexico City emissions on air quality and chemistry

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Abstract. The impact of Mexico City (MCMA) emissions is examined by studying its effects on air quality, photochemistry, and on ozone production regimes by combining model products and aircraft observations from the MILAGRO experiment during March 2006. The modeled influence of MCMA emissions to enhancements in surface level NO_x , CO, and O_3 concentrations (10–30% increase) are confined to distances <200 km, near surface. However, the extent of the influence is significantly larger at higher altitudes. Broader MCMA impacts (some 900 km Northeast of the city) are shown for specific outflow conditions in which enhanced ozone, NO_y , and MTBE mixing ratios over the Gulf of Mexico are linked to MCMA by source tagged tracers and sensitivity runs. This study shows that the “footprint” of MCMA on average is fairly local, with exception to reactive nitrogen, which can be transported long range in the form of PAN, acting as a reservoir and source of NO_x with important regional ozone formation implications. The simulated effect of MCMA emissions of anthropogenic aerosol on photochemistry showed a maximum regional decrease of 40% in $\text{J}[\text{NO}_2 \rightarrow \text{NO} + \text{O}]$, and resulting in the reduction of ozone

production by 5–10%. Observed ozone production efficiencies are evaluated as a function of distance from MCMA, and by modeled influence from MCMA. These tend to be much lower closer to MCMA, or in those points where modeled contribution from MCMA is large. This research shows that MCMA emissions do effect on regional air quality and photochemistry, both contributing large amounts of ozone and its precursors, but with caveat that aerosol concentrations hinder formation of ozone to its potential due to its reduction in photolysis rates.

1 Introduction

Megacities are defined as metropolitan areas with more than 10 million people (Gurjar and Lelieveld, 2005). This high concentration of urban dwellers generates challenging conditions for government: traffic congestion, solid and liquid waste disposal, drinking water supply, and bad air quality (Mayer, 1999; Lynn, 1999). Projections suggest that these problems will worsen as more people are migrating from rural to urban areas (Saier, 2008), especially in the developing world, where most new megacities are located. In addition to the adverse impacts on ecosystems and human health at local scales, megacities may also have noticeable impacts on



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regional air quality and photochemistry (Mage et al., 1996; Guttikunda et al., 2001, 2003; Madronich, 2006).

Mexico City Metropolitan Area (MCMA), with a population of 20 million, is located at 2200 m above sea level and has complex constraining topography. Its physical properties generate conditions for low combustion efficiency and entrainment of pollutants. Additionally traffic congestion, a relatively old vehicular fleet, and the concentration of economic and industrial activity make it one of the most polluted cities in the world (Molina and Molina 2002; Molina et al., 2007). Megacities such as MCMA are also the source of considerable amounts of particulates, which affect photochemical reactions involved in ozone production and destruction (Carslaw and Carslaw, 2001) mainly by altering the amount of radiation involved in photochemical reactions. For example a study in the MCMA showed that observed photolysis rates of NO_2 at an urban site were 10–30% lower than at a nearby rural site (Raga et al., 2001). Model sensitivity studies in Asia and North America have shown that regional photochemistry can be altered by as much as 20% in the presence of aerosol (Tang et al., 2003; Li et al., 2005).

An intensive atmospheric chemistry measurement campaign denominated MILAGRO (Megacity Initiative: Local and Global Research Observations) (Fast et al., 2007; Molina et al., 2008) was carried out during March, 2006, collecting chemical and meteorological data in the MCMA and surrounding areas. Besides the use of ground-based measurement platforms, the campaign included multiple aircrafts: DC-8, and J-31 supported by NASA, C-130 supported by NCAR-NSF, G-1 and B200 supported by DOE, and a Twin Otter supported by the US Forest Service and NSF. This paper focuses on the analysis of observations from the C-130 and DC-8. The spatial coverage, maneuverability and sampling strategies for these aircrafts were different. The C-130 sampled near and within the city, performing zig-zags and spirals at multiple altitudes with an altitude range from 0–6.5 km above mean sea level (MSL), with most measurements being taken in Mexico City or the Gulf of Mexico. The DC-8 performed direct flights from Houston, TX, with limited spirals, and a higher altitude range (0–12 km above MSL). With the different spatial coverage, these aircraft sampling strategies intended to evaluate the regional influence of MCMA emissions on a regional (C-130), and continental (DC-8) scale.

In this paper we examine the impact of MCMA's emissions on ozone production focusing on interactions between ozone precursor concentrations and photochemistry. Using a combination of observations and model sensitivity analyses we estimate MCMA's "air quality" footprint. The area required to provide water, food, energy and waste management services to a megacity has been defined as the ecological footprint, which has been estimated to be significantly larger than the city itself (Valentine and Heiken, 2000). Here we extend such analysis to look at the "air quality footprint" of the MCMA emissions, which we define as the geograph-

ical extension of discernible enhancements (contributions) of concentrations and photochemical rates attributable to the MCMA.

2 Methods

2.1 Model description

2.1.1 Meteorological and chemical transport model

In this campaign we used the STEM-2K3 model (Carmichael et al., 2003) to carry out air quality forecasting simulations to provide support and context to aircraft observations during the campaign. This chemical transport model (CTM) features full chemistry calculations using a lumped species SAPRC99 chemical mechanism (Carter, 2000) in conjunction with the TUV(Tropospheric Ultraviolet Radiation Model) (Madronich et al., 1992) on-line photolysis model and the SCAPE II (Simulating Composition of Atmospheric Particles in Equilibrium aerosol solver(Kim et al., 1993, 1995, 2003). The calculation of the photolysis was two-way coupled with other chemical modules, including inputs of concentrations of aerosols and gas phase species, and outputs of 30 photolysis rates defined by the SAPRC99 mechanism. The STEM-2K3 model was driven by the WRF V2.1.2 (Weather Research Forecasting) meteorological model (Grell et al., 2005), using the NCEP (National Center for Environmental Prediction) Global Forecasting System (GFS) $1^\circ \times 1^\circ$ analyses for meteorological boundary conditions (NCEP, 2003). For this study the model simulations used two nested domains, the outer using a 60 km resolution, 100 cells in East-West direction, and 64 cells in North-South direction, with an inner domain using a 12 km resolution, 110 cells East-West, and 95 cells North-South. The model had 21 vertical layers, extending from the surface to 100 hPa, with 10 of the levels representing altitudes from surface to 1 km above the terrain. The Grell-Devenyi ensemble cumulus parameterization (Grell, 1995), the YSU planetary boundary layer parameterization (Hong et al., 2006), and the NOAH land surface model (NOAA, 2005) were used for the WRF runs. In addition, tagged non-reactive tracer calculations were performed to qualitatively assess the contribution of geographical areas and source categories (MCMA, Northern Mexico, Southern Mexico, Central America, Texas, Central America, Asia, South East United States, biomass burning, large point sources, total anthropogenic CO, biogenic CO) to CO mixing ratios.

The simulation period was March 2006, with an additional one week spin-up.

2.1.2 Emissions inventories and boundary conditions

The model runs used a high resolution emissions inventory for the MCMA developed by Universidad Nacional Autónoma de México (UNAM) for 1999 (Tie et al., 2007),

Table 1. Summary of total emissions for MCMA for base model run (modified from UNAM emissions inventory 1999).

Sector	PM ₁₀	PM _{2.5}	SO ₂	CO	NO _x	VOC's	NH ₃
Industry	3916	651	3284	6443	9869	62 756	196
Services and area	10 801	1962	41	7731	5831	119 109	13 543
Transportation	4768	3748	3321	1 777 907	73 986	101 144	3775
Vegetation	1201	261	N/A	N/A	313	9683	N/A
Total	20 686	6622	6646	1 792 081	89 998	292 692	17 514

which included speciated VOC which were lumped into the SAPRC99 VOC categories. Table 1 shows a summary of the emissions used in this study separated by sector. For surrounding areas of MC, south of 21° N the Emission Database for Global Atmospheric Research (EDGAR 2.0) 1° resolution anthropogenic area emissions inventory for 1990 was used (Olivier et al., 2002). For the six states in Mexico that share a border with the US (Baja California, Sonora, Chihuahua, Coahuila, Nuevo León, and Tamaulipas) a 36 km resolution anthropogenic emissions inventory from the BRAVO (Big Bend Regional Aerosol and Visibility Observational) study (Kuhns et al., 2005) was used. Biomass burning emissions of CO were provided by the an ecosystem based wild fire emissions processor (Pierce et al., 2007) which is part of the RAQMS (Regional Air Quality Modeling System) model (Pierce et al., 2005). Biogenic emissions were estimated using BEIS 2 (Biogenic Emissions Inventory System) (Geron et al., 1994) that generates time-variable isoprene and monoterpene emissions driven by meteorological variables, in this case from WRF simulations

Chemical boundary conditions were obtained from the Model for Ozone and Related Chemical Tracers (MOZART) (Horowitz et al., 2003), which was run by NCAR at a 0.7° resolution using MOPITT assimilated CO concentrations for the same modeling period (March 2006) (Pfister et al., 2005).

2.2 Interpolation of observations

Aircraft measurements provide a unique opportunity to collect three dimensional observations of multiple chemical and meteorological parameters. Plotting these observations along flight tracks allow us to infer outflow patterns from the cities. However, interpolating these observations horizontally highlight these patterns even more. In this study we interpolated aircraft observations using kriging (Krige, 1951) for the nearest 50 observations and an isotropic exponential semi-variogram. Interpolations were performed using ArcView 9.2 Geostatistical Analyst. This method, which has been used before in interpolating point bias estimations for aircraft observations during INTEX-NA (Mena-Carrasco et al., 2007), is only valid if the uncertainty of the method (subject to the sampling strategy, and the data) is significantly lower than the range of the interpolated values, which was the case for the analysis presented below.

3 Results and discussion

The regional impacts of MCMA are analyzed in two different categories: impact on air quality levels (again, focusing on O₃, NO₂, and CO) and impact on photochemistry. While interrelated, these impacts are analyzed separately to estimate various aspects of the air quality “footprint” of MCMA emissions, which we define as the geographical extension of discernible enhancements (contributions) on concentrations of pollutants, or on photochemical rates. Before any conclusions or results can be presented, it is necessary to establish that the model performance is adequate.

Table 2 shows a summary of some species correlation coefficients for ozone and some of its precursors support the conclusions drawn from this study. The base model run used in this study was the result of improvements of model performance after the forecast stage of the campaign, focusing on updating boundary conditions from global models and analyzing model bias associated to emissions inventory errors (and how ozone precursor errors were correlated to ozone errors) (Mena, 2007). In general the model does a good job in predicting the ambient levels of the trace gases (correlation coefficients of ~0.6 for all science flights), and in capturing the variability (i.e., the predicted and observed variability are very similar as measured by the ratio of the standard deviation divided by the mean values). Further details on the model performance can be found in a separate paper that focuses on model performance during the forecast stage, and improvements made by updating emissions inventories and boundary conditions. Indeed, it is apparent that there is room for further improvement. However, for the specific analyses illustrated in this paper, model performance was substantially better ($R > 0.8$) providing better support for our findings.

3.1 Modeled contribution of MCMA to pollutants

There are many ways to estimate the impact of the emissions from the MCMA. For this project the effects of MCMA on the regional distributions of pollutants calculated as the difference between mixing ratios of modeled species with and without MCMA emissions, normalized to mixing ratios without MCMC emissions. Figure 1 shows the mean daytime contribution of MCMA to near surface mixing ratios