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Design and characterization of a sequential cyclone system for the collection of bulk particulate matter

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In this paper, we describe the design, development and characterization of a high-volume sequential cyclone system for the collection of size-segregated PM in dry bulk form from the ambient environment in sufficient quantity for physical, chemical and toxicological characterization. The first stage of the system consists of a commercially available high volume PM₁₀ inlet. The second stage cyclone was designed by us to collect inhalable coarse particles (<10 µm and >2.5 µm). When tested individually with a challenge aerosol, a *D*₅₀ cut-size of this stage was found to be 2.3 µm at a flow rate of 1 m³ min⁻¹. The third stage, a commercially available cyclone designed for surface dust sampling, had a *D*₅₀ cut-size of 0.3 µm when tested at the same flow rate. The purpose of the third stage is to collect the fine particle portion of PM_{2.5} or accumulation mode (PM <2.5 µm and >0.1 µm). Thus, the sequential cyclone system will collect bulk samples of both the inhalable coarse particles and the fine particle portion of PM_{2.5}. The operation and maintenance of the new system are straightforward and allow for reliable collection of dry bulk ambient PM at relatively low cost.

1. Introduction

Airborne particulate matter (PM) is a complex mixture that includes particles in a wide range of sizes and with significantly different composition. Numerous epidemiological and toxicological studies have found associations between increases in morbidity and mortality for respiratory and cardiovascular illness and elevated concentrations of ambient PM.^{1–3} Toxicological studies exploring mechanisms which might explain these epidemiological findings typically use model particles such as carbon black,^{4,5} or residual oil fly ash (ROFA).^{6,7} The National Research Council⁸ identified the need to better understand the relationship between toxicity and the size and composition of PM as an important research priority. The epidemiological evidence suggesting that PM toxicity is size-dependent was

recently reviewed by Pope and Dockery.¹ In the early 1990s research and policy focused on health effects related to PM₁₀ (PM smaller than 10 µm in aerodynamic diameter). The focus shifted to PM_{2.5} (PM <2.5 µm in aerodynamic diameter) in the late 1990s. However, since PM_{2.5} is a subset of PM₁₀, there is growing interest in specifically exploring the impact on health from exposure to inhalable coarse particle fraction (particles with aerodynamic diameter between 2.5 µm and 10 µm, or PM_{10–2.5}). To date, few studies have examined this question^{9,10} and even fewer have begun to assess the impact of PM composition.^{11,12}

Surrogates of ambient PM such as carbon black and ROFA used in toxicological studies are only partially representative of ambient particulate matter. To allow for a true exploration of why ambient PM may be responsible for increased mortality and morbidity, a method for the collection of ambient PM samples for use in toxicological studies is needed. Existing samplers that collect PM in large quantity typically utilize filter-based or virtual inertial impaction techniques.^{13,14} The recently developed high volume cascade impactor or HVC¹⁵ size-fractionates and collects PM on polyurethane foam, which minimizes particle bounce. However, the presence of any collection matrix complicates attempts to harvest the PM in a dry state and to obtain a comprehensive chemical characterization that could inform toxicological assays.

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Environmental impact

Toxicological studies typically require milligram quantities of particulate matter (PM) for studies conducted *in vivo* and when complementary chemical characterization of PM constituents is needed. The goal of this project was to build upon current cyclone technology to design and develop a cyclone system that allows for the collection of ambient PM in gram quantities without the need for a collection substrate. PM is segregated in two size fractions and collected in bulk for use in toxicological studies that explore the mechanisms of injury associated with exposure to ambient particulate matter.

Non-media systems such as fine and ultrafine particle concentrators have also been developed,^{16,17} which generate concentrated ambient PM (CAP) for direct exposure in toxicological and human studies.^{18,19} Ultrafine particle concentrators typically separate the particles from the air by condensation with water prior to virtual impactor separation. Concentrated particles are then restored to their original sizes using thermal or diffusion dryers. Concentrators are not ideal for field collection of bulk PM samples because of their high relative cost and need for continuous maintenance.

An alternative to impactor or filter-based methods is high volume cyclones, which allow for the bulk collection of PM within the body of the cyclone. Unlike impactors, cyclones are easy to operate and maintain, are not subject to particle bounce, and their cost is low in comparison to other collection devices.²⁰ High volume cyclones are routinely used in agricultural and manufacturing industries for the continuous, cost-effective removal of PM from process air.^{21,22} On a smaller scale, cyclones have been extensively used for low flow rate size-selective air sampling in the occupational and environmental health settings.^{23,24} Cyclones have also been used as collectors to sequentially size-segregate particles; a low volume, 5-stage cyclone system for high particulate concentration stack sampling²⁵ has been used to characterize exposure to beryllium aerosols^{26,27} and another low-flow 2-stage sequential cyclone was recently developed for biological sample collection.²⁸

The goal of this project was to build upon current cyclone technology to design and develop a cyclone system that would allow for the collection of ambient PM in gram quantities without the need for a collection substrate. The system would take advantage of previous work utilizing a single-stage high-volume cyclone for the bulk collection of environmental PM samples.^{29–35} The new system would incorporate multiple stages that allow for size selectivity in PM capture. This paper describes the design and development of a high-volume sequential cyclone system (HVSCS) for collection of size-segregated dry bulk PM from the ambient environment in sufficient quantity for physical, chemical and toxicological characterization.

2. Materials and methods

2.1 Design overview

A framework for the sequential system is provided in Fig. 1. The first stage removes PM $>10\ \mu\text{m}$, allowing PM $<10\ \mu\text{m}$ to pass to the second stage collector. The second stage, with a theoretical D_{50} cut-size (the aerodynamic diameter of particles that are

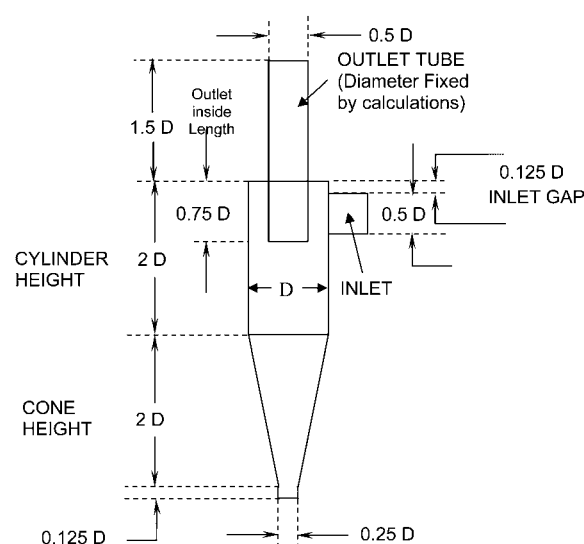


Fig. 2 Dimensions for the Stairmand high efficiency cyclone (Moore and McFarland, 1993).

collected with 50% efficiency) of $2.5\ \mu\text{m}$, will collect the PM passing through the first stage providing a coarse PM sample (PM_{10–2.5}). Finally the third stage should have a D_{50} as low as possible to provide the most representative sample of PM_{2.5}.

Stage 1 is a high-volume PM₁₀ inlet (HI-Q Environmental, San Diego, CA) with a D_{50} equal to $10\ \mu\text{m}$ at a flow rate of $1.1\ \text{m}^3\ \text{min}^{-1}$ ($40\ \text{cfm}$) $\pm 10\%$ (1.0 to $1.2\ \text{m}^3\ \text{min}^{-1}$).³⁶ Stage 2 is a cyclone designed as part of this project with a target D_{50} of $2.5\ \mu\text{m}$. The final stage, Stage 3, is a commercially available cyclone (model HVS3, CS3 Inc., Sandpoint, ID) designed as part of a vacuum carpet dust sampler for collection of settled dust to be analyzed for lead and pesticides.³⁷ Calculations based on the design equations suggest that this Stage 3 cyclone will have a D_{50} of $\sim 0.8\ \mu\text{m}$ when operated at $1\ \text{m}^3\ \text{min}^{-1}$. We have used this cyclone as a single stage collector for PM toxicology studies.^{30,34} We report in this paper the laboratory-based D_{50} for both Stage 2 and Stage 3 cyclones first tested separately and then tested when assembled as a system. The sampling pump used for the system is a BRL-3300M (HI-Q Environmental, San Diego, CA), a commonly used brushless pump for high volume environmental sampling.

2.2 Stage 2 cyclone design

The performance of cyclones can be estimated using semi-empirical equations. Cyclone design is therefore often an

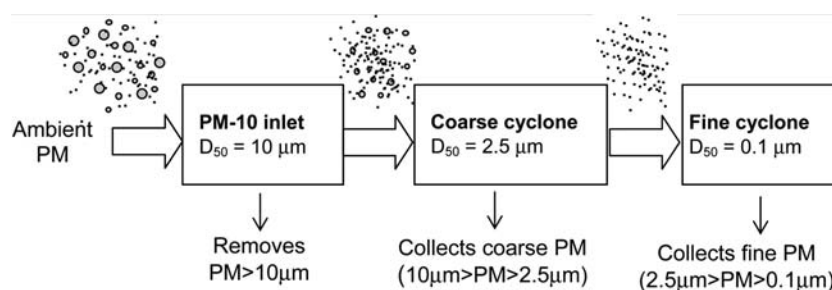


Fig. 1 Conceptual design of the 3-stage Sequential PM Collection System.

iterative process where the equations define the gross behavior, with fine-tuning of dimensions often needed to get more precise collection characteristics. The specifications of the Stage 2 cyclone, to be described from here forward as the coarse fraction cyclone, were based on the design of a Stairmand high efficiency cyclone³⁸ (Fig. 2). The target dimensions of the coarse fraction cyclone were calculated for a D_{50} of 2.5 μm at 1.1 $\text{m}^3 \text{min}^{-1}$ (same flow rate as the PM_{10} pre-selector), following the model described by Moore and McFarland,³⁹ summarized in eqn (1)–(3). As shown in eqn (1)–(3), the diameter of the cyclone (D) is dependent on the flow Reynolds number (Re_f), which cannot be solved without a value for D . Therefore, D must be obtained by an iterative process after assuming a D_o .

$$U_i = Q / \pi \times \left(\frac{D_o}{2} \right)^2 \quad (1)$$

$$\text{Re}_f = \frac{\rho \times U_i \times (D - D_o)}{2 \times \mu} \quad (2)$$

$$D = \frac{D_{50}}{\exp(\ln(a) + b \ln(\text{Re}_f))} \quad (3)$$

where U_i = inlet velocity (cm s^{-1}); Q = flow rate ($\text{cm}^3 \text{s}^{-1}$); D_o = diameter of cyclone's outlet (cm); π = pi; Re_f = flow Reynolds number; ρ = air density (g cm^{-3}); D = cyclone's body diameter (cm); μ = fluid viscosity ($\text{g cm}^{-1} \text{s}^{-1}$); D_{50} = aerodynamic particle cut point diameter (cm); $\ln a = -2.933 \pm 0.042$; and $b = -0.81719 \pm 0.017$.

The starting requirement for the coarse fraction cyclone was that the diameter of the outlet needed to interface directly to the diameter of the inlet of Stage 3.

Penetration efficiency tests (described later) of an initial prototype coarse cyclone showed a relatively sharp cut point of 1.5 μm , below the targeted D_{50} of 2.5 μm (Fig. 5). A second prototype of the coarse fraction cyclone was re-designed to move the D_{50} closer to 2.5 μm , resulting in an increase in D by 1.7 cm. The rest of the dimensions changed accordingly (Fig. 2). The penetration efficiency of this new cyclone was also tested in a laboratory setting, using monodisperse particles of different sizes at two flow rates: 1.1 and 1.0 $\text{m}^3 \text{min}^{-1}$. These flow rates

were chosen to represent the midpoint and lower edge of acceptable flow ranges for the Stage 1 PM_{10} inlet.

2.3 Laboratory characterization of cyclone performance

Collection efficiencies were tested first for each cyclone individually and then for each cyclone when operated as a system using fluorescently tagged particles, one size at a time, as described below.

2.3.1. Determination of collection efficiency of cyclones tested individually. Individual cyclones were tested using two different aerosol generation methods: (1) an ink-jet aerosol generator (IJAG)⁴¹ that allows testing outside a chamber for particles ranging from 2 to 12 μm and (2) polystyrene latex (PSL) monodisperse particles ranging from 0.1 to 5 μm nebulized inside a chamber.⁴⁰ The combined systems, described below in detail, generate solid fluorescent particles of sizes ranging from 0.1 to 12 μm (one size at a time). Characterization of each cyclone's performance efficiency was accomplished by measuring: (1) fluorescence of the total mass of particles entering the cyclone, using a reference filter (RF); (2) fluorescence of particles penetrating through the cyclone using an outlet filter (OF); and (3) fluorescence of particles retained by the cyclone's collection cup (CC) and body (Fig. 3b and c). Both reference and outlet filters were 8" \times 10" AE glass fiber (Pall Corp., East Hills, NY).

Fluorescence was extracted from the filters, body and cup by washing with a known volume of an appropriate recovery solution (see below for details). Fluorescence was then determined by pipetting a 3 mL aliquot from each wash in a cuvette and analyzing on a fluorometer (Model 450, Sequoia-Turner, Mountain View, CA). Because of the small number of total samples collected, and the high variability of the fluorescence from blank filters, between 40 and 60% filter blanks were analyzed to determine fluorescence background. Blanks of the recovery solution were read every 3 samples to correct for baseline drift.

To test individual cyclones, particles with Aerodynamic Equivalent Diameter (AED) $>2 \mu\text{m}$ were generated with an IJAG particle generation system (Fig. 3a), in which monodisperse particles are produced by desiccating liquid droplets of sodium hydroxide (NaOH) mixed with sodium fluorescein

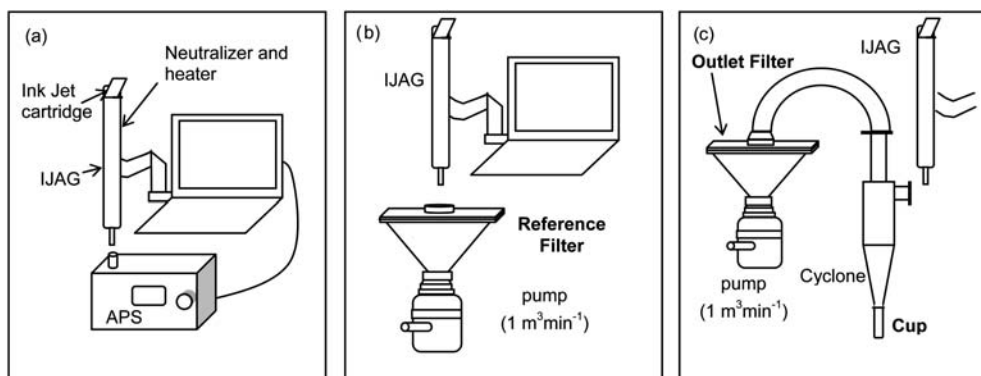


Fig. 3 Cyclone tests using an Ink-Jet Aerosol Generator (IJAG). (a) Verification of size and concentration generated with the IJAG; (b) collection on a reference filter; and (c) test of cyclone collection efficiency.

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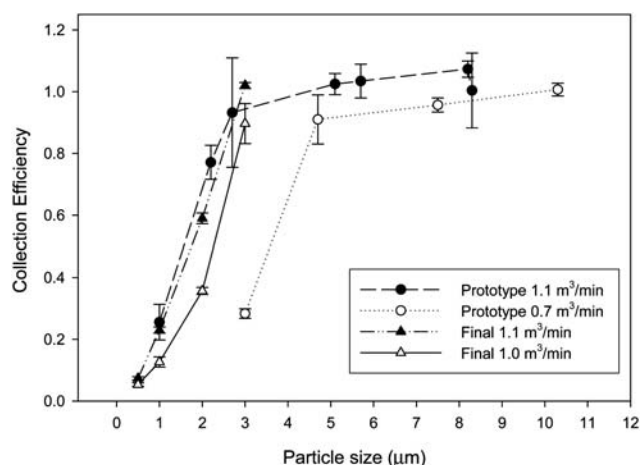


Fig. 5 Collection efficiency of the Stage 2 cyclone (prototype and final model) tested at different flow rates. Error bars represent standard deviations.

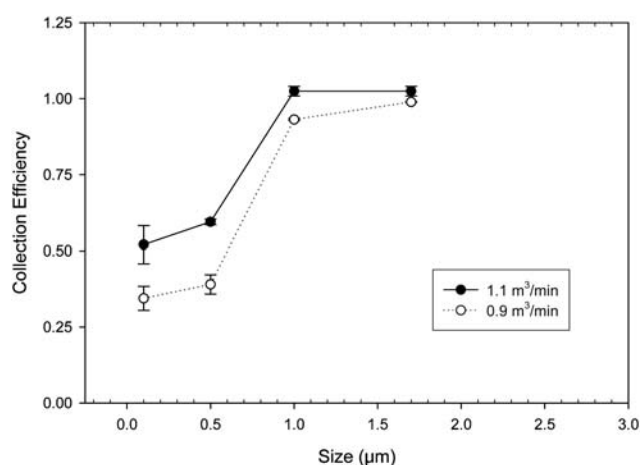


Fig. 6 Collection efficiency of the Stage 3 cyclone, measured at $0.9 \text{ m}^3 \text{ min}^{-1}$ and at $1.1 \text{ m}^3 \text{ min}^{-1}$. Error bars represent standard deviations.

from 1 to $5 \mu\text{m}$. The process was very similar to the one described above for PSL particles inside a chamber, except that there was no outlet filter to capture the smallest particles.

Flow rates through the cyclone system and reference filters were calibrated before each particle size. Tests for each particle size were conducted in triplicate. All cyclones and transitions were washed with ethyl acetate at the end of each size, and the wash was analyzed for fluorescence as previously described.

2.3.3. Validation of extraction from filters. Extraction efficiency from the AE filters was evaluated for each of the two fluorescence materials: Na-Fl particles generated by the IJAG system and polystyrene latex beads generated in the chamber. Extraction efficiency of Na-Fl from the AE filters was determined by spiking 8 filters with 1 mL of a known concentration of Na-Fl. The filter was placed on a filter holder connected to a pump drawing $1.1 \text{ m}^3 \text{ min}^{-1}$ through the filter for 5 minutes. The Na-Fl laden filters were folded and soaked in centrifuge tubes with 40 mL of recovery solution (0.1% ammonium hydroxide in deionized water). Two filter blanks were also

soaked in 40 mL recovery solution. The average extracted fluorescence from the blank filters was subtracted from that of the spiked filters, and then compared to 40 mL recovery solution spiked with 1 mL of the same Na-Fl that was used to spike the filters.

Validation of PSL extraction was performed by spiking 1 mL of a 1 : 1000 dilution of stock PSL on AE filters connected to a pump running at $1.1 \text{ m}^3 \text{ min}^{-1}$. The PSL beads were extracted from the filters by soaking the filters in 40 mL ethyl acetate. The filters were then placed on a shaker at 150 rpm to remove any remaining PSL. Blank filters were also soaked in ethyl acetate and the fluorescence subtracted from that of spiked filters. The resulting fluorescence was compared to a mixture of 40 mL ethyl acetate spiked with 1 mL of the same 1 : 1000 PSL solution used to spike the filters. Readings of fluorescence were conducted three times each hour for three hours to determine if the percent recovery of the PSL changed over time.

3. Results

3.1 Particle recovery from the filters

The mean Na-Fl recovery from the filters was $84 \pm 10\%$ (Table 1). Recovery of PSL particles from the filters ranged from 97–99.5% ($\pm 15\%$). Cyclone testing results using the IJAG generator with Na-Fl particles were adjusted using the 84% recovery value.

Validation of the method to recover PSL beads from the filters indicated that over time there was a loss in recovery percentages. Recovery after 1 hour was 97%, whereas recovery after 3 hours was 94%. Therefore during testing, filters collecting PSL particles were extracted and analyzed within one hour of sample collection.

3.2 Cyclone design

The target design dimensions of the Stage 2 coarse fraction cyclone were calculated for a D_{50} of $2.5 \mu\text{m}$ at $1.1 \text{ m}^3 \text{ min}^{-1}$,

Table 1 Summary of experiments to validate fluorescence recovery from filters

Sample type	Type of spike	Fluorescence units (FU)					Recovery (%)
		Average	Std dev.	CV (%)	Blank corr.		
Reference solution ^a	2 $2.5 \mu\text{m}$ Na-Fl	800	4	1			
Filter blank ^b	3 None	144	47	33			
Spiked filter ^b	8 $2.5 \mu\text{m}$ Na-Fl	815	64	8	671	84	
Reference solution ^c	2 $1 \mu\text{m}$ PSL	1294	1.4	0.10			
Filter blank ^b	4 None	3	1.2	38			
Spiked filter ^b	8 $1 \mu\text{m}$ PSL	1258	56.2	4.50	1255	97 ^d	
Reference solution ^c	2 $0.1 \mu\text{m}$ PSL	114.5	0.7	0.60			
Filter blank ^b	4 None	3	0.8	27			
Spiked filter ^b	6 $0.1 \mu\text{m}$ PSL	117	13.8	12	114	99.5 ^d	

^a 0.1% ammonium hydroxide in deionized water. ^b All filters are $8'' \times 10''$ AE glass fiber. ^c 100% ethyl acetate. ^d Fluorescence determined within 1 hour.

Table 2 Final design of a Stairmand high efficiency cyclone based on body diameter (D) (refer Fig. 2)

Dimension	Identifier	Stairmand proportions	Final iteration (cm)
Body diameter	D		9.91
Outlet diameter	D_o		5.5
Inlet gap		0.125D	1.24
Exit diameter	D_e	0.25D	2.48
Modified exit diameter	D_e^a	0.33D	3.303
Inlet diameter	D_i	0.5D	4.96
Outlet inside length		0.75D	7.4
Outlet outside length		1.5D	14.87
Cylinder length		2D	19.82
Cone length		2D	19.82

^a Based on Baker and Hughes, 1999.

following the model described by Moore and McFarland.³⁹ The resulting dimensions are summarized in Table 2.

3.3. Cyclone performance

Two prototype coarse fraction cyclones were tested. Tests of the first prototype at $1.1 \text{ m}^3 \text{ min}^{-1}$ using particles from 1 to $12 \mu\text{m}$ showed a sharp cut point (D_{50}) at $1.5 \mu\text{m}$ with 100% efficiency after about $3 \mu\text{m}$ (Fig. 5). A second version of the coarse fraction cyclone modified to move the D_{50} closer to $2.5 \mu\text{m}$ was tested using particles ranging in size from 0.1 to $5 \mu\text{m}$ at 1.1 and $1.0 \text{ m}^3 \text{ min}^{-1}$. Cyclone collection efficiency results are presented in Fig. 5. Based on the logistic function model, the D_{50} of the second prototype coarse cyclone was calculated to be $1.8 (\pm 0.5) \mu\text{m}$ at $1.1 \text{ m}^3 \text{ min}^{-1}$. The collection efficiency curve shifted to the right at $1.0 \text{ m}^3 \text{ min}^{-1}$, providing a D_{50} of $2.3 (\pm 0.2) \mu\text{m}$. Both models were statistically significant at $p < 0.05$. Collection efficiency results at each particle size were found to be highly reproducible with coefficients of variation ranging from 2.5% to 13%.

Results for the collection efficiency testing of the Stage 3 cyclone are presented in Fig. 6. Based on the logistic function model, the D_{50} at $0.9 \text{ m}^3 \text{ min}^{-1}$ is $0.5 \mu\text{m}$ ($p < 0.05$) and at $1.1 \text{ m}^3 \text{ min}^{-1}$ is $0.1 \mu\text{m}$ ($p < 0.05$). Extrapolating between these two

curves would result in a D_{50} close to $0.3 \mu\text{m}$ at a flow rate of $1.0 \text{ m}^3 \text{ min}^{-1}$. Particles with diameters of $0.1 \mu\text{m}$ were collected with 30% efficiency at $1.1 \text{ m}^3 \text{ min}^{-1}$. Coefficients of variation ranged from 0.1% to 11.5%.

Tests of the assembled cyclone system at $1 \text{ m}^3 \text{ min}^{-1}$ are presented in Fig. 7. Tests with PSL beads indicate that when assembled as a system, the Stage 2 cyclone retains particles between 10 and $3.5 \mu\text{m}$, while the Stage 3 cyclone collects particles between 3.5 and $\sim 0.3 \mu\text{m}$.

Limited environmental sampling to date confirms that the cyclone system collects hundreds of milligrams of ambient PM. In the first sites of deployment the cyclone has collected between 400 and 1500 mg of “fine” PM and between 110 and 1200 mg of “coarse” PM over the course of 5 weeks on each site.

4. Discussion

The cyclone system was developed for the collection of bulk quantities of ambient particulate matter without the complications of a sampling substrate. The goal of this sampler is to collect size segregated dry bulk PM samples in large enough quantity to be useful for toxicological studies, as well as for a range of chemical characterizations without the need to collect multiple collocated samples or to remove the PM from a sampling substrate. The final design, depicted in Fig. 8, is a sequential system that consists of a high-volume PM_{10} inlet, a custom-designed coarse fraction cyclone and a cyclone that collects particles down to $\sim 0.3 \mu\text{m}$. By eliminating particles with mean aerodynamic diameter $> 10 \mu\text{m}$ from the air stream, the Stage 2 cyclone, as currently designed, collects particles representative of the coarse fraction PM (particles with aerodynamic equivalent diameter equal to or smaller than $10 \mu\text{m}$ and larger than approximately $3.5 \mu\text{m}$ $\text{PM}_{10-3.5}$). Subsequent designs will be manufactured and tested to shift the cut point closer to $2.5 \mu\text{m}$. The PM collected by the Stage 3 cyclone is representative of fine PM, composed of particles with mean aerodynamic diameter smaller than $3.5 \mu\text{m}$ and larger than $0.3 \mu\text{m}$ ($\text{PM}_{3.5-0.3}$). Each element of the sampler is coupled to the next with smooth connectors to minimize PM losses, and are connected using quick release V-clamps (MHLA, Kaestner Company, Baltimore, MD) for easy assembly.

The major limitation of this system is that cyclone technology does not allow for the collection of ultra-fine particles, which may be associated with significant health consequences due to their high particle number, surface area and bioavailability. As inertial collectors cyclones are inefficient collectors of particles with low mass (*i.e.*, ultrafines). Recovery of the ultrafine particles was considered in the developmental stages of the design process. The use of an after-filter was tested, resulting in a large pressure drop; since no pump could be found to overcome the increase in pressure at the target flow rate, this option was abandoned. Another option explored in the design stage was the use of an electrostatic precipitator. However, this technology is not well developed for the designed flow rate nor for the size range and low concentrations found in environmental samples. In addition, the mass of particles smaller than $0.1 \mu\text{m}$, if collected, would not have been adequate for toxicological tests and characterization evaluations. From our laboratory tests of the cyclone system characterization, the Stage 3 cyclone collects between 30% and

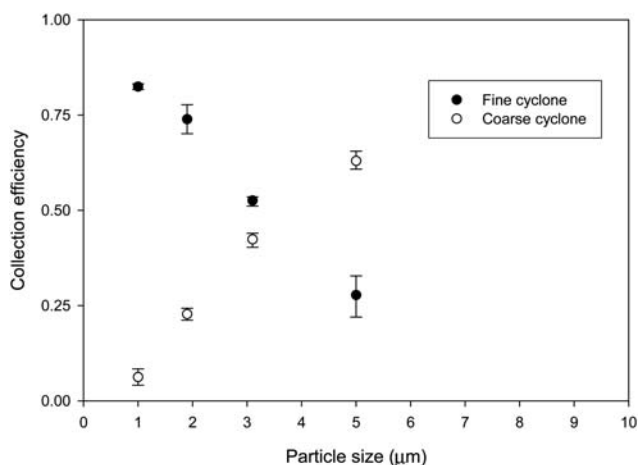


Fig. 7 Collection efficiency for each of the cyclones when tested as a system, at $1 \text{ m}^3 \text{ min}^{-1}$.

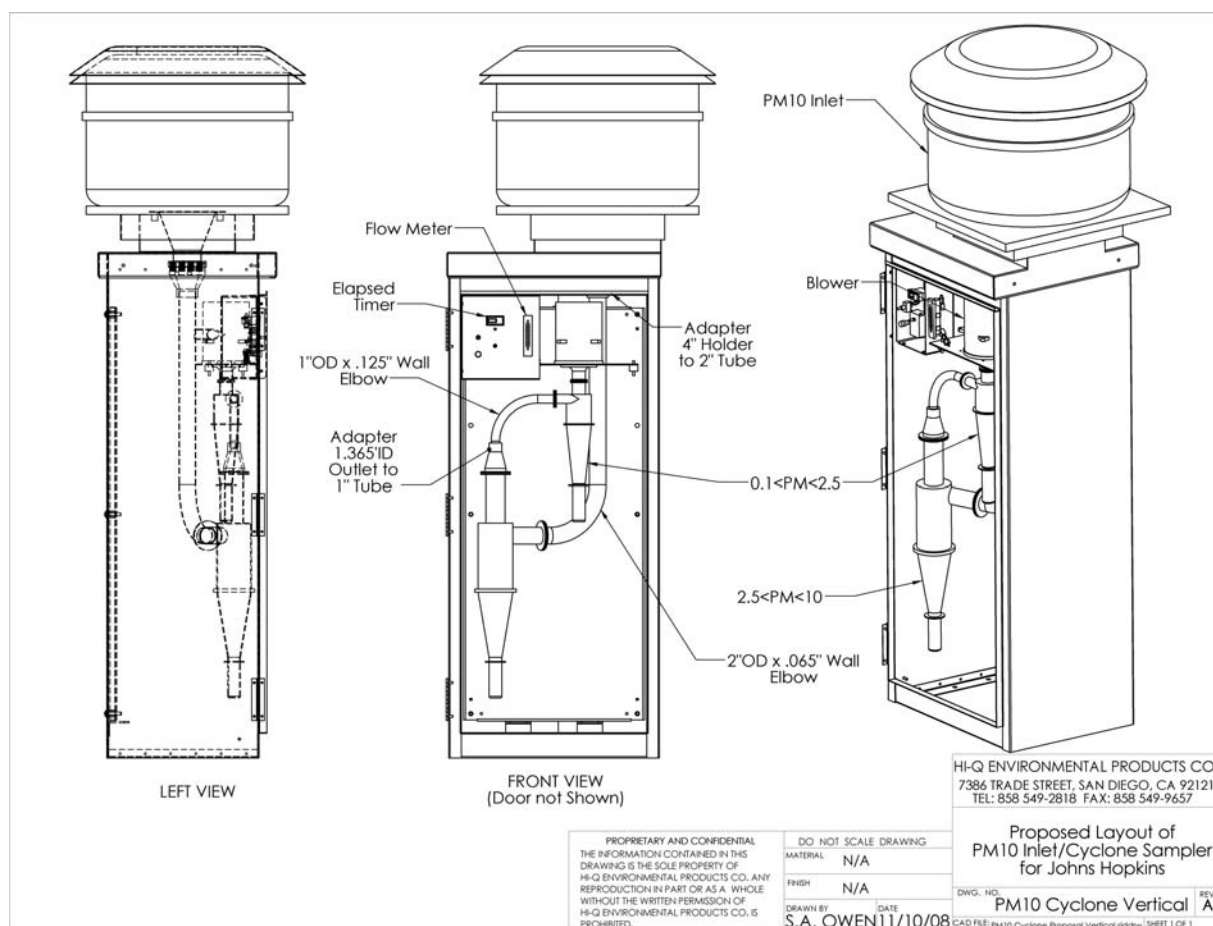


Fig. 8 Schematic of the Sequential Cyclone Sampler. Final prototype.

50% of particles with $0.1\ \mu\text{m}$ in diameter (Fig. 6). This means that our system exhausts most particles smaller than $0.1\ \mu\text{m}$ in diameter. Based on published results by Wallace and colleagues,⁴² we have calculated that PM smaller than $0.1\ \mu\text{m}$ comprises 13% to 20% of the fine fraction mass. The cyclone system, therefore, collects ~80 to 87% of typical fine PM.

The primary advantage of this system is the absence of a sampling substrate that must be removed before delivery of the PM to any toxicological or chemical assay. For toxicological studies exploring the mechanisms of injury associated with exposure to ambient particulate matter, milligram quantities of particulate matter can be needed if the studies are being conducted *in vivo*. This system allows for the collection of hundreds of milligrams of ambient PM over relatively short periods of time. In an urban environment such as Baltimore, MD, where weekly average $\text{PM}_{2.5}$ concentrations during the summer months can be on the order of $20\ \mu\text{g m}^{-3}$, one week of sampling will yield approximately 200 mg bulk fine PM. A month-long sampling effort would yield adequate mass for complementary chemical characterization for constituents including trace elements, elemental carbon, and non-volatile inorganic and organic compounds.

This system as designed is being used in a nation-wide study of PM toxicity. PM samples from cities across the US are being collected to compare toxicity in *in vivo* and *in vitro* models.

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