

Determination of Black Carbon in Fine Particles Using a Semi-Continuous Method at Two Sites in the City of Guadalajara, Mexico, During 2007

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Received: 3 January 2011 / Accepted: 1 June 2011 / Published online: 15 June 2011
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Abstract The black carbon is a pollutant species primarily emitted from the combustion of fossil fuels (diesel). Their concentrations associated to PM_{2.5} were monitoring at two sites in the city of Guadalajara. From January to May (except April), downtown site shown 2.7, 2.6, 4.0 and 2.3 times higher monthly concentrations. The dry season two showed higher concentrations respect to at least one of the others seasons ($p < 0.0001$) at each site, probably due to atmospheric conditions less favorable for the dispersal of pollutants. During the 24 h period were observed at the year two peaks of concentrations: the highest morning peak and lower night peak, both probably related to anthropogenic activity.

Keywords Black carbon · PM_{2.5} · Aethalometer · Guadalajara

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The carbon in airborne particles can be classified as organic carbon, carbonate and elemental carbon (EC). This latter also is known as black carbon (BC) (Jiang et al. 2005). The black carbon is a pollutant species primarily and the principal emission sources in cities are the incomplete combustion of diesel used in vehicular transport, electrical power plants and diverse industrial processes, production of residential heat from the burning of wood and wild fires (Sasaki and Sakamoto 2006). Black carbon aerosols play an important role in the atmosphere due to their porous and adsorptive nature that provides sites for some important chemical reactions (Latha and Badrinath 2004), as well as in the global climate change by affecting solar intensity (Hansen et al. 2000). Zhang et al. (2005) indicate that in the big cities' carbonaceous aerosols can include a significant proportion of nano-particles, which are dangerous to human health due to their high concentrations and great degree of penetration into the respiratory tract. Likewise, studies like those of Hitzenger et al. (2006) and Viidanoja et al. (2002) show that black carbon is more abundant in fine particles less than 2 μm . In addition, due to the porosity of carbonaceous particles and their enormous surface area, they can adsorb a great quantity of mutagenic and carcinogenic organic compounds, such as polycyclic aromatic hydrocarbons (PAH) and derivatives (Finlayson-Pitts and Pitts Jr 2000; Lighty et al. 2000). Therefore, black carbon is a risk to human health because of its ability to carry carcinogens into the lungs (Jiang et al. 2005).

The goal of this study was to generate one of the first set of information about black carbon associated with PM_{2.5}, as well as contribute to knowledge of the variation of their concentrations (daily, monthly and seasonal) at two sites of the city of Guadalajara, Jalisco, Mexico.

Materials and Methods

The city of Guadalajara, Jalisco, Mexico, together with its metropolitan zone has nearly 4 million people and 812,000 motor vehicles circulating (INEGI 2003), and the city has frequent air pollution problems. The first sampling site was located at the Centro station of the Atmospheric Monitoring Automatic Network that is operated by the Secretary of the Environment for Sustainable Development for the State of Jalisco (SEMADES for its Spanish abbreviation). The site is a commercial zone characterized by a high population density with heavy flow from both gasoline vehicles and public transport buses that operate on diesel. The second site is located inside the Center for Research and Assistance in Technology and Design of the State of Jalisco (CIATEJ for its Spanish abbreviation), almost 2.5 km northwest of the first site. It is a primarily residential area with shops and near the intersection of major arterials with high vehicular flow at certain hours of the day.

Continuous monitoring was carried out between January and December 2007. The concentration of black carbon in this study was determined at 5 min intervals using an Aethalometer operated at the recommended default settings specified by the manufacturers (Magee Sci model AE21), with attenuation cross section of $16.6 \text{ m}^2 \text{ g}^{-1}$ of black carbon at a flow of 2.4 L min^{-1} . The aethalometer converts light attenuation transmitted through a filter to black carbon. At a constant air stream velocity, the rate of deposition of black carbon on the filter is proportional to its concentration in the aerosol ($\mu\text{g m}^{-3}$) and gives a corresponding rate of increase of optical attenuation. The Aethalometer includes an air mass flow meter with an output signal voltage that is calibrated in terms of standard liters per minute, defined as 20°C temperature and 1,013 mb pressure. The instrument flow rate was verified periodically. Five-minute aethalometer black carbon measurements were averaged for each hour and the hourly averages were used to obtain the 24 h average. Monthly data (average and median) was obtained from 24 h period. The 90th and 10th percentiles during period 24 h were used to detect outliers.

Non-parametric tests such as Mann–Whitney U and Kruskal–Wallis were used for comparisons between sites and seasons, respectively. The linear regression model for least-squares of residuals estimated the degree of explanation for the variation between months and hour concentrations at the two sites. All these tests were carried out using the software STATISTICA 6.

Results and Discussion

Table 1 shows a summary of the descriptive statistics of the concentrations of black carbon, with differences

between Centro and CIATEJ sites ($p < 0.0001$). The highest monthly concentrations at Centro were found between January to March and November to December ($p < 0.01$ in all cases), with the lowest concentrations in August and September ($p > 0.05$). At CIATEJ the highest concentrations occurred from November to December ($p < 0.01$ in both cases) and the lowest in February, March and May ($p > 0.05$ in all cases). There were significant differences between these sites from January to May (except April), with higher levels of black carbon at Centro ($p < 0.0001$ in all cases) with 2.7, 2.6, 4.0 and 2.3 times higher concentrations than in CIATEJ, respectively, but not from June to December ($p > 0.05$ in all cases). Figure 1a shows the monthly concentrations at the study sites. Important variations were observed between these two sampling sites, which could be due to intense activity of emission sources or atmospheric conditions less favorable for the dispersal of pollutants at the Centro site during the first months. The Centro site is in an area which has a large number of diesel powered buses and it has been well documented that these sources generate large amounts of black carbon (Park et al. 2002). In 2003 there were at least 5,826 buses (public and private) in the city of Guadalajara and nearly 807,000 vehicles circulating throughout Metropolitan Zone of Guadalajara (MZG) (INEGI 2003), and which surely contributed to the levels of this pollutant.

With respect to black carbon levels in other cities in the world using only Aethalometers, it is observed that the CIATEJ and Centro sites' annual concentrations of black carbon are equivalent to those reported for Vienna, Austria ($2.2 \mu\text{g m}^{-3}$) (Hitzenberger et al. 2006) and different from Xi an, China ($16.5 \mu\text{g m}^{-3}$) (Yang et al. 2006), Pune, India ($4.1 \mu\text{g m}^{-3}$) (Safai et al. 2007) and Seoul and Kwangju, Korea (7.3 and $4.9 \mu\text{g m}^{-3}$, respectively) (Park et al. 2002), whose levels were higher. It is important to note that the monitoring method (Aethalometer) for determining the black carbon can show slight differences in the results with respect to other methods (Hitzenberger et al. 2006). Viidanoja et al. (2002) indicate that the main source of carbonaceous aerosol is vehicular flow. Considering the characteristics of high vehicular flow at the sampling sites, we can suggest that this activity is a major source of emissions for the current black carbon concentrations in the city of Guadalajara.

The study period was divided into two dry seasons; the first from January to May (DS1) and a second from November to December (DS2). June to October corresponds to the rainy season (RS) (CONAGUA 2010). Figure 1b shows the behavior of the seasonal concentrations at each site: at Centro DS2 had higher concentrations with respect to the other two seasons (at least $p < 0.0001$, in both cases), and DS1 was higher than the RS ($p < 0.0001$). At the CIATEJ site DS2 also showed higher

Table 1 Monthly variation in concentrations of black carbon ($\mu\text{g m}^{-3}$) at Centro and CIATEJ sites, both located downtown in the city of Guadalajara in 2007

	n	Average ^a	SD	Median	Percentile 90th	Percentile 10th	Ratio ^c
<i>Centro</i>							
January	30	4.9	2.1	4.7	8.0	2.1	3.8
February	16	4.1	1.6	3.7	6.8	2.2	3.1
March	27	3.7	1.3	3.6	5.5	2.2	2.5
April	30	2.4	1.0	2.3	3.6	1.3	2.7
May	30	2.8	0.7	2.8	3.9	2.0	2.0
June	30	2.2	0.6	2.2	2.9	1.5	1.9
July	17	2.2	0.8	2.2	3.3	1.4	2.3
August	31	1.8	0.4	1.9	2.3	1.3	1.8
September	15	1.9	0.5	1.9	2.7	1.3	2.1
October	30	2.4	0.9	2.3	3.5	1.3	2.7
November	21	4.6	2.0	4.1	7.4	2.5	2.9
December	29	5.6	2.3	5.5	7.9	2.7	3.0
Annual	306	3.2	1.8	2.7	6.0	1.5	4.0
<i>CIATEJ</i>							
January	8	1.8	0.9	1.7	3.1	0.7	4.6
February	28	1.5	0.4	1.4	2.0	1.1	1.9
March	28	0.9	0.3	0.9	1.5	0.5	2.8
April ^b	–	–	–	–	–	–	–
May	10	1.5	1.0	1.2	3.1	0.7	4.5
June	30	2.4	0.6	2.4	2.9	1.7	1.8
July	31	2.0	0.8	2.0	2.7	1.2	2.2
August	31	1.8	0.4	1.9	2.3	1.3	1.8
September	30	2.1	0.6	2.0	2.9	1.3	2.3
October	31	2.9	1.4	2.8	4.5	1.2	3.9
November	30	4.8	1.8	4.6	7.7	2.4	3.2
December	30	5.7	1.8	5.4	7.9	3.0	2.6
Annual	287	2.6	1.8	2.1	5.2	1.0	5.3

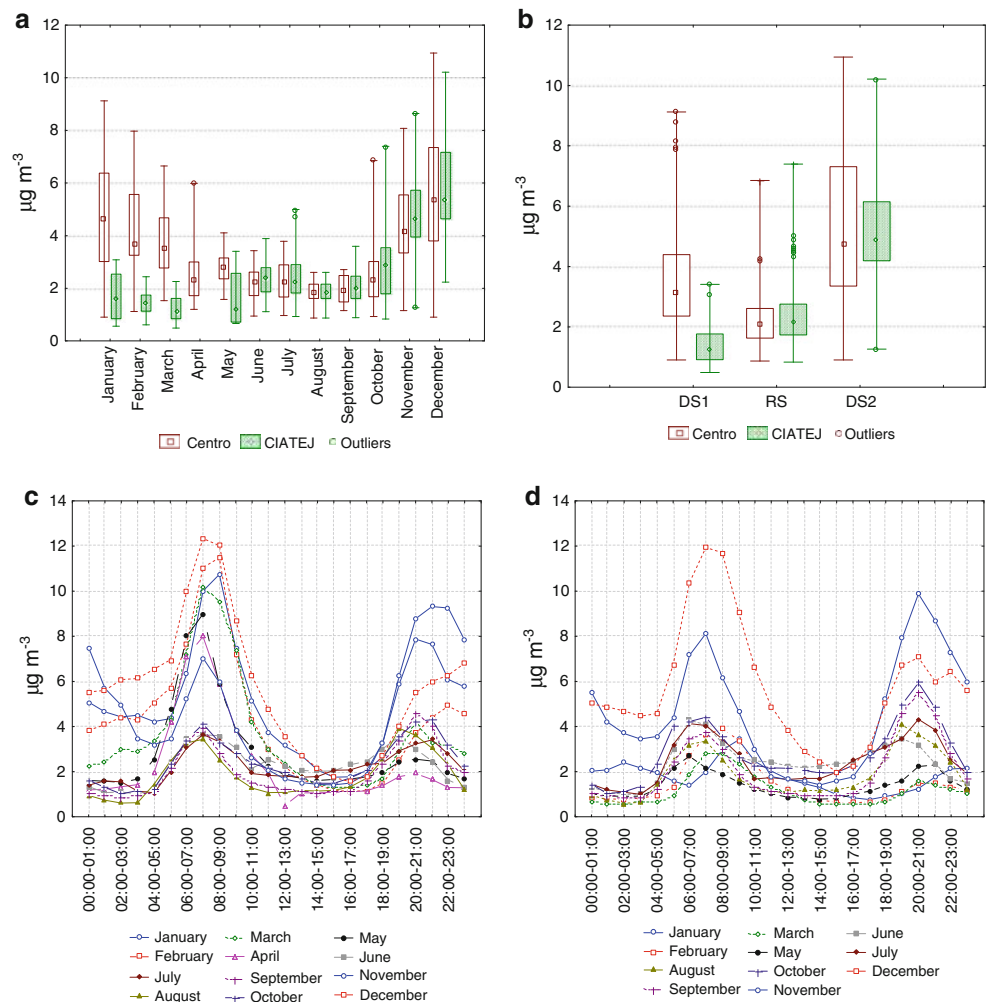
SD, Standard Deviation; n, Days of monitoring

^a January and October without outliers and extremes^b April at CIATEJ without data^c Percentile 90th/10th

concentrations than the other seasons ($p < 0.0001$, in both cases), but RS was higher than DS1 ($p < 0.0001$), probably due to conditions more favorable to the dispersion during DS1. In general, these results suggest that the lower temperatures and conditions less favorable to the dispersion of pollutants during November and December resulted in higher concentrations of black carbon at both sites. Furthermore, during this season there is a greater consumption of fuels for power generation (PJMCA 2007), which also could contribute to the variations observed in this study. Indeed DS1 also has some months characterized by low temperatures (January and February), which partly explains the high concentrations of black carbon at Centro. In November and December (winter) there are fronts of cold and dry air masses that penetrate through the northern

region of the country and move towards the central region, going as far as the MZG. The cold air masses cause lower temperatures, some frost, stratification of the atmospheric layers and intensifies the thermal inversions and stagnation of pollutants (PJMCA 2007). Devara et al. (2002) indicated that during the winter months the rate of ventilation is low because of lower mixing layer height and lower wind speeds. This leads to a smaller dispersion of pollutants, particularly fine particles where carbonaceous material are abundant (Viidanoja et al. 2002; Safai et al. 2007; Harrison and Yin 2008). Marley et al. (2004) argue that for the city of Chicago (US) during the summer vacation period the concentrations of carbonaceous material can be reduced by a factor of two to three, due to a low use of diesel vehicles. In the city of Guadalajara during July and August, the

Fig. 1 **a** Monthly variation in concentrations of black carbon at Centro and CIATEJ sites in the city of Guadalajara during 2007. The first 5 months (except April) showed higher concentrations at Centro ($p < 0.0001$), with respect to CIATEJ. In the rest of the year there were no significant differences ($p > 0.05$), CIATEJ did not have data for April; **b** variation between seasons with significant differences in at least one of them ($p < 0.0001$) at each site; **c** monthly average concentrations by hour of black carbon at Centro and **d** CIATEJ sites. *open square*—Median, *box*—percentile 10–90th, *maximum- and minimum-bar*, *open circle*—outliers



summer vacation period, some of the lower concentrations were observed, mainly at Centro. In these months the activities and movements of large numbers of people in the city are considerably reduced. Furthermore, during the summer season the city receives warm and moist air from the Pacific Ocean, the Gulf of Mexico and the Caribbean Sea, generating higher temperatures that cause vertical movement of the air, leading to a dilution and dispersion of pollutants. Also the higher moisture in this season causes intense rain. It is probably that this condition could contribute to observed seasonal variation.

Variations in the concentrations of black carbon during 24 h periods were also studied. Figure 1c, d illustrates this situation of monthly concentrations per hour at each site, except for April at CIATEJ. Throughout the year there was a marked pattern with two peaks. At both sites from January to May the highest peak (peak 1) was present in the early hours of the morning (06:00–10:00 a.m.) and a lower peak (peak 2) occurred at night (19:00–24:00 p.m.), although at CIATEJ the latter event was not as marked. At the Metropolitan Zone of the

Valley of Mexico (ZMVM) a similar higher level of pollutants was observed during 24 h periods, especially between 06:00 to 11:00 a.m. (IE-ZMVM 2008). From June to August the peak 1 and peak 2 did not exceed $4 \mu\text{g m}^{-3}$. It is probable that the rain and relative humidity during these months determine this behavior. September and October represented a transition period at both sites, during which the peak 2 was slightly higher than the peak 1, because in November the peak 2 showed concentrations greater than the peak 1 at both sites. For December, the pattern was the same as in the first months of the year, with the highest concentrations in the early hours of the morning. A similar tendency of the black carbon hourly concentrations was observed in studies conducted in the cities of Seoul and Kwangju (Korea) and Paris (France) (Favez et al. 2007; Park et al. 2002). Moore et al. (2007) observed that the peak in the morning was related to the times when the population travels to work and stated that concentrations observed in the morning are more affected by nearby sources than in the afternoon. At midday the concentration of black carbon

decreases because the boundary layer rises, causing a dilution of the accumulated black carbon that occurs much faster than any increase in concentration due to the emission rate at that time (Favez et al. 2007). It is probably that the black carbon peak 2 may be related to the return of people to their homes and vehicular generated emissions, although the level is lower possibly because of better conditions for the dispersion of pollutants. The lowest concentrations of black carbon at Centro and CIATEJ occurred around 15:00 p.m., similar to those found by Safai et al. (2007) for the city of Pune, India (16:00 p.m.). However, at CIATEJ during part of the rainy season (August to October), declines were observed in the early morning (02:00 a.m.). Moore et al. (2007) mentioned that the decline in the levels of PM_{2.5} and black carbon in this lapse of time is due to the greater height of the mixing layer. In the same way, in this period there is an increase in the convective activity of the winds which also tend to decrease levels of black carbon (Safai et al. 2007). Table 1 lists the monthly ratios between the 90th percentile and 10th percentile of black carbon concentrations at Centro and CIATEJ; the annual average ratio suggested the great variation of black carbon in periods of 24 h to which the population of the surrounding areas were exposed. A great variation throughout the day and between sites has been described for other air pollutants by Ito et al. (2004) and Safai et al. (2007) in studies on chemical components of PM_{2.5}.

We chose to correlate the concentrations of black carbon at both sites to determine whether one site affected the concentrations of black carbon at the other. For this, linear least squares regression analysis with residuals was carried out for concentrations per hour and month (except April), with CIATEJ as an independent variable because it is located to the north of the city and the wind direction at Centro was from east and northeast during DS2 and RS (dates not shown), respectively, situation that probably contributed with transport of pollutants from CIATEJ and other sites. Considering the value of r^2 : in January CIATEJ black carbon values only explained 32% of the dependent variable (without outliers CIATEJ explained 80%). March, July and October to December values were between 80% and 87%, while May and February were 55% and 76%, respectively (May without outliers explained 80%). In June, August and September, concentrations at CIATEJ explained more than 90% of the variation observed at Centro. Throughout the year at two sites in the downtown section of the city of Guadalajara, evidence has shown that for only a few months, the variation in concentrations of black carbon at CIATEJ had a high influence on the observed levels of black carbon at Centro, and satisfactorily explains an important part of its variation ($r^2 > 90\%$). During the rest of the year emission

patterns of the sources and environmental conditions in both places probably defined the differences in the variations in their concentrations. These results suggested that it is not possible estimate with precision the exposure of the total population in a large geographic area (e.g., as great a city as Guadalajara), when only a few points of monitoring are available. The results can mask those months (or components of particulate matter) that show high relationships between sites (Ito et al. 2004). In general, estimation of exposures is based on the use of average concentrations (of the contaminant of interest) in a city, assuming a homogeneous exposure within the study area (Hoeck et al. 2002). This assumption may result in a significant error, which occurs because the relationship between actual exposure and monitored levels is invalid (Moon 2001). Several studies have reported significant variations in air pollution levels on a small scale within cities, particularly related to the proximity to vehicular flow and the distance to city downtowns (Raaschou-Nilsen et al. 2000). As indicated before, it is important to increase the number of monitoring sites which would allow a more reliable and representative determination of the exposure concentrations in the population in the city of Guadalajara, not only to black carbon, but also to other pollutants.

At the Centro site annual average concentrations per hour (January to November) were obtained for some criteria pollutants (SINAICA 2010) and was then correlated with corresponding levels of black carbon. During the year very similar behavior was observed throughout the day between the concentrations of black carbon versus CO, NO₂ and SO₂ ($r^2 = 0.89$, $r^2 = 0.75$ and $r^2 = 0.80$, respectively, at least $p < 0.0001$ in all cases), but not with O₃. The similarity of the variation in the 24 h concentrations of these pollutants and black carbon suggests that they may come from common sources, mainly diesel vehicles in the roads near the site, since the black carbon within urban areas is considered an important component of these emissions (Park et al. 2002). Park et al. (2002) also mentions that the relationship between carbonaceous aerosol and CO is similar because both are produced in abundance during inefficient combustion processes, and even suggest that the levels of NO_x are a good indicator of vehicle emissions due to the high correlation between NO_x and carbonaceous material concentrations. Harrison and Yin (2008) for the City of Birmingham (UK) found good correlation between carbonaceous material with CO and NO_x. For O₃, there were significant correlation with black carbon ($p < 0.0001$), but they were negative. Ozone is an indicator of secondary photochemical activity which, along with hydroxyl radicals in the atmosphere, is involved in the production of numerous semi-volatile organic species that are most abundant in the particulate phase (Moore et al.

2007). The results of this study indicate that O₃ comes from a different source than the black carbon.

In summary, the median of the annual black carbon concentration at the Centro site was higher. From January to May levels of black carbon were higher at Centro (except April), with proportions of concentrations 2.3–4.0 times higher with respect to CIATEJ, which could be due to intense activity of emission sources or atmospheric conditions less favorable for the dispersal of pollutants at the Centro site. Moreover, at the Centro station the annual average ratio between the 90th–10th percentile of black carbon concentrations was 4.0, while at the CIATEJ this ratio was 5.3, which indicated the great variation of black carbon in 24 h to which the population of the surrounding areas was exposed. The high variation was repeated between seasons and highlights the importance of environmental conditions on concentration changes, especially during DS2 (winter). Low temperature and a higher frequency of slow wind speed probably contributed to this result. Variations of black carbon over a 24 h period indicate the existence of two peaks, one during the morning, peak 1, and the other in the night, peak 2. The peak 1 is related to hours of high vehicular flow (probably mainly due to diesel vehicles), but the peak 2 may be influenced by better conditions for dispersion. Annual average concentrations per hour of some criteria pollutants correlated with the daily fluctuations of the black carbon at Centro. This was especially true with CO and SO₂ and to a lesser degree with NO₂, suggesting sources in common. This has been reported in other cities in the world, where it was also suggested that diesel vehicular flow was the main source of black carbon. Concentrations of black carbon and the respective analysis of linear least squares regression with residuals between sites suggests that during the year, sources that affect the CIATEJ site have a relative impact on concentrations at Centro, what implies it is not possible to determine with precision the concentrations to which the population is exposed to in a large geographic area (e.g., a city such as the city of Guadalajara), when only a few points are used to estimate the exposure for the entire population of a city. These results demonstrate the necessity of monitoring black carbon and other air pollutants in different sites in the city and obtaining a database representative of the entire metropolitan area.

Acknowledgments The authors would like to express their appreciation to Sandra Bravo and Rosalva Cuevas for their technical assistance, to Dana Erickson and Winston Smith of the Peace Corps for the revision of this paper, to the SEMADES, for allowing the installment of the equipment in their locations and to Edmundo Zendejas of the Water National Commission (CONAGUA) for giving us pluvial precipitation data. Special thanks also to Consejo Nacional de Ciencia y Tecnología (CONACyT) for the financial support in this project.

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