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Development of a Near-Continuous Monitor for Measurement of the Sub-150 nm PM Mass Concentration

Bhabesh Chakrabarti, Manisha Singh, and Constantinos Sioutas
Civil and Environmental Engineering, University of Southern California, Los Angeles, California

Population exposures to ambient particulate matter (PM) have recently received considerable attention due to the association between ambient particle concentrations and mortality. Recent toxicological studies suggest that ultrafine PM (diameter <100 nm) may be responsible for the observed health effects. However, even though ultrafine mass concentrations vary drastically over short time scales in the atmosphere, no monitor currently measures ultrafine PM mass continuously. The need for monitors that can perform ultrafine particle concentration measurement in shorter time intervals is of paramount importance to environmental health, as such a monitor can lead to substantial improvements in population exposure assessment to ambient ultrafine PM.

In this study, a modified BAM (Beta Attenuation Monitor) is employed to measure near-ultrafine (i.e., <0.15 μm or $PM_{0.15}$) particulate mass concentration. The BAM is preceded by a 0.15 μm cutpoint impactor, which is designed to have very low pressure drop. The BAM is operated in a 2 h cycle at a downwind receptor site in the Los Angeles Basin in Claremont. Among the other instruments colocated with the BAM are scanning mobility particle sizer (SMPS), an aerodynamic particle sizer (APS), and a Micro-Orifice Uniform Deposit Impactor (MOUDI).

Our results indicate that the $PM_{0.15}$ mass concentrations obtained by means of the modified BAM and MOUDI are in excellent agreement. The $PM_{0.15}$ SMPS-to-BAM concentration ratio is generally smaller than 1 and follows a rather distinct diurnal profile, with a maximum towards the middle of the day and minima during the early morning and nighttime periods, presumably due to the classification of fractal-structured ultrafine particles in the accumulation mode by the SMPS. The lack of correlation between $PM_{2.5}$ and $PM_{0.15}$ mass concentrations further corroborates the need for developing monitors such as the modified BAM for the documentation of the short-term variation of ultrafine mass measurements.

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Address correspondence to Constantinos Sioutas, Civil and Environmental Engineering, University of Southern California, 3620 South Vermont Avenue, Los Angeles, CA 90089, USA. E-mail: sioutas@usc.edu

INTRODUCTION

Increased concentrations of airborne particulate matter (PM) have been positively correlated with mortality and morbidity rates by a variety of epidemiological studies (Dockery et al. 1993; Pope et al. 1995). Atmospheric ultrafine particles (often defined as those smaller 100 nm) have recently received significant attention because recent toxicological investigations have indicated their potential for eliciting adverse health effects (Oberdörster et al. 1992, 1995; Donaldson and MacNee 1998).

To date, there has been rapidly increasing epidemiological evidence linking respiratory health effects and exposures to ultrafine particles. Epidemiological studies conducted by Heyder et al. (1996) and Peters et al. (1997) have demonstrated a stronger association between health effects and exposure to ultrafine particles as compared to fine or coarse particles. A study by Pekannen et al. (1997) showed associations between fine and ultrafine particles and the incidence of asthma in children. Laboratory studies by Ferin et al. (1991) indicate that, for deposition of the same amount of PM in the lung, toxicity seems to increase as the particle size decreases. More recent studies by Li et al. (2002, 2003) demonstrate that when epithelial cells from human airways were exposed to the different modes of atmospheric PM, based on an equal mass basis, ultrafine PM caused a greater degree of response.

These emerging findings from both the toxicological and epidemiological studies further strengthen the need for developing instruments that could measure ultrafine PM mass concentrations. However, despite the fact that ultrafine mass concentrations vary drastically over short time intervals in the atmosphere, no monitor currently measures ultrafine PM mass concentration continuously or near continuously (in 1–3 h intervals).

Although the scanning mobility particle sizer (SMPS; TSI Inc., St. Paul, MN, USA) measures particle number concentrations near continuously, it suffers from many shortcomings when it comes to measurement of ultrafine particle mass. A recent study by Shen et al. (2002) found that SMPS underestimates ultrafine PM mass concentrations due to effect of shape and density of fractal-like particles typically generated by vehicular emissions. Apart from the effects of particle shape and density, the validity of the assumption that charged particles in the SMPS follow a charge distribution that can be predicted by the Boltzmann equilibrium may also contribute to errors in the measurement of ultrafine PM concentrations (Sioutas et al. 1999).

Ultrafine particles have been typically defined as those smaller than 0.1 μm. Unlike the case of coarse and fine particles, which are naturally divided by a cutpoint of 2.5 μm, there is no clear cutpoint that separates the ultrafine from the accumulation mode PM. This is because contrary to coarse and fine (accumulation plus ultrafine) PM, which have distinctly different origins, a major fraction of accumulation-mode PM originates from the ultrafine mode. It should be noted that in the aforementioned epidemiological studies linking exposures to ultrafine PM and health effects ultrafine particles were not defined as those having physical or aerodynamic diameters below 0.1 μm but rather as particles which dominate the number-based size distribution of ambient aerosols. No attempt was made (or considered necessary) to separate population exposures to particles above and below 0.1 μm. This definition is probably appropriate when referring to urban aerosols, for which the number median diameters typically range from 20–50 nm (Kim et al. 2002; Morawska et al. 2002). In the context of the present study, however, we prefer to set the cutpoint at 0.15 μm for the following reasons. If "ultrafine PM" are defined as those originating mostly from vehicular emissions and accounting for over 90% of the number-based particle concentrations, this cutpoint between the ultrafine and accumulation modes should ensure the accuracy and integrity of this definition. In our recent studies of the Southern California Supersite funded by the US EPA, we have found that the ambient aerosol number median diameters in the inland valleys of the Los Angeles Basin (areas with some of the worst PM pollution levels in the US) are in the 90–150 nm range during the warmer months of the year (Kim et al. 2002; Fine et al. 2003), which incidentally is when PM levels are the highest. We thus believe that setting the cutpoint at 0.15 μm may provide a more unambiguous separation between the ultrafine and accumulation-mode PM, at least in receptor areas of the Los Angeles Basin, which are also of considerable epidemiological interest given their high PM concentration levels.

This article presents the development of a modified beta attenuation monitor (BAM, Model 1020, Met One instruments, Inc., OR, USA) for near-continuous ($\sim$2 h) measurement of near-ultrafine PM mass concentrations. The BAM is preceded by a very low pressure drop, 0.15 μm cutpoint inertial impactor to remove all but particles below that size from the air sample. Sampling was conducted in the outdoor environment of Claremont, a receptor site of the Los Angeles Basin. The BAM is colocated with micro-orifice uniform deposit impactor (MOUDI; MSP Corp. Minneapolis, MN, USA; Marple et al. 1991), a SMPS (TSI Model 3936), and an aerodynamic particle sizer (APS; TSI Model 3320). The focus of the study is to document short-term variation in the ambient near-ultrafine ($PM_{0.15}$) mass concentrations and to compare the BAM output with the time-integrated $PM_{0.15}$ mass concentration from the SMPS. The ratios of SMPS to BAM mass concentrations are studied for different time periods of the day. The correlations, or lack thereof, between ultrafine number concentrations from SMPS and mass concentrations from BAM are investigated for specific time periods of the day, as well as for 24 h averaged data. A similar exercise is carried out for $PM_{0.15}$ mass concentrations from BAM and the $PM_{2.5}$ mass concentrations provided by the SMPS–APS tandem.

METHODS

Instrumentation

The monitor for measuring near-ultrafine PM mass concentrations near continuously consists of a standard BAM (BAM, Model 1020, Met One instruments, Inc., OR, USA) preceded by a 0.15 μm cutpoint impactor (Figure 1). Ambient aerosols are drawn through the BAM at a sampling flow of 16.7 l min^{-1} via a vertical aluminum tube 245 cm long and 3.3 cm in outer diameter. The lower end of the inlet tube is inserted directly into the top of BAM housing, the other end points horizontally upward above the roofline. The 0.15 μm cutpoint impactor is mounted on the upper end of the inlet tube. The modified BAM is operated in a 2 h cycle.

The 0.15 μm cutpoint impactor is a single, rectangular (slit) jet geometry conventional impactor made of aluminum operating at a flow rate of 16.7 l min^{-1} and under a very low pressure drop (8 in H_2O or 1.99 kPa). Particles larger that the cutpoint are collected on narrow strips of quartz filter substrate (Tissuquartz, Pall Gelman, Ann Arbor, MI, USA). The impactor has a rectangular nozzle 5 cm in length and 0.011 cm in width (W). The distance between the exit of the acceleration nozzle plate and the impaction substrate is 0.0254 cm. The average jet velocity (U) corresponding to a flow of 16.7 l min^{-1} is about 50 m/s.

The principal parameter determining particle capture is the Stokes number of a particle having a 50% probability of impacting, St, defined as the ratio of the particle stopping distance to the width of the acceleration slit nozzle of the impactor (Hinds 1992):

$$\mathrm{St} = \frac{\tau U}{W}\frac{\rho_p C_c d_p^2 U}{9\,\mu W}, \qquad [1]$$

where W is the width of the impactor's nozzle, U is the average velocity of the impactor jet, ρ_p is the particle density, μ is the

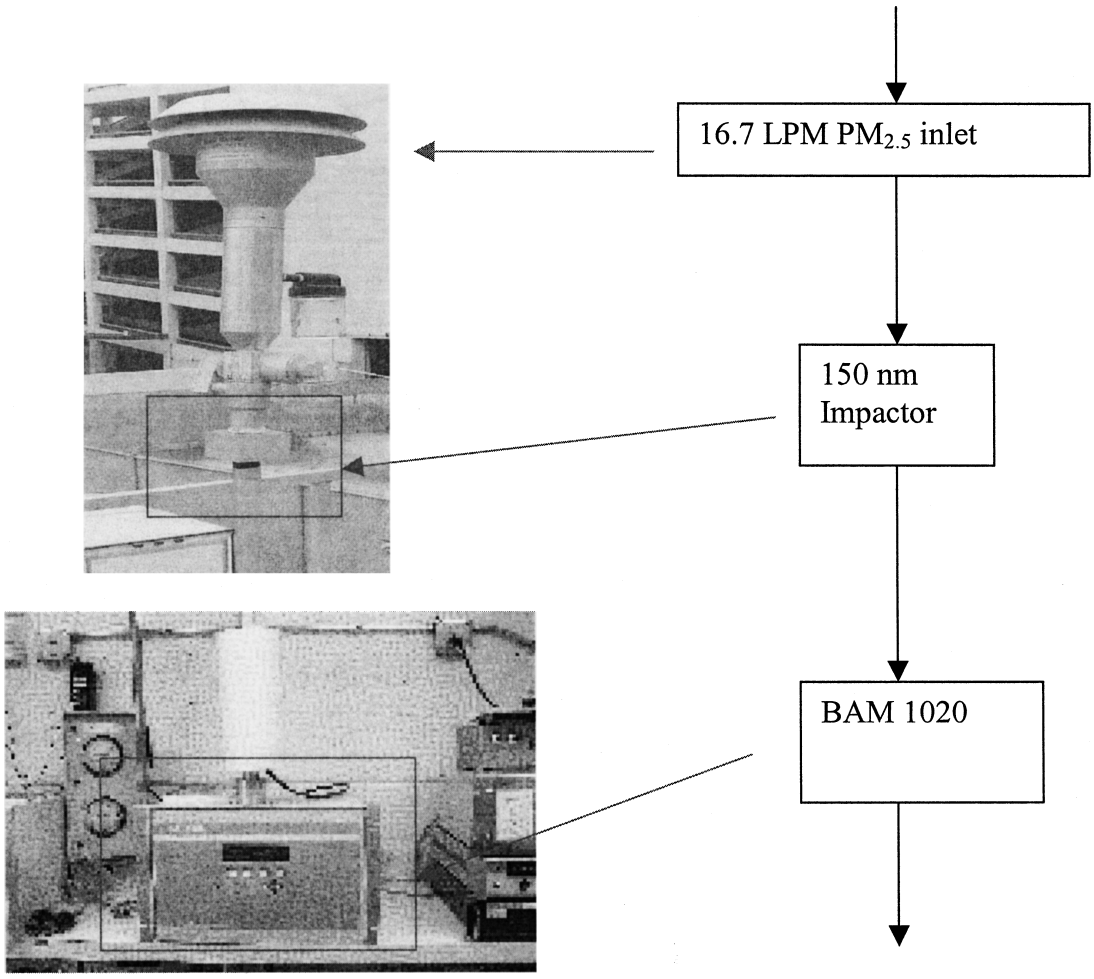


Figure 1. Modified BAM monitor for continuous near-ultrafine ($PM_{0.15}$) mass concentrations measurement.

dynamic viscosity of the air, and C_c is the Cunningham slip correction factor, given by the following equation (Hinds 1982):

$$C_c = 1 + \frac{2}{Pd_p}\left[6.32 + 2.01e^{(-0.1095Pd_p)}\right], \qquad [2]$$

where P is the pressure at the location of the particle in the flow (in cm Hg) and d_p is the particle diameter in μm. For the aforementioned values of W and U, the Stokes number corresponding to 0.15 μm was approximately 0.14.

The modified impactor was characterized in the laboratory using polydisperse ammonium sulfate aerosols generated by means of a nebulizer (HOPE, B & B Medical Technologies, Inc., Orangevale, CA, USA). The experimental setup is shown schematically in Figure 2. Aqueous ammonium sulfate solutions (roughly 1 mg of ammonium sulfate in 1 mL of deionized water) were nebulized using room air at 20 psi. The generated aerosol passed through a 1 l chamber with ten Polonium 210 ionizing units (Staticmaster, NRD Inc., Grand Island, NY, USA) to reduce particle charge close to the Boltzmann equilibrium. After the neutralizer, the aerosol was mixed with particle-free room air (relative humidity: 20–30%) in a 35 l chamber and passed through the specific impaction stage. For particles in the size range of 0.015 to 0.50 μm, penetration (or 1 – collection efficiency) was determined by measuring their number concentration upstream and downstream of the impactor by means of the SMPS (Model 3936, TSI). The SMPS sampled 0.3 l min^{-1} of the total flow rate of 16.7 l min^{-1} through the impactor.

Sampling Location and Field Tests

The performance of BAM was evaluated in a field study conducted in July and early August 2002 in Claremont, CA. The BAM was collocated with a MOUDI, SMPS (Model 3936, TSI Inc), and APS (Model 3320, TSI Inc.). Direct comparisons were made between the time-integrated averaged $PM_{0.15}$ mass concentrations measured by the BAM and those of the MOUDI corresponding to particles smaller than 0.16 μm in aerodynamic diameter. All instruments were placed inside a

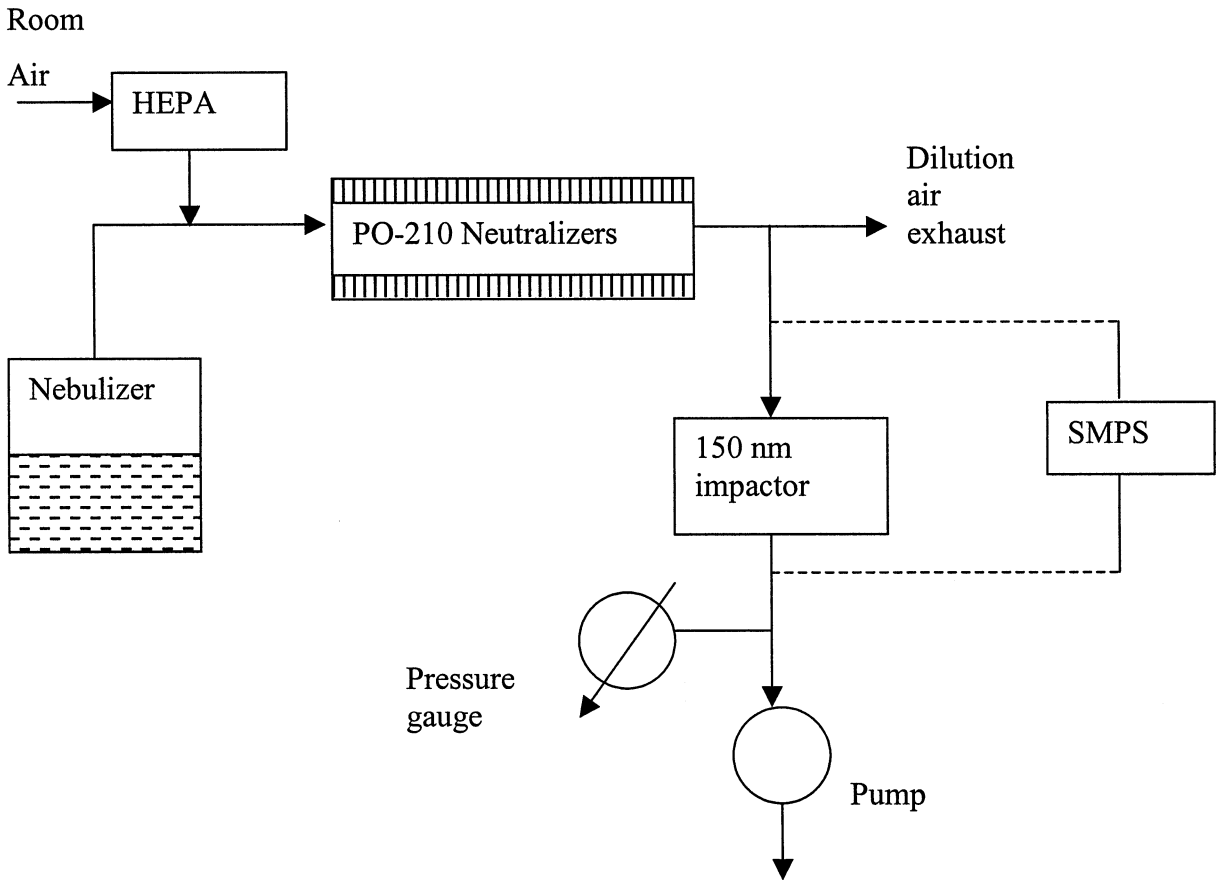


Figure 2. Laboratory evaluation of 150 nm cutpoint impactor.

mobile laboratory trailer that was developed through funds provided by the US Environmental Protection Agency and is being currently used in large-scale field studies that are part of the Southern California Particle Center and Supersite activities. Sampling was conducted at Claremont, a downwind receptor site in the Los Angeles Basin located approximately 45 miles east of downtown Los Angeles. Claremont is a receptor site of the eastern inland valleys of the Los Angeles basin in which aerosol plume generated by the millions of vehicles, mostly west of downtown Los Angeles, is advected by the predominant westerly winds after aging for several hours to a day (Pandis et al. 1992). Ambient aerosols were drawn through the SMPS and MOUDI via vertical stainless steel tubes, 250 cm long and 2.0 cm in diameter, equipped with elbow-shaped inlets to prevent entrainment of rain droplets. The resulting particle residence time was about 1.6 and 7.5 s for MOUDI and SMPS, respectively, thus small enough to avoid diffusional losses of ultrafine PM.

The MOUDI operated at 30 l min^{-1} and sampled for 4 h time intervals. Forty-seven-millimeter Teflon filters (PTFE, Gelman, 2 μm pore, Ann Arbor, MI, USA) were used as impaction substrates. Particles smaller than 0.16 μm were collected on a 37 mm Teflon after-filter. Teflon filters of MOUDI^{TM} were pre- and postweighed using a Mettler Microbalance (MT5, Mettler-Toledo, Inc, Hightstown, NJ, USA) after 24 h equilibration under controlled humidity (35–40%) and temperature (22–24°C) to determine particle mass concentrations. The SMPS and APS number-based concentrations were converted to mass concentrations using an algorithm described by Sioutas et al. (1999), assuming that particles are perfectly spherical with a density of 1.6 g cm^{-3}. The SMPS concentrations were adjusted to take into account the experimentally determined particle penetration from the 0.15 μm impactor of the BAM, in lieu of assuming a step-function change in particle penetration at 0.15 μm.

RESULTS AND DISCUSSION

Laboratory Evaluation of the 150 nm Cutpoint Impactor

Results from the laboratory evaluation tests of the impactor are summarized in Figure 3. Particle collection efficiency data are plotted as a function of aerodynamic particle diameter. The results plotted in Figure 3 indicate that the experimentally determined 50% collection efficiency cutpoint is approximately 148 ($\pm$10) nm in aerodynamic diameter. Particle collection

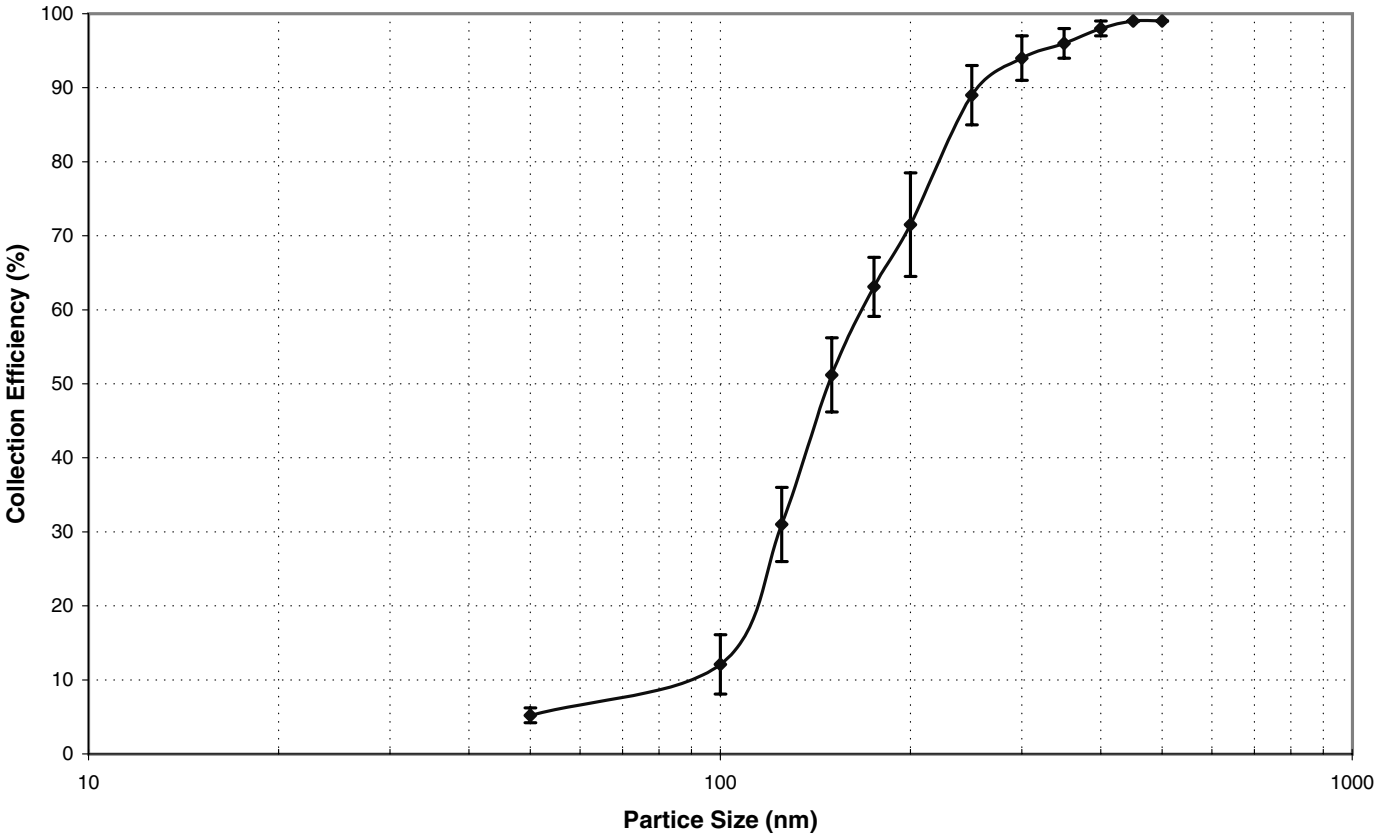


Figure 3. 0.15 μm cutpoint impactor—collection efficiency as a function of particle size.

efficiency increases rapidly with particle size to values exceeding 90% as particles become larger than about 250 nm in aerodynamic diameter. The ability to separate $PM_{0.15}$ from the rest of the aerosol under a very low pressure drop (1.99 kPa) is a particularly attractive feature of this impactor because it eliminates the need for using a high vacuum pump, thus making it compatible with real-time (or continuous) instruments, such as the BAM, which typically operate using light, low-powered pumps or blowers.

The sharpness of the collection efficiency curve of an impactor can be defined in terms of the geometric standard deviation (σ_g), which is the square root of the ratio of the aerodynamic particle diameter corresponding to 84% collection efficiency to that corresponding to 16% efficiency (Marple and Willeke 1976). Generally, lower σ_g values indicate higher precision in particle separation characteristics of a given impaction stage, a highly desirable feature for an impactor because it leads to a finer resolution in the size distribution of an aerosol. Based on this definition, the value of σ_g was estimated to be approximately 1.38 (i.e., roughly the ratio of 0.23 to 0.12 μm, the particle sizes corresponding to 84% and 16% collection efficiency, respectively), thereby indicating that the impactor achieves reasonably sharp particle separation.

Field Evaluation of BAM

$PM_{0.15}$ particle mass concentrations (integrated over 4 h periods) measured by the BAM were compared with those obtained with MOUDI. Mass concentrations ranged from 1.50 $\mu g/m^3$ to 8.98 $\mu g/m^3$ for the BAM and 1.51 $\mu g/m^3$ to 9.30 $\mu g/m^3$ for the MOUDI, respectively. Intercomparisons between BAM and MOUDI indicate an overall excellent agreement, with an average BAM-to-MOUDI concentration ratio of 0.92 ($\pm$0.12). Figure 4 depicts the plot between $PM_{0.15}$ mass concentrations obtained with BAM and those obtained with MOUDI, along with the linear regression line and the regression coefficient. As is evident from the figure, the BAM concentrations are highly correlated with those of MOUDI with $R^2 = 0.92$.

Comparisons Between $PM_{0.15}$ Mass Concentrations Obtained from BAM and SMPS

The average ratio of the SMPS-to-BAM $PM_{0.15}$ mass concentration, along with standard error, is plotted in Figure 5. The plotted mean ratios correspond to 2 h data collected during July and early August of 2002. The SMPS-to-BAM concentration ratio is smaller than 1 for most of the time during the day, indicating that the mass concentration of sub-150 nm PM estimated

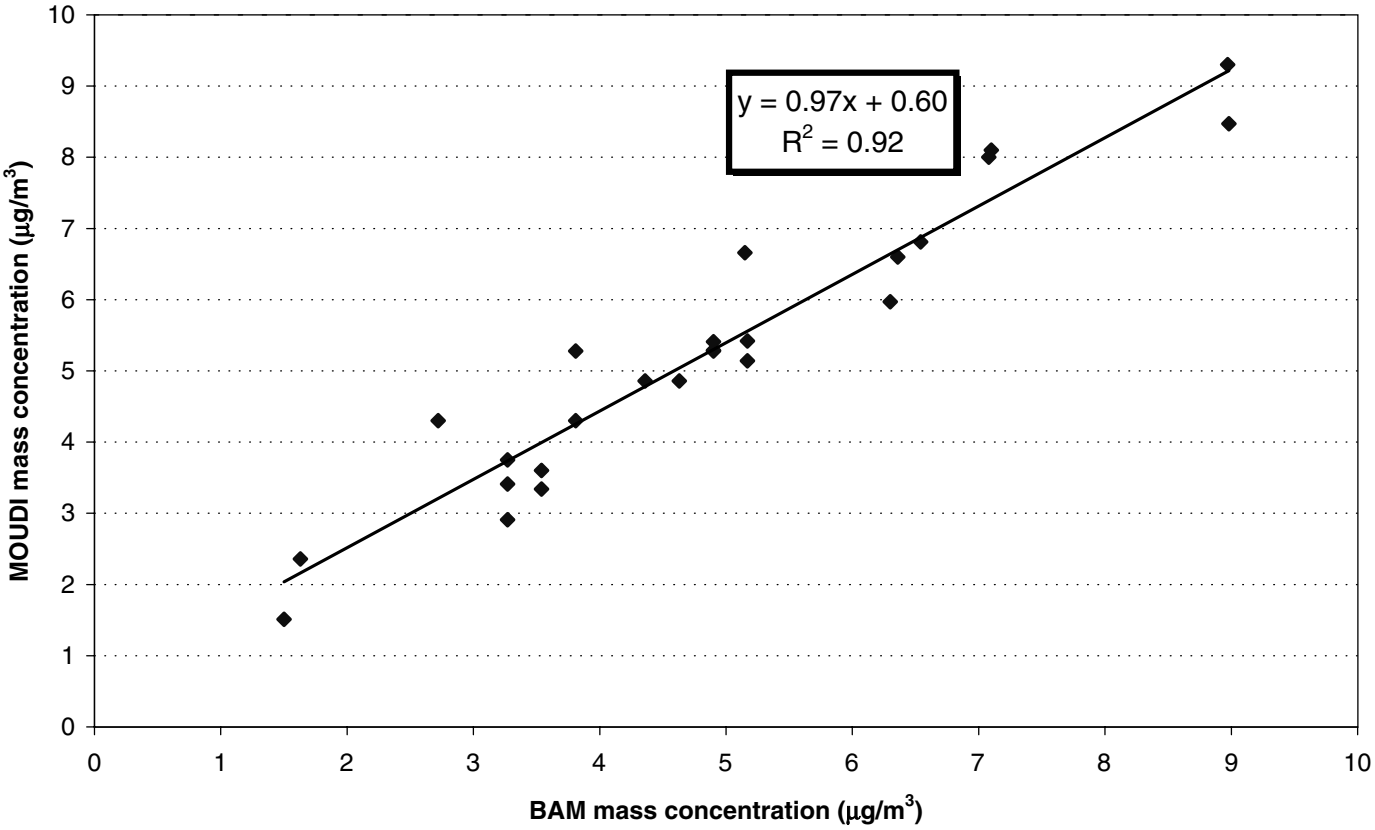


Figure 4. Plot of BAM versus MOUDI $PM_{0.15}$ concentrations in Claremont, CA.

from SMPS is always lower than those obtained from the BAM. The SMPS-to-BAM ratio resides between 0.4–0.5 from midnight to 9 am, then increases rapidly to a range of 0.6–0.8 up to about 4 pm, and then again decreases to 0.4–0.5 during the late afternoon and evening hours. The maximum value of this ratio was observed at around noon and was about 0.77 ($\pm$0.04). This distinctive diurnal profile is representative of all the days of the sampling period.

We believe that the reduced SMPS-to-BAM concentration ratio is due to the following reason. The SMPS classifies particles according to their mobility diameter, which to a first approximation depends on the surface area of the particles (Hinds 1992). When conversion from number to mass is determined, the particles typically are assumed to be perfect spheres ($\chi = 1$, $\rho_p = 1.6$ g cm^{-3}) given that no other information on the morphological properties of ambient particles is available. Previous studies have found that a significant fraction of the $PM_{0.15}$ particles in urban areas such as Los Angeles are agglomerate structures rather than spherical (Friedlander 2000; Kim et al. 2001). These particles are primarily generated from high-temperature combustion sources such as motor vehicles. By their very nature, agglomerate structures have higher surface areas than spherical particles with the same equivalent diameter and are generally less dense (Weber et al. 1995). Because of their low density, a substantial fraction of these particles would be classified by an inertial separator (such as the MOUDI or the impactor preceding the BAM) as $PM_{0.15}$, whereas the SMPS would classify these irregular particles in larger size ranges because of their high surface area, hence, mobility. Similar observations have been made in a recent study by Park et al. (2003) in which the effective density of diesel particles was measured by relating the mobility-measured diameter of combustion particles to their aerodynamic diameter. That study demonstrated that as the mobility size increases, the effective density tends to decrease, presumably because of the surface irregularities of the larger particles.

It should be expected that the relative abundance of these fractal-like agglomerates would vary, depending on the sampling location(s) as well as the time of day, in order to account for the effect of vehicular emissions. This probably explains the lower values of the SMPS-to-BAM concentration ratio during the nighttime and early morning periods, during which sub-150 nm particles originate from motor vehicle emissions. As the day progresses, the decrease in traffic reduces the contribution of vehicular emission to the sub-150 nm mass. Particles formed during the early morning traffic period grow to more homogeneous structures due to further agglomeration and condensation of vapors on their surface. Better agreement between the BAM

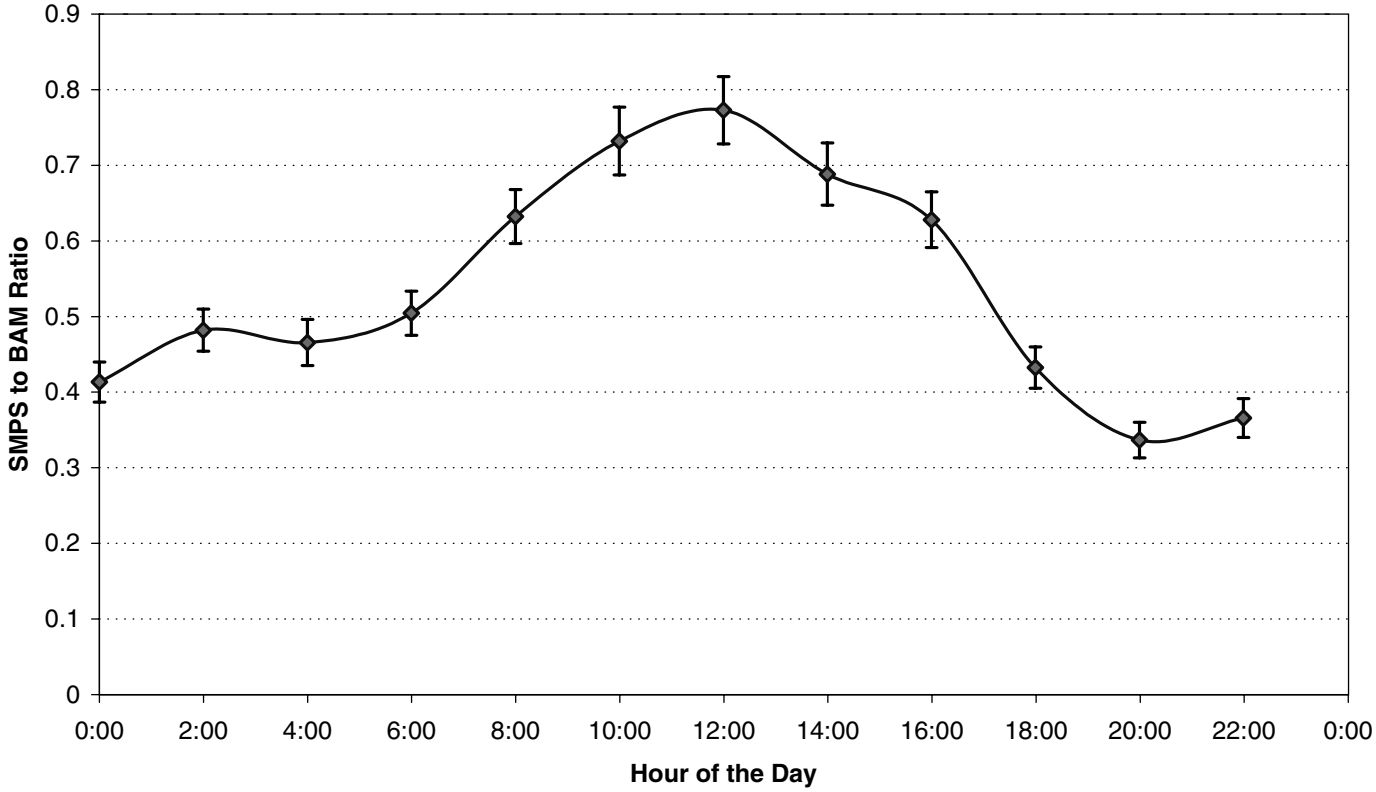


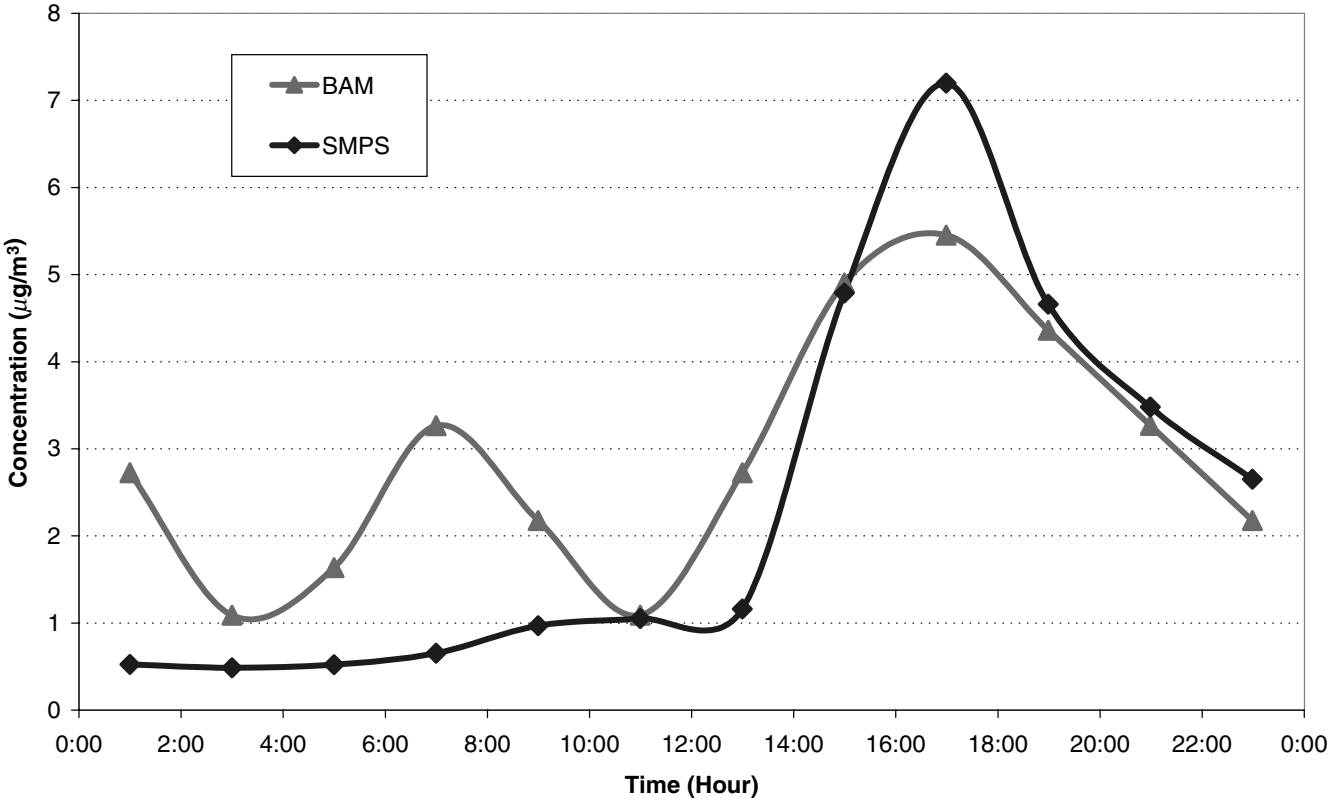
Figure 5. Average diurnal profile of SMPS-to-BAM ratio of $PM_{0.15}$ mass concentrations in Claremont, CA (07/01/2002–08/07/2002).

and SMPS $PM_{0.15}$ concentrations is thus observed during the period between 9 am to 3 pm. The SMPS-to-BAM ratio decreases again later in the afternoon and evening, presumably due to the contribution of the evening traffic to the overall sub-150 nm PM mass.

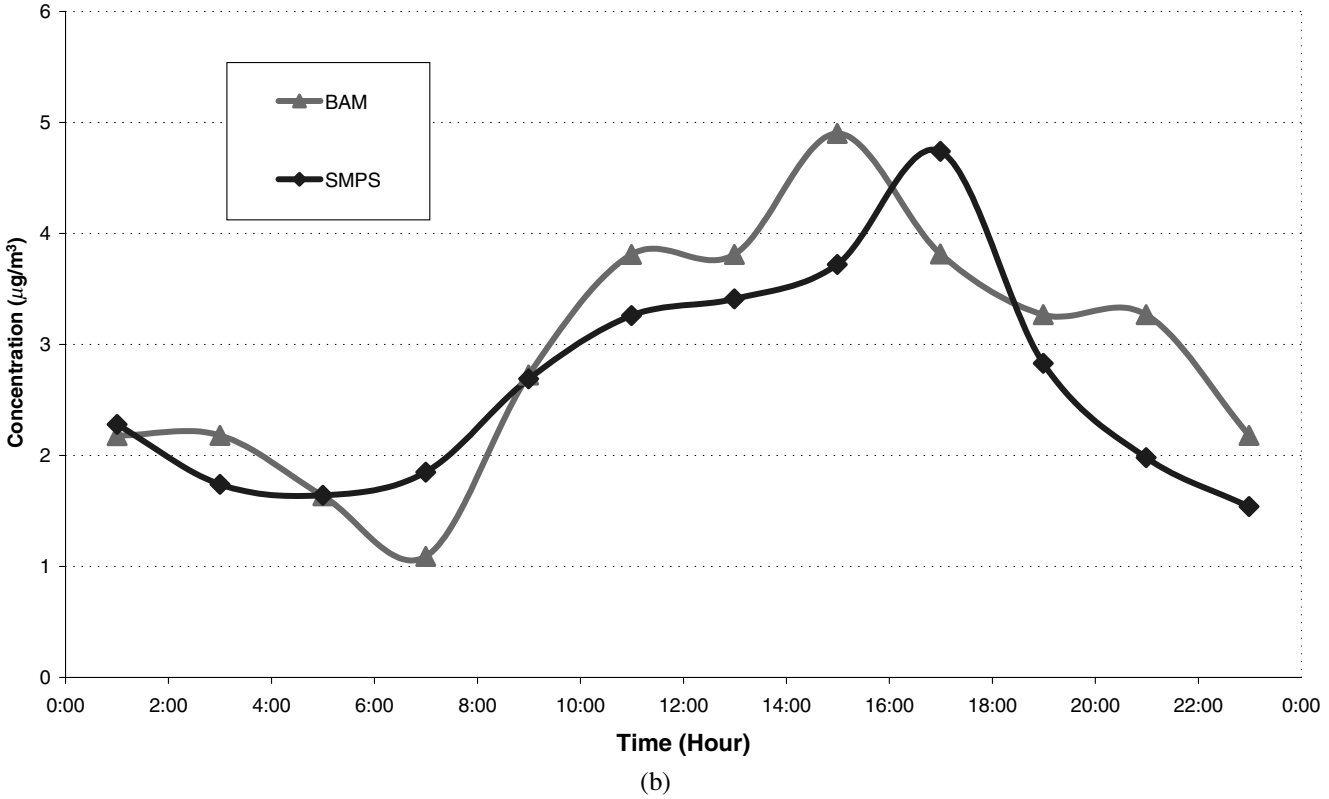
Further evidence on the dependence of the SMPS-to-BAM concentration ratio on the contribution of vehicular emissions can be seen in Figures 6a–c, showing the $PM_{0.15}$ mass concentrations obtained by these two methods for a typical weekday, weekend, and a special case (4 July 2002), respectively. Though there are differences in the mass concentration of sub-150 nm PM obtained from the SMPS and BAM monitors, these concentrations closely follow each other on 21 July 2002 (Sunday). By comparison, the SMPS concentrations do not seem to follow the increase in BAM concentrations observed during the morning traffic hours on 18 July 2002 (Thursday), as shown in Figure 6b.

The data plotted in Figure 6c (4 July 2002) are of particular interest. The unusually high mass concentration of sub-150 nm particles documented by BAM between 9 pm to midnight is likely due to the celebration of the Independence Day and the burning of fireworks. It is conceivable that the aerosol generated by fireworks may contain a high fraction of fractal-like agglomerates, similar to the diesel particles formed primarily by incomplete combustion. The increased nighttime PM emissions are almost undetected by the SMPS, while they cause a large increase in the response of the BAM. These results further corroborate the ability of the modified BAM to capture pollution peaks due to the $PM_{0.15}$ particles produced as a result of fresh emissions.

The diurnal profiles of sub-150 nm PM mass concentrations (measured by the BAM) for weekend and weekdays in Claremont for the month of July 2002 are shown in Figure 7. The plotted concentration data for each hour represent averages of the entire sampling period. The effect of the morning traffic during weekdays on $PM_{0.15}$ concentrations is evident in Figure 7, in which near-ultrafine PM concentrations are notably higher for weekdays compared to weekends. For either weekdays or weekends, however, the highest $PM_{0.15}$ concentrations are observed in the afternoon, between 2 to 5 pm, at which time these concentrations reach values in the range of 5–6 $\mu g/m^3$. The apparent similarity between the weekdays and weekends suggest that at least a substantial fraction of these particles may not originate from vehicular emissions. Previous studies have shown that secondary formation from photochemical reactions, as well as eastward advection of air parcels that originate from the urban areas of Los Angeles, are important sources of sub-150 nm PM in the inland (or receptor) areas of the Los Angeles Basin, (Pandis et al. 1992; Kim et al. 2002). Our results shown in Figure 7 appear to be consistent with these studies.

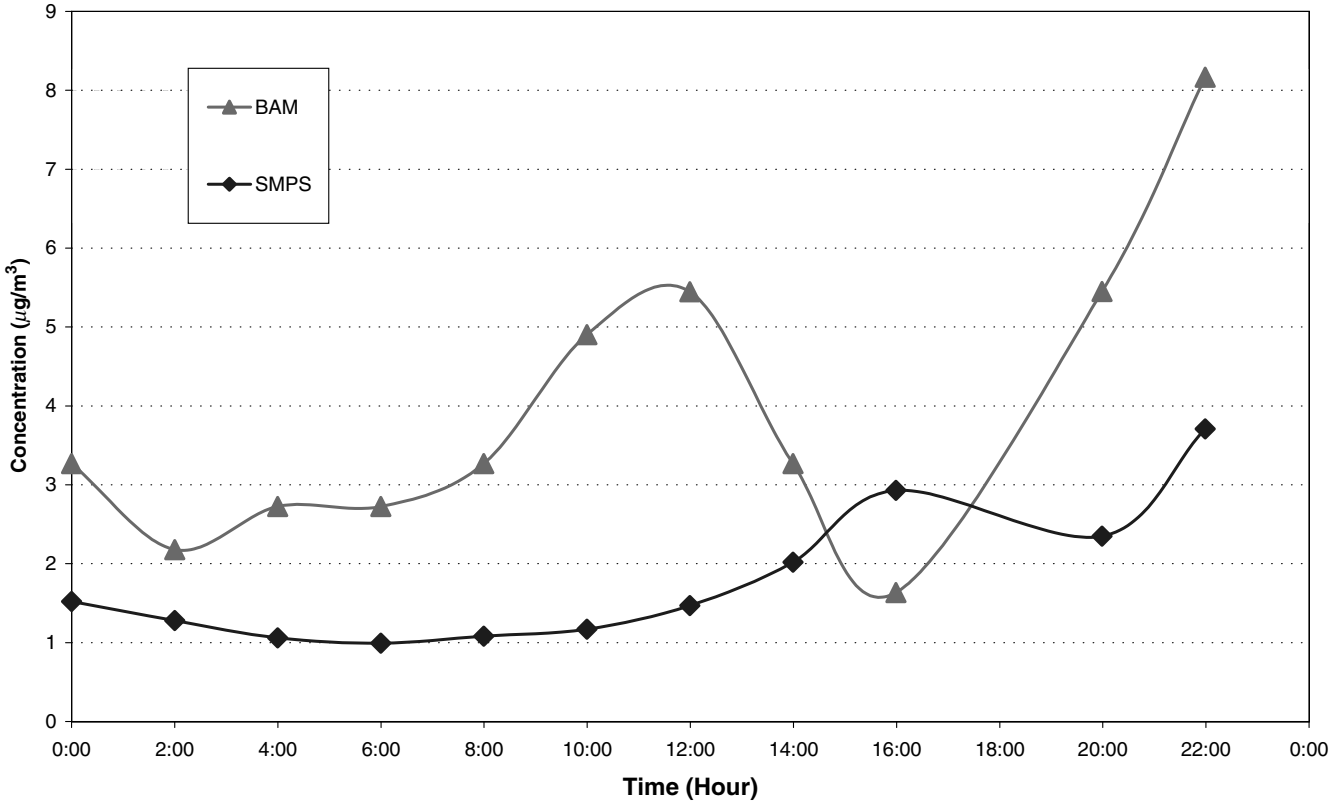


(a)



(b)

Figure 6. Combined plot of BAM and SMPS sub-150 nm mass concentrations for specific days: (a) weekend (Sunday, 21 July 2002), (b) weekday (Thursday, 18 July 2002), and (c) episodic day (Thursday, 4 July 2002—independence day). *(Continued)*



(c)

Figure 6. (Continued)

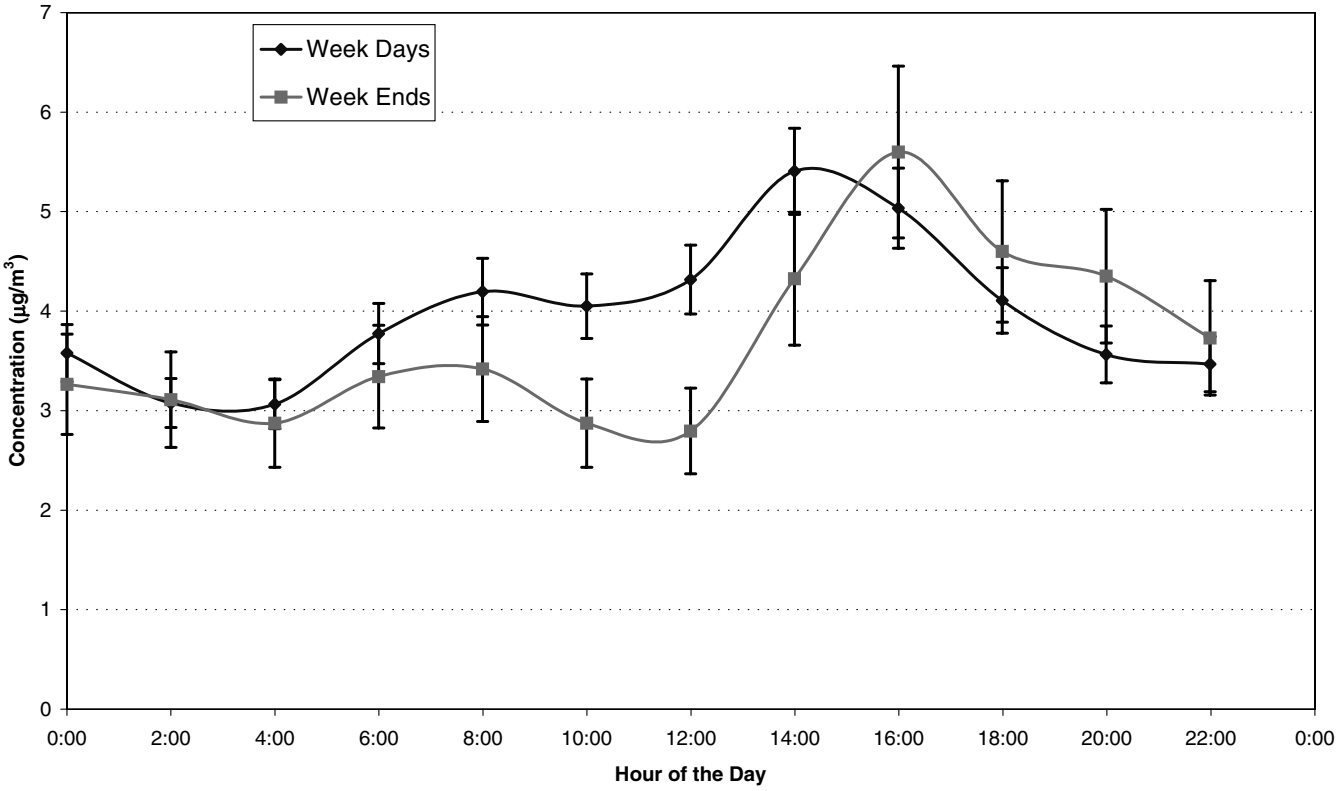
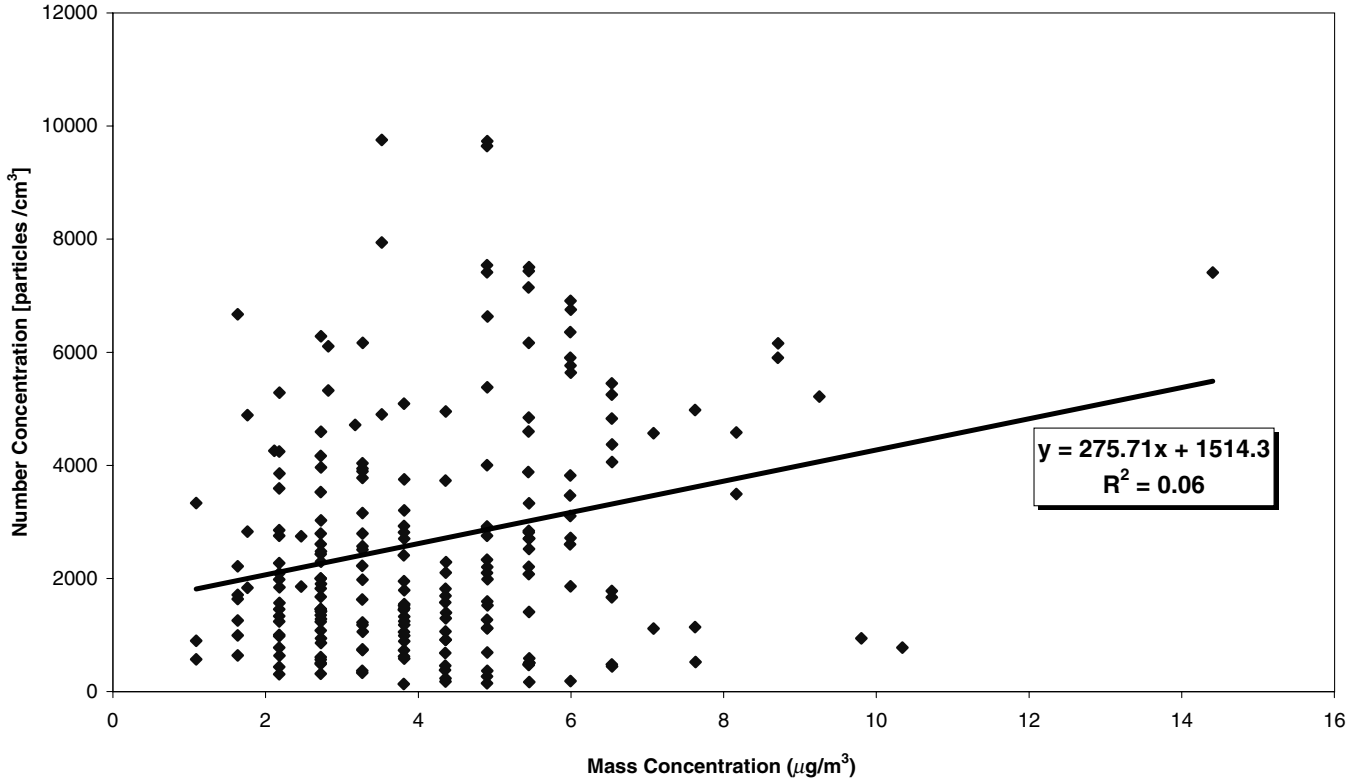


Figure 7. Average diurnal profile of sub-150 nm mass concentration from BAM for the weekdays and weekends.



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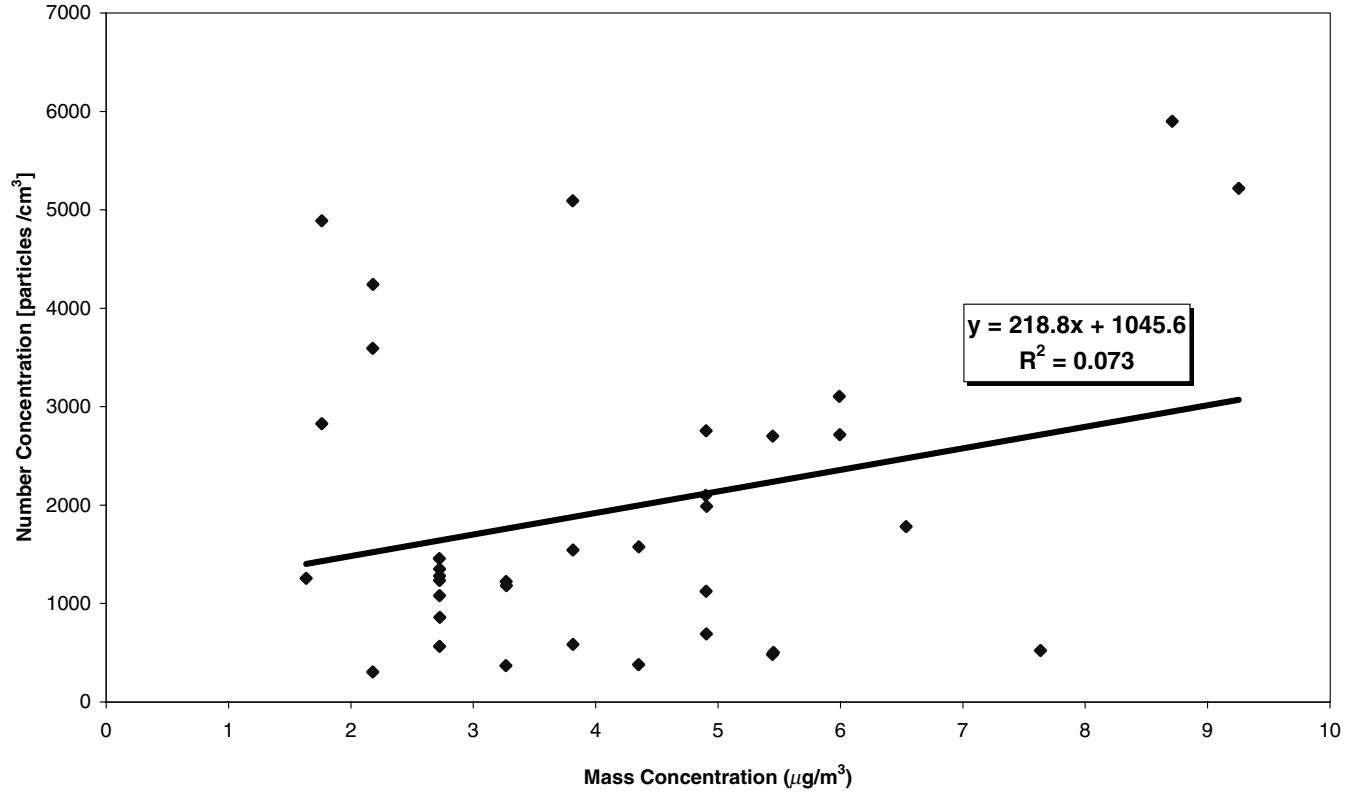
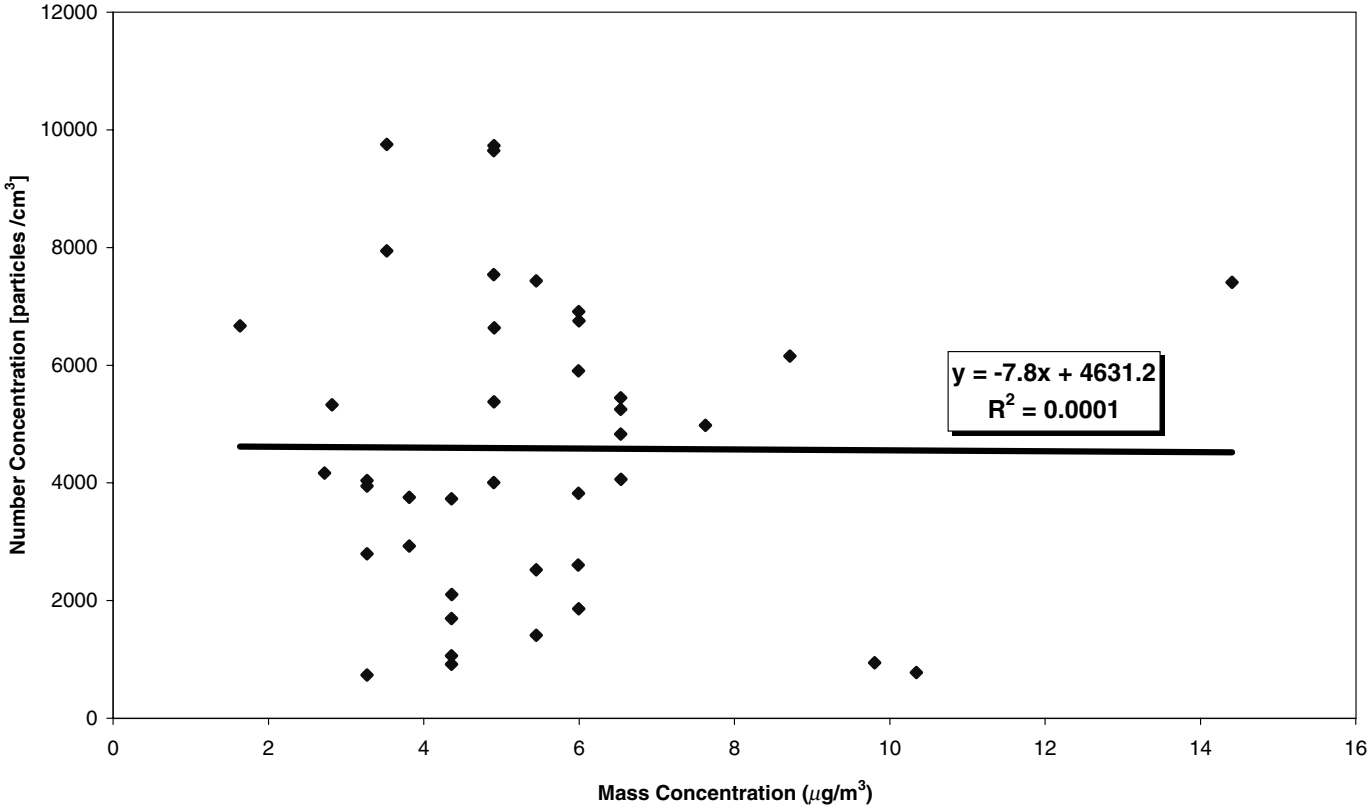


(b)

Figure 8. Correlation between number concentrations from SMPS and mass concentrations from BAM for sub-150 nm particles for (a) 24 h, (b) morning traffic hours (6–9 am), and (c) hours of photochemical reactions (2–6 pm). *(Continued)*



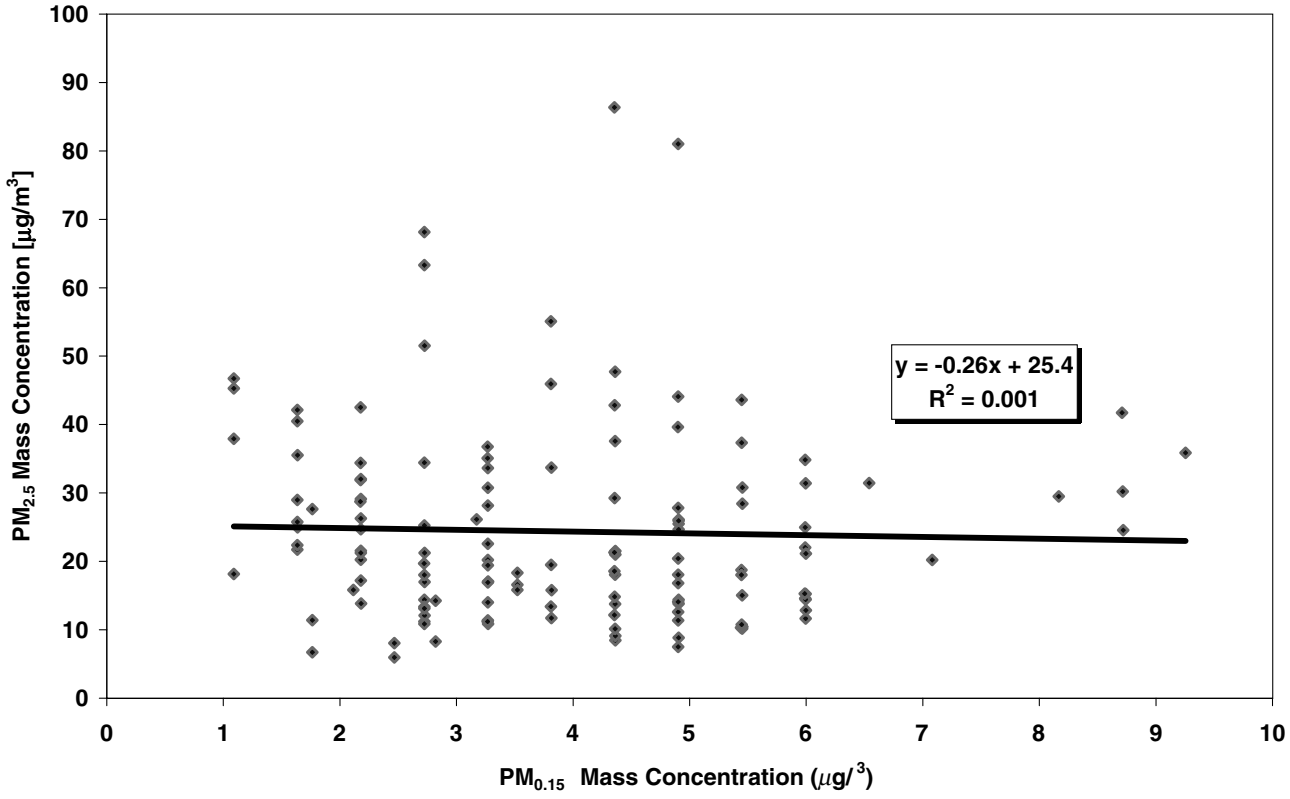
(c)

Figure 8. (Continued)

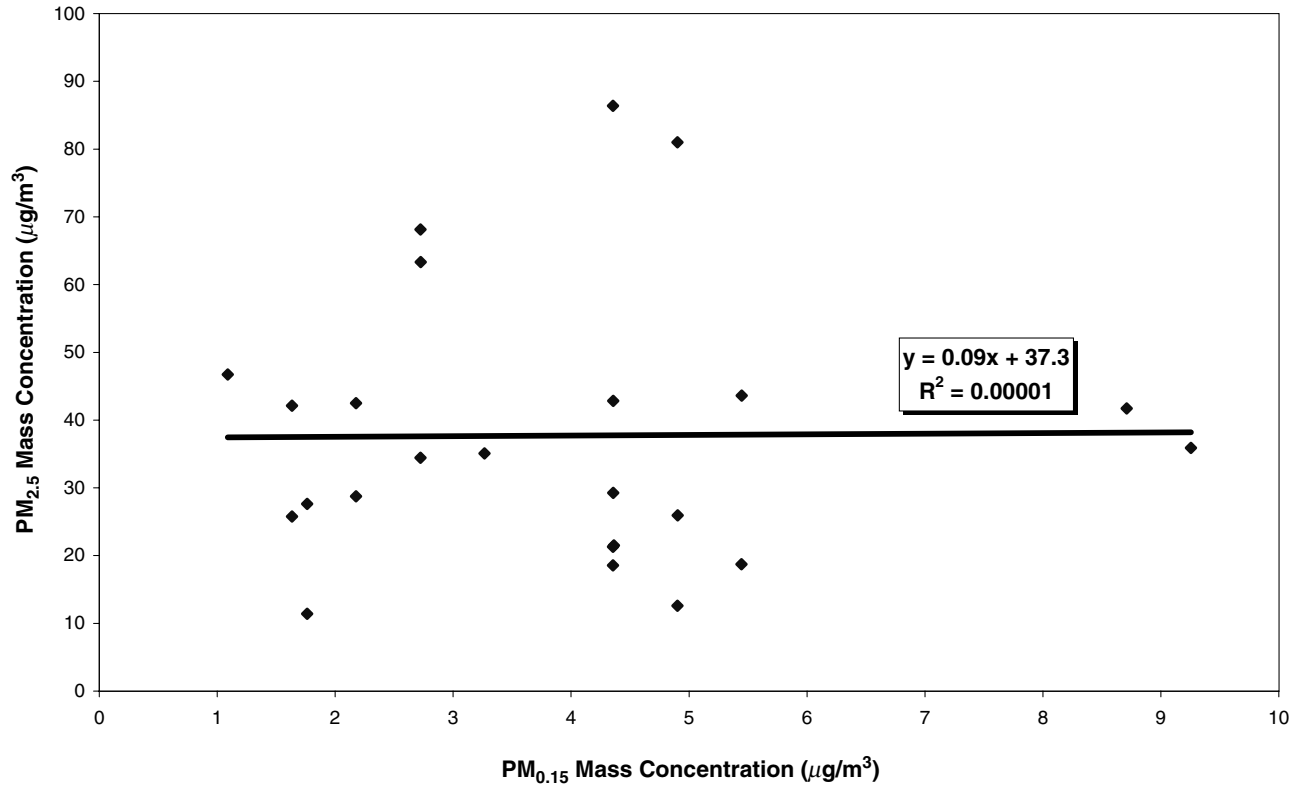
Correlations Between Sub-150 nm Particle Number, Mass, and $PM_{2.5}$ Concentrations

As part of our investigation, we examined the relationship between the total particle number concentrations (determined by integrating the number-based concentrations of the SMPS), the $PM_{0.15}$ mass concentration (determined by the BAM), and the $PM_{2.5}$ mass concentrations measured by the SMPS. Results for these investigations are shown in Figures 8a–c. Based on data generated over the period of a month, no correlation seems to exist between the number and mass concentration of sub-150 nm PM, with the R^2 value being only 0.06. Figure 8a shows the overall correlation between the SMPS number concentration (number of particles/cm^3) and BAM mass concentration (μg/m^3) for sub-150 nm PM. Two time segments of the day were chosen to further investigate whether this correlation improves during the morning hours (6–9 am) when the traffic rush is very high, and/or in the afternoon (2–6 pm) when photochemical reactions are the main PM sources in receptor areas of the Los Angeles basin, such as Claremont (Kim et al. 2001). Figures 8b and c depict these two cases and reveal that no significant correlation exists in the morning and evening hours, with R^2 values being 0.07 and 0, respectively. This lack of correlation between number and mass concentrations may have regulatory implications, because is suggests that if current or future epidemiological or toxicological studies demonstrate health effects based on mass or chemical composition of ultrafine PM, monitoring their number concentrations (as it has been proposed) may not adequately protect the public.

A similar lack of correlation was observed between the $PM_{2.5}$ mass concentrations measured by the SMPS–APS and the $PM_{0.15}$ mass concentration monitored by the BAM. Figure 9a largely indicates that $PM_{2.5}$ and $PM_{0.15}$ concentrations are independent of each other ($R^2 = 0.001$). Further investigations of the correlation between the two concentrations at any specific period of the day, for example, morning hours (6–9 am) when traffic rush is at the maximum and evening hours (2–6 pm) when the photochemical reactions play the key role in particle formation, did not improve this relationship, with R^2 values of 0.001 and 0.13, respectively (Figures 9b and c). These findings imply that $PM_{2.5}$ mass is a poor surrogate of the sub-150 nm fraction of $PM_{2.5}$, thus necessitating the need for developing continuous or near-continuous monitoring system for sub-150 nm particle mass measurements if these particles prove to be responsible for health effects.

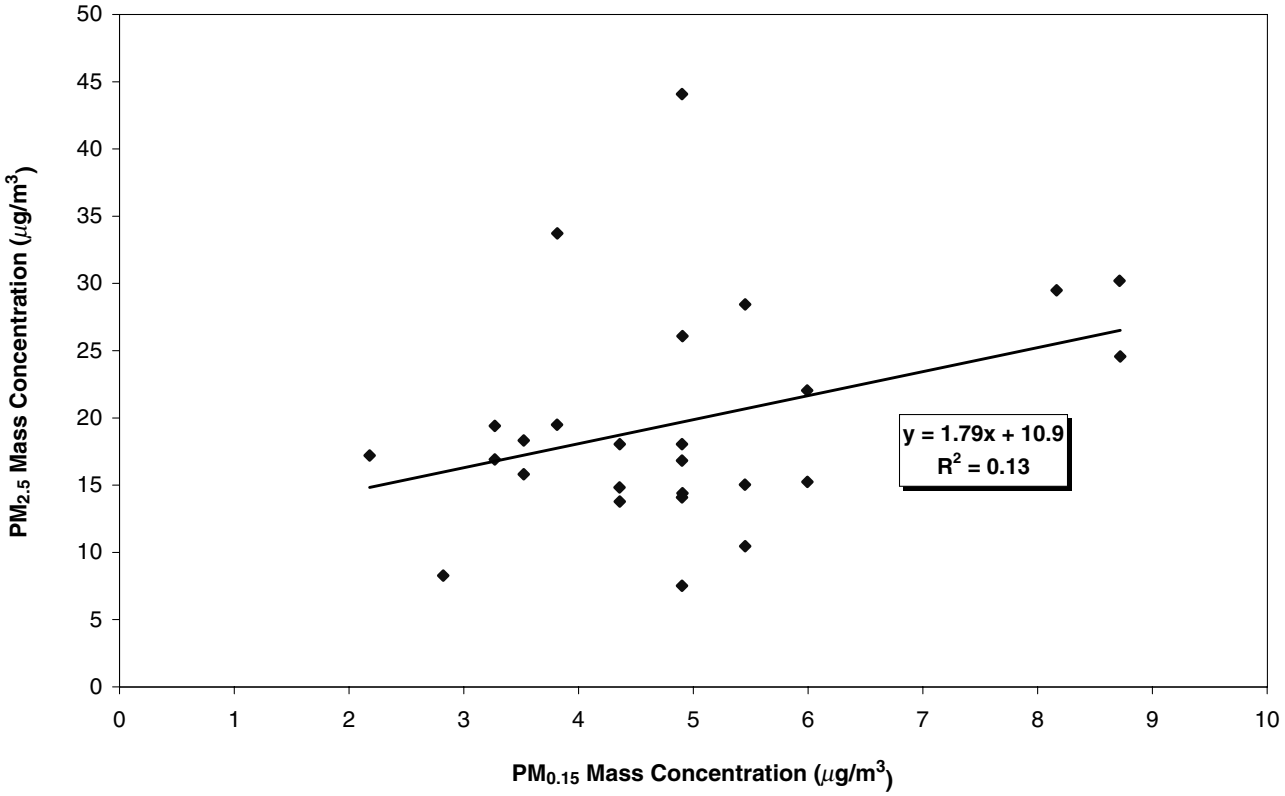


(a)



(b)

Figure 9. Correlation between $PM_{2.5}$ mass concentrations from SMPS-APS and $PM_{0.15}$ mass concentrations from BAM for (a) 24 h, (b) morning traffic hours (6–9 am), and (c) hours of photochemical reactions (2–6 pm). *(Continued)*



(c)

Figure 9. (Continued)

SUMMARY AND CONCLUSIONS

This article describes the development and field evaluation of a near-continuous monitoring system for measuring near-ultrafine (sub-150 nm) particle mass concentrations in ambient air. The BAM is preceded by a 0.15 μm cutpoint impactor, which is designed to operate at 16.7 l min^{-1} under a very low pressure drop. The laboratory evaluation of the impactor indicated sharp separation characteristics and confirmed the cutpoint at 0.15 μm.

Findings from the field study ascertain that the present BAM setup can be used to measure $PM_{0.15}$ mass concentrations reliably, based on the excellent correlation between the BAM and the MOUDI ($R^2 = 0.92$) concentrations of particles smaller than 0.15 μm in aerodynamic diameter. Our study reveals that BAM is more accurate and efficient in measuring the sub-150 nm PM mass concentration compared to the SMPS. In virtually every sampling day of our study, the morning traffic peak in sub-150 nm PM mass concentrations measured by the modified BAM was undetected by the SMPS, possibly due to the high content of fractal-like particles which tend to be classified in higher size ranges by the SMPS. The very poor correlation between the SMPS number concentrations and the BAM mass concentrations implies that further studies are necessary to determine whether the number or the mass of ultrafine PM are the best surrogates of their toxic effects. From the daily profile of the SMPS and BAM mass ratio, it can be inferred that the "effective" particle densities used for the calculation of the sub-150 nm PM mass from SMPS may vary over the course of the day. Finally, the poor correlation between the $PM_{2.5}$ and $PM_{0.15}$ mass concentrations demonstrate the need of a continuous monitoring requirement for sub-150 nm particles to ensure effective assessment of the short-term variations in their mass concentration.

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