Development of a High-Volume PM$_{2.5}$ Particle Sampler Using Impactor and Cyclone Techniques

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ABSTRACT

Detailed mechanisms of the cellular biochemical reactions associated with the toxicity of PM$_{2.5}$ have not been elucidated well so far, because it is difficult to collect a sufficient amount of PM$_{2.5}$ particles to carry out toxicity assays using cells. A high-volume PM$_{2.5}$ particle sampler using the impactor and cyclone techniques has been developed in this study. The 50% cut-points of the impactor and cyclone for ambient aerosols at 1,100 L/min of air flow were 2.0 $\mu$m and 0.15–0.18 $\mu$m, respectively. The separation characteristics of the impactor were similar to those of the WINS impactor. The contents of EC, which is an aggregate/agglomerate of primary particles with sizes less than 100 nm, in the cyclone particles was similar to that in the filter particles. The contents of organic matter and ammonium in the particles collected using the cyclone system were lower than those in the particles collected using traditional filter sampling. We propose that the high-volume PM$_{2.5}$ particle sampler developed in this study can collect a large amount of aerosol particles in a “powder form” and can also avoid certain sampling artifacts caused by the adsorption of volatile organic compounds and gaseous ammonia to particles on the filter media. The sampling device developed in this study allows researchers to collect a sufficient amount of PM$_{2.5}$ particles for cell exposure studies without the use of filters.

Keywords: Ambient aerosols; Cyclone; Inertial impactor; In vitro bioassay; Particle form.

INTRODUCTION

Exposure to atmospheric aerosols is a serious concern for human health (Dockery et al., 1993; Pope et al., 1995; IARC, 2013). PM$_{2.5}$ (particulate matter that passes through a size-selective inlet with a 50% efficiency cut-off at an aerodynamic diameter of 2.5 $\mu$m) has been regulated worldwide over the last ten to twenty years (European Parliament and of the Council, 2008; Ministry of Environment, Japan, 2009; USEPA, 2013). These regulations have been set based on numerous epidemiological studies, and many of which have reported adverse health effects due to PM$_{2.5}$. However, detailed mechanisms of the cellular biochemical reactions associated with the toxicity of PM$_{2.5}$ have not been elucidated well so far. The reasons for that are as follows: (1) PM$_{2.5}$ consists of thousands of chemical species that need to be examined to determine their biological effects; (2) A wide variety of cells may be affected; (3) There are many bioassay methods for the examination of the toxicological effects of PM$_{2.5}$; (4) It is difficult to isolate a sufficient amount of PM$_{2.5}$ particles to carry out toxicity assays using cells. This is a critical limitation in exposure studies. Some studies have used aerosol concentrators in order to carry out exposure studies on laboratory animals (Maciejczyk et al., 2005). However, while this method is effective for studies conducted on animals that respire actively, it is not suitable for static in vitro cell exposure studies, such as feeding particles on a multi-well plate where cells are seeded. This is because it is uncertain whether the amount of particles that the cells (which are often seeded on plates) are exposed to is linearly correlated with the concentration of aerosols in an exposure chamber. Generally, cell exposure studies for PM$_{2.5}$ particles have been conducted using PM collected by vibrating aerosol-loaded filters (Lichtveld et al., 2005; Ogino et al., 2014). However, the particles used for the exposure study may not be the same as those present in ambient air. Possible contamination from the filter material should also be considered. In addition, sample handling to obtain particles for exposure experiments is highly complicated and time-consuming. Besides, the amount of particles that are collected on a filter is often insufficient to perform an exposure studies. Hence, the development of techniques that allow researchers...
to collect a sufficient amount of PM$_{2.5}$ particles for exposure studies without the use of filters is needed. In this study, a high-volume PM$_{2.5}$ particle sampler was developed using the impactor and cyclone techniques, in order to facilitate the collection of PM$_{2.5}$ particles.

METHODS

Design and Operation of the High-Volume PM$_{2.5}$ Particle Sampler Using the Impactor and Cyclone Techniques

The high-volume PM$_{2.5}$ particle sampler developed in this study is shown in Fig. 1. The single-stage impactor, which used round jets, was designed for collecting PM$_{2.5}$ at an air flow rate of 1,100 L/min, based on the design in JIS-K0302 (1989) (Fig. 2). The air flow rate was determined by the requirement of the cyclone, which was connected to the impactor. There were 140 nozzles, and the round-jet nozzle diameter ($D_0$) was 2.88 mm. In this case, the Stokes number that gives 50% collection efficiency ($Stk_{50}$) for particles of size 2.5 μm was calculated following Eq. (1):

$$Stk_{50} = \frac{\rho_p d_p^2 C_c v}{9\eta D_0}$$

where $\rho_p$ is the standard density (1,000 kg/m$^3$), $d_p$ is the particle diameter, $C_c$ is the slip correction factor, $v$ is the velocity of the gas, and $\eta$ is the dynamic gas viscosity ($1.833 \times 10^{-5}$ Pa·s). In this case, $Stk_{50}$ was 0.289, so that the impactor could separate PM$_{2.5}$ (Ranz and Wong, 1952). The nozzle Reynolds number, as expressed in Eq. (2):

$$Re = \frac{\rho_g v D_0}{\eta}$$

where $\rho_g$ was the standard air density (1.205 kg/m$^3$), was calculated to be 3,800. The length of the inlet nozzle was 5 mm, and the distance between the inlet nozzle and the impaction plate was 4 mm.

A glass fiber filter (2500AO, thickness 0.5 mm, Pall Corp., USA) immersed in silicone oil was placed on an aluminum plate below the inlet nozzles as an impaction surface, in order to prevent particle rebound. A cyclone (HVS3, CS3 Inc., USA), which had been reported as being able to collect PM$_{0.1}$ at air flow rate of 1,100 L/min (Rule et al., 2010), was connected to the impactor downstream. An amber bottle (I-Chem 100 wide mouth amber glass jar, 250 mL, Thermo Fisher Scientific Inc., USA), which functioned as a particle collector, was connected to the cyclone. The air was pumped out using a blower (U2S-150, Showa Denki Co. Ltd., Japan). The flow rate of the air was controlled by an inverter (FRN2.2C1S-7J12, Fuji Electric Co. Ltd., Japan) and monitored using a mass flow meter (CMG400, Azbil Corp., Japan).

Fig. 1. The high-volume PM$_{2.5}$ particle sampler developed in this study. (a) Schematic diagram and (b) photographs of each device used in the system.
The pressure drop was measured using an electronic manometer (testo 506, testo AG, Germany). The pressure drops at an air flow rate of 1,100 L/min were 10.5 hPa at the cyclone inlet (the impactor outlet) and 82.1 hPa at the cyclone outlet. The high-volume PM$_{2.5}$ particle sampler was installed on the rooftop of a six-story building at Keio University, Yokohama, Japan. Ambient aerosol particles were typically collected for three weeks. After sampling, the particles in the amber bottle were collected and weighed using an electronic microbalance (ME236S, Sartorius AG, Germany). More than 100 mg of the particles were typically collected per sampling period in a glass bottle attached to the cyclone.

**Scanning Electron Microscope (SEM) Images of the Collected Particles**

SEM images of the collected particles were obtained to measure their geometric size distribution. The particles collected in the amber jar were transferred to a glass plate, and then the sample surface was coated with osmium. The projected area diameter was obtained using FE-SEM (JSM-
In this paper, we refer to the ambient air around the impactor as “Ambient”, the area between the impactor and cyclone as “Middle”, and the outlet of the cyclone as “Bottom”. The particle size distribution at each of these locations was measured using an aerodynamic particle sizer (APS spectrometer, Model 3321, TSI Inc., USA) and a scanning mobility particle sizer (SMPS, Model 3910, TSI Inc., USA). Note that the APS measures the aerodynamic diameter whereas the SMPS measures the electrical mobility diameter. The SMPS can measure particle size distributions with high resolution in a nanometer range in comparison to other instruments such as an electrical low pressure impactor (ELPI) (Marić et al., 2010). Besides, the particle size distributions of the ambient aerosols measured using the SMPS and ELPI exhibited similar features even though the aerodynamic diameter was essentially different from the electrical mobility diameter by a factor of the square root of the projected area diameter of the measured silica particles was 0.48 μm and 0.06 μm, respectively; the median was 0.49 μm, the geometric mean was 0.48 μm and the geometric standard deviation was 1.2 (n = 365).

**Evaluation of the Separation Characteristics of Particles for the Impactor and Cyclone**

Penetration for the Impactor and Cyclone

In Eq. (3):

\[
\text{Penetration}_{\text{Impactor}}(d_p) = \frac{N_{\text{Bottom}}(d_p)}{N_{\text{Middle}}(d_p)}
\]

where \(d_p\) is the particle diameter and \(N\) is the number concentration at each sampling point. \(N_{\text{Middle}}\) and \(N_{\text{Ambient}}\) were measured by attaching/detaching the impactor. Similarly, the penetration of particles at the cyclone was measured as the ratio of the concentration at Bottom to that at Middle, as expressed in Eq. (4):

\[
\text{Penetration}_{\text{Cyclone}}(d_p) = \frac{N_{\text{Bottom}}(d_p)}{N_{\text{Middle}}(d_p)}.
\]

\(N_{\text{Bottom}}\) and \(N_{\text{Middle}}\) were measured by connecting/bypassing the cyclone. The evaluation of the impactor was based on the APS results, and that of the cyclone was based on the SMPS results.

**Parallel Sampling Using the Cyclone and Filter**

In order to compare the chemical composition of particles collected using the cyclone and filter, parallel sampling using the high-volume PM2.5 particle sampler developed in this study and a traditional filter sampling system was carried out from the 10th to the 28th of November, 2014. The high volume cyclone sampler was operated under the condition mentioned above. The filter sampling was carried out using two low-volume air samplers equipped with a multinozzle impactor (Okuda et al., 2007). A polytetrafluoroethylene (PTFE) filter (Whatman 7592-104, General Electric Company, USA) and a quartz fiber (QF) filter (Advantec QR-100, Toyo Roshi Kaisha, Ltd., Japan) were used for particle collection. The air flow rate was set at 5 L/min for each sampling line. The collected samples were subjected to chemical analysis mentioned below.

**Chemical Analysis of the Collected Particles**

The elemental carbon (EC) and organic carbon (OC) contents in the particles were measured following the IMPROVE-TOR protocol using a Sunset ECOC analyzer (Okuda, 2013). Water-soluble inorganic ions in the particles were extracted by mechanical shaking with deionized water, and analyzed using ion chromatography (Okuda et al., 2013a). Elemental analysis of the particles was carried out using an energy-dispersive X-ray fluorescence spectrometer (EDXRF, EDXL300/NEX CG, Rigaku Inc., Japan) without any pretreatments. The details of the analytical conditions of the EDXRF measurements for the filter samples have been described elsewhere (Okuda et al., 2013a, b; Okuda and Hatoya, 2013; Okuda et al., 2014). We modified the protocol in order to analyze the particles collected in the “powder form” without using filters. In brief, 50–100 mg of particles were transferred to the sample hole (12 mm-i.d. and 1 mm depth) of a sample plate and sealed using thin film (Prolene Film, 4.0 μm in thickness, Chemplex Industries, Inc., USA). The sample plates were placed in the EDXRF instrument and analyzed under vacuum (1 Pa). The analysis time was 15 min per sample. In order to evaluate the analytical performance, we analyzed six reference materials, and compared the measured values of the elemental contents to the certified/reference ratios. The measured-to-certified/reference ratio ranged from 0.82 to 1.24 (Table 1). The reference materials used in this study were as follows: JA-2 (igneous rock, andesite), JB-3 (igneous rock, basalt), and JSd-1 (riverine sediment) were provided by National Institute of Advanced Industrial Science and Technology (AIST), Japan; CRM#28 (urban aerosols collected in Beijing, China) was provided by National Institute for Environmental Studies (NIES), Japan; SRM1649a (urban dust collected in Washington, D.C., USA) and SRM1944 (New York/New Jersey Waterway Sediment) were provided by National Institute of Standards and Technology (NIST), USA.

**RESULTS AND DISCUSSION**

**SEM Images of the Collected Particles**

A representative SEM image and size distribution of the collected particles are shown in Figs. 4 and 5, respectively. The distribution of the projected area diameter for 2,147 aerosol particles collected in April 2013 had a maximum peak at 0.05–0.06 μm. The median of the projected area diameter was 0.48 μm.
diameter of these particles was 0.12 μm, the geometric mean was 0.16 μm and the geometric standard deviation was 2.4. Particles larger than 2.5 μm in size were not found. Therefore, the separation of larger particles at the impactor was achieved successfully.

Separation Characteristics of the Impactor and Cyclone

The penetration curves for the impactor and cyclone are shown in Fig. 6. The 50% cut-point of the impactor for ambient aerosols at an air flow rate of 1,100 L/min was 2.0 μm. Fig. 6 also shows the penetration curve for the USEPA WINS impactor (Peters and Vanderpool, 1996; Peters et al., 2001) obtained using the same test protocol, but at an air flow rate of 16.7 L/min. The separation characteristics of the impactor installed in the high-volume PM<sub>2.5</sub> particle sampler were quite similar to those of the WINS impactor. Therefore, the separation of larger particles at the impactor was achieved successfully.

The separation characteristics of the cyclone for ambient aerosols at an air flow rate of 1,100 L/min were examined on two different days (Run #1: 4<sup>th</sup> of December, 2013; Run #2: 5<sup>th</sup> of November, 2014). The penetration curves for the two runs were slightly different, while the 50% cut-points were similar (0.15–0.18 μm). Rule et al. (2010) stated that the 50% cut-point of the same type of cyclone was 0.1 μm, but they did not show the detailed separation characteristics for the submicron-nanometer range of the particles that we showed here in this study. Hence, the high-volume PM<sub>2.5</sub> particle sampler developed in this study can collect submicron particles effectively.

Comparison of the Chemical Composition of Particles Collected Using the Cyclone and Filter

The chemical composition of the particles collected using the high-volume PM<sub>2.5</sub> particle sampler developed in this study (hereafter referred to as the cyclone particles) and the particles collected using the traditional filter sampling system (hereafter referred to as the filter particles) are shown in Fig. 7. The 62% of the mass of particles collected using the cyclone were identified compared with 83% for the filter. The detailed chemical compositions are also presented. Here, organic matter (OM) was calculated as OM = 1.6 × OC (Turpin and Lim, 2001), and the oxide minerals were
Fig. 4. Representative images of the collected particles. (a) A representative SEM image, and (b) corresponding analysis image of the same view obtained using Image-J software.

Fig. 5. Representative particle size distribution using the projected area diameter of the particles collected in April 2013 using FE-SEM with Image-J analysis.

Median = 0.12 μm
GeoMean = 0.16 μm
GSD = 2.40
expressed as a weight percent of the major oxides of the measured elements. The major finding was that the contents of OM and ammonium in the cyclone particles were lower than those in the filter samples. On the contrary, the content of oxide minerals in the cyclone particles was considerably higher than that in the filter particles. We suggest five possible reasons for these results as follows.

1. A possible reason for the difference in the OM and ammonium contents in the two types of particles is that the cyclone collection can avoid some sampling artifacts, which occur due to the adsorption of volatile organic compounds (VOCs) and gaseous ammonia to the particles on the filter media. Contents of organic carbon and ammonium measured on filters are frequently overestimated because organic vapors/ammonia adsorb to the particles and/or the filter media (Pathak and Chan, 2005; Turpin et al., 2000; Watson et al., 2009). These artifacts occur when air containing such VOCs and ammonia passes through the filter media. It is believed that the particles collected using the cyclone are less affected by the sampling air than the particles collected using filter media.

2. One potential reason for the difference in the content of oxide minerals between the cyclone particles and the filter particles might be a rebound of larger particles at the impactor. This may not be a major reason, especially at the beginning of the sampling because the glass fiber filter used as an impaction surface was immersed in silicone oil to prevent particle rebound. In addition, SEM images showed that rebound of larger particles did not occur. The separation characteristics of the impactor in this study were similar to those of the WINS impactor. Therefore, the separation of larger particles at the impactor appeared to be successful. However, the contents of possible crustal elements (Al and Fe) in the cyclone particles were one order of magnitude higher than those in the filter particles. This suggests possible contamination of larger crustal particles in the cyclone particles. Even though the number of larger

![Fig. 6. Penetration curves for the impactor and cyclone that were installed in the high-volume PM$_{2.5}$ particle sampler developed in this study (@1,100 L/min). The penetration curve for the USEPA WINS PM$_{2.5}$ impactor is also shown (@16.7 L/min).](image)

![Fig. 7. The chemical composition of the particles collected using the high-volume PM$_{2.5}$ particle sampler developed in this study, and that of the PM$_{2.5}$ collected on filter media.](image)
particles that penetrated into the cyclone section was small, the effect of these particles on the chemical composition of the particles collected at the cyclone would be significant, as larger particles have a relatively higher mass of elements compared with smaller particles.

(3) It is possible that relatively smaller particles could not be collected using the instrument developed in this study because the actual 50% cut-point of the cyclone was 0.15–0.18 μm. However, the contents of EC, which is an aggregate/agglomerate of primary particles with sizes less than 100 nm, in the cyclone particles was similar to that in the filter particles. Therefore, this may not be a major reason for the difference in the chemical composition of the particles collected using the cyclone and filter.

(4) In this study, ambient aerosol particles were collected in a glass bottle attached to the cyclone. However, the particles that adhered to the inner wall of the cyclone were not collected. It is possible that the chemical composition of the particles that adhered to the cyclone differed from that of the particles collected in the bottle due to the deliquescent ammonium salt particles adhering to the cyclone wall. To investigate this, the cyclone wall was washed with deionized water, and analyzed using ion chromatography. The results indicated that the contents of nitrate and sulfate in the cyclone-washed water accounted for 77% of the total ions, and the ammonium content was less than 10%. This ionic composition is similar to that of the cyclone particles. Therefore, particle adherence to the cyclone wall occurs, but it is not the major reason for the difference in the chemical composition between the cyclone and filter particles.

(5) Certain physical and/or chemical reactions may have occurred during the sampling period, which lasted several weeks, resulting in the loss of certain chemical species. For example, ammonium nitrate appears to sublime over time. However, the ammonium content of the particles in this study was consistently low, even for the particles collected during winter. In fact, the chemical compositions of the cyclone particles collected from April 2013 to November 2014 (n = 22) were similar to each other when compared with the difference in the chemical composition between the cyclone and filter particles. Therefore, the gas/particle partitioning is not a major reason for the difference in the chemical composition between the cyclone and filter particles.

We propose that the high-volume PM$_{2.5}$ particle sampler developed in this study can collect a large amount of aerosol particles in a “powder form” and can also avoid certain sampling artifacts caused by the adsorption of VOCs and gaseous ammonia to particles on the filter media. Reduction of the particle rebound at the impactor is a future challenge. One possible option is the use of a virtual impactor (Marple et al., 1990) so that particle rebound can be eliminated. Further studies are necessary to determine the effect of different sample collection methods on the chemical composition of the particles.

CONCLUSIONS

A high-volume PM$_{2.5}$ particle sampler using the impactor and cyclone techniques has been developed. The 50% cut-points of the impactor and cyclone for ambient aerosols at an air flow rate of 1,100 L/min were 2.0 μm and 0.15–0.18 μm, respectively. The separation characteristics of the impactor were similar to those of the WINS impactor. The contents of EC, which is an aggregate/agglomerate of primary particles with sizes less than 100 nm, in the cyclone particles was similar to that in the filter particles. The contents of organic matter and ammonium in the particles collected using the cyclone system were lower than those in the particles collected using traditional filter sampling. We propose that the high-volume PM$_{2.5}$ particle sampler developed in this study can collect a large amount of aerosol particles in a “powder form” and can also avoid certain sampling artifacts caused by the adsorption of VOCs and gaseous ammonia to particles on the filter media. The sampling device developed in this study allows researchers to collect a sufficient amount of PM$_{2.5}$ particles for cell exposure studies without the use of filters. It was possible that particle rebound occurred at the impactor because a significant amount of oxide minerals was found in the cyclone particles. However, the separation characteristics at the impactor, tested using APS, showed that the separation of larger particles was achieved successfully. Possible future work should investigate the use of a virtual impactor so that particle rebound can be eliminated.

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