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Sampling and Characterization of PM-fractions of Ambient Particulate Matter in Bangkok Utilizing a Cascade Virtual Impactor

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ABSTRACT

A cascade virtual impactor was developed and utilized for sampling and measurement of ambient aerosols in size classes demarcated by particle aerodynamic diameters of 10μm, 2.5μm and 1μm. Virtual impaction diminishes the flow of ambient air through the collected sample by 90%, consequently reducing the chance of sampling artifacts, such as extensive evaporation of semi-volatile compounds, or chemical cross-reactions after sample deposition. An omni-directional sampling head with an adjustable upper particle cut-off size was designed and numerically investigated for its suitability for PM sampling. The total flow rate through the instrument was 12.5 lpm; thus, with about five hours sampling, a total mass on the order of a few hundred micrograms in urban areas can be collected for air pollution determination purposes. Measurements were performed in Bangkok, Thailand to obtain mass concentrations, PAHs and carbon content of the sampled material. They show the feasibility of utilizing cascade virtual impaction for simultaneous sampling of various PM fractions and subsequent chemical analysis in air quality research and also give insight into the air pollution burden of this city.

Keywords: environmental measurement, cascade virtual impactor, aerosol sampling, PM-fractions, chemical composition

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INTRODUCTION

Environmental aerosols and related health impacts are strongly associated with their size distributions and concentrations, as well as with the chemical composition of ambient matter. A number of epidemiological studies document links between ambient particulate matter concentrations, air pollution levels, and adverse health effects (Dockery et al., 1994; Hall and Wynder, 1984; Schwartz and Dockery, 1992). Environmental aerosols undergo many physical, physico-chemical and chemical alterations during their atmospheric residence time. As a result, one encounters a polydispersed particulate system of chemically heterogeneous particles with complex toxic potential. These aerosols exist as an aerodispersed system of solid and liquid particles with different chemical compositions and sizes spreading over more than four orders of magnitude (Wilson and Suh, 1997). For urban ambient particle size distributions, typically about half the total ambient mass can be related to so-called fine aerosols, particles with diameters smaller than 1 μm. Coarse particles, with diameters larger than 1 μm, are usually present in the ambient atmosphere due to various mechanical particle-production processes, and are likely to have chemical characteristics different from fine particles. Particles can be characterized in several ways to evaluate their health consequences (CEN, 1993; Wolff, 1996).

The term particulate matter (PM) has been used for describing air quality. Rules for PM_{10} and $PM_{2.5}$ size categories were established by the U.S. Environmental Protection Agency (USEPA, 1997, 2001). PM_{10} refers to particles with aerodynamic diameters smaller than 10 μm, at which value the sampler has 50% sampling efficiency. $PM_{2.5}$ represents particles with diameters smaller than 2.5 μm. Based on these definitions further classification defines the coarse fraction as 2.5 to 10 μm of total mass and the fine fraction as less than 2.5 μm in aerodynamic diameter (Wilson and Suh, 1997). There are opinions that $PM_{1.0}$ could be a more suitable size cut off than $PM_{2.5}$ for health-related aerosol sampling. However, the problem is that relatively limited amounts of data are available for the $PM_{1.0}$ fraction.

Virtual size separation has been successfully used for inertial separation of airborne particles; but usually a dichotomous separation into only two size classes has been performed (McFarland et al., 1978; Loo and Cork, 1988). In a few cases, a cascade system using this technique has been utilized that allows parallel sampling of more than two size fractions (Haglund et al., 2002; Szymanski and Liu, 1986).

Based on the rationale outlined above we developed a cascade virtual impactor (CAVI) with cut-off sizes of 10, 2.5, and 1.0 μm, corresponding to PM_{10} and $PM_{2.5}$, with a further stage cut off at 1.0 μm. Particles are collected on filters corresponding to these size fractions, with the final filter collecting particles less than 1.0 μm ($PM_{1.0}$). All filters can be extracted easily from the CAVI for determining mass concentration and for further chemical analysis. An omni-directional sampling head was designed for the CAVI based on earlier work (Liu and Pui, 1981; Podgorski et al., 2003). Because of some design changes and dimensions, the sampling head's performance was investigated numerically for its suitability to

separate and transport sampled environmental particles into the CAVI. This system consisting of sampling head, transfer tube and CAVI was used to perform ambient PM measurements near a major urban traffic road in Bangkok and analyzed for PM fractions, polycyclic aromatic hydrocarbons (PAHs), and organic and elemental carbon content. Frequently, organic carbon (OC) and elemental carbon (EC) constitute a large fraction of the PM mass in ambient air and are related to adverse health effects and urban haze, and can be used to correlate ambient particle concentrations with likely sources (Sinlanpää et al., 2005). The ratio of OC to EC is also a useful indicator for secondary organic aerosol formation. PAHs are primary organic aerosols and belong to a group of organic compounds composed of two or more carbon rings derived from benzene. They degrade slowly in the environment and some are suspected to be harmful (Santodonato et al., 1981). As a rule, when PAH compounds grow in molecular weight, their solubility in water decreases; a fact which may be important under very humid conditions. Although the interaction of PAHs and their metabolites with the human body is not yet completely understood, first investigations show that some PAHs are probable carcinogens. Results presented here show distributions of the above-named chemical compounds in PM_{10}, $PM_{2.5}$ and $PM_{1.0}$ classes.

QUALITY OF SIZE SEPARATION IN A CASCADE VIRTUAL IMPACTOR

Size separation by a virtual impactor stage is based on the inertia of accelerated and decelerated particles in a gas (air) flow. The principle of operation of a separation stage and the major parameters governing the performance are shown in Figure 1. More detailed discussion of this subject can be found in Marple and Chien (1980). Only key aspects of the technique are given here.

The separation stage consists in its basic configuration of a coaxially oriented acceleration and collection nozzle, indicated by the diameters D_0 and D_1. The particle-laden air enters the nozzle and accelerates, depending on the acceleration nozzle diameter and the total flow rate, and is directed to the collection nozzle. The flow rate through this nozzle is called minor flow which is a fraction of the total flow rate (10% in this design). The major fraction, or major flow, is redirected and by-passes the collection nozzle. Consequently, particles above a certain aerodynamic size (cut-off size) follow the minor flow, penetrate the collection nozzle and can be collected, in this case, on a filter. Particles smaller than this cut-off size will follow the major flow and can be directed into another stage where a subsequent separation may take place; thus forming a fractionating cascade. The performance of a separation stage is characterized by a collection efficiency curve. Due to the specifics of this separation process, there will always be a residue of particles larger than the minor flow cut-off size and particles smaller than major flow cut-off. A key design issue is to minimize this effect. It is well-known that internal losses, particularly on the collection nozzles might be substantial (Marple and Chen, 1980). The CAVI presented here is a laminar flow unit, with overall internal losses typically below 10% per stage.

For the design of the separation stages in this work, $S = D_0 = L$ and the value of the $Stk_{50} = 0.44$ were chosen based on earlier studies (Szymanski and Liu, 1986). Nominal cut-off sizes (D_{50}) were calculated using the standard model approach (Marple et al., 1993):

$$D_{50} = \sqrt{\frac{9\pi\mu {D_0}^3 (Stk_{50})}{C(D_A)\rho Q}} \tag{1}$$

where μ is the viscosity of air at NTP conditions; $\rho = 1000$ kg/m^3 (because the inertial cut-off diameter is given in terms of an aerodynamic diameter); $C(D_A)$ is the Cunningham slip correction factor (Allen and Raabe, 1985); Q and D_0 are flow rates per nozzle and acceleration nozzle diameter, respectively. Multiple nozzles per stage were used in order to maintain a pressure drop as low as possible across the whole unit, which totaled to about 10% of the ambient pressure. The PM_{10} and $PM_{2.5}$ stages were equipped with six nozzles each. The 1.0 μm stage consisted of three nozzles in order to achieve the appropriate flow velocity and necessary separation characteristics. The key issue in such a design is to ensure that multiple nozzles combined into a cascading operation maintain the separation characteristics of a single nozzle for which the design calculations (eqn 1) were made.

In each separation stage, particles from the minor flow are collected onto an internal filter, and the major flow is redirected to the next separation stage. Consequently, the separated and collected matter is exposed only to the small fraction of the overall flow entering the device; hence, minimizing possible chemical modification of the sampled material. The sampling system described and used for the study consists of the inlet section with the omni-directional sampling head and the transfer tube 50 cm long and a 5-cm inner diameter, followed by the CAVI where size-separated sampling takes place.

MODELLING OF THE INLET SECTION OF THE CAVI – THE OMNI-DIRECTIONAL SAMPLING HEAD WITH THE TRANSFER TUBE

The sampling head was designed to provide laminar flow, allowing sampling independent of the direction of incoming particulate matter and having the option of removing particles with aerodynamic diameters larger than a certain size. Removal was achieved by means of a variable-width slit and a particle-removal surface at the entrance of the sampling head. The opening can be calculated using eqn 1 with $Stk_{50} = 0.59$, as suggested for slit impactors (Marple et al., 1993). Because the CAVI is equipped with multiple nozzles equidistantly spaced on a 20-mm diameter circle, the sampled matter after the collection through the sampling head must spread evenly over a certain area to provide a uniform distribution of the sample over all nozzles of the first impactor stage.

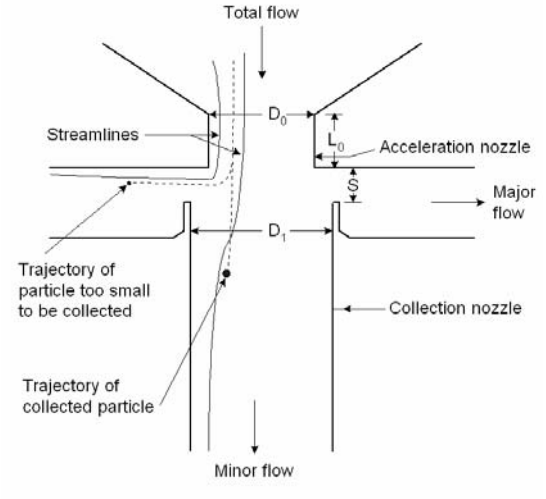

Figure 1. Schematic diagram of a virtual impactor stage showing the principle of operation and indicating the most critical design parameters.

The evaluation of the air-flow structure and particle paths within the sampling system under conditions dictated by the CAVI (total flow of 12.5 lpm) were numerically investigated. Modeling was performed using the FLUENT 6.0 flow modeling software (Fluent, Inc., Lebanon, NH 03766, USA, 2002). First, the system of Navier-Stokes and continuity equations was solved numerically for a standard k-epsilon model (Launder and Spalding, 1972). This approach was chosen because it was not assumed a priori that the flow in all parts of the system must be perfectly laminar. Consequently, trajectories of particles entering the sampler were determined using the Lagrangian method of description of a disperse system. Particle positions and velocities were calculated as a function of time. Calculations were done for the air-flow field at NTP. The results are summarized in Figure 2. It can be seen that the entire flow field is basically laminar with the exception of a very short distance at the entrance slit, where laminar and slightly transient flow character is present (Fig. 2a).

The colored scale in Figure 2a represents velocity vectors, which for this flow situation are within 0.2-1.3 m/s. Figures 2b and 2c show particle locations within the calculated domain, and show particle positions and residence times from the inlet section up to the entrance into the CAVI. For model calculations in Figures 2b and 2c, trajectories of 1,000 particles spread evenly through the inlet slit were determined. Here the colored scale represents the time, so not only the particle trajectory, but also the transit time through the system can be observed. It is evident that under these conditions particle residence

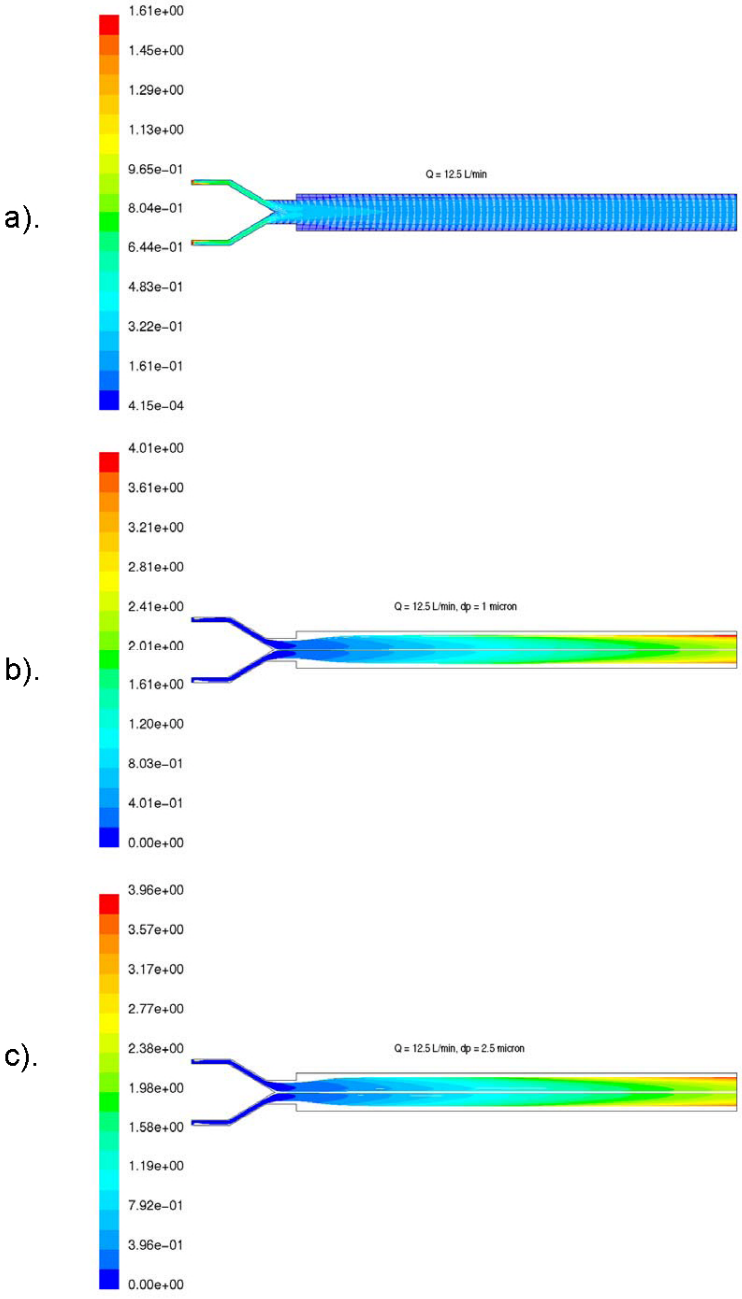

Figure 2. Results of modeling of the flow pattern and particle trajectories in the inlet and transport tube section of the measuring system for the flow rate of 0.75m^3/h: a) color scale shows the local flow velocity vectors in [m/s]; b) color scale shows for particles with diameter D_p= 1μm, their residence time in [s] and their location; c) same as b) except for particles with diameter D_p=2.5 μm.

times in the tube before entering the CAVI are fairly uniform through the exit cross-section, with typical values of transport times of 2.5-4 s. Moreover, it can be seen that virtually no contact with walls occurs, providing advantageous conditions for a representative particle sampling. Similar conditions with regard to particle trajectory and locations can be achieved for flow rates up to 50 lpm. It is notable that particle-laden air is quite evenly distributed across the exit of the transfer tube entering the CAVI.

SEPARATION PERFORMANCE OF THE CAVI

The sizing performance of any inertial separation device is determined in general by its collection efficiency curves. As mentioned before, the separation characteristics of a single nozzle, or a single stage should be maintained when these are assembled to a multi-nozzle cascade system. The CAVI performance was determined using liquid particles (oleic acid aerosols tagged with uranine) from 1-20 μm in size and solid polystyrene (PSL) particles 0.2-3 μm in diameter. Liquid particles were generated with a vibrating orifice generator. The particle size was monitored with an aerodynamic particle sizer. Following the deposition on a filter they were washed off (NaOH) and detected with a fluorometer. The PSL particles were generated by pneumatic atomization and measured by means of a laser aerosol spectrometer. In this case the ratio of upstream-to-downstream number concentrations provided a measure for the size separation efficiency η of the impactor stage.

$$\eta = \frac{C_N}{C_N + C_F} \tag{2}$$

where C_N (N = 1 - 3) and C_F (final stage) are concentrations (either mass or number) collected on a given stage. The expected cut-off diameters of 10, 2.5 and 1.0 μm were confirmed by the analysis of the separation curves for each stage yielding 9.94, 2.53 and 1.05 μm, respectively.

The steepness of the size separation curves can be described by a means of a dimensionless parameter $\Phi = D_{84} / D_{50} \approx D_{50} / D_{16}$, which are aerodynamic diameters corresponding to separation efficiency values of 84%, 50% and 16%, respectively. For the CAVI unit those Φ values were found to be 2.2, 1.3 and 1.5 for stages with 1, 2.5 and 10 μm cut-off size, respectively; being also comparable with values provided by U.S. federal reference methods (FRMs) (USEPA, 2001). Further examination of the separation effectiveness of the CAVI was delivered by means of optical image evaluation of particles deposited on filters at each stage. Size-separated particles for this microscopic, optical inspection were collected on polycarbonate filters. Images of the sampled material generated with the SEM, were transferred with the SEM calibration information into a computer and analyzed using OLYSIA imaging software (Olympus Corp.). The obtained projected area diameters agreed well with the geometric interval mid-point diameter.

Table 1 shows very satisfactory agreement between the sampling size classes given by the CAVI's cut-off sizes.

Table 1. Results of the optical analysis of sampled particles obtained from the SEM images. Mean deposited particle size in each size range has been determined based on the evaluation of the projected area diameter.

Impactor Stage	Classified Size Range [μm]	Mean Particle Size [μm]	Abs. Standard Deviation [μm]
1	10 - 50	14.1	3.38
2	2.5 - 10	3.39	1.40
3	1 – 2.5	1.61	0.55
4	0.1 - 1	0.26	0.12

MEASUREMENT RESULTS AND CHARACTERIZATION OF AMBIENT PARTICULATE MATTER

The system described above was utilized to sample and specify certain characteristics of the ambient aerosol on an urban major traffic route in Bangkok, Thailand. The sampling system was located at roadside with the sampling head positioned at 1.5 m above the ground. First, the sampling was performed for about six hours in order to examine the particulate matter deposition pattern on filters at all sampling stages of the CAVI. Sampled particles were collected typically on glass fiber filters (Pall-Gelman, 37 mm). Figure 3 shows results of this sampling. It can be seen that the sampled particulate matter is quite evenly distributed over the entire active filtration area. This is convenient because, if needed, a partition of the filter is possible for determining various chemical compounds in different analysis methods in order to establish the elemental composition of the sample.

The corresponding mass fractions are presented in Figure 4 showing a normalized measured mass size distribution (CAVI data). The horizontal arrows indicate the PM classes which can be retrieved conveniently from such a measurement. A probability density function (PDF) was then fitted to the collected mass distribution data assuming the log-normal particle size distribution (O'Shaughnessy et al., 2000; Raabe, 1978). This procedure resulted in MMD = 4.1 μm and σ_g = 2.50. The particle mass concentration for the PM_{10} fraction determined from this measurement was 75 $\mu g/m^3$, a rather typical value for this sampling location. A series of measurements was then performed over the dry season at various times during the day in order to establish the levels of PM and the $PM_{2.5}/PM_{10}$ ratio in Bangkok, which are summarized in Figure 5. The linear regression coefficient of $R^2 = 0.88$ shows good correlation between both data sets. PM pollution values collected during light traffic conditions were lowest, indicating the major cause of the environmental burden. The ratio values of $PM_{2.5}/PM_{10}$ are about 0.7 which is comparable with data from other major cities (Sillanpää et al., 2005). A comparison of PM

fractions between the wet and dry season shows that $PM_{1.0}$ roadside concentrations were nearly unaffected by seasonal environmental conditions (Fig. 6). However, the larger size fractions, $PM_{2.5}$ and the $PM_{10,}$ shown in Figure 6, indicate an obvious trend with the seasons. It is apparent that the influence of wet season conditions decreases the overall concentrations in those larger size fractions by about 40%, whereas the amount of fine particles with diameters below 1 μm remains constant and constitutes a very substantial fraction of the larger PM classes.

Besides size-segregated mass fractions the carbon content of ambient Bangkok aerosols was also addressed. Samples were measured using a microelemental analyzer (Vario EL-III, Elementar Amerias, Inc.). Appropriately prepared probes (Lim et al., 2003; Watson et al., 2005) were burned in a combustion tube, where the total carbon (TC) is defined as carbon burned at 1,150 °C for 90 s in a 0.1 oxygen-to-helium environment. The OC is defined by burning at 350 °C in pure helium. EC can be obtained by subtracting OC from TC. The carbon data shown in Figure 7 corresponds to the dry season sampling. It is interesting to note that in presented results the EC in all PM classes constitutes about one third of TC. However, one has to be careful with conclusions drawn from thermally obtained fractions (Turpin et al., 2000). The presence of some inorganic salts, or the charring of insoluble organic materials, can change the combustion rate of certain compounds (Kirchstetter et al., 2001; Yu et al., 2002).

Finally, the content of selected polycyclic aromatic hydrocarbons (PAHs) was also determined. In this study, PAHs were grouped in two classes. The first contained PAHs with three or less aromatic rings: naphthalene, acenaphthene, fluorene, phenanthrene and anthracene. The other contained PAHs with four or more aromatic rings: fluoranthene, pyrene, benz-a-anthracene, chrysene, benzo-b-fluoranthene, benzo-k-fluoranthene, benzo-a-pyrene, dibenz-anthracene, benzoperylene and idenopyrene. These compounds were analyzed with high performance liquid chromatography (HPLC) combined with a fluorescence detector (Furuta and Otsuki, 1983; Santodonato et al., 1981). Samples were prepared by condensing using a vacuum rotary evaporator after ultrasonically dissolving the samples in a 1:3 ethanol/benzene solution. The data is summarized in Figure 8. PAHs with four or more rings are quite uniformly spread over the investigated size classes; whereas, compounds with three or fewer rings peak between 1 and 2.5 μm. The overall determined PAH concentration burden ranges from 1-10 ng/m^3, values which are comparable with those found in different urban areas, as well as with Bangkok data obtained by other researchers (Norramit et al., 2005; Tongsanit et al., 2003).

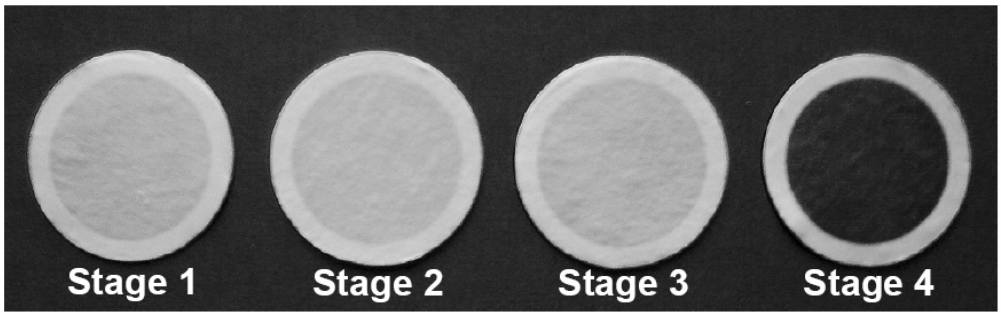

Figure 3. Photographs of deposited particulate matter in each size fraction showing uniform deposition pattern across the filter area: Stage 1 (D_p>10μm), Stage 2 (10μm <D_p>2.5μm), Stage 3 (2.5μm<D_p>1.0μm), Stage 4 (D_p<1.0μm)

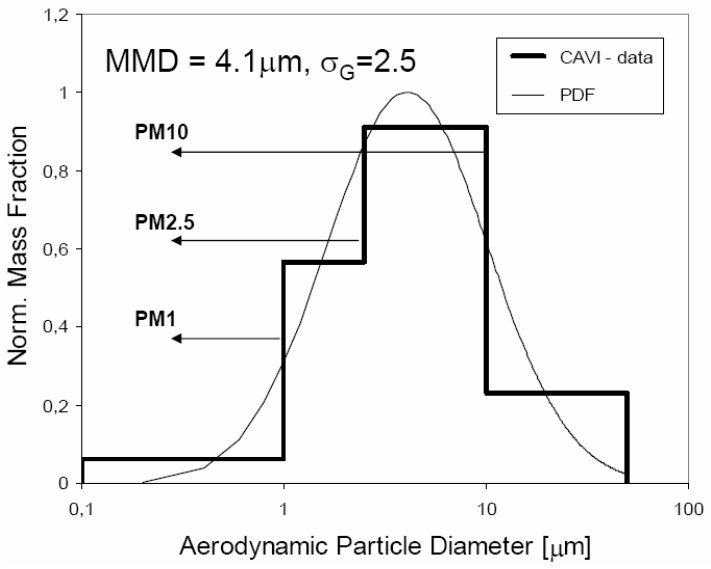

Figure 4. Diagram of a measured particle mass size distribution indicating the PM fractions.

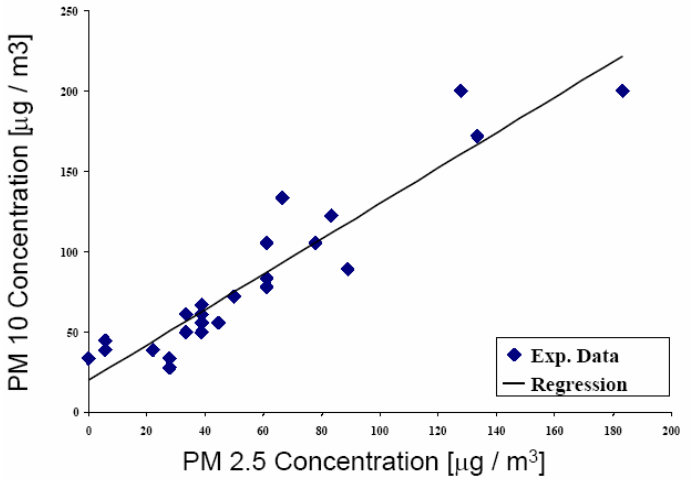

Figure 5. Summary of PM_{10} and $PM_{2.5}$ data collected in Bangkok over the dry season period.

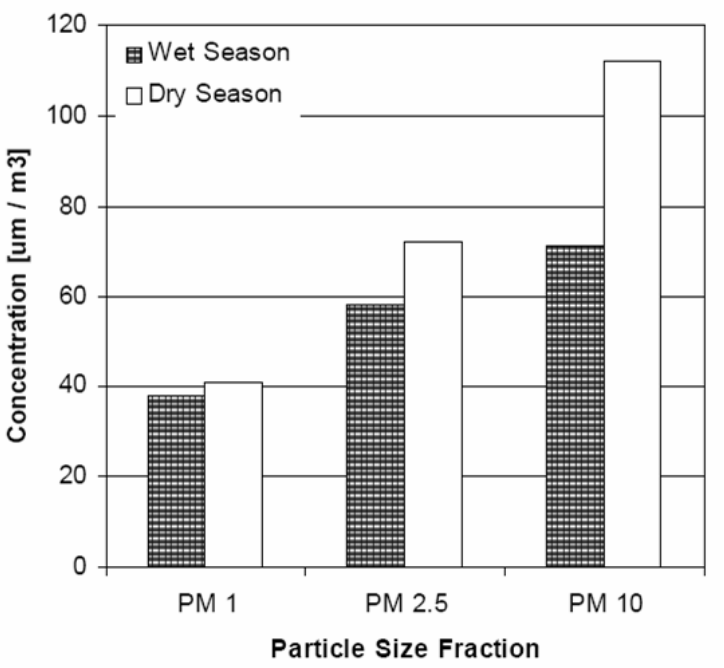

Figure 6. Seasonal influence on the concentration levels of measured PM fractions.

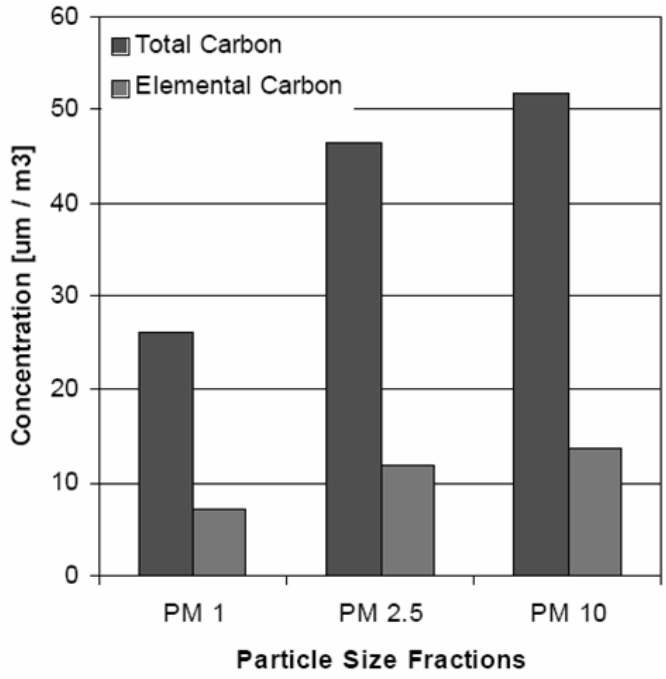

Figure 7. Carbon content of measured PM fractions of airborne particulate matter.

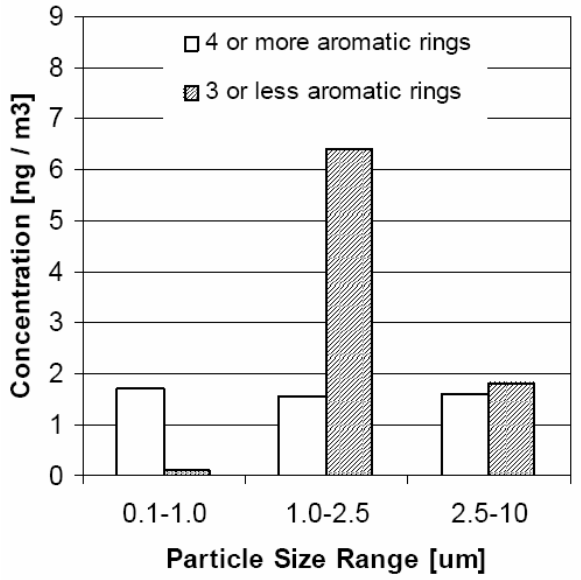

Figure 8. Concentrations of measured PAHs contained in collected PM fractions.

SUMMARY AND CONCLUSIONS

The described system designed for sampling of ambient particulate matter has been characterized and its feasibility for size-selective measurement of ambient aerosols has been proven in various field applications. The omni-directional sampling head allows a representative collection of aerosols and their transport for size fractionation in a virtual cascade impactor, but it also could be used in connection with any other aerosol measuring device, providing that, for the total flow rate, laminar flow conditions are maintained. For this study, the head cut-off diameter was set to 50 μm. For higher flow rates, however, the width of the inlet slit of the omni-directional sampling head has to be adjusted in order to provide the passage of particles of a required size into the transport tube. In the first stage, particles with sizes larger than 10 μm in diameter were collected. The particles sampled in the following stages were in the size classes PM_{10}, $PM_{2.5}$ and $PM_{1.0}$, and were collected on glass-fiber filters (Pall-Gelman, 37 mm). The homogeneous distribution of the sampled matter across the filter area allows determination of mass concentrations and a meaningful post-sampling investigation of chemical composition of each fraction, which can be performed from a part of each filter. Moreover, due to the fact that only a small fraction of the ambient air (minor flow) passes through the sampled material the sampling artifacts caused by possible chemical modification of the sampled material are minimized. The Thailand National Air Quality Standard was exceeded for a number of air samples investigated; although at this stage it is difficult to judge the long-term burden. Long-term measurements at multiple sampling locations are needed to address this issue. However, certain trends related to seasonal factors were observed that

clearly indicate the increase of ambient $PM_{2.5}$ and PM_{10} fractions during the dry season. Notably, the $PM_{1.0}$ fraction stays practically constant and not influenced by the change of seasons. A possible toxicological issue is indicated by measurements showing that a substantial carbon and PAH fractions are in the $PM_{2.5}$ and even $PM_{1.0}$ range. Further measurements especially correlated with local meteorology would allow more insight into the formation of secondary organic aerosols. The significance of data shown for health assessment can be addressed only in scope of a broader study.

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