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Comparison of continuous and filter based mass measurements in Mexico City

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Abstract

US EPA federal equivalent method (FEM), the tapered element oscillating microbalance (TEOM) (Rupprecht and Pattachnick, Albany, NY) for measuring continuous hourly PM_{10}, was collocated with a filter-based federal reference method (FRM), the PM_{10} sequential filter sampler (SFS), at five sites in Mexico City during February and March, 1997. A PM_{10} mass comparison showed significant differences between instruments that exceeded the expected uncertainties. In general, the TEOM measured higher and more variable PM_{10} than the SFS. It was found that when averaging 24 h of PM_{10} concentrations, exposures to very high levels were missed. The TEOM measurements exhibited much less spatial variations across different sites than the similar comparison of SFS measurements. The mass and chemical composition of the SFS measurements showed that the majority of the PM_{10} mass can be explained by the measured elemental, ionic, and carbon concentrations. This analysis shows that TEOM and filter-based PM_{10} cannot be used interchangeably to determine temporal and spatial distributions in Mexico City during 1997.

Keywords: Particulate matter; Particulate matter measurements; Mexico City; Reference methods; Equivalent methods

1. Introduction

During the past 100 years, Mexico City (MC) has experienced a continuous growth in population and territory, as well as in productive activities and energy consumption. MC currently contains almost 20 million inhabitants with a yearly population growth of 3.3%. MC comprises more than 30% of all of Mexico's national industry with most large factories located in the northern sectors of MC.

The demographic, topographic and meteorological characteristics of the city make it one of the most polluted cities in the world. The inhalable fraction of airborne particulate matter (PM) has gained the attention of the MC authorities. In addition to the filter-based, 24-h integrated total suspended particles (TSP, particles less than 30–60 μm) and PM_{10} (particles with aerodynamic diameters less than 10 μm) monitoring in the compliance monitoring network since 1988, the Mexican environmental authorities deployed in 1995 an automated network with continuous hourly PM_{10} Tapered Element Oscillating Microbalance (TEOM) samplers to have a real-time indicator of air quality in the Automatic Atmospheric Monitoring Network (RAMA). This network currently measures hourly PM_{10} at 13 sites.

In the US, reference and equivalent methods are designated for PM_{10} compliance networks (Code of Federal Regulations, 1988). The sequential filter sampler (SFS) is designated as federal reference method (FRM) (EPA, 1997). The TEOM is designated as a federal equivalent method (FEM) (EPA, 1990). Differences

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between TEOM and filter PM_{10} measurements have been reported (e.g., Green et al., 2001; Cyrys et al., 2001; Pang et al., 2002; Anderson et al., 2002). The most frequent cause of discrepancies is loss of semi-volatile, inorganic and organic material from the TEOM filter due to heating of the sampling stream to 50°C. Some of the volatile material may also evaporate from filters while they are in the sampler, during transport and storage. Filters in this experiment were removed within a day after sampling and kept cool before weighing.

The objectives of this work are to examine spatial variations of PM_{10} across the MC, and to determine the equivalence of TEOM PM_{10} as operated in the RAMA during 1997 with PM_{10} from an independent filter measurement.

2. Characteristics of Mexico City

MC, covering an area of approximately 1300 km^2, lies on the southeastern part of a basin located at 19° north and at an altitude of 2240 m above sea level. The basin is naturally open to the north and surrounded on the three other sides by mountains with an average height of 1000 m above the valley floor. It is a dry region of moderate year-round temperature with prevailing winds from the northwest and the northeast. The rainy season lasts from June to October. PM_{10} concentrations are highest from December through April, dry months where days are shorter and morning temperature inversions are frequent and intense. Mexican O_3 and PM_{10} standards are routinely exceeded during winter.

Most of the TSP originates from the resuspended dust in the northern and eastern parts of MC and from traffic-caused suspension prior paved and unpaved roads. TSP emissions up to 93% are estimated to derive from fugitive dust (Aldape et al., 1991; CAM, 1999).

From 1988 to 2001 (SIMAT, 2002) TSP was $>260\,\mu g/m^3$ (the Mexican 24 h standard) during wintertime over 90% of the time at the Xalostoc station, one of the northern most in the RAMA. A similar behavior was observed for PM_{10} with the 24-h air quality standard of $150\,\mu g/m^3$. From 1990 to 2001, the Mexican annual standards for TSP and PM_{10} (75 and $50\,\mu g/m^3$, respectively) were often exceeded with values up to four times higher than those for all monitoring stations.

Environmental authorities of MC promulgated the Integral Program against Atmospheric Pollution (PICCA, 1990) in 1990. This document emphasized for the first time the importance of PM from both natural and anthropogenic origin. The PICCA program was the beginning of a series of activities that focused on the implementation of a strategy for attaining and maintaining an air quality standard in MC in order to protect human health and welfare.

In 1996, environmental authorities initiated a program (Proaire, 1997) in an effort to improve the air quality in MC. Among other topics, this program emphasized the frequent number of violations of the PM_{10} standard, and the lack of effective control measures that could be adopted to reduce those violations.

One of the most important issues is to determine how well the RAMA PM_{10} measurements provide values suitable for compliance as well as for evaluating spatial and temporal distributions and models results.

3. Reference (SFS) vs. equivalent (TEOM) method studies

Several studies compare filter-based and TEOM PM_{10} measurements (Allen et al., 1997; Cyrys et al., 2001; Green et al., 2001). The most frequently identified cause of difference is loss due to heating of the TEOM sampling stream to 30°C or 50°C. The default 50°C temperature for TEOM prevents water vapor condensation and provides a standard sampling condition, but it volatilizes most of the ammonium nitrate and some of the semi-volatile organic compounds in atmospheric particles (Pang et al., 2002). In many urban areas a significant fraction of the PM_{10} or $PM_{2.5}$ (particles with aerodynamic diameter less than 2.5 μm) consists of organic compounds that might be semi-volatile (Chow et al., 2002a). Monitoring sites with high levels of ammonium nitrate and organic particulate mass does not always yield a reasonable correspondence between time-integrated TEOM and collocated filter measurements.

A comparison between TEOM and two manual gravimetric samplers (Micro Orifice Uniform Deposit Impactor (MOUDI, St. Paul, Minnesota) and a solar-powered low volume aerosol sampler (Solar-Vol 1100) was carried out in Australia (Ayers et al., 1999). The results showed systematically lower results than the TEOM by an average of >30% due to evaporation of semi-volatile aerosol components from the heated filter.

By comparing data from collocated TEOM and Partisol (Rupprecht and Patashnick, Albany, NY) samplers at UK non-urban sites King et al. (2000) found that the agreement between TEOM and Partisol measurements was not consistent and that volatile material may be relatively more prevalent when 24 h averaged PM_{10} concentrations are close to the upper limit of $50\,\mu g/m^3$.

Salter and Parson (1999) showed that in areas principally affected by geological material, the use of the TEOM is supported. However, TEOM sampling in regions with high volatile PM fraction, may produce readings significantly lower than the true values.

High-volume size-selective inlets (Hi-Vol-SSI) were compared with TEOM PM_{10} measurements from 1995 to 1999 (Retama and Castillejos, 2000) at the Tlalnepantla, Xalostoc, La Merced, Cerro de la Estrella and

Pedregal sites from the RAMA network. The results showed PM_{10} from the TEOM to be less than that from the Hi-Vol-SSI. The reason for the large discrepancies was not suggested, although the lack of information with respect to the organic and inorganic chemical composition of PM_{10} and its possible influence in TEOM performance was mentioned.

Retama (2002) showed that comparability varied among sites depending on the sampling periods. The Hi-Vol measured higher PM_{10} concentrations most of the times, consistent with the UK and Australian studies mentioned before.

In general, the previous studies indicate that the correspondence between continuous, in-site hourly TEOM and integrated 24 h TEOM PM_{10} is less than filter-based in regions with a high proportion of volatile components.

4. Sampling methodology

TEOM units from the local Mexican government network and SFS (United States Desert Research Institute, Reno, Nevada), were collocated at five sites within MC (Watson and Chow, 2001a; Chow, 1995). Samples were taken from 23rd February to 22nd of March of 1997 as part of the project "Investigación sobre Materia Particulada y Deterioro Atmosférico-Aerosol and Visibility Evaluation Research" (IMADA-AVER) (Edgerton et al., 1999). SFS samples were collected daily, four times a day at Xalostoc (XAL), La Merced (MER) and Cerro de la Estrella (CES) with sampling periods of 6 h (0:00–6:00, 6:00–12:00, 12:00–18:00, 18:00–24:00 h), giving in total 348 samples (Chow et al., 2002b; Vega et al., 2002). At Tlalnepantla (TLA) and Pedregal (PED), sampling periods were daily 24 h (0:00–24:00 h) with a total of 58 SFS PM_{10} samples. TEOM PM_{10} inlets were cleaned prior to the sampling campaign and 1 l/min was drawn through the TEOM filter, in contrast to the 3 l/min specified for US PM_{10} equivalent. TEOM measurements were available on an hourly basis at all five stations. Whereas RAMA gaseous measurements were the subject of regular external audits, TEOM and meteorological measurements were not subjected to independent verification prior to the IMADA-AVER study. The results of PM_{10} mass concentration obtained with both samplers were compared by the statistical methods specified below.

5. Equipment

The Environmental Protection Agency (EPA) in USA designates TEOM as a FEM for 24-h PM_{10} measurements (EPA, 1990). Particles are continuously collected on a filter mounted on the tip of a glass element, which oscillates in an electric field (Patashnick and Rupprecht, 1990; Patashnick and Rupprecht, 1991). The glass element is hollow, with the wider end fixed and air is drawn through the filter and glass element. The oscillation frequency of the glass element is maintained based on the feedback signal from an optical sensor. The resonant frequency of the element decreases as mass accumulates on the filter, directly measuring inertial mass, i.e., the change in mass of particles collected on the filter changes the resonant oscillation frequency of the tapered tube. Temperatures are maintained at a constant value, typically 30°C or 50°C, to minimize thermal expansion of the tapered element. The TEOM sampler used in this study heated the inlet air stream to a constant 50°C to stabilize the measurement process. It is very sensitive to changes in mass concentrations and can provide precise measurements for sample duration of less than one hour. The Andersen 246 PM_{10} size-selective inlet (Watson and Chow, 2001b) was equipped with each TEOM sampler. The effect of heating on volatilizable species was discussed above, more recent TEOM designs conditions the sample to minimize this volatilization.

The SFS has been designated as a PM_{10} FRM (Code of Federal Regulations, 1988; EPA, 1997) when equipped with an Andersen SA254 size-selective inlet (Smyrna, GA). The PM_{10} inlets provide their specified 50% cut-points at a flow rate of 113 l/min. The flow rate was controlled by maintaining a constant pressure across a valve with a differential pressure regulator (Chow et al., 1993; Chow, 1995). This sampling system, first applied in Portland Aerosol Characterization Study (Watson, 1979) has been used in over a dozen of major studies in US over the last decade (Chow and Watson, 2001; Gertler et al., 1993; Chow et al., 1996; Watson et al., 1998). The device can be programmed for up to six days of unattended operations and allow automatic filter—sequencing as filter overloading occurs. For the IMADA-AVER Study during 1997, the timer was set up to take daily 24 or 6-h samples. Dual sampling channels on each SFS were configured with parallel Teflon-membrane filter and quartz-fiber/sodium chloride impregnated cellulose-fiber filters at a flow rate of 30 l/min. A subset of samples was presented to chemical analysis for ions, carbon, and elements (EPA, 1997; Chow et al., 1996; Chow et al., 2002b). Before gravimetric analyses, Teflon filters were equilibrated for 6 weeks in a controlled environment with a relative humidity of 25–35%, and temperature 21.5 ± 0.5°C to minimize particle volatilization and aerosol liquid water bias (Chow, 1995; Chow and Watson, 1998).

6. Site descriptions

The five sampling sites (shown in Fig. 1) encompass residential, industrial and mixed-use settings. The

northwest station, Tlalnepantla (TLA), is located in a mixed, medium income residential and industrial area. The northeast station, Xalostoc (XAL), is located in a highly industrialized area, and has shown historically the highest concentrations of TSP recorded in MC. La Merced (MER) station is located in the commercial and administrative district downtown. The southwest station is located in Jardines del Pedregal (PED), in a high-income neighborhood, and the southeast station located in Cerro de la Estrella (CES) is a mixed, medium income residential and commercial area. XAL and CES stations are located east of the Texcoco Lake.

7. Results and discussion

The data collected from the TEOM and SFS were used to calculate 24-h average PM_{10} concentrations in the XAL, CES, MER, TLA and PED stations. In the case of XAL, CES and MER, the 6-h measurements from the SFS were averaged to match the 24-h sampling intervals.

7.1. Hourly concentrations

The hourly TEOM PM_{10} measurements showed similar diurnal variations at all stations as shown in Fig. 2. Hourly PM_{10} distributions of TEOM measurements can be assumed to be approximately log-normal, with a geometric mean between 74 $\mu g/m^3$ for MER and 105 $\mu g/m^3$ for PED. The figure shows clear diurnal peaks around 09:00 and 1800–1900 h at all sites. It can be observed that diurnal variations are less pronounced at Pedregal. For SFS monitors, no hourly values were available.

Table 1 shows the percentage distribution of hourly TEOM PM_{10} concentrations as a function of concentration level at all five stations. High hourly concentrations ($>500\ \mu g/m^3$) were often observed, even at non-industrialized sites. All stations observed a maximum concentration exceeding 700 $\mu g/m^3$. Nine cases above

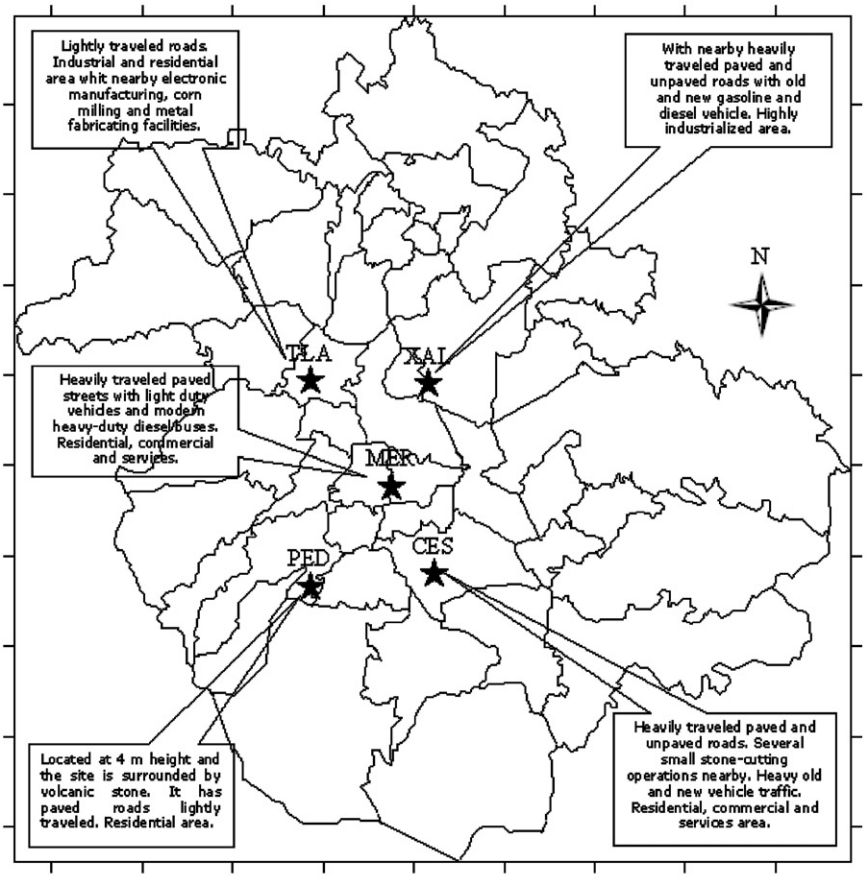

Fig. 1. Map showing Mexico Basin, measurement sites and description.

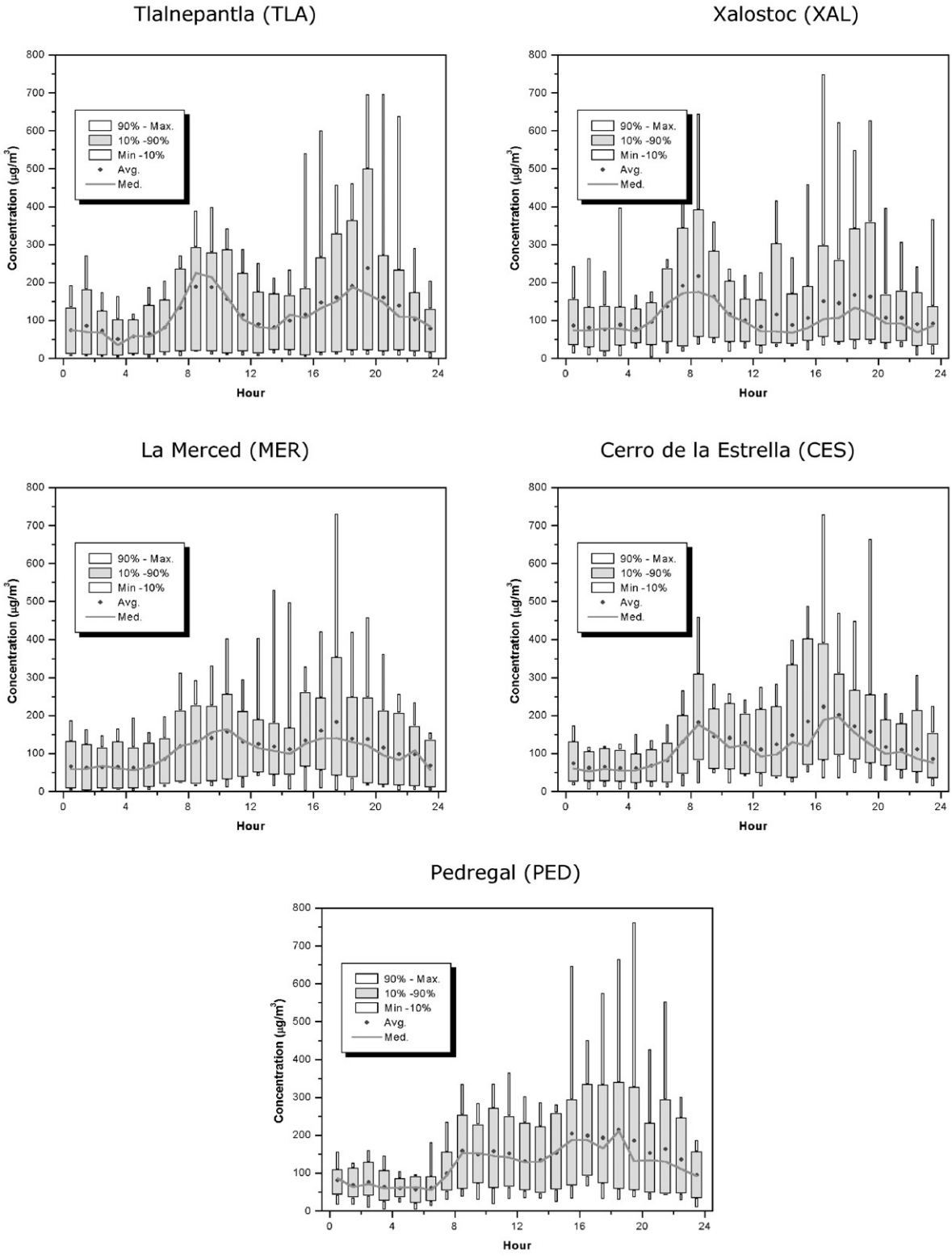

Fig. 2. Diurnal variations of hourly PM_{10} concentrations at the five sites for samples acquired during February and March 1997.

Table 1
Percent distribution of hourly TEOM PM_{10} concentrations as a function of concentration level acquired from 23rd February to 22nd March 1997

Station/PM_{10} mass ($\mu g/m^3$)	<149	150–199	200–300	301–400	401–500	>500
Tlalnepantla	68.8	13.1	13.1	2.6	1.4	1.0
Xalostoc	76.9	9.7	8.1	3.3	1.0	1.0
La Merced	75.9	13.2	8.1	1.4	1.0	0.4
C. Estrella	72.9	11.8	11.3	2.5	1.0	0.5
Pedregal	69.4	13.1	12.9	2.8	0.5	1.3

500 $\mu g/m^3$ were recorded in PED, followed by XAL and TLA with 8 and 7 cases, respectively. A high frequency of values below 149 $\mu g/m^3$ was also observed in MER and XAL. Elevated PM_{10} concentrations occurred in different times and at different locations. While local fugitive dust emissions might affect the nearby station and PM_{10} concentration, it may also be the case that some of the dust collected by the PM_{10} inlet was resuspended and measured as PM_{10} when it was actually much larger. The low 1 l/min flow rate would amplify the effect of this reentrainment as the TEOM mass.

7.2. Daily values

Twenty-four hour PM_{10} concentrations from the TEOM and SFS are plotted in Fig. 3; they can be shown to follow approximately a normal distribution. Arithmetic average TEOM PM_{10} was between 110 $\mu g/m^3$ for MER and 131 $\mu g/m^3$ for PED, with maximum values ranging from 193 $\mu g/m^3$ at CES to 241 $\mu g/m^3$ at XAL. For the SFS-measurements 24-h average ranged from 39 $\mu g/m^3$ at PED to 104 $\mu g/m^3$ at XAL. Maximum-values for SFS ranged from 60 $\mu g/m^3$ at PED to 181 $\mu g/m^3$ at XAL. Daily averaged 24 h PM_{10} concentrations showed that six TEOM and one SFS concentrations were above 200 $\mu g/m^3$.

Homogeneity of variance between stations was tested for each method. Daily averaged TEOM values have comparable variability among all five stations; TEOM measurements have considerably higher variances than SFS measurements. For the SFS PM_{10} mass, only PED and TLA showed similar variance, the other stations had considerably higher variability.

Table 2 presents linear regression statistics for intercepts b, slopes m and correlation coefficients r, for TEOM vs. SFS PM_{10}. Except for the CES station, the TEOM and SFS PM_{10} were reasonably correlated ($0.75 < r < 0.84$). Intercepts close to 0 and slopes close to 1 are needed for the methods to be considered equivalent (Watson and Chow, 2002). For XAL, a slope of 0.62 was obtained, but the other stations showed an average slope of 0.21–0.33. Intercepts are large for all comparisons. TEOM and SFS PM_{10} are not equivalent. SFS PM_{10} is consistently lower than the TEOM PM_{10}. Inferences with Spearman correlation coefficients lead to the same conclusions. The lower slope is partially affected by the large intercept (21–39 $\mu g/m^3$ except for 9 $\mu g/m^3$ at PED). These large intercepts suggest a systematic bias between the two samplers.

The correlation at CES was especially low ($r = 0.13$). The 95% confidence interval for the slope contains the 0-value indicating that the TEOM and SFS measurements can be considered independent and not correlated. Similar results were found when paired measurement tests were carried out.

An experimental blocked design was used to determine the 24-h PM_{10}-measurement differences among stations. Different days were considered as blocks to filter out possible non-random effects due to, for example, meteorological conditions. This test showed that the TEOM measurements could be considered statistically the same for all 5 monitoring stations. Due to the inhomogeneity of variances among stations, this analysis could not be applied for the SFS measurements. A high fraction (89%) of the TEOM PM_{10} was higher than the SFS values. In general the TEOM concentrations were twice or even 4 times higher than the SFS concentrations. Descriptive statistics for the TEOM and SFS 24-h averages are presented in Fig. 4, indicating maximum and minimum values and 25%, 50% (median) and 75% percentiles. For SFS PM_{10}, the highest median value was measured at XAL (99.8 $\mu g/m^3$), with the lowest value, 39.4 $\mu g/m^3$, at PED. Median PM_{10} concentrations varied across the sites by about 60%. Median values for CES, MER and TLA were similar (59.0, 59.9 and 60.0 $\mu g/m^3$, respectively). Medians for TEOM PM_{10} were similar for all sampling sites, ranging from 107.5 to 130.6 $\mu g/m^3$ (Fig. 4). The SFS PM_{10} is more consistent with site characteristics and results from the 31-site network of Chow et al. (2002b).

7.3. 6-h concentrations

Comparisons of 6-h PM_{10} averages of both instruments at the XAL, CES and MER stations are shown in Fig. 5. The frequency of PM_{10} levels above 200 $\mu g/m^3$ is

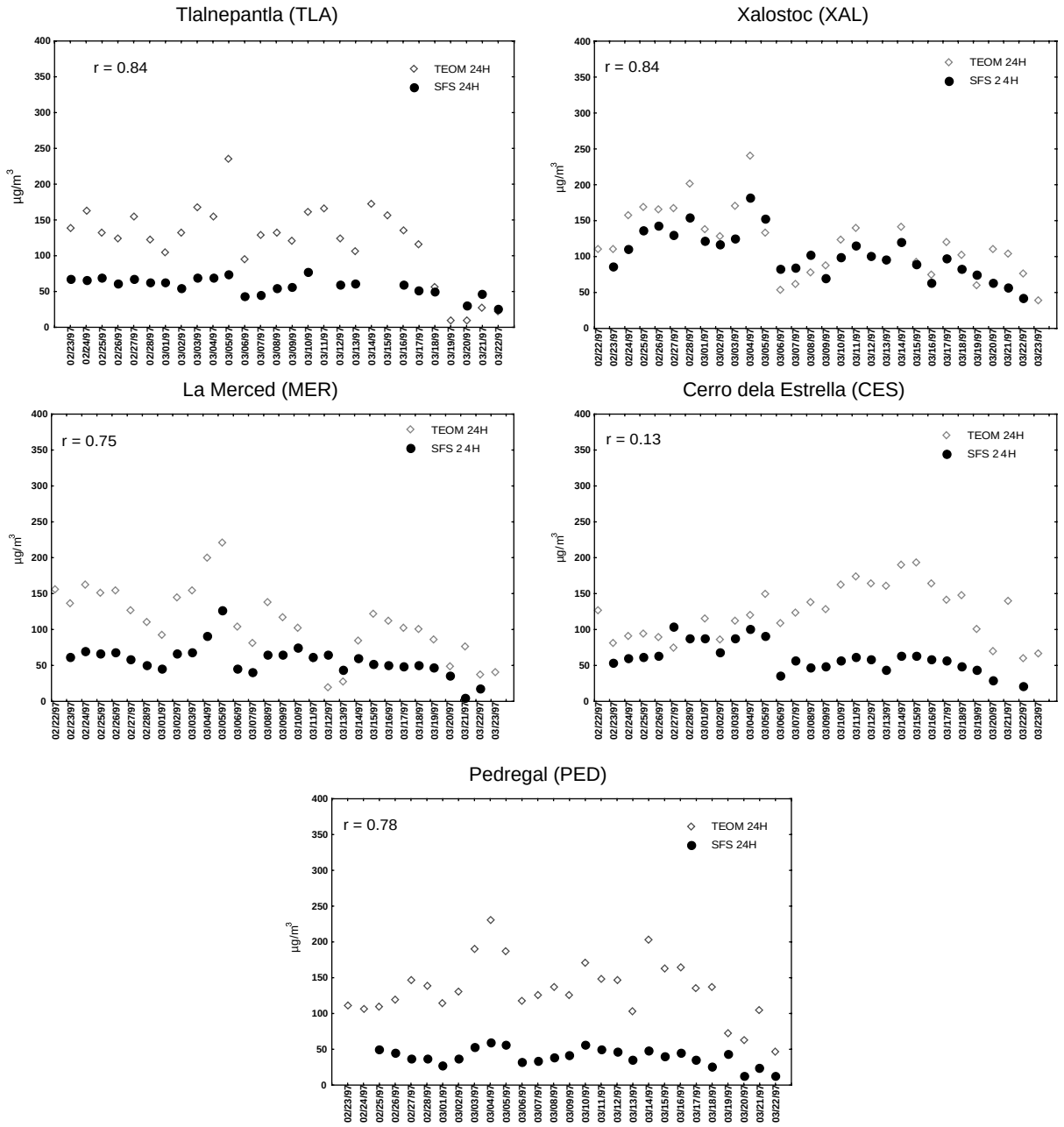

Fig. 3. Averages of 24 h PM_{10} concentrations ($\mu g/m^3$) for TEOM and SFS.

higher for the 6 h averaging periods, with 38 cases for TEOM and 9 cases for SFS PM_{10}.

TEOM and the SFS 6-h average concentrations were often more similar than the 24-h comparisons (Fig. 3), but TEOM PM_{10} were typically higher than SFS PM_{10}. At the XAL station, PM_{10} and its variation are similar for both TEOM and SFS (Fig. 3). In the case of MER, the peaks and valleys are similar for both measurements with consistently higher TEOM concentrations. For the 24 h averages shown in Fig. 3, PED and TLA stations show the largest variations in concentration (in some cases the TEOM PM_{10} is five times higher than the SFS PM_{10}). CES shows a different trend for the TEOM and SFS; for most days (e.g., 6–26th of March) there are large differences in PM_{10} from the two methods. In the new TEOM units a Nafion dryer has been incorporated

Table 2
Linear regression statistics for SFS (depended variable) and TEOM (independent variable) PM_{10}

Station	Correlation coefficient		Linear regression		
	r	95% confidence interval for r	b	m	95% confidence interval for m
Tlalnepantla	0.84	0.66–0.93	32.4	0.21	0.15–0.27
Xalostoc	0.84	0.68–0.92	28.5	0.62	0.46–0.78
La Merced	0.75	0.52–0.88	20.9	0.33	0.21–0.45
Cerro de la Estrella	0.13	−0.37–0.57	38.9	0.32	−0.67–1.30
Pedregal	0.78	0.55–0.89	9.4	0.22	0.14–0.29

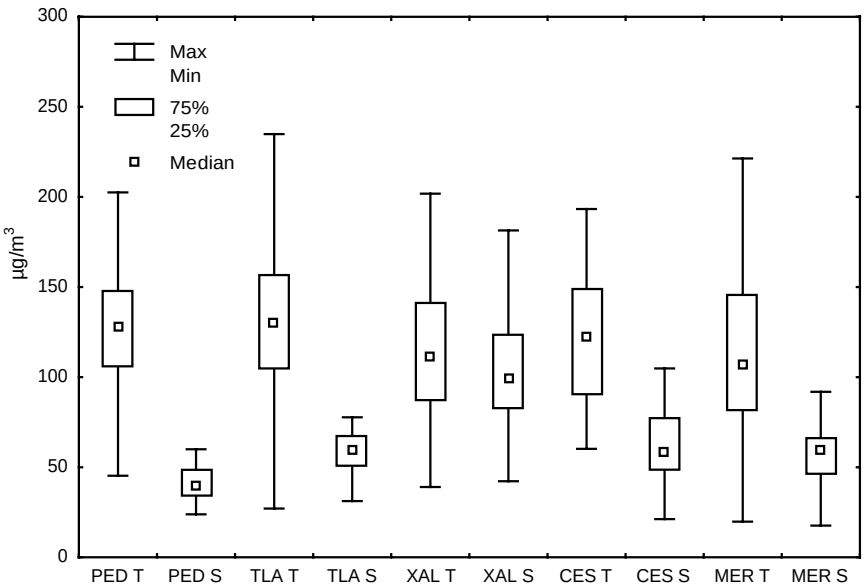

Fig. 4. Basic statistics for 24 h averages of PM_{10} concentrations using TEOM and SFS samplers. (Site codes ending with T designating TEOM, and S designating SFS measurements.)

to minimize particle loss, and continuously conditions both the sample and bypass flows of the TEOM monitor (Meyer et al., 2000).

The statistically significant differences between TEOM and SFS measurements encountered in this study are opposite to the results expected from previous studies (i.e., TEOM $PM_{10}<$ filter-based PM_{10}). Chemical components (i.e., elements, ions, and carbon) from the SFS samples showed mass closure between the mass and chemical measurements (Chow et al., 2002b, c), lending confidence to the SFS PM_{10}. With existing information, it is not possible to precisely identify the cause of the discrepancy. The most likely cause is that mentioned previously; reentrainment of large dust from infrequently cleaned inlets.

8. Conclusions

TEOM and SFS PM_{10} measurements for five sites in MC were compared for the February–March 1997 IMADA-AVER study period, nevertheless, the applicability to other time periods or sites is not known. It is important to mention that for the February to March 1997 IMADA-AVER study period at five sampling locations, their applicability to other time periods and at other locations is not known.

SFS PM_{10} concentration was non-uniformly distributed spatially, the highest and the lowest median values for SFS were observed at XAL (99.8 $\mu g/m^3$) and PED (39.4 $\mu g/m^3$), respectively. Median values varied across sites by about 60%, but the levels at CES, MER and TLA were similar. This is consistent with the survey of

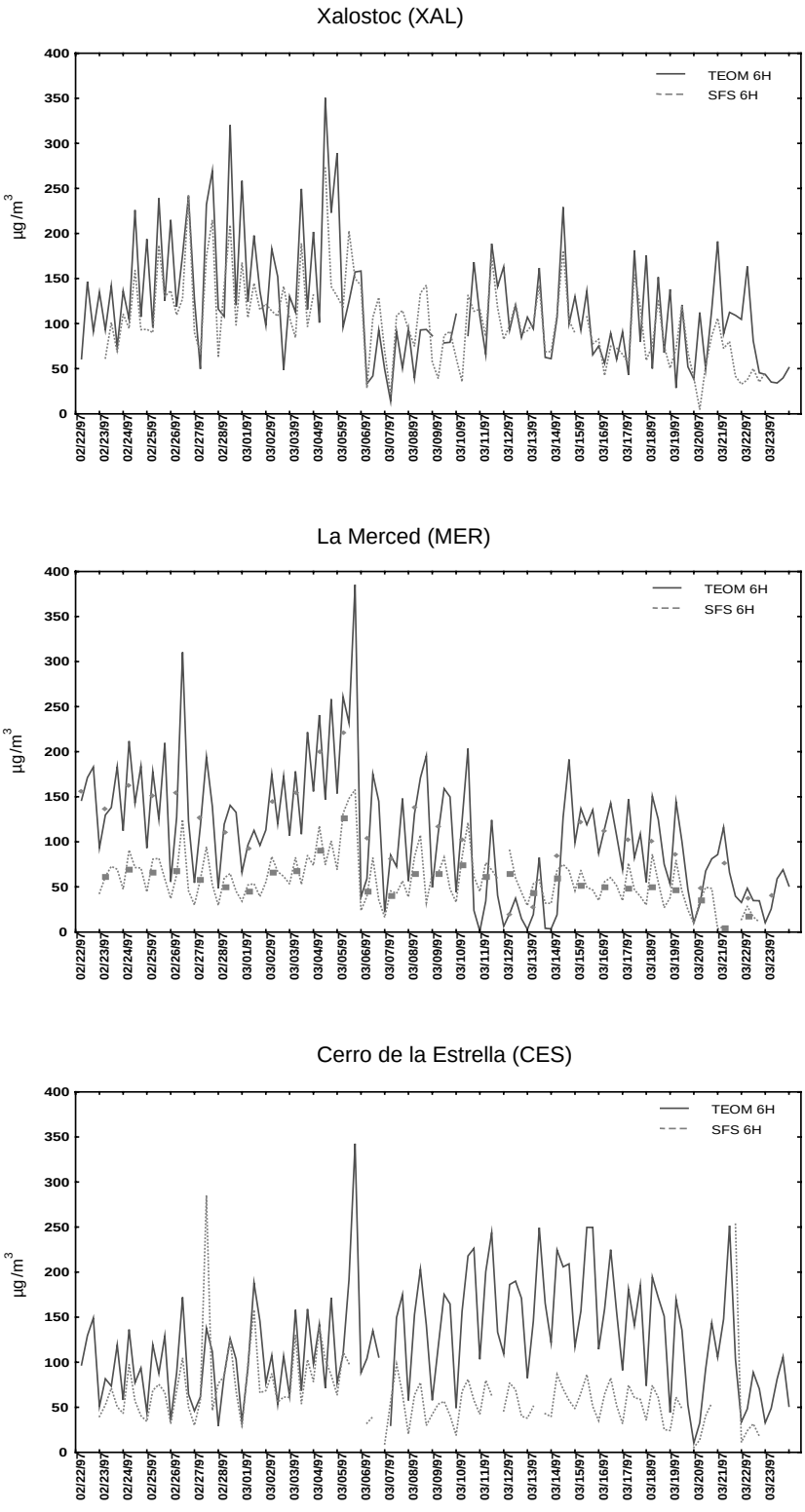

Fig. 5. Averages of 6 h PM_{10} concentration ($\mu g/m^3$) using TEOM and SFS.

local emitters near the monitors. Some monitoring sites are influenced by nearby sources and represent neighborhood rather than urban-soot exposure. This is especially the case for the CES samplers that are located near dust sources that contribute to the majority of coarse particles to atmospheric loadings. XAL is another site that is influenced by emission from different industries and unpaved roads. These sources have a regional as well as local effect, but they may overestimate the PM_{10} mass concentrations measured when the sampler is very close to the emission sources and may bias exposure estimates. Much less spatial variability was found for the TEOM averages.

An experimental blocked design was used to determine the PM_{10}-measurement differences among stations. It was found that the TEOM measurements could be considered statistically the same for all 5 monitoring stations.

In this study it was found that the relationship between reference gravimetric methods and the TEOM varied depending on location of site, and range of particle concentrations. Different statistical methods showed that TEOM and SFS samplers were not equivalent and in general, the SFS concentrations were consistently lower than the TEOM concentrations. This is opposite to the bias expected owing to the evaporation of volatile substances and most likely due to poor cleaning of the inlets.

The lack of comparison indicates that TEOM and SFS PM_{10} concentrations cannot be used interchangeably for modeling and for spatial or temporal data analyses.

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