Asian Journal of Atmospheric Environment
Vol. 12, No. 1, pp. 78-86, March 2018
doi: https://doi.org/10.5572/ajae.2018.12.1.078
ISSN (Online) 2287-1160, ISSN (Print) 1976-6912

Technical Information

Development of a High-Volume Simultaneous Sampler for Fine and Coarse Particles using Virtual Impactor and Cyclone Techniques

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ABSTRACT

Filter-based sampling techniques are the conventional way to collect particulate matter, but particles collected and entangled in the filter fibers are difficult to be removed and thus not suited for the following cell- and animal-based exposure experiments. Collecting aerosol particles in powder form using a cyclone instead of a filter would be a possible way to solve this problem. We developed a hybrid virtual-impactor/cyclone high-volume fine and coarse particle sampler and assessed its performance. The developed system achieved 50% collection efficiency with components having the following aerodynamic cut-off diameters: virtual impactor, 2.4 μm; fine-particle cyclone, 0.18-0.30 μm; and coarse-particle cyclone, 0.7 μm. The virtual impactor used in our set-up had good $PM_{2.5}$ separation performance, comparable to that reported for a conventional real impactor. The newly developed sampler can collect fine and coarse particles simultaneously, in combination with exposure testing with collected fine- and coarse- particulate matter samples, should help researchers to elucidate the mechanism by which airborne particles result in adverse health effect in detail.

Key words: Cyclone, Virtual impactor, Coarse particles, $PM_{2.5}$, High-volume air sampler, Powder form

1. INTRODUCTION

Ambient particulate matter has been concerned around the world because of its adverse health impact when inhaled. In particular, $PM_{2.5}$ (fine particles with 50% collection efficiency at an aerodynamic cut-off diameter of 2.5 μm) has been paid much attention for its health hazard by short- and long-term exposure (Pope *et al*., 1995; Dockery *et al*., 1993). The OECD predicts that atmospheric pollution will be the single leading cause of early death by 2050, with cases involving particulate matter expected to be most severe, rising to 3.6 million deaths per year, and more than double 2010 levels (OECD, 2012). Concerns of adverse health effect due to particulate matter have led several countries to establish environmental quality standards and regulations for $PM_{2.5}$ (USEPA, 2013; Ministry of Environment, Japan, 2009; EU, 2008). The definitions of $PM_{2.5}$ utilized in these regulations depend solely on particle aerodynamic diameter, and do not distinguish based on differences in chemical composition. However, the toxicity of particulate matter likely depends strongly on its physical and chemical characteristics. This makes it imperative to assess its health impact based not only on the mass of particles, but also on differences in chemical composition, if we are to elucidate the mechanism by which it causes adverse health effect. Much research to date has evaluated the hazards of $PM_{2.5}$ by exposing cells and/or animals to extracts or individual components (Kumagai and Taguchi, 2007; Yanagisawa *et al*., 2006; Hiyoshi *et al*., 2005; Takano *et al*., 1997). At present, however, few studies directly evaluate health effects by exposing the entire particulate matter (particles themselves) in the ambient air. Filter-based sampling techniques are the conventional way to collect particulate matter, but particles collected and entangled in the filter fibers are difficult to be removed and thus not suited for the following cell- and animal-based exposure experiments. Moreover, even if sufficient quantities are recovered using physical (scraping the filter surface or vibrating the filter) or chemical (extracting using organic- or inorganic solvents) means, the

effects of contamination from the filter itself are unavoidable (Van Winkle *et al*., 2015). Clearly, several problems stand in the way of assessing the true effects of particulate matter in the air by using filter for particle collection. Another major problem with filter-based collection is the unexpected artifacts – adsorption of gaseous substances, chemical reactions, volatilization of semi-volatile compounds, etc. – which can alter the levels of the components in samples from those present in the ambient air (Hasegawa, 2016; Kameda *et al*., 2010).

Aiming to solve these problems, our laboratory has developed a device in which $PM_{2.5}$ is collected in powder form using a cyclone instead of a filter (Okuda *et al*., 2015a). This device separates $PM_{2.5}$ from air using a real impactor having an impaction plate at the inlet (Kaneyasu and Yamamoto, 2016; Kaneyasu, 2010) and collects them using a cyclone downstream (Rule *et al*., 2010). Particles removed from the airflow by the cyclone are collected in a collection bottle, meaning that unlike filter-based devices, the airflow does not continue to pass over particle surfaces after their collection. Mice exposure experiments using $PM_{2.5}$ powder obtained using this device caused airway inflammation and hypersensitivity, suggesting $PM_{2.5}$ may raise the risk of developing asthma (Ogino *et al*., 2017).

While this cyclone-based $PM_{2.5}$ sampling system is useful, there is much room for improvement. For instance, real impactors utilize an impaction plate, from which particles can rebound and move downstream. This could potentially be prevented by replacing it with a virtual impactor at the inlet of the system. A virtual impactor contains two paths for air flow. A major portion of the flow is diverted 90° away from the inlet nozzle, then small particles (in this study, $PM_{2.5}$) with low inertia follow the major flow streamlines. Large particles with greater inertia deviate from the flowlines and continue moving axially in their forward path to the downstream with the minor flow, which allow users to collect coarse particles from ambient air. Coarse particles like yellow sand are known to cause inflammation in the body separate from the effects of fine particles like $PM_{2.5}$ (Song *et al*., 2016; Ichinose *et al*., 2008). Accordingly, the ability to simultaneously collect coarse and fine particles, in combination with exposure testing with collected samples, should help researchers to elucidate the mechanism by which airborne particles result in adverse health effect in detail.

In the present study, a hybrid virtual-impactor/cyclone high-volume simultaneous sampler for fine and coarse particles was developed, and its characteristics of separation and collection of the particles was evaluated.

2. MATERIALS AND METHODS

2.1 Design and Development of a Hybrid Virtual-impactor/cyclone High-volume Simultaneous Sampler for Fine and Coarse Particles

The sampling system was designed as detailed below. Particle diameter with 50% collection efficiency (Dp_{50}) was calculated according to Equation 1 (JIS Z7152, 2013; ISO 13271, 2012).

$$Dp_{50} = \sqrt{\frac{9 \times \mu \times Stk_{50} \times N \times \pi \times (D_0)^3}{4 \times C \times Q_0 \times \rho_{0.P}}} \quad (1)$$

Stk_{50}: Stokes number corresponding to Dp_{50}
N: Number of particle acceleration nozzles
D_0: Inner diameter of nozzles [m]
C: Cunningham correction factor
($= 1.063$ when $Dp_{50} = 2.5 \times 10^{-6}$ m)
$\rho_{0.P}$: Density of particle, unit density ($= 1{,}000$ kg/m^3)
μ: Viscosity of air ($= 1.8 \times 10^{-5}$ Pa·s)
Q_0: Suction flow rate [m^3/s]

The separation performance of a virtual impactor is governed by the inertia of the particles in the air stream. This concept is represented by the Stokes number, a dimensionless number that represents particles' affinity to the expected fluid flow. In the context of particle samplers, devices with higher Stokes numbers can separate finer particles. Equation 2 gives Stk_{50}, the Stokes number corresponding to Dp_{50}.

$$Stk_{50} = \frac{\rho_{0.P} \times C \times v \times (Dp_{50})^2}{9 \times \mu \times D_0} \quad (2)$$

Dp_{50}: Particle diameter with 50% collection efficiency
($= 2.5 \times 10^{-6}$ m)
v: Gas velocity in the particle acceleration nozzle
($= 4 \times Q_0 / \pi \times Dc^2$ m/s)

Dp_{50} was set equal to 2.5 μm, corresponding to the diameter of $PM_{2.5}$. However, Stk_{50}, suction flow Q_0, and particle acceleration nozzle number N and inner diameter D_0 are undefined, and must be configured separately when designing the actual system. JIS Z7152 establishes a reference range for Stk_{50} of 0.4-0.5, and many past studies have successfully used a Stk_{50} of approximately 0.5 for virtual impactors for $PM_{2.5}$ (Wang *et al*., 2013; Wada *et al*., 2009; Loo and Cork, 1988). Our set-up therefore adopts $Stk_{50} = 0.5$, the upper bound of the standard range prescribed by JIS. Next, total flow rate (Q_0) was determined. In a similar system with a cyclone installed after the main flow stream, good performance was observed for $Dp_{50} = 0.1$-0.3 μm at a flow rate = 1,100 L/min (Okuda

and Isobe, 2017; Okuda *et al*., 2015a). The flow rate of the major flow (Q_1) was therefore set to 1,100 L/min. Similarly, good separation performance has been observed in a single-stage virtual impactor having a major flow rate (Q_1) of 55 L/min and minor flow rate (Q_2) of 5 L/min (i.e., $Q_0 = 60$ L/min) (Okuda *et al*., 2015b). These values were multiplied by 20 to establish the default settings for the current system (Q_0: 1,200 L/min, Q_1: 1,100 L/min, and Q_2: 100 L/min). Therefore, the virtual impactor used in this study had 20 nozzles (i.e., $N = 20$). Nozzle inner diameter D_0 was then calculated as 4.7 mm using Eq. 2, assuming $Q_0 = 1{,}200$ L/min and $N = 20$. Linear velocity of the gas flow within the nozzles was 58 m/s, a value comparable to previous studies (Kim *et al*., 2002). With this, the design of the major components governing the separation ability of virtual impactors was completed.

Next, we determined additional specifications for the system. JIS Z7152 specifies that the ratio of the inner diameter of the particle-sampling nozzle (D_1) to that of the acceleration nozzle (D_0) should be about 1.33. The system was therefore configured with $D_1 = 6.35$ mm ($D_1/D_0 = 1.35$). Similarly, it stipulates that the ratio of nozzle length l_0 to D_0 should be less than 2.5. The system was therefore configured with $l_0 = 10$ mm ($l_0/D_0 = 2.13$). Moreover, it prescribes a range of 0.8-2 for the ratio of the distance between the outlet of the accelerator nozzle and the inlet of the sampling nozzle s (simply "nozzle distance" below) to D_0. We assumed this would be variable in the system, and set a range of $1.9 < s < 12.4$ mm ($= 0.4 < s/D_0 < 2.6$).

A stainless-steel cyclone was connected to the system further down the main flow stream for collecting fine particles in powder form ("fine-particle cyclone" below). The model used has a 50% cut-off diameter of 0.23 μm at a flow rate of 1,100 L/min, and an average inner-wall surface roughness Ra of 0.08 μm (Okuda and Isobe, 2017). A circular cone attachment was fitted between the bottom of the cyclone and the collection bottle in order to prevent particles collected in the bottle from resuspending up into air stream (Okuda and Isobe, 2017). $PM_{2.5}$ was collected in a wide-mouth amber glass jar attached to the bottom of the cyclone (Thermo Fisher Scientific Inc., I-Chem 100, 250 mL volume). A back-up filter holder and mass flow meter (Azbil Corp., CMG400) and blower (Showa Denki Co. Ltd., U2S-150) were connected downstream of the fine-particle cyclone, with blower flow rate controlled by an inverter (Fuji Electric Co. Ltd., FRN2.2C1S-7J12). In addition, a commercially available PTFE-coated aluminum cyclone was connected to the system further down the minor flow stream for collecting coarse particles in powder form (URG Corp., URG-2000-30EH: "coarse-particle cyclone" below). According to manufacturer specifications, $Dp_{50} = 2.5$ μm at a flow rate of 16.7 L/min, and $Dp_{50} = 1.0$ μm at 50 L/min. Coarse-particle powder was collected in an amber glass jar attached to the bottom of the cyclone. A back-up filter holder and flow meter (Azbil Corp., CMS200) and vacuum pump (Nitto Kohki Co., Ltd., VP0940T) were connected downstream of the coarse-particle cyclone. The flow rate of the secondary stream was controlled by a valve (Fujikin Inc., DH-12). Fig. 1 shows a schematic diagram of the hybrid virtual-impactor/cyclone high-volume simultaneous sampler for $PM_{2.5}$ and coarse particles developed in this study. The system was installed on the rooftop of a six-floor building on the Yagami campus of Keio University (35.56°N, 139.65°E, 30 m above the ground).

For purposes of comparison, $PM_{2.5}$ were also collected using a conventional high-volume air sampler equipped with a 2.5 μm-cut impactor (SIBATA HV-RW) and quartz-fiber filter, operated at a flow rate of 740 L/min (Kaneyasu and Yamamoto, 2016; Kaneyasu, 2010). $PM_{2.5}$ collected by cyclone in our system were compared with those collected using the conventional method, in terms of their chemical properties.

2.2 Separation Performance of Impactor and Cyclones

An aerodynamic particle sizer (APS, TSI Inc., Model 3321) and scanning mobility particle sizer (SMPS, TSI Inc., Model 3910) were used to measure particle number concentration in five locations in the system: before the system inlet, and (in both flow streams) between the virtual impactor and the cyclone, and after the cyclone. Isokinetic sampling probes were used to adjust the velocity of the air going into the sampling probe nozzle to that of the undisturbed air flow at that point. Appropriate diameter of nozzles were selected according to the flow rates of the target flow and those of measurement instruments. The penetration at the virtual impactor was calculated as the ratio of particle number at the inlet air versus that in the major flow of virtual impactor; cyclone penetration was calculated similarly by comparing the inlet to outlet. APS data was used to evaluate the performance of the virtual impactor and the coarse-particle cyclone, while SMPS data was used to evaluate that of the fine-particle cyclone. Virtual impactor and coarse-particle cyclone performances were evaluated using ambient aerosol particles. The fine-particle cyclone performance was evaluated using ambient aerosol particles, and with atomized polystyrene latex particles (PSL) and NaCl generated by an atomizer (TSI Inc., Model 3079) through a diffusion dryer (RH of the outlet air was less than 10%).

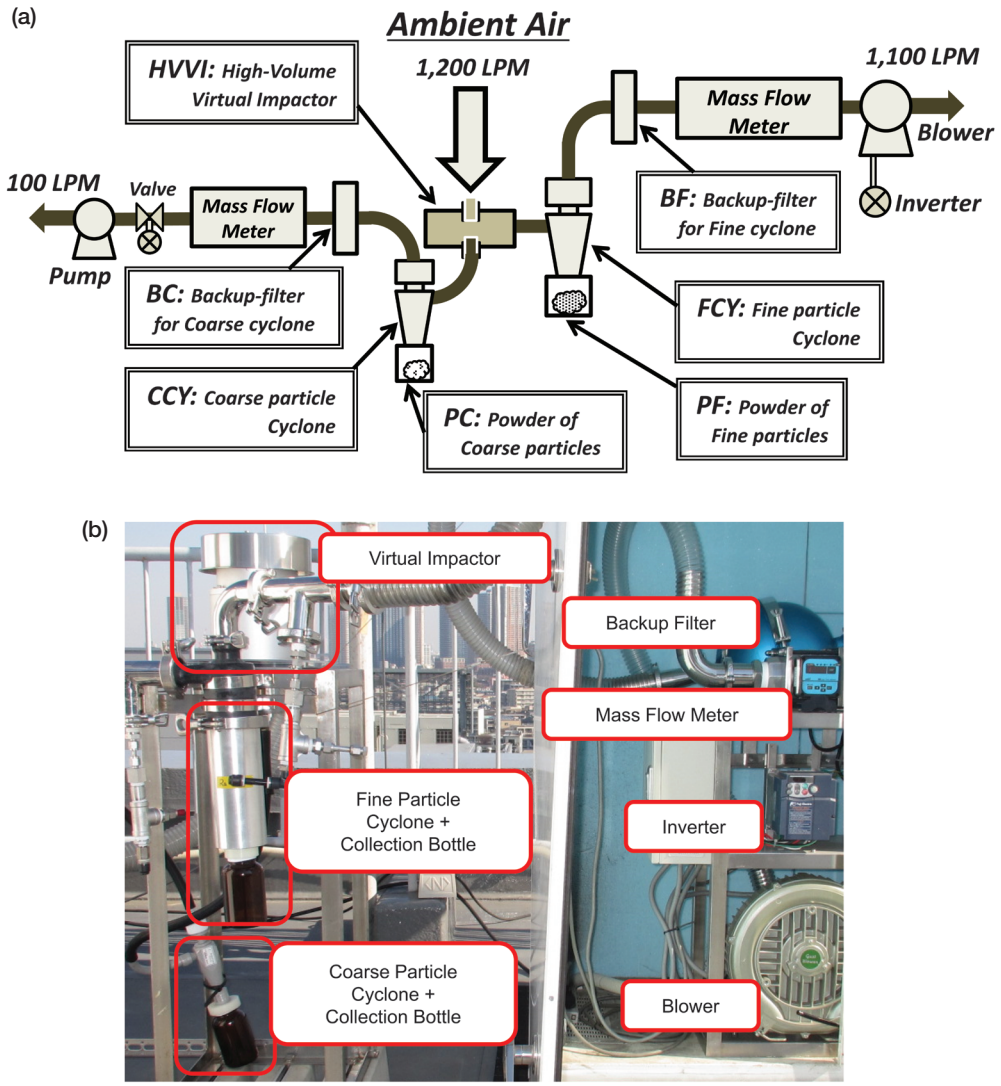

Fig. 1. High-volume simultaneous sampler for $PM_{2.5}$ and coarse particles; (a) schematic diagram, and (b) photographs of each device in the system.

2. 3 Chemical Analysis of Collected Particles

Aerosol particles collected by the cyclones were subjected to chemical analysis according to the methods below. Elemental carbon (EC) and organic carbon (OC) were measured by thermal/optical carbon analysis using an ECOC analyzer (Sunset Laboratory Inc.) (Okuda, 2013a). Water-soluble ionic components were extracted with deionized water and measured by ion chromatography (Okuda *et al*., 2013b). Elemental composition was measured using an energy-dispersive X-ray fluorescence analyzer (EDXRF, Rigaku Corp., EDXL300/NEX CG) (Okuda *et al*., 2014; Okuda *et al*., 2013a, b, c; Okuda and Hatoya, 2013d). Approximately 20 mg of the collected powder was compressed with ~100 mg of cellulose powder (Funacel II, Funakoshi Co., Ltd.) at 50 kN to form two-layer pellet, then the samples thus prepared were analyzed by EDXRF in vacuum (approx. 1 Pa). The quantitative method used was Fundamental Parameter coupled with O-Balance method, and the analysis time was set at 15 min per sample. The certified reference material of urban aerosol, NIES CRM#28 was analyzed together with test samples to check the validity of the analysis data. The ratio of measured and certified values for each element was usually in the range of 0.8-1.2, and measurement precision – represented as the coefficient of variation (= standard deviation / mean) – was less than 20% (Table 1). Analytical methods for the filter samples were almost the same as mentioned above except for elemental analysis that was carried out by using

Table 1. Measured and Certified (or Reference) values of elements in a reference material (NIES CRM#28, Urban Aerosols) obtained by EDXRF analysis (n = 5).

Element	Certified/ Reference value [ppmw]	Measured value [ppmw]	SD	Measured/ Certified ratio
Mg	14000	12643	3537	0.90
Al	50400	59693	9007	1.18
Si	149000	158451	26761	1.06
P	1450	923	211	0.64
S	39100	57638	11196	1.47
Cl	8070	6639	2559	0.82
K	13700	11180	2601	0.82
Ca	66900	63612	14428	0.95
Ti	2920	2688	649	0.92
V	73.2	67.2	16.6	0.92
Cr	56.5	60.8	14.6	1.08
Mn	686	562	126	0.82
Fe	29200	22243	4732	0.76
Ni	63.8	55.5	11.7	0.87
Cu	104	82.8	15.8	0.80
Zn	1140	932	178	0.82
Pb	403	386	67	0.96

EDXRF with FP for thin film (filter) samples (Okuda *et al*., 2014, 2013c, d; Okuda and Hatoya, 2013d).

3. RESULTS AND DISCUSSION

3. 1 Separation Performance of Virtual Impactor

Penetration curves for the virtual impactor for different nozzle distances are shown in Fig. 2a ($s = 1.9$, 4.9, and 7.9 mm). Note that the penetration did not reach 1.0 for the smaller particles since the air flow volume per given time for the major flow of virtual impactor (1,100 L/min) was essentially smaller than that for the inlet (1,200 L/min), and the smaller particles also go along with the minor flow (100 L/min). Dp_{50} was 2.0 μm when s was 1.9 mm, and the slope of the line given by Dp_{20}/Dp_{80} was 2.3. This failed to meet JIS Z8851 reference standards ($Dp_{50} = 2.5 \pm 0.2$ μm; slope ≤ 1.5). When s was 4.9 mm, however, Dp_{50} was 2.4 μm and Dp_{80}/Dp_{20} was 1.4. This performance met the JIS Z8851 standards. When s was 7.9 mm, Dp_{50} was 2.7 μm and Dp_{80} was indeterminate. The nozzle distance $s = 4.9$ mm was therefore chosen as the optimal configuration value. In this case, the ratio of nozzle distance to the diameter of inlet nozzle (s/D_0) was 1.0.

Next, the virtual impactor's performance at $s = 4.9$ mm was compared with a real impactor, for which suitable $PM_{2.5}$ separability has been reported at a flow rate of 740 L/min (Kaneyasu, 2016, 2010) (Fig. 2b). The virtual impactor's separation curve when operated at a flow rate of 1,200 L/min was nearly identical to the real impactor's curve when operated at a flow rate of 740 L/min, both meeting JIS Z8851 standards. The results show that the virtual impactor used in our system performs comparably to a real impactor used in conventional sampling systems.

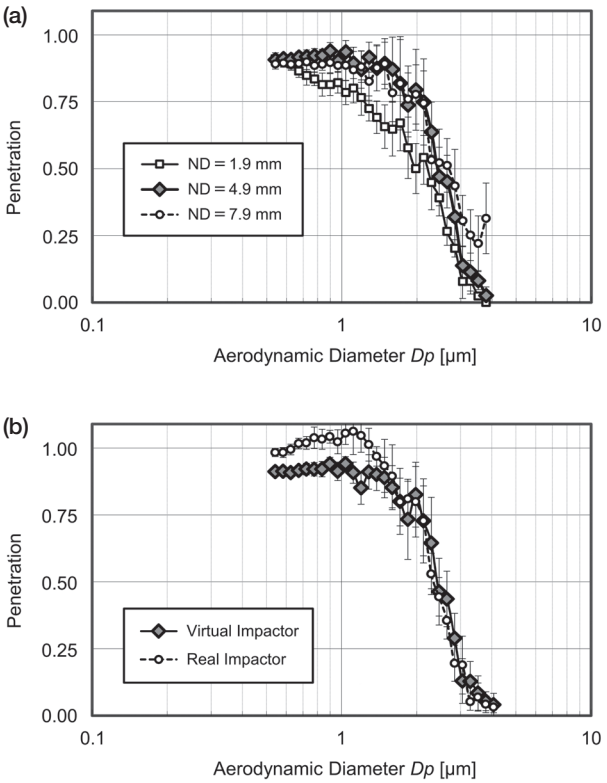

Fig. 2. Penetration curves for (a) the virtual impactor with varying nozzle distance, and (b) virtual impactor and real impactor, using ambient aerosol particles.

3. 2 Separation Performance of Fine-particle Cyclone

Penetration curves for the fine-particle cyclone are shown in Fig. 3. Dp_{50} varied from 0.18 μm to 0.30 μm. These values agree well with the values for ambient aerosol particles (Okuda *et al*., 2015a) and NaCl particles (Okuda and Isobe, 2017) observed in past researches. No significant differences in the separation characteristics of the cyclone among using PSL, NaCl and ambient aerosol particles. The SMPS used in this study was a simple and portable model so that the size resolution may not be sharp enough for obtaining more clear penetration curves for the cyclone. In any case, the cyclone installed for fine particle collection in the present system had comparable separation performance to that observed in previous reports.

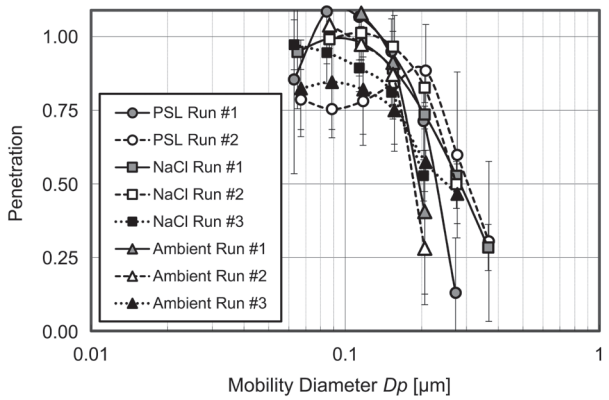

Fig. 3. Penetration curves for the fine-particle cyclone using polystyrene latex (PSL), NaCl and ambient aerosol particles.

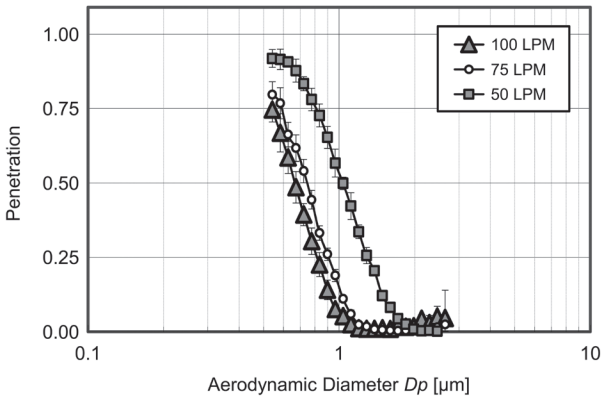

Fig. 4. Penetration curves for the coarse-particle cyclone using ambient aerosol particles.

3. 3 Separation Performance of Coarse-particle Cyclone

Penetration curves for the coarse-particle cyclone at variable flow rates are shown in Fig. 4 (50, 75, 100 L/min). Dp_{50} was 1.0 μm when flow rate = 50 L/mm, agreeing with published manufacturer specifications. Moreover, Dp_{50} fell with increasing air flow, falling to 0.68 μm when flow rate = 100 L/min. The collection efficiency for 1-μm-diameter particles was calculated to be about 95% when flow rate = 100 L/min. According to the data presented the separation performance of the virtual impactor used in this study, nearly all particles larger than 1 μm in diameter are led to the minor flow and go into the coarse-particle cyclone (see section 3.1). Our results suggested that our set up is capable of collecting sufficient quantities of coarse particles via the minor flow stream using an URG-2000-30EH cyclone (the coarse-particle cyclone) with a flow rate of 100 L/min.

3. 4 Chemical Composition of Collected Particles: Proposed vs. Conventional Sampling

Fig. 5a shows the results of chemical analysis of particulate matter samples collected using the developed system and a conventional filter sampling from April to May 2016. Note that the results were expressed as ppmw in each particle mass, meaning these values will be affected by other components that were not analyzed in this study (such as water content). This makes it difficult to compare these values each other.

The chemical compositions of particulate matter collected from the fine- and coarse-particle cyclones were compared. Na^+, Ca^{2+}, Si, and Fe are thought to exist mainly in coarse particles in air: these elements comprised a larger percentage of collected particulate matter in the coarse-particle cyclone samples. In contrast, EC, Zn, and Pb are thought to exist mainly in fine particles in air: these compounds comprised a larger percentage of collected particulate matter in the fine-particle cyclone samples. Thus, it seems that the coarse and fine particles separated by the system had chemical compositions characteristic of their respective sizes. However, the concentrations of several components such as ammonium, nitrate, sulfate, and vanadium were high in the coarse-particle cyclone samples, whereas these were generally considered distributing in much smaller particle size. It was very difficult to explain this discrepancy convincingly. One possible reason would be that these components adhered onto the inner wall of the cyclones. The fine-particle cyclone was larger than the coarse-particle cyclone, therefore the larger surface area of cyclone inner wall may retain much more adherable components.

The chemical compositions of particulate matter collected with the fine-particle cyclone were compared with those of $PM_{2.5}$ collected in the quartz-fiber filter using the conventional method. Levels of individual compounds and elements varied somewhat between the samples, with some comparable and others dissimilar. Notably, levels of NH_4^+ and SO_4^{2-} were markedly higher in filter-collected samples, while chloride and nitrate were more deplete in the case of filter sampling. Again, this discrepancy was difficult to explain, but one possibility was that the cyclone had a 50% cut-off diameter of 0.18-0.30 μm, so it may not have completely collected smaller aerosol particles such as ammonium sulfate. However, EC content did not decrease markedly in the particulate matter collected in the fine-particle cyclone, even though EC is aggregate of soot particles only a few tens of nanometers in diameter. Another possible explanation for the difference

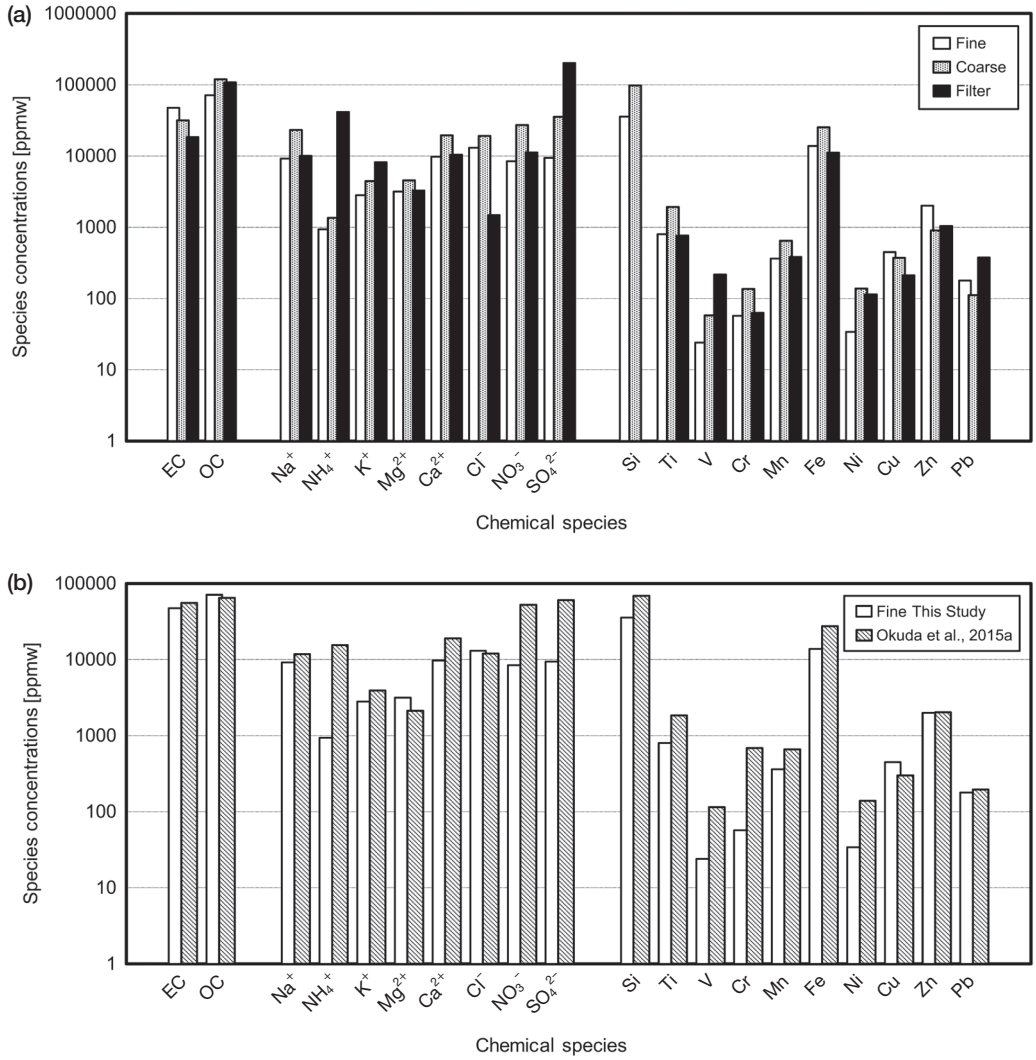

Fig. 5. (a) Chemical composition of particulate matter collected by the fine- and coarse-particle cyclones, and a filter sampling by conventional method, and (b) comparison of the results between this study and Okuda *et al.* (2015a).

would be positive sampling artifacts. During the sampling, particles collected by the cyclone are isolated from the dynamic air flow passing through it. On the other hand, particles remain trapped in the filter in the conventional method, which is in constant contact with the dynamic air flow. Volatile organic compounds (VOC), acidic and basic gases, and other gaseous components are not removed from the air flow: they are constantly fed through the filter, which means they could have been adsorbed into the filter during sampling. It could also have been the opposite reason: the influence of negative sampling artifacts. The systems were run continuously for 2-3 weeks to collect the aerosol particles, and it is possible that the particulate matter lost some of its specific constituents due to physical or chemical reactions in the meantime. VOC, for example, are likely to be lost: in this case, this rationale would not apply to sulfate particles, but it could apply to ammonium salts such as ammonium nitrate. It seems that the negative artifact would be a possible reason for the depletion of chloride and nitrate in the particles collected on the filter.

Finally, chemical composition of particulate matter collected by the fine-particle cyclone was compared to that presented by Okuda *et al.* (2015a), in order to evaluate whether the particle rebound from the real impactor could reduce when using the virtual impactor (Fig. 5b). Some components such as Ca^{2+}, Si, Ti and Fe were low in this study, suggesting the rebound of larger particles might be reduced. However, the concentrations of several components such as ammonium, nitrate and sulfate were also reduced in this study. It

was very difficult to compare the results of this study to the previous paper since the sampling period was different. It needs further experiments to understand properly the chemical characteristics of the particles collected by the virtual impactor-cyclone system presented in this study.

4. CONCLUSIONS

We developed a hybrid virtual-impactor/cyclone high-volume fine and coarse particle sampler and assessed its performance. The developed system achieved 50% collection efficiency with components having the following aerodynamic cut-off diameters: virtual impactor, 2.4 μm; fine-particle cyclone, 0.18-0.30 μm; and coarse-particle cyclone, 0.7 μm. The virtual impactor used in our set-up had good $PM_{2.5}$ separation performance, comparable to that reported for a conventional real impactor. The exposure testing with collected fine- and coarse-particulate matter samples should help researchers to elucidate the mechanism by which airborne particles result in adverse health effect in detail.

The chemical compositions of particulate matter collected with the fine- and coarse-particle cyclones were compared with those of $PM_{2.5}$ collected in the quartz-fiber filter using the conventional method. It seems that the coarse and fine particles separated by the system had chemical compositions characteristic of their respective sizes. However, the concentrations of several components such as ammonium, nitrate and sulfate were low in the fine-particle cyclone samples. It was very difficult to explain this discrepancy convincingly, therefore it needs further experiments to understand properly the chemical characteristics of the particles collected by the virtual impactor-cyclone system presented in this study.

ACKNOWLEDGEMENT

This study was supported partly by the Environment Research and Technology Development Fund (5-1651) of the Environmental Restoration and Conservation Agency, the Matching Planner Program from Japan Science and Technology Agency, JST (MP28116789653), JSPS KAKENHI Grant Numbers JP17H04480 and JP26340010, JKA foundation (27-127), Steel Foundation for Environmental Protection Technology, Grant for Environmental Research Projects from The Sumitomo Foundation, “Super Egg Program” System for the Development of Future Scientific Technologies of Keio University, the Keio Leading-edge Laboratory of Science and Technology (KLL) Specified Research Projects, and Keio Gijuku Academic Development Funds.

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(Received 30 August 2017, revised 16 December 2017, accepted 18 December 2017)