J 1.2 A COMPARISON OF AEROSOL OPTICAL PROPERTIES AT T1 AND T0 DURING THE MILAGRO FIELD STUDY

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1. INTRODUCTION

Megacities, large urban and suburban centers whose populations exceed ten million inhabitants, are steadily increasing worldwide with the most rapid growth in the tropical areas of South America and Asia. The Mexico City metropolitan area (MCMA) is the largest urban center in North America and the second largest megacity worldwide. It occupies ~3540 km^2 with a population of ~19 M (CAM, PROAIRE 2002-22010). In general, megacities suffer from poor air quality due to the cumulative effects of population growth, industrialization, increased vehicle usage and total energy consumption. However, the topography of the MCMA also strongly affects the poor air quality (Fast et al., 2007; Fast and Zhong, 1998; Doran et al., 1998). Mexico City is located in a basin on the central Mexican plateau at an altitude of 2240 m and latitude of 19^o N. The basin is surrounded on the west, south, and east by mountain ranges that rise up to 1000 meters above the basin floor. This serves to inhibit dispersion of primary pollutants within the basin and the high levels of incoming solar radiation at this latitude and elevation promotes atmospheric photochemical reactions that rapidly form secondary pollutants (Whiteman et al., 2000).

The presence of highly absorbing carbonaceous aerosols in Mexico City leads to a reduction in solar flux of 17.6 % locally (Raga et al., 2001b). The mass of these absorbing aerosols exported from this megacity into the surrounding region is estimated to be 6,000 metric tons per day or 2 mega-tons per year (Gaffney et al., 1999). Since freshly formed black carbon aerosols are hydrophobic, they are expected to be more resistant to washout and have longer lifetimes than more hygroscopic aerosols such as sulfate and nitrate (Gaffney and Marley 2005; Dua et al., 1999). In addition, since these aerosols are introduced into the atmosphere at altitudes that would be considered to be in the free troposphere 300 km away they are assumed to have longer lifetimes than aerosols released at lower altitudes (Raga_et al., 2001b). The MCMA is therefore a major source of carbonaceous aerosols to the surrounding regions and the release of these highly absorbing aerosols will have an impact on the radiative balance and climate on a regional scale.

In order to better understand the evolution and transport of pollutant aerosols and gasses from the Mexico City basin and their resulting impacts on regional climate, a multiagency field campaign was undertaken called the Megacities Initiative: Local and Global Research Observations (MILAGRO). The MILAGRO study was composed of five collaborative field experiments. Two of the components of the MILAGRO study focused a major part of their efforts on aerosol emissions. The Megacity Aerosol Experiment, Mexico City 2006 (MAX-Mex) was sponsored by the U.S. Department of Energy (DOE) to investigate the direct radiative effect of aerosols in the Mexico City plume as a function of time, location and processing conditions. The MCMA-2006 study, supported by various Mexican institutions, the U.S. National Science Foundation (NSF) and the DOE deployed ground-based instrumentation to examine fine particles and secondary aerosol precursor gas emissions within the Mexico City Basin. As part of these two MILAGRO components, aerosol scattering and absorption measurements were obtained at a site located at the Instituto Mexicano del Petroleo (IMP), in the northwestern part of the Mexico City center. This site, known as T0, was chosen to represent the fresh emissions in the MCMA. Measurements were also obtained at the Technological University of Tecamac, located 18 miles northwest of T0. This site, known as T1, was expected to represent a mixture of fresh and aged pollutants as they exit the basin.

The evolution of absorbing aerosols downwind of Mexico City has been described previously in a comparison of measurements obtained at site T1 with those obtained at site T2 (Rancho La Bisnaga), located 35 km (22 mi) north-northeast of T1 (Doran et al., 2007a, 2007b; Doran, 2007). This study focused primarily on the changes in elemental carbon (EC) and organic carbon (OC) content of the aerosols and the resulting effects on the aerosol mass specific absorption coefficients. It was concluded from this work that emission sources outside the MCMA, including biomass-burning sources, are important contributors to the regional aerosol burden. A comparison of the measurements of aerosol absorption, aerosol scattering and single scattering albedo obtained at sites T0 and T1 during the MILAGRO campaign will be presented here. These data are used to estimate the distribution and evolution of aerosol optical properties within the surrounding region of the Mexico City basin.

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2. METHODOLOGY

Measurements were obtained from March 10 to March 29, 2006 at the IMP laboratories. This site, known as T0, is located in the north central part of Mexico City at latitude 19° 29' N, longitude 99° 09' W, and at an altitude of 2240 m above sea level. The aerosol instrumentation was located on the rooftop of Building No. 32 (Héctor Lara Sosa Building) 15 m above ground level. The sample inlets were designed to collect aerosols in the 0.1 to 2 micron size range. Aerosol scattering was measured with a three wavelength integrating nephelometer (TSI Model 3563) operating at 450, 530, and 660 nm. Results at 530 nm are reported here.

Aerosol absorption was measured at site T0 with a Multi-angle Absorption Photometer, or MAAP (Thermo Electron Model 5012). The aerosols in the air sample are collected within the instrument by continuous filtration through a glass fiber tape strip. The instrument uses a multiple detector setup to simultaneously measure the 670 nm light intensity both transmitted and scattered from the particle-loaded filter tape. The instrument then uses a two-stream-approximation radiative transfer scheme to determine aerosol absorption. This explicit treatment of light scattering effects caused by the aerosol and filter matrix in the radiative transfer scheme improves the determination of aerosol absorption considerably over methods that rely on the measurement of transmission or reflection alone (Petzold, et al., 2005). The instrument routinely calculates the black carbon content automatically from the aerosol absorption measurements by assuming black carbon to be the main absorbing aerosol species in the samples with a mass specific absorption coefficient of 6.6 m^2/g at 670 nm. However, these results are easily reconverted to the initial aerosol absorption measurement.

Aerosol optical measurements were obtained from March 1 to March 29, 2006 at the Technological University of Tecamac, 30 km (18 mi) north of Mexico City. This site, known as T1, is at latitude 19° 43' N and longitude 98° 58' W at an altitude of 2340 m above sea level. The sample inlet was located at a height of 10 m above ground level and collected particles having diameters less than 2 um at a flow rate of 16.7 L/m. Aerosol scattering was measured with a portable integrating nephelometer (Radiance Research Model 903) operating at 530nm.

Aerosol absorption was obtained at site T1 by a particle soot absorption photometer, or PSAP (Radiance Research). This method is also a filter based measurement technique. The particle laden air stream is first passed through a primary filter and the change in light intensity from a 567 nm LED is measured to determine aerosol absorption. The clean air stream is then passed through a second filter adjacent to the primary filter. This second filter is used as a reference in order to ensure that the observed change in primary filter transmittance is not due to changes in the intensity of the LED light source. Corrections were made to the measurements for the magnification of the absorption by the filter medium and for nonlinearities in the response of the instrument as the filter is loaded with particulates. These corrections have been determined empirically by the manufacturer (Bond et al., 1999).

3. RESULTS

The results of fine aerosol absorption and scattering at sites T0 and T1 are shown in Figure 1. Aerosol absorption at T0 ranged from 6 – 93 Mm^{-1} with an average of 31 Mm^{-1} and followed a diurnal pattern that reached a maximum at around 06:30 (range of 05:00 – 08:00) local standard time (LST) and a minimum at 13:00 (range of 12:00 – 14:00) LST. Aerosol Absorption measurements at site T1 ranged from 2 – 160 Mm^{-1} with an average of 27 Mm^{-1}. The same diurnal pattern observed at T0 was also evident at site T1 (maximum at 06:30 and minimum at 13:00 LST). Forty-six minute averages of aerosol absorption obtained with a photoacoustic spectrometer have been reported previously for site T1. These varied from 1 – 50 Mm^{-1} (Doran et al., 2007a). While the daily maximum at T1 reported here exceeded those at T0 on most days, these high levels were of much shorter duration, lasting only about 1-2 hours as compared to 7-9 hours of peak levels at site T0. This indicates a local source of

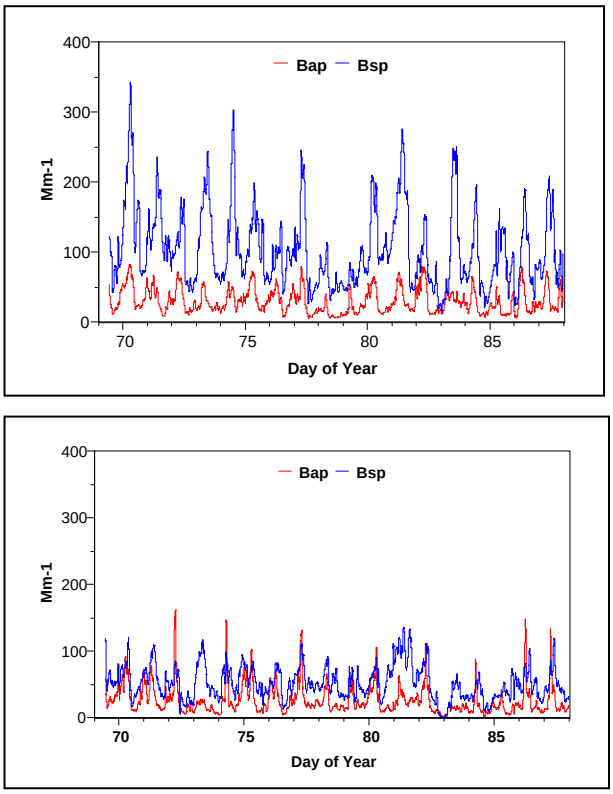

Figure 1. Measurements of fine (<1 micron) aerosol absorption (Bap) and scattering (Bsp) obtained at sites T0 (top) and T1 (bottom) from March 10 (day 69) to March 29, (day 88) 2006 during the MILAGRO field campaign.

absorbing aerosols at site T1. In addition, the minimum aerosol absorption levels at site T1 routinely fell below those observed at site T0. These minimum levels are an indication of the regional background levels in the MCMA.

Aerosol scattering at site T0 ranged from 27 – 344 Mm^{-1} with an average of 128 Mm^{-1}. Scattering values generally reached a maximum at 10:30 (range of 07:30 – 13:00) LST. Aerosol scattering values at T1 were in general much lower than at T0 ranging from 6 – 136 Mm^{-1} with an average of 62 Mm^{-1} and values generally reached their maximum at 08:30 (range of 06:00 – 13:30) LST, earlier than at T0. On 9 of the 18 days reported here the aerosol maximum absorption surpassed the aerosol scattering values at T1.

The aerosol single scattering albedo (SSA) determined from the absorption and scattering measurements at T0 and T1 are shown in Figure 2. The lower aerosol scattering observed at site T1 translates into lower values for aerosol SSA at T1 with a range of 0.35 – 0.87 and an average of 0.69 compared to the SSA determined at T0 of 0.57 – 0.93 with an average of 0.77.

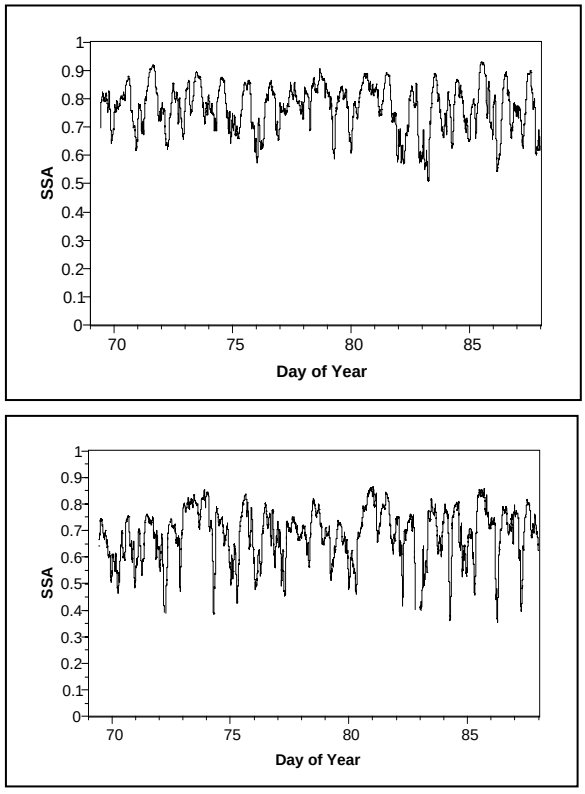

Figure 2. Aerosol single scattering albedo (SSA) determined at site T0 (top) and T1 (bottom) from March 10 (day 69) to March 29, (day 88) 2006 during the MILAGRO field campaign.

Doran et al. have calculated forward and back trajectories of air masses at 1000 m above ground level (AGL) over site T1 during daylight hours (06:00 – 18:00 LST) over a 20-day period during the month of March 2006 (Doran et al. 2007a). The most favorable conditions for transport from site T0 to site T1 were seen to occur on days 69, 70, 77, 78, 79, 81, 83, 86 and 87. On days 71 – 76 and day 82 the trajectories indicate that transport would have likely been from site T1 towards Mexico City and site T0.

Aerosol absorption and scattering measurements shown here were averaged over the daylight hours corresponding to the back trajectories calculated by Doran et al. for sites T0 and T1. The daily absorption averages were found to be similar at both sites ranging from 13 – 44 Mm^{-1} with an overall average value of 31 Mm^{-1} at T0 and a range of 15 – 40 Mm^{-1} with an overall average of 28 at site T1. The daily scattering averages varied from 61 – 197 Mm^{-1} with an overall average of 125 Mm^{-1} at site T0. The daily averages of aerosol scattering were lower and more constant at site T1 with a range of 38 – 105 Mm^{-1} and an overall average of 57 Mm^{-1}.

The aerosol SSAs determined from the daily averages of absorbance and scattering measurements at site T0 and T1 are shown in Figure 3. The average SSA at site T0 ranged from 0.74 – 0.85 with an overall average of 0.8 while the average SSA at T1 was lower and ranged from 0.64 – 0.79 with an overall average of 0.7. Doran et al reported daily average aerosol SSAs at T1 determined from Multi-filter Rotating Shadow-band Radiometer (MFRSR) data on days 71, 78 and 86 as 0.84, 0.85 and 0.89 (Doran et al. 2007). The corresponding average SSA reported here are 0.70, 0.74, and 0.69 as determined from the aerosol

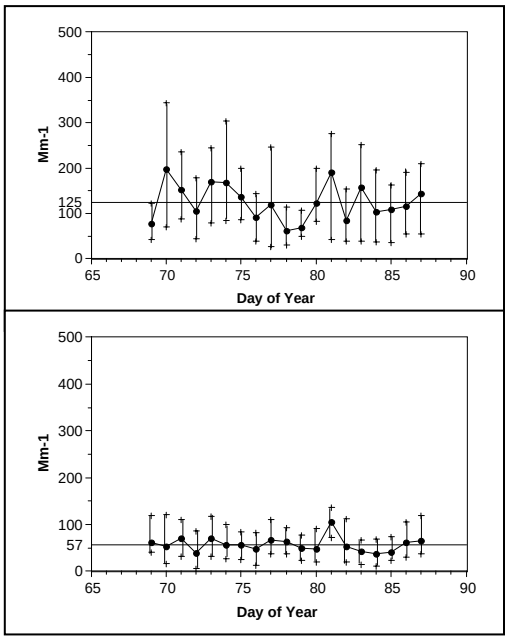

Figure 3. Daily average aerosol scattering (06:00 – 18:00 LST) and scattering ranges measured at sites T0 (top) and T1 (bottom) from March 10 (day 69) to March 29, (day 88) 2006 during the MILAGRO field campaign.

absorbance and scattering measurements. The differences in the reported values arise from the differences in measurement methods used. The MFRSR values are a total column measurement that includes all particles present in the atmosphere. While the values calculated in this work represent a surface measurement of fine aerosol particles only. The fine aerosol fraction is more highly absorbing than the much larger mineral dust particles, which are included in the MFRSR measurements.

4. CONCLUSIONS

Aerosol absorption at T0 ranged from 6 – 93 Mm^{-1} with an average of 31 Mm^{-1} and followed a diurnal pattern that reached a maximum at around 06:30 (range of 05:00 – 08:00) local standard time (LST) and a minimum at 13:00 (range of 12:00 – 14:00) LST. Aerosol Absorption measurements at site T1 ranged from 2 – 160 Mm^{-1} with an average of 27 Mm^{-1}. The daily maximum aerosol absorption at T1 exceeded those at T0 on most days. However, these high levels were of much shorter duration, lasting only about 1-2 hours as compared to 7-9 hours of peak levels at site T0. This indicates a local source of absorbing aerosols at site T1.

Aerosol scattering at site T0 ranged from 27 – 344 Mm^{-1} with an average of 128 Mm^{-1}. Scattering values generally reached a maximum at 10:30 (range of 07:30 – 13:00) LST. Aerosol scattering values at T1 were in general much lower than at T0 ranging from 6 – 136 Mm^{-1} with an average of 62 Mm^{-1} and values generally reached their maximum at 08:30 (range of 06:00 – 13:30) LST, earlier than at T0. On 9 of the 18 days reported here the aerosol maximum absorption surpassed the aerosol scattering values at T1.

The lower aerosol scattering observed at site T1 translates into lower values for aerosol SSA at T1 with a range of 0.35-0.87 and an average of 0.69 compared to the SSA determined at T0 of 0.57-0.93 with an average of 0.77.

5. ACKNOWLEDGEMENT

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