### BLACK CARBON IN URBAN AREAS: MEASUREMENTS ON HOLIDAYS DEMONSTRATE THE IMPACT OF DIESEL SOOT

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# 1. INTRODUCTION

Megacities (population > 10 million) are important sources of fine aerosols and trace gases that can influence the radiative balance of the atmosphere on urban, regional, and global scales. Mexico City, one of the world's largest megacities, has high levels of air pollution because of its large emissions and its topography. Despite advancing technology, Mexico City still has significant numbers of older automobiles and trucks with inadequate emission controls. Chicago, Illinois, and surrounding area, with a population of about 8 million, is an example of a technologically advanced mini-megacity (population 5–10 million) with stronger vehicle controls.

Black carbon (BC) was recently identified as a key aerosol species with significant potential effects on the radiative balance of the atmosphere. We previously presented a paper on the significance of megacities as major sources of BC and the roles that BC can play in radiative forcing through both direct and indirect effects (1). Use of heavy diesel engines in megacities has been identified as a significant source of diesel soot, an important contributor to BC levels in urban centers and to primary BC emissions into the atmosphere on regional and global scales.

Here we present a comparison of BC levels in Mexico City and Chicago during two periods before and during holidays when diesel fuel use was reduced. In April 2003, as part of the Mexico City Metropolitan Area (MCMA) 2003 air quality study, we measured the abundance of BC at a time resolution of 2–5 min by using a seven-channel aethalometer (Thermo Anderson). Data were collected before and during the Easter holiday to assess changes in aerosol loadings as a function of vehicle traffic levels.

We made similar measurements in Chicago before, during, and after the July 4 holiday weekend in 2004 to examine diesel soot inputs from city traffic and from construction activity on the campus of the University of Chicago and on Lake Shore Drive. The data clearly indicate the presence of BC emitted from uncontrolled heavy diesel engines on normal workdays. The data for the two cities are compared and discussed in light of potential impacts of BC on urban heating, air quality, and regional and global radiative balance.

# 2. METHODOLOGY

Measurements of BC were made in Mexico City in April 2003 at a rooftop laboratory at the Centro Nacional de Investigación y Capacitación Ambiental (CENICA) in Iztapalapa, during a collaborative field project between the Department of Energy's Mexico City Megacity 2003 field study and the MCMA 2003 project. The measurements were coordinated by Drs. Mario and Luisa Molina of the Massachusetts Institute of Technology and our research group.

Black carbon measurements were made in Chicago in 2004 at the Center for Environmental Science, Urban Atmospheres Observatory, on the fifth floor of the Hinds Laboratory at the University of Chicago, in the southern part of the city. The BC content of fine aerosols was measured by using a seven-channel aethalometer (Andersen) with a sample inlet designed to collect aerosols in the size range 0.1- $2\,\mu\text{m}$ . The aerosols in the air sample are collected within the instrument by continuous filtration through a paper tape strip. The optical transmission of the deposited aerosol particles is then measured sequentially at seven wavelengths (370, 450, 520, 590, 660, 880, and 950 nm). Because BC is a strongly absorbing aerosol species with a relatively constant absorption coefficient over a broad spectral region, the instrument can automatically calculate the BC content from the transmission measurements if BC is assumed to be the main absorbing aerosol species in the samples with a mass-specific absorption coefficient of  $19 \text{ m}^2 \text{ g}^{-1}$  (2,3).

The instrument is operated by an embedded computer with a display screen and keypad that control all functions. Data are automatically recorded to a builtin 3.5-in. floppy diskette. Data were recorded for each of the seven channels at 2-min time resolution. In addition, the analog output of the 520-nm channel was monitored continuously, and 1-min averages from this channel were recorded separately.

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As the sample is deposited on the paper tape strip, light attenuation increases steadily. At high sample loadings, the high absorptions cause detection limits to decrease. To prevent this, the instrument automatically advances the tape to a new sample spot when light attenuation becomes severe. After the tape advance, a background measurement is taken at each of the seven wavelengths to correct for variations in filter surfaces and source light intensities. The instrument also collects some sample on the filter before taking sample measurements to minimize artifacts due to light scattering from the clean filter surface. The instrument was operated in a dilution mode to minimize downtime due to excessively frequent tape advances caused by the high BC loading observed in both urban areas

Unlike other absorbing aerosol species (such as humic-like substances), the absorption of BC is relatively constant from the ultraviolet to the infrared (2). Thus, comparison of results from the different channels can give independent validation of the assumption that BC is the main absorbing species in the samples. For our sampling periods, all of the seven channels were in excellent agreement, with variation of 1-2%, indicating that BC was indeed the major light-absorbing material present in the aerosol, if not the only one.

Here we present only the data obtained at 880 nm for the two holiday periods. Results for Good Friday 2003 in Mexico City and the 4 July 2004 holiday in Chicago are compared with data for normal workdays before and after the holidays.

#### 3. RESULTS FOR MEXICO CITY

Measurements of BC in Mexico City are shown in Figure 1 for three consecutive Fridays: 11 April, 18 April (Good Friday), and 25 April 2003 (Julian Days 101, 108, and 115). The 24-hr averages and the maximum and minimum values for these dates, calculated for the 24-hr period from midnight to midnight, are given in Table 1.

TABLE 1. Black carbon concentrations  $(\mu g m^{-3})$  obtained in Mexico City with the 880-nm aethalometer channel on three consecutive Fridays, including Good Friday (18 April 2003). Other channels gave similar results.

		Concentration ( $\mu$ g m <sup>-3</sup> )		
Date	Julian Day	Max	Min	Avg
11 April	101	17.3	1.1	6.0
18 April	108	5.5	0.9	2.1
25 April	115	20.5	2.5	6.1

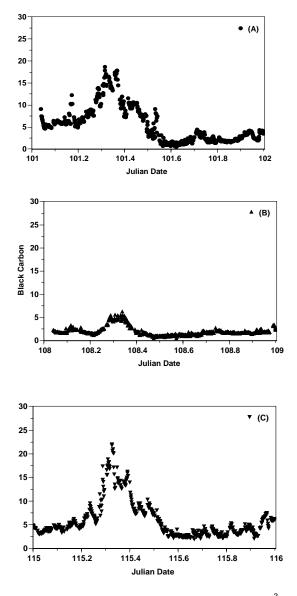


FIGURE 1. Black carbon measurements ( $\mu g m^{-3}$ ) in Mexico City on (A) 11 April, (B) 18 April (Good Friday), and (C) 25 April 2003 from the 880-nm aethalometer channel.

The other aethalometer channels were in good agreement and gave similar results for BC loadings, indicating that BC was the predominant absorbing aerosol species. The large BC peaks seen in the morning hours on 11 April and 25 April (Figures 1A and 1C) are consistent with the expected heavier traffic on those days, coupled with lower boundary layer heights. Values drop off after the peak traffic periods to give a BC level of about  $1-2 \,\mu g \,m^{-3}$ . The data for Good Friday (Figure 1B) show dramatically reduced soot levels, consistent with a reduction in heavy-duty diesel

traffic. In addition, the daily average values for BC (Table 1) are quite consistent for the two non-holiday Fridays at  $6 \ \mu g \ m^{-3}$ . The average BC levels for Good Friday show approximately a threefold reduction compared to the other Fridays.

The Mexico City data clearly indicate that although diesel activity was significantly reduced during the holiday period, BC was not completely absent, with a background level remaining at about 1  $\mu$ g m<sup>-3</sup>. During the month of April satellite data indicated that smoke from biomass fires in the Yucatan Peninsula were affecting the Mexico City area (4). We also have preliminary <sup>14</sup>C data indicating that a substantial amount of the background BC affecting Mexico City was due to long-range transport of soot from the biomass burning event, which lasted throughout the month of April 2003. Other local burning activities, including wood burning and incineration of garbage, could have contributed to the BC and other organic aerosol concentrations observed. This would also contribute to <sup>14</sup>C loadings for the organic aerosols.

#### 4. RESULTS FOR CHICAGO

For comparison, we made measurements of BC in Chicago before and during the 4 July 2004 holiday period (Sunday 4 July and Monday 5 July). Figure 2 shows the BC data collected in Chicago during this holiday period (Julian Days 186 and 187). For comparison, data collected the previous Sunday and Monday (Julian Days 179 and 180) are shown in Figure 3. The 24-hr averages and the maximum and minimum values for these dates, calculated for the 24-hr period from midnight to midnight, are in Table 2.

The BC levels were generally lower by a factor of 10 in Chicago (Figures 2 and 3) than in Mexico City (Figure 1). Thus, it is apparent that because of its higher population and larger emissions, combined with meteorological patterns (5), Mexico City has much higher levels of BC than Chicago. Although the maximum BC levels for Mexico City on Good Friday (the cleanest day we observed there during

TABLE 2. Black carbon concentrations  $(\mu g m^{-3})$  obtained in Chicago with the 880-nm aethalometer channel on the holidays Sunday 4 July and Monday 5 July 2004, as well as on Sunday 27 June and Monday 28 June 2004. Other channels gave similar results.

		Concentration (µg m <sup>-3</sup> )		
Date	Julian Day	Max	Min	Avg
27 June	179	2.3	0.06	1.0
28 June	180	3.0	0.30	1.4
4 July	186	1.0	0.02	0.4
5 July	187	1.8	0.30	0.8

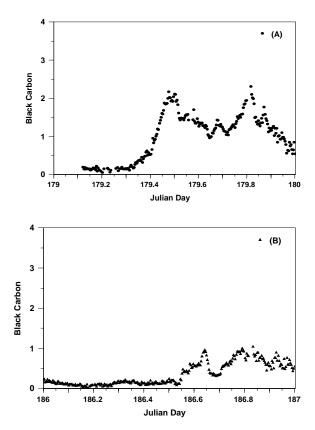


FIGURE 2. Black carbon measurements in Chicago on (A) Sunday 27 June 2004 and (B) the holiday Sunday 4 July 2004 from the 880-nm aethalometer channel.

April 2003) were 5.5  $\mu$ g m<sup>-3</sup>, the maximum levels for BC in Chicago on 27–28 June 2004 were 2.3 and 3.0  $\mu$ g m<sup>-3</sup>, respectively. The maximum BC a week later on 4 July was 1.0  $\mu$ g m<sup>-3</sup>. These values are lower by a factor of 2–5 than the lowest concentrations observed in Mexico City and lower by a factor of about 7–12 than the typical weekday values in Mexico City.

The maximum BC measured in Chicago on 4 July 2004 was 1  $\mu$ g m<sup>-3</sup>, while the maximum the previous Sunday was 2.3  $\mu$ g m<sup>-3</sup>. The general traffic level in the city on the holiday was high because of the crowds attending citywide celebrations. However, BC levels were low because of decreased heavy-duty diesel traffic. Maximum values for BC on Monday 28 June were 3  $\mu$ g m<sup>-3</sup>, as compared to the Monday 5 July levels of 1.8  $\mu$ g m<sup>-3</sup>. Diesel emissions clearly increased on 5 July over 4 July, but they were still below the value for the normal Monday, 18 June. Note also that the minimum values for the Mondays were about 0.3  $\mu$ g m<sup>-3</sup>, while Sunday minimum levels were much lower at 0.06–0.02  $\mu$ g m<sup>-3</sup>.

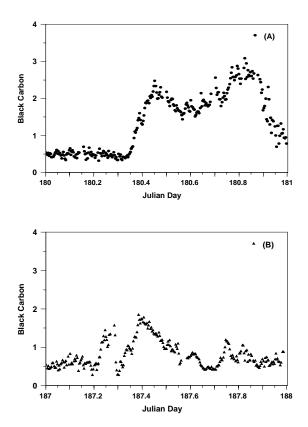


FIGURE 3. Black carbon measurements in Chicago on (A) Monday 28 June 2004 and (B) the holiday Monday 5 July 2004, from the 880-nm aethalometer channel.

Comparison of the Chicago data is more complex than comparison of the Mexico City data. Mexico City has a relatively constant meteorology from day to day (5), while meteorological situations in the Chicago area can change dramatically from day to day and week to week. In the case of 28 June 2004, morning winds prevented the BC levels from reaching their usual peak until later in the morning.

A comparison of the average and maximum BC levels in Chicago for the two periods leads to an estimate of the holiday reduction of BC of about a factor of 2–3. This is in surprisingly good agreement with the results for Mexico City and is probably due to the reduction in use of diesel engines, a significant contributor to the BC levels, during the holiday period, as anticipated.

Diesel engine emissions of BC are not well controlled at this time in either Mexico City or Chicago. Thus, with implementation of controls on diesel engines expected in the United States in the next few years, urban BC levels should fall by a factor of 2–3 when the controls are in full force. A similar reduction would be expected in Mexico City when diesel emission controls are implemented. However, the higher background BC

levels in Mexico City than in Chicago (by about a factor of two) indicate that biomass burning and other factors are significantly affecting the regional air.

## 5. ACKNOWLEDGEMENT

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