DEVELOPMENT AND EVALUATION OF A CONTINUOUS COARSE (PM_{10} - $PM_{2.5}$) PARTICLE MONITOR

Chandan Misra, Michael D. Geller, Pranav Shah, Constantinos Sioutas*

Civil and Environmental Engineering

University of Southern California

3620 South Vermont Avenue

Los Angeles, CA 90089

Paul A. Solomon

U.S. EPA

National Exposure Research Laboratory

944 East Harmon Avenue

Las Vegas, NV 89119

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*** Author to whom correspondence should be addressed**

ABSTRACT

In this paper, we describe the development and laboratory and field evaluation of a continuous coarse (2.5 – 10 μm) particle mass (PM) monitor that can provide reliable measurements of the coarse mass (CM) concentrations in time intervals as short as 5-10 minute. The operating principle of the monitor is based on enriching CM concentrations by a factor of about 25 by means of a 2.5 μm cutpoint round nozzle virtual impactor, while maintaining fine mass, i.e., mass of $PM_{2.5}$ (FM) at ambient concentrations. The aerosol mixture is subsequently drawn through a standard TEOM™, the response of which is dominated by the contributions of the CM, due to concentration enrichment. Findings from the field study ascertain that a TEOM™ coupled with a PM_{10} inlet followed by a 2.5 μm cutpoint round nozzle virtual impactor can be used successfully for continuous CM concentration measurements. The average concentration-enriched CM concentrations measured by the TEOM™ were approximately 26-27 times higher than those measured by the time-integrated PM_{10} samplers (MOUDI™ and Partisol™ sampler), and highly correlated. CM concentrations measured by the concentration-enriched TEOM™ were independent of the ambient FM-to-CM concentration ratio, due to the decrease in ambient coarse particle mass median diameter (MMD) with an increasing FM-to-CM concentration ratio. Finally, our results illustrate one of the main problems associated with the use of real impactors to sample particles at relative humidity (RH) values lower than 40%. While PM_{10} concentrations obtained by means of the MOUDI™ and Partisol were in excellent agreement, CM concentrations measured by the MOUDI™ were low by 20%, while FM concentrations were high by a factor of 5, together suggesting particle bounce at low RH.

IMPLICATIONS

Several researchers have raised the issue of the quality of CM concentrations data used in PM exposure assessment and epidemiological studies. Poor CM precision could lead to potential biases in exposure-health effect models that include both FM and CM exposure variables, and make it more difficult to properly assess the spatial correlations of CM over metropolitan areas. Since these issues may be important in evaluating the health effects of CM relative to PM_{10} or $PM_{2.5}$, it is desirable to have CM measurements that are sufficiently precise to resolve the uncertainty surrounding existing PM studies that include CM data. This paper describes the development and performance evaluation of a CM monitor that can provide reliable measurements in time intervals as short as 5 minutes. The simplicity and reliability of this monitor makes it ideal for use in large scale monitoring networks.

INTRODUCTION

Ambient particles in the size range 2.5 to 10 μm are referred to as coarse particles or coarse mode (CM) aerosols. Coarse particles may consist of several potentially toxic components, such as resuspended particulate matter from paved and unpaved roads, industrial materials, brake linings, tire residues, trace metals, and bioaerosols. Since a considerable fraction of these particles may deposit in the upper airways and to a lesser extent into the lower airways, they may be responsible for the exacerbation of asthma. Recent data from a small number of epidemiological studies indicate that, apart from--or in addition to—the fine fraction (FM) of particulate matter (also called $PM_{2.5}$), health effects also may be closely associated with the CM fraction and sometimes even to a larger extent than FM [1-3]. In vitro studies with human monocytes show that cellular toxicity and inflammation also may be associated with the CM and its biological components [4-6].

Several researchers have raised the issue of the quality of CM concentrations data used in PM exposure assessment and epidemiological studies[7-9]. These researchers state that poor CM precision could lead to potential biases in exposure-health effect models that include both FM and CM exposure variables, and make it more difficult to properly assess the spatial correlations of CM over metropolitan areas. Since these issues may be important in evaluating the health effects of CM relative to PM_{10} or $PM_{2.5}$, it is desirable to have CM measurements that are sufficiently precise to resolve the uncertainty surrounding existing PM studies that include CM data.

According to the Federal Reference Method (FRM), current measurements of both the PM_{10} and $PM_{2.5}$ mass concentrations are based on gravimetric analysis of

particles collected on filters over a period of 24 hours (Federal Register [10]). Gravimetric analysis was selected because most of the particle data used for the epidemiological studies investigating associations between mortality and morbidity outcomes and ambient particle exposures are based on PM concentrations [11,12]. Typically, a time-integrated sample (e.g., over 24 hours) is collected on the filter, which is later equilibrated at designated temperature and RH conditions, and subsequently weighed to determine the mass of the deposited PM. Dividing by the amount of air sample yields the atmospheric concentration. Since the values of atmospheric parameters influencing ambient particle concentration, hence human exposure, such as the emission strengths of particle sources, temperature, RH, wind direction and speed and, mixing height, fluctuate in time scales that are substantially shorter than 24 hours, a 24-hour measurement may not reflect an accurate representation of human exposure. Thus, more accurate, better quality data on the physico-chemical characteristics of particles are needed to understand their atmospheric properties and health effects.

Methods that are capable of providing continuous or near continuous measurements (i.e. 1-hour average or less) are highly desirable because they can provide accurate information on human exposure and atmospheric processes in short timer intervals. Over the past decade, a significant number of state-of-the-art methods were developed for continuous PM_{10} and $PM_{2.5}$ mass concentration measurements. These include the Tapered Element Oscillating Microbalance (TEOM™ 1400A; Rupprecht and Patashnick, Albany NY), a host of nephelometers, such as the DataRAM™ (RAM-1, MIE Inc., Billerica, MA), and the DUSTTRACT™ (Model 8520, TSI Inc., St. Paul, MN), and the Continuous Ambient Mass Monitor[13] (CAMM™, Thermo Andersen, Smyrna,

GA). The latter method can only provide measurements of FM. Mass concentration measurements using photometers or nephelometers are based on light scattering, and are dependent on particle size and chemical composition [14, 15, 16] showed that variations in particle size and chemical composition may introduce considerable errors in predicting the response of nephelometers such as the DataRAM.

The TEOM™ measures either PM_{10} or $PM_{2.5}$ (but not directly CM) by recording the decrease in the oscillation frequency of a particle-collecting element due to the increase in its mass associated with the depositing particles. In its standard configuration, the TEOM™ collects particles at a flow rate of 2-4 liter per minute (lpm) on an oscillating filter heated to 50 °C. The TEOM™ filter is heated to eliminate interferences from changes in RH that can change the amount of particle-bound water associated with the collected PM [17]. Determining CM concentrations by difference, as currently proposed by EPA [18] introduces significant uncertainties in cases where FM account for a large fraction of the PM_{10}. Moreover, since much of the semi-volatile particulate matter (which is mostly associated with FM) is expected to be lost from the TEOM™ filter during and after collection at 50 °C, there is the potential for a substantially different measurement of PM_{10} mass between the TEOM™ and FRM. This is most likely to occur in urban areas (or areas affected by urban plumes) where volatile compounds, such as ammonium nitrate and organic compounds can comprise a substantial fraction of the FM. Heating is not likely to affect the mostly non-volatile constituents of coarse particles, thus the accuracy of CM concentrations determined as the difference between PM_{10} and $PM_{2.5}$ will be compromised by the generally random loss of volatile compounds from FM.

In theory, continuous measurements of CM concentrations also could be conducted by means of optical, electrical, and time-of-flight monitors. These monitors measure size-resolved particle concentrations based on particle numbers, which could be subsequently converted to volume concentrations assuming spherical particles and an assumption about particle density; both assumptions are required to convert particle volume to mass concentrations. As in most air sampling applications, information on particle density is generally not available and assumptions about its value will introduce uncertainties in the resulting mass concentrations estimates. A far more important limitation of the aforementioned particle number-based monitors results from the sharply decreasing number of ambient particles with increasing particle size. The ambient particle size distribution, by number, is dominated by ultrafine particles (i.e., smaller than 0.1 μm). As well, when converting a number to volume distribution, a 1.0 μm particle weighs as much as 10^3 times a 0.1 μm particle and 10^6 times a 0.01 μm particle. Consequently, counting errors associated with this conversion, which may be substantial for large particles, due to their relatively low numbers combined with electronic noise, may lead to significant uncertainties in volume and consequently mass as a function of particle size. This was demonstrated in a recent study by Sioutas et al [19], which showed that the mass concentrations obtained with the Scanning Mobility Particle Sizer/Aerodynamic Particle Sizer system (SMPS, Mode 3936, TSI Inc., St. Paul, MN; APS, Model 3320, TSI Inc., St. Paul, MN) were higher by 70-200% than those determined with a reference gravimetric method.

In this paper, we describe the development and laboratory and field evaluation of a Continuous Coarse Particle Monitor (CCPM) that can provide reliable measurements of

the CM concentrations in time intervals as short as 5-10 minute. The operating principle of the monitor is based on enriching the CM concentrations by a factor of about 25 while maintaining FM at ambient concentrations. The aerosol mixture is subsequently drawn through a standard TEOM™, the response of which is dominated by the contributions of the CM due to concentration enrichment. This paper also presents a comparison between the CM and FM concentrations obtained different time-integrated samplers (i.e., filters and impactors), which was conducted during the field evaluation study of the CCPM.

METHODS

Description of the Continuous Coarse Particle Monitor

The CCPM, shown schematically in Figure 1, operates at an intake flow of 50 lpm, and consists of three main components: a) a PM_{10} inlet; b) a 2.5 μm cutpoint round nozzle virtual impactor (or, coarse particle concentrator), and; c) TEOM™.

Particles are drawn at 50 lpm through a circular nozzle, 1.1 cm inside diameter, attached to a 90^{o} aluminum duct elbow, 3.2 cm in diameter. The nozzle protrudes 3 cm from the rest of the inlet section of the continuous monitor and extends up to a distance of 1.5 cm from the inside wall of the 90o elbow, as shown in Figure 1. The nozzle has been designed with a cutpoint of approximately 10 μm aerodynamic diameter (AD). During the field tests, a thin layer (approximately 1 mm) of silicon grease (Chemplex ™ 710, NFO Technologies, Kansas City, KS) was applied periodically to the inside wall of the elbow to prevent particle bounce.

The collection efficiency of the PM_{10} inlet was evaluated in field tests by measuring the mass-based concentrations of ambient particles in the 2.5 to 20 μm range

by means of an APS™. For these tests, the TEOM™ was disconnected from the virtual impactor and the minor flow was drawn directly the APS™. The sampling flow of the APS™ is 5 lpm, thus higher than the minor flow of the CCPM (2 lpm). Since the cutpoint of the PM_{10} inlet does not depend on the minor-to-total flow of the virtual impactor but on the total aerosol flow entering the impactor-inlet, the major flow of the virtual impactor was adjusted to 45 lpm in order to maintain the total flow entering the PM_{10} inlet and virtual impactor at 50 lpm.

The concentration of particles in the 2.5 to 20 μm (enriched by a factor of approximately 10) was obtained for a sampling period of 3 minutes. Subsequently, the PM_{10} inlet was removed and the mass-based concentration of 2.5 to 20 μm particles was obtained for a period of 3 minutes. The above test sequence was repeated five times. Particle penetration through the PM_{10} inlet was determined for each size by dividing the average concentration (based on five tests) obtained with the PM_{10} inlet connected to the sampler to the concentration without the inlet. The wind speed (a crucial parameter in for the performance evaluation of the inlet) was recorded during these experiments and varied from 1 to 7 miles per hour (mph), which is a typical range for Los Angeles.

Particles smaller than 10 μm in AD are drawn through the virtual impactor, which was designed to have a theoretical 50% cut point at about 2.5 μm for an intake flow rate of 50 lpm. This is single-stage, round-jet nozzle virtual impactor with an acceleration nozzle diameter of 0.37 cm and collection nozzle diameter of 0.56 cm. The distance between the acceleration and collection nozzles is 0.7 cm.

The flow field in a virtual impactor is determined by the Reynolds number, which, is defined as:

$$\mathrm{Re} = \frac{U\,W\,\rho}{\mu} \qquad (1)$$

where U is the average jet velocity through the acceleration nozzle of the impactor, W is the diameter of that nozzle, and μ and ρ are the dynamic viscosity and density of air, respectively. The value of Re corresponding to the operating configuration of the virtual impactor is 18,927. Coarse particles follow the minor (concentrated) flow, while particles smaller than the cutpoint of the virtual impactor follow the major flow. The minor flow in these experiments was set at 2 lpm to achieve a nominal enrichment factor of 25. Concentrated CM, including a small fraction of FM (about 4%) are drawn through the TEOM, whose flow was adjusted to 2 lpm. In its most common configuration, the aerosol is heated to 50 °C before collection on the TEOM™ filter, which is attached to the oscillating element. Our experiments were performed at sample temperatures of 50 ^{o}C and 30 ^{o}C to determine whether differences in these temperatures would result in significant differences in the response of the CCPM. While the standard configuration of the TEOM™ is to operate it at 50 ^{o}C, due to loss of semi-volatile species at this temperature, many TEOMs are being operated at 30 ^{o}C with a nafion dryer to remove water vapor prior to the collection substrate. No nafion dryer was used in our configuration. The remaining 48 lpm (major flow) through the virtual impactor is drawn through a separate, lightweight, rotary vane pump (Gast, Model 1023, Gast Mfg. Corp., Benton Harbor, MI). The pressure drops across the major and minor flows of the virtual impactor are 5.8 and 0.25 kPa, respectively.

Laboratory Evaluation of 2.5 μm Cutpoint Round Nozzle Virtual Impactor

The first series of experiments were conducted in the laboratory to investigate the relationship between the concentration enrichment achieved by the 2.5 μm cutpoint round nozzle virtual impactor as a function of particle size. Briefly, monodisperse aerosols in the size range of 1 to 10 μm were generated by atomizing dilute aqueous suspensions of fluorescent polystyrene latex particles (Polysciences Inc., Warrington, PA) with a constant output nebulizer (HEART™, VORTRAN Medical Technology, Inc., Sacramento, CA). The generated particles were mixed with dry room air in a 1-liter bottle to remove the excess moisture . The dry aerosol was then drawn through a tube containing ten Po-210 neutralizers that reduced particle charges prior to entering the virtual impactor. For each of the monodisperse particles in the range of 1 to 5 μm, the DataRAM was used to first measure the mass concentration of the generated aerosols prior to entering the 90° elbow of virtual impactor. The DataRAM was subsequently connected downstream of the minor flow of the virtual impactor to measure the mass concentration of the aerosols after concentration enrichment. The measurements were repeated at least three times, and the average concentration enrichment was determined as a function of particle size. The contributions from background ambient concentrations before and after the enrichment were recorded and subtracted from those of the input and concentrated aerosols prior to determining the collection efficiencies at the given particle size. It should be noted that indoor air levels were on the order of 7 – 15 $\mu g.m^{-3}$, and substantially smaller than those of the generated aerosols (prior to concentration enrichment), which varied from 170 to about 500 $\mu g.m^{-3}$. Therefore the contributions of

the indoor aerosol to the overall concentrations measured upstream of- and in the minor flow of the virtual impactor were considered negligible.

Concentration enrichment for 5 to 10 μm particles was determined by comparing the mass collected on a glass fiber filter (2 μm pore, Gelman Science, Ann Arbor, MI) connected to the minor flow of the virtual impactor, and the mass of a similar glass fiber filter in parallel to the test system to measure the concentration of the monodisperse aerosol. The filter sampling in parallel was connected to a pump operating at 30 lpm. At the end of each run, each glass fiber filter was placed in 5 ml of ethyl acetate to extract the fluorescent dye from the collected particles. The quantities of the fluorescent dye in the extraction solutions were measured by a Fluorescence Detector (FD-500, GTI, Concord, MA) to determine particle concentration. Concentration enrichment for each particle size was defined as the ratio of the concentration measured in the minor flow to that of the aerosol immediately upstream of the virtual impactor inlet.

Field Study

Following the completion of the laboratory experiments, the performance of the CCPM was evaluated in a field study which was part of the Los Angeles Supersite project at the Rancho Los Amigos National Rehabilitation Center in Downey, CA. Situated near the Los Angeles "Alameda corridor", Downey has some of the highest inhalable PM_{10} concentrations in the US, very often exceeding the 24-hour National Ambient Air Quality Standard for PM_{10} of 150 $\mu g.m^{-3}$. The field experiments were performed during the period of October to December 2000.

Concentrated CM were provided directly to the TEOM™ from the minor flow (2 lpm) of the 2.5 μm cutpoint round nozzle virtual impactor. Measurements of concentration-enriched CM measured by the TEOM™ were compared to direct measurements with a co-located Microorifice Uniform Deposit Impactor (MOUDI™, MSP Corp. Minneapolis, MN) and Dichotomous Partisol-Plus™ (Model 2025 Sequential Air Sampler, Rupprecht and Patashnick Co. Inc., Albany, NY). The MOUDI™ sampled at 30 lpm. Instead of using all available MOUDI™ stages , only those having cut-points of 10 μm and 2.5 μm were used. Thus the first MOUDI™ stage (2.5-10 μm) was used as a reference sampler for CM concentrations and the last stage (i.e., the after-filter) was used to determine the ambient FM concentrations. Teflon filters with diameters of 4.7 and 3.7 cm (2 μm pore size, Gelman Science, Ann Arbor, MI) were used to collect CM and FM in the two MOUDI™ stages, respectively.

The Partisol™ uses a PM_{10} inlet operating at 16.7 lpm to remove particles larger than 10 μm in AD. The remaining PM_{10} aerosol is drawn through a virtual impactor, or, "dichotomous splitter", located after the inlet. Two separate flow controllers maintain the CM at 1.67 lpm and the FM stream at 15 lpm. CM and FM are collected on two 4.7cm Teflon filters, placed in the minor and major flows of the Partisol virtual impactor, which are housed in reusable cassettes.

The Teflon filters of both MOUDI™ and Partisol™ samplers were pre- and post-weighed using a Mettler Microbalance (MT5, Mettler-Toledo, Inc, Hightstown, NJ) after 24-hour equilibration under controlled humidity (35-40%) and temperature (22-24 ^{o}C).

The experiments were performed with simultaneous sampling from the TEOM™ and the MOUDI™ and/or the Partisol™. The sampling time varied from 90-minute to 210

minute depending on the ambient concentrations to allow sufficient mass to be collected on the time-integrated samplers. The majority of the experiments were for sampling periods of 120-minute. The volume concentration of ambient CM also was recorded in 15-minute intervals using an APS™ for a number of experiments. In addition, in selected experiments, the time-weighed mass median diameter (MMD) of the ambient coarse particles was determined by means of the APS™. Temperature and RH data, for each experiment were also measured continuously by the Partisol™ and recorded automatically by the systems software. The mass concentration of the CCPM was determined both by the 1- or 2-hour time integrated TEOM™ readings and by directly dividing the mass deposited on the TEOM™ filter by the total air volume sampled. In all experiments, these two concentrations differed by less than 5%. CM and FM concentrations of the MOUDI™ were determined by dividing the total PM collected on the MOUDI™ substrates by the total sampled air volume. The CM concentration of Partisol was determined after dividing by the appropriate sample flow and subtracting 10% of FM concentration from it, which corresponded to the ratio minor flow to the total flow of the Partisol™l virtual impactor.

RESULTS AND DISCUSSION

Evaluation of the PM_{10} Inlet

Particle penetration values through the PM_{10} are plotted as a function of AD in Figure 2. The data plotted in this figure indicate that particle penetration is 90% or higher for particles in the range of 2.5 to 8 μm. Penetration decreases sharply to about 50% at 10 μm and further to less than 10% for particles larger than 12 μm in AD. The sharpness of

the particle penetration 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 particle AD corresponding to 16% penetration to that corresponding to 84 % penetration [20]. Based on this definition, the value of σ_g is approximately 1.2 (roughly the ratio of 11 μm / 8 μm) for the PM_{10} inlet, thereby indicating reasonably sharp aerodynamic particle separation characteristics.

Laboratory Evaluation of the 2.5 μm Cutpoint Round Nozzle Virtual Impactor

Figure 3 presents the concentration enrichment of the 2.5 μm cutpoint round nozzle virtual impactor as a function of particle AD. The data in Figure 3 confirm the rise of the enrichment factor as a function of particle AD. As seen from the figure, the enrichment factor increases sharply up to its ideal value of 25, as predicted based upon the intake and minor flow rates of 50 and 2 lpm, respectively. The plotted data correspond to the averages of at least three experiments per particle size, whereas the error bars represent the standard deviation in the enrichment values. The concentration enrichment factor increases sharply from about 2 to 23 as particle AD increases from 2 to 3 μm. The enrichment factor is practically the same for particles in the AD range of 3 to 9 μm. The data shown in Figure 3 also indicate that the 50% cut point of the virtual impactor, defined as the aerodynamic particle size at which the enrichment factor is half of its ideal value (i.e. about 12.5) is approximately 2.4 μm. [The enrichment factor measured at 2.5 μm is about 15]. The overall high concentration efficiencies of 9 μm particles, proves that there are no significant losses of these particles in the 90° elbow of the PM_{10} inlet. More importantly, these tests imply that the size distribution of concentrated CM before

entering the TEOM is the same as that of the ambient air, since the concentration enrichment factor does not depend on particle size—at least for particles larger than 2.5 μm in AD.

Field Evaluation of the Continuous Coarse Particle Monitor

The results of the field evaluation of the CCPM are shown in Figures 4 to 7 for experiments performed at a TEOM™ temperature of 50 ^{o}C. Figure 4 shows the comparison between the TEOM™ and MOUDI™ CM concentrations at 50 ^{o}C. As indicated, the data are highly correlated (R^2=0.88) with a slope of 25 and a near zero intercept. The ratio of concentrations equal to 26.1 (± 3.6) also is close to the expected value.. Figure 5 shows the comparison between the TEOM™ and Partisol™ CM concentrations at 50 ^{o}C. Again, these datas are highly correlated (R^2=0.88) with a slope of 24 and a near zero intercept. The ratio of concentrations equal to 25.8 (± 4.1) also is close to the expected value. It is worthwhile noting, that the TEOM™ concentrations are not corrected for the contributions of the FM, which is present in the inlet stream. The purpose of concentrating the CM by a factor of 25 is to eliminate the need for knowing *a priori* the FM concentration. Ideally, the mass concentrations measured by the CCPM are related to the actual ambient CM concentrations as follows:

$$CCPM = 25\ CM + FM \qquad (2)$$

Thus a 1:1 FM-to-CM concentration ratio would result in the CCPM being 26 times higher than the actual CM concentration.

An important implication of equation (2) is that unusually high (but not impossible) FM-to-CM concentration ratios (i.e., 4 - 6) would lead to a positive bias (or overestimation) of the CM concentration by the CCPM, if the concentrations are not corrected to account for the contribution of FM. To investigate the effect of the FM-to-CM concentration ratio on the response of the CCPM, the ratio of the concentration-enriched TEOM™-to-MOUDI™ and TEOM™-to-Partisol™ concentrations were plotted as a function of the FM-to-CM concentration ratio. The results, shown in Figure 6, clearly indicate that the ratio of TEOM™-to-MOUDI™ CM concentration and the ratio of TEOM™-to-Partisol™ CM concentration are, under the conditions of this experiment independent of the ratio of ambient FM-to-CM concentrations. (R^2= 0.0064). This independence can be further explained by the data plotted in Figure 7, which shows the decrease in the ambient MMD (determined by the APS) as the FM-to-CM concentration ratio increases. There is a marked shift in MMD from 4.8 – 5 μm to 2.8 – 3 μm as the ratio of FM-to-CM concentration increases from 1 to 5 respectively. The highest values of FM-to-CM concentrations, ranging from about 3.5 to 4.6, were obtained on October 20 and 21, 2000. During these two days, stagnation conditions occurred in Downey, with the average wind speed during the sampling periods being less than 1 miles per hour (mph). 2-hour averaged FM concentrations measured by either the MOUDI or Partisol™ during these two days ranged from 80 to 146 $g.m^{-3}$. These conditions are expected to result in high FM concentrations in locations such as Downey, which is primarily impacted by vehicular emissions from nearby freeways, while the relatively low CM concentrations may be explained by the lack of sufficient wind velocity to either generate or transport coarse particles. As the virtual impactor-particle concentrator

preceding the TEOM™ has a 50% cutpoint at about 2.5 μm, particles in the 2.5 – 3 μm AD range would be concentrated somewhat less efficiently than those larger than 3 μm. For example, the laboratory evaluation of the 2.5 μm cutpoint virtual impactor (Figure 3) indicated that 2.5 to 3 μm particles are concentrated by a factor ranging from 16 to 22, compared to particles in the 3 – 10 μm range that are concentrated by a factor of 25. This slightly uneven concentration enrichment, combined with the intrinsic relationship between the coarse particle MMD and the FM-to-CM concentrations ratio, brings the CCPM-to-CM concentration ratio closer to the range of 25-26, and thus, compensates for the increase in the FM-to-CM concentration ratio. As a result, the CCPM can be used efficiently for measuring the ambient CM concentrations even in cases where the ratio of FM-to-CM concentration is unusually high.

The results of the field experiments conducted at a TEOM™ temperature at 30 ^{o}C are presented in Figures 8 to 11. Similar to the 50 ^{o}C configuration, highly correlated data (R^2=0.85) are obtained for the comparison of the TEOM™ and Partisol™ CM concentrations as shown in Figure 8. The ratio of concentrations is 27.4 (± 3.7), which is slightly higher, but not statistically different (p=0.69) than that at 50 ^{o}C.

No comparisons between the CCPM and the MOUDI™ concentrations were conducted for the 30 ^{o}C TEOM configuration, although MOUDI™ data were collected concurrently to the continuous monitor and the Partisol™. This is because the ambient RH was unusually low (even by the standards of the generally arid climate of the Los Angeles Basin), often below 20 to 30 %. As a result, while the comparison between TEOM™ and Partisol™ CM concentrations is robust, the CM concentrations measured by the MOUDI™ were low, resulting in unrealistically high ratios between the TEOM™ and

MOUDI™ CM concentrations. This is confirmed by plotting the CM concentration ratio of Partisol™-to-MOUDI™ vs RH, as shown in Figure 9. From the data plotted in Figure 9 there is a well-defined inverse relationship between this ratio and the RH. This ratio achieves an ideal value of 1 as the RH reaches 45-50 %. For lower RH, this ratio increases sharply and becomes as high as 5 when the RH reaches the 10 to 15% range. To confirm that this phenomenon is related to particle bounce, which would be more pronounced at lower RH, the ratio of FM concentration of Partisol™-to-MOUDI™ vs RH was plotted, as shown in Figure 10. The reverse trend is observed, with the ratio of the FM concentration of the Partisol™-to-MOUDI™ increasing from 0.2 to about 1, as the RH increases from 10 to 50 %. Further, the total PM_{10} Partisol™-to-MOUDI™ ratio was 0.99 (± 0.13) based on 30 field experiments, thereby suggesting that since both samplers agreed well for PM_{10}, the only difference is in the FM and CM concentrations measurements, that is, CM concentration is low and FM concentration is high at low RH, suggesting particle bounce. These field observations illustrate one of the main drawbacks of impactors, and raises serious implications on the appropriateness of using impactors with uncoated substrates to obtain the size distributions of aerosols under low (< 30%) RH conditions.

Experiments at a TEOM™ temperature setting of 30 ^{o}C also showed independence of the ratio of the TEOM™-to-Partisol™ CM concentrations to the ambient FM-to-CM concentration ratio (Figure 11). Data plotted in Figures 6 and 11 indicate that the mass concentration ratio of the concentration-enriched TEOM™ to either the MOUDI™ or Partisol™ is independent of the FM-to-CM concentration ratio over a range of values extending from about 0.2 to 5, thereby covering a broad spectrum of ambient

sampling conditions, and thus, strengthening the applicability of the CCPM to other locations and times of the year.

During these experiments, ambient PM data for a few selected runs were recorded using an APS™. Figure 12 shows the time series in CM concentrations measured by the TEOM™ and the APS™ during one day of the field experiments. A particle density of 1.6 g/m^3 was assumed in the APS™ data. The TEOM™ CM concentrations were converted to ambient CM concentrations by dividing by 26. Direct comparison between the actual concentrations measured by the two monitors cannot be made, since knowledge of the real (as opposed to an assumed) density of ambient coarse particles is required in order to convert the APS™ concentrations to actual mass concentrations. However, the data plotted in Figure 12 clearly show that very good overall agreement is observed in the time series of the CM concentrations obtained by means of the two samplers.

SUMMARY AND CONCLUSIONS

This paper describes the development and laboratory and field evaluation of a CCPM that is based on enriching the CM concentrations by a factor of 25, while maintaining FM concentration at ambient concentrations. The aerosol mixture is subsequently drawn through a standard TEOM™, the response of which is dominated by the contributions of the CM due to enrichment of the coarse particles. The laboratory evaluation of the 2.5 μm cutpoint round nozzle virtual impactor confirms the rise in the enrichment factor as a function of particle AD. The concentration enrichment factor increases sharply from about 2 to about 25 as particle AD increases from 2 to 3 μm. The enrichment is the same, with in the error of the measurement, for particles in the AD range of 3 to 9 μm.

Findings from the field study ascertain that the TEOM™ coupled with a 2.5 μm virtual impactor can be used successfully for continuous CM concentration measurements. The results indicate excellent correlation between the concentration-enriched TEOM™ and time integrated samplers (MOUDI™ and Partisol™), with the average TEOM™ CM concentration being approximately 26-27 times higher than those measured by the time-integrated samplers. No substantial differences in the response of the concentration-enriched TEOM™ are observed between TEOM™ operating temperatures of 30 and 50 °C. Results from the field experiments also show that the CM concentrations measured by the concentration-enriched TEOM™ are independent of the ambient FM-to-CM concentration ratio. This is due to the decrease in ambient coarse particle MMD with increasing FM-to-CM concentration ratio, as might be expected, since FM concentrations tend to increase and coarse particle loadings tend to decrease during stagnation conditions. This also strengthens the applicability of the CCPM in cases where the FM-to-CM concentration ratio is very high. Finally, our results illustrate one of the main problems associated with the use of impactors to sample particles under conditions of RH values lower than 40%. While PM_{10} concentrations obtained by means of the MOUDI™ and Partisol™ are in excellent agreement, CM concentrations measured by the MOUDI™ are as low as 20% compared to those measured by the Partisol™, while MOUDI™ FM concentrations were high by as much as a factor of 5, together suggesting particle bounce at low RH.

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DISCLAIMER

The U.S. Environmental Protection Agency through its Office of Research And Development collaborated in the research described here. It has been subjected to Agency review and approved for publication. Mention of trade names or commercial products does not constitute an endorsement or recommendation for use

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FIGURE CAPTIONS

Figure 1. Schematic of the Continuous Coarse Particle Monitor.

Figure 2. Particle Penetration Through the PM_{10} Inlet.

Figure 3. Concentration Enrichment Factor as a Function of Particle Aerodynamic Diameter. Total Flow Rate; 50 LPM. Minor Flow; 2 Lpm.

Figure 4. TEOM™ vs MOUDI™ CM Concentrations. TEOM™ at 50 ^{o}C.

Figure 5. TEOM™ vs Partisol™ CM Concentrations. TEOM™ at 50 ^{o}C.

Figure 6. Dependence of TEOM™-MOUDI™ and TEOM™-Partisol™ Ratio on FM-to-CM concentration ratio. TEOM™ at 50 ^{o}C.

Figure 7. Relationship Between Coarse Particle Mass Median Diameter (MMD) and FM-to-CM concentration ratio.

Figure 8. TEOM™ vs Partisol™ CM Concentrations. TEOM™ at 30 ^{o}C.

Figure 9. Plot of the Partisol™-to-MOUDI™ CM Concentrations as a Function of Ambient Relative Humidity.

Figure 10. Plot of the Ratio of Partisol™-to-MOUDI™ FM Concentrations as a Function of Relative Humidity

Figure 11. Dependence of TEOM™-Partisol™ Ratio on FM-to-CM concentration ratio. TEOM™ at 30 ^{o}C.

Figure 12. Time-series of TEOM™ and APS™ CM Concentrations.

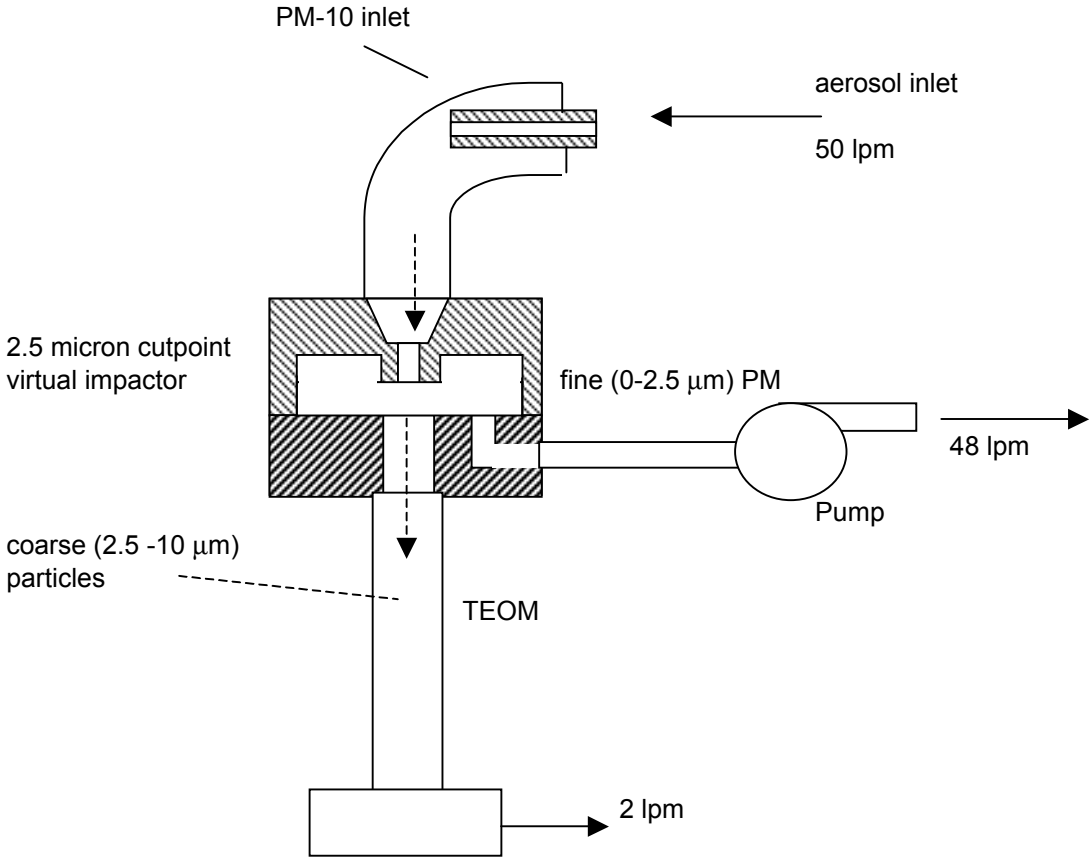

Figure 1. Schematic of the Continuous Coarse Particle Monitor

Figure 2. Particle Penetration Through the PM10 Inlet

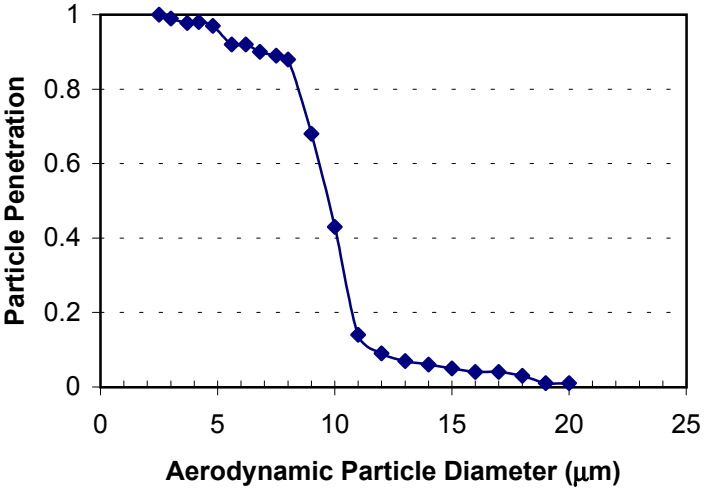

Figure 3. Concentration Enrichment as a Function Particle Aerodynamic Diameter. Total Flow Rate; 50 LPM. Minor Flow; 2 LPM

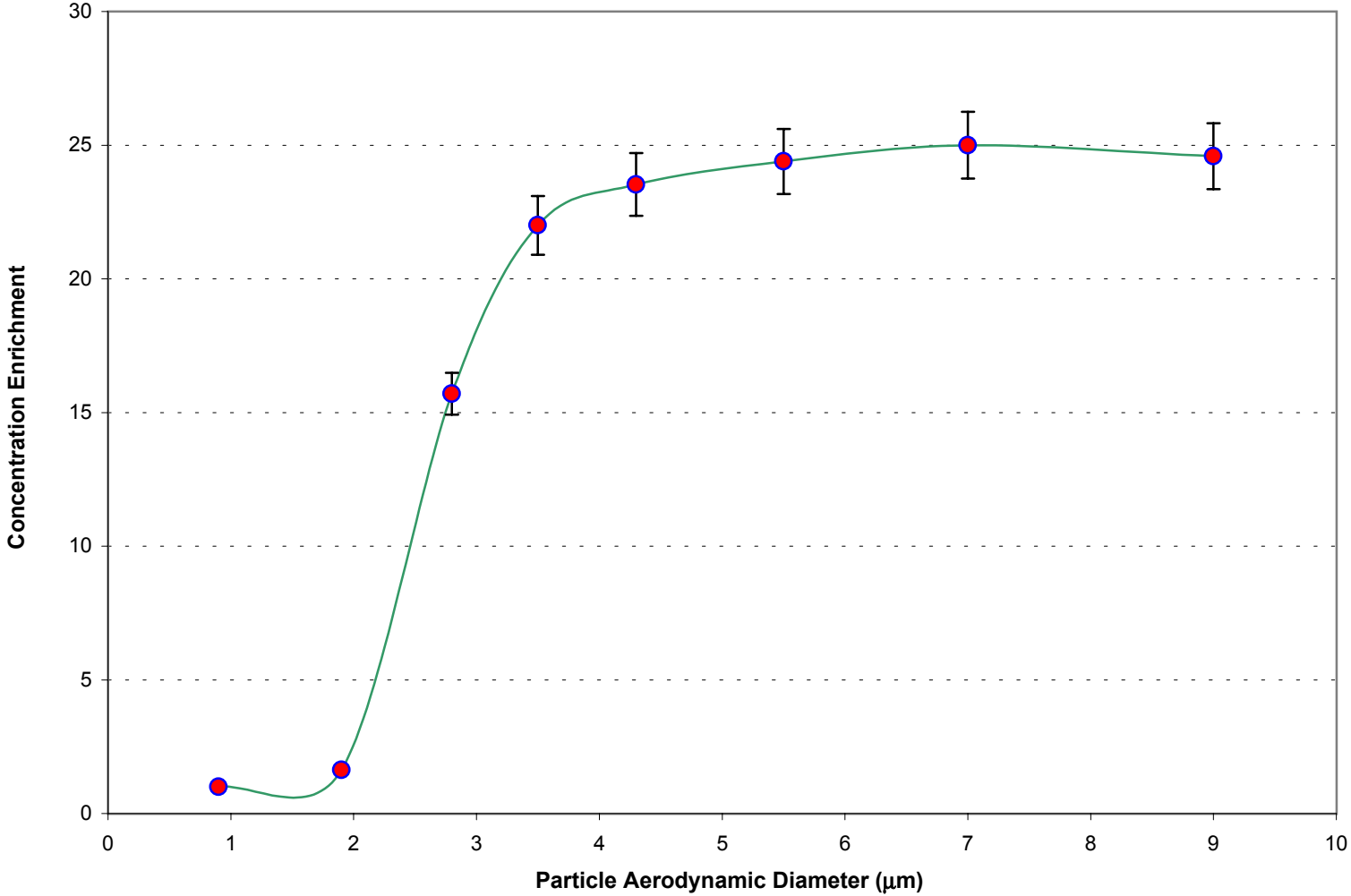

Figure 4. TEOM vs MOUDI Coarse PM Concentrations. TEOM at 50 deg C

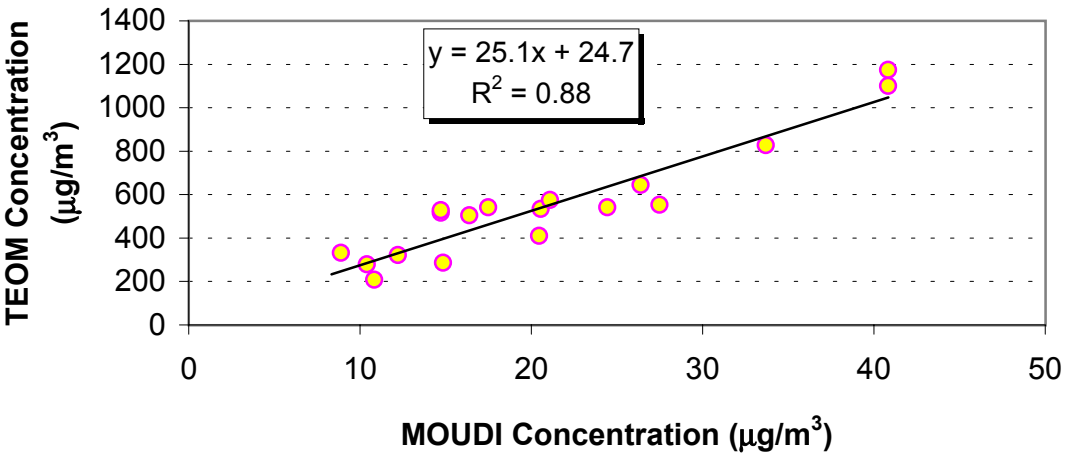

Figure 5. TEOM vs Partisol Coarse PM Concentrations. TEOM at 50 deg C

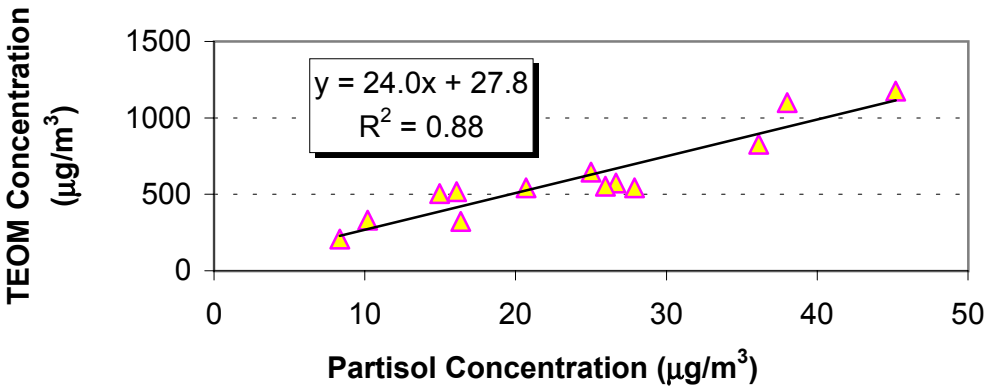

Figure 6. TEOM-MOUDI and TEOM-Partisol Concentration Ratio as a Function of Fine/Coarse PM ratio. TEOM at 50 deg C

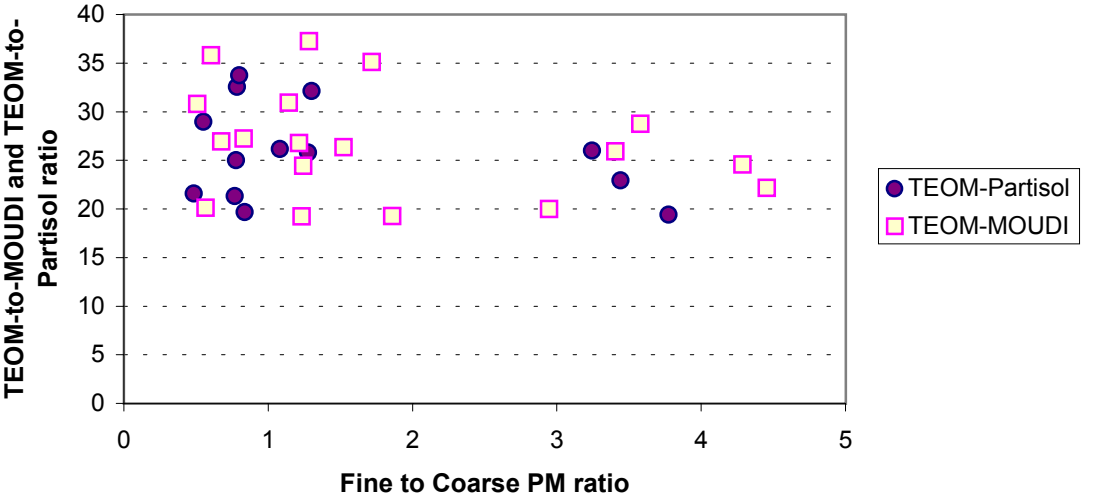

Figure 7. Relationship Between Coarse Particle Mass Median Diameter (MMD) and Fine-to-Coarse PM ratio

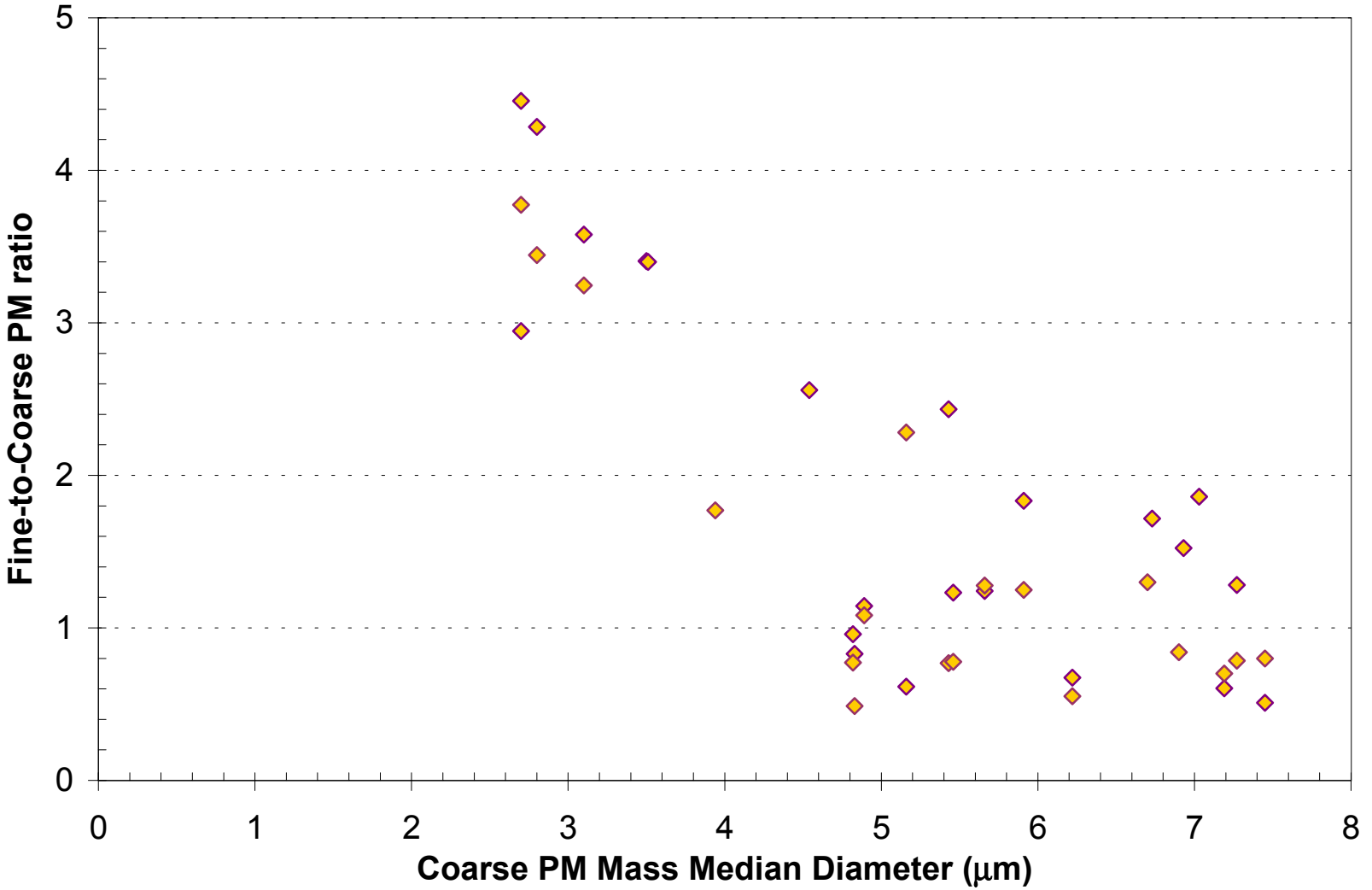

Figure 8. TEOM vs Partisol Coarse PM Concentrations. TEOM at 30 deg C

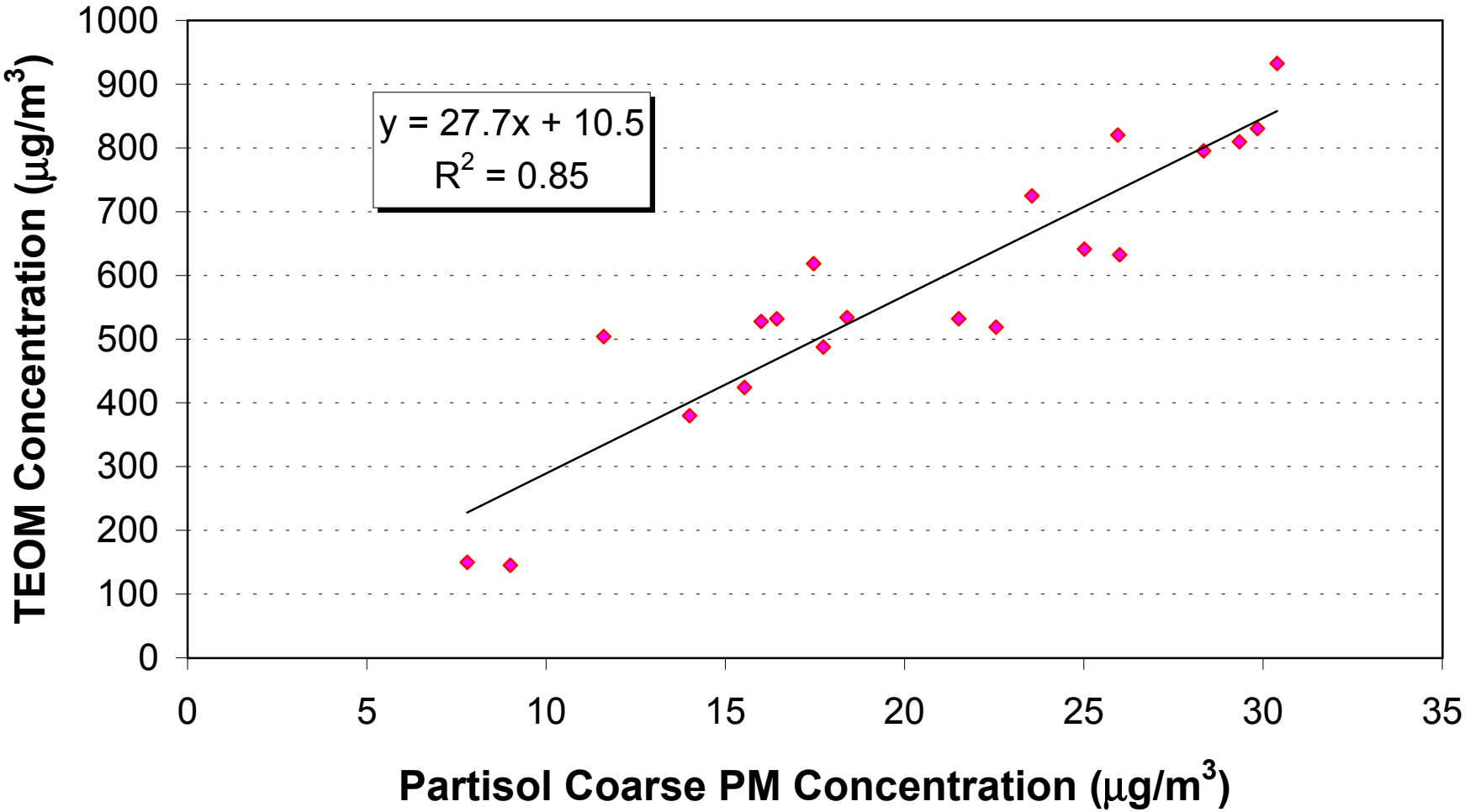

Figure 9. Plot of the Partisol-to-MOUDI Coarse Particle Concentrations as a Function of Ambient Relative Humidity

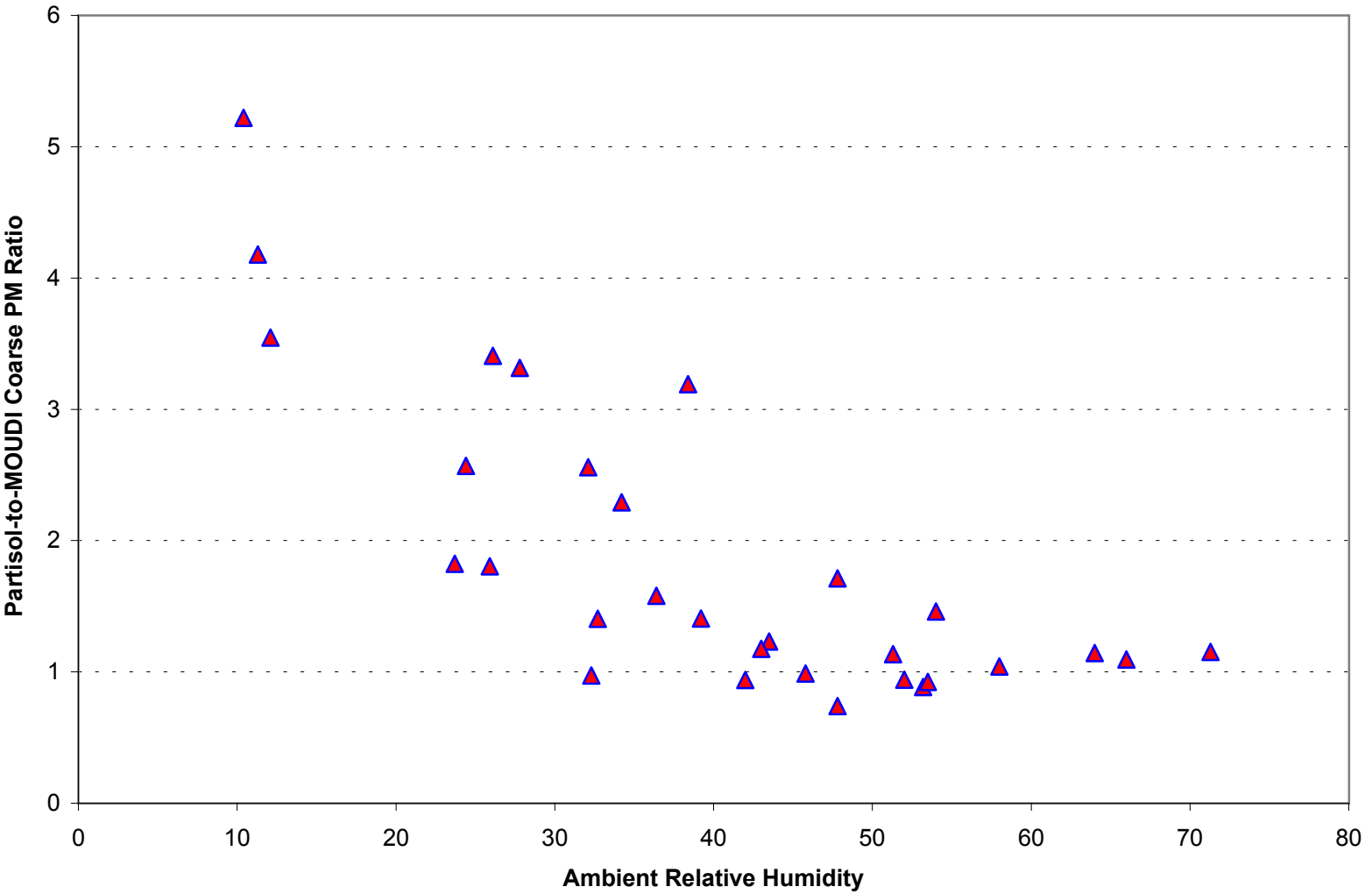

Figure 10. Plot of the Ratio of Partisol-to-MOUDI Fine PM Concentrations as a Function of Relative Humidity

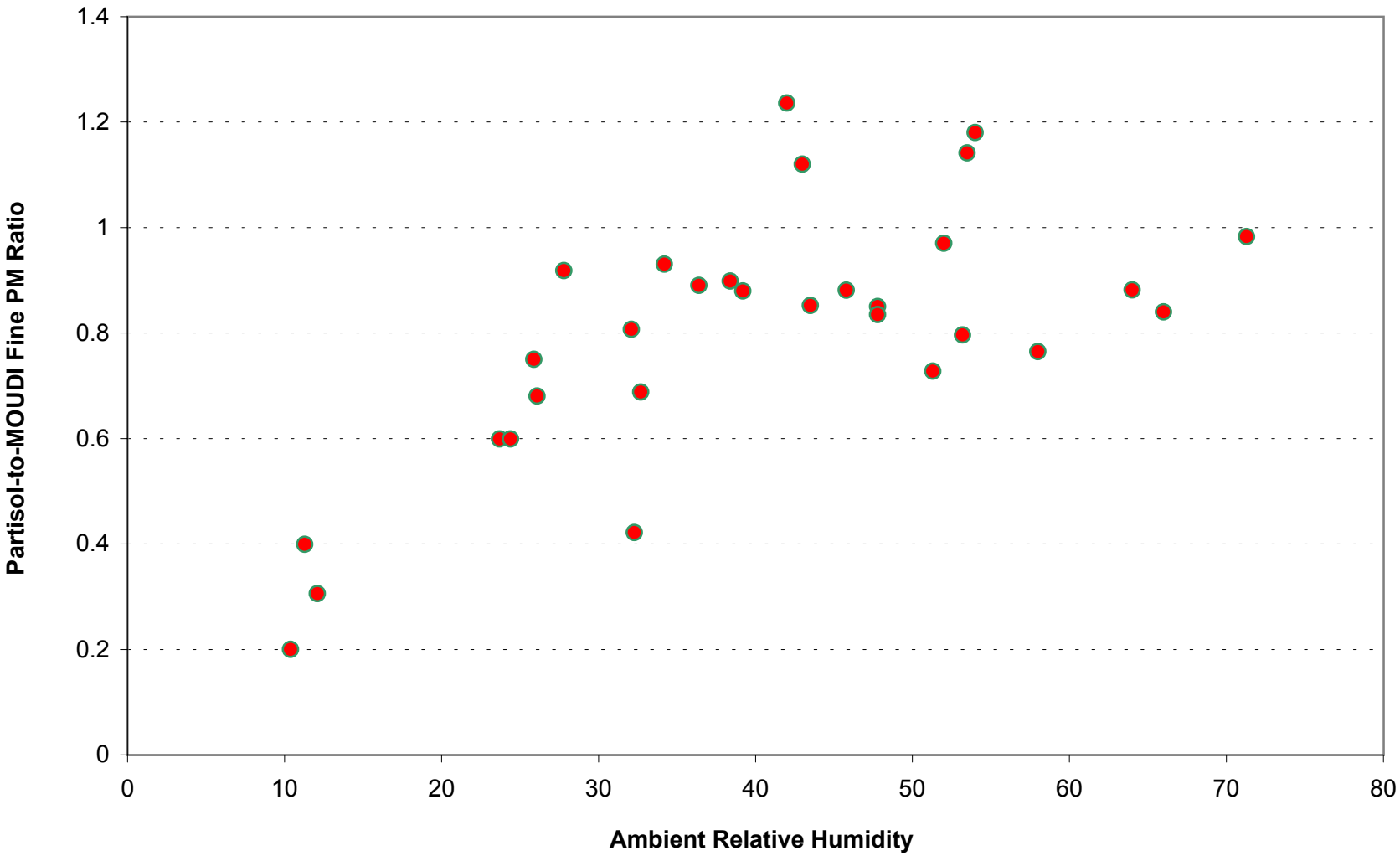

Figure 11. Dependence of TEOM-Partisol Ratio on Fine/Coarse PM ratio. TEOM at 30 deg C

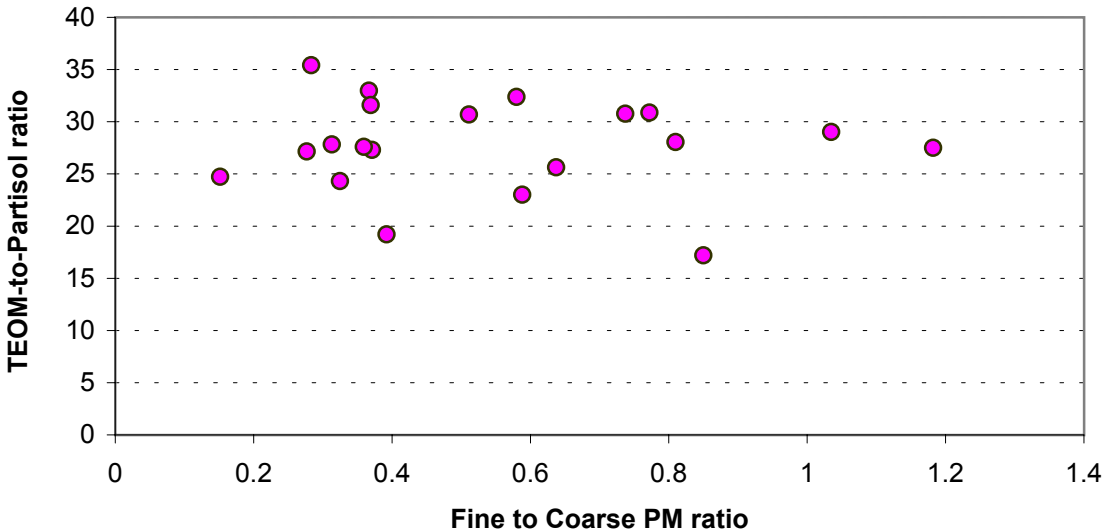

Figure 12. Time-series of TEOM and APS Coarse Particle Concentrations