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Development of $PM_{0.1}$ Personal Sampler for Evaluation of Personal Exposure to Aerosol Nanoparticles

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ABSTRACT

A $PM_{0.1}$ sampler for the evaluation of the personal exposure to nanoparticles was designed based on a novel approach to a layered mesh inertial filter. Applications to practical environments would include roadsides and highly contaminated workplaces. The separation performances of $PM_{0.1}$ sampler consisting of a layered mesh inertial filter and pre-separators for the removal of coarse particles were evaluated. The influence of particle loading on the pressure drop and separation performance, which is important from a practical standpoint, was also discussed. The novel personal sampler recorded a cutoff size of 100 nm with a small pressure drop of ~5 kPa. Through the combination of a layered mesh inertial filter for the $PM_{0.1}$ and pre-cut impactors for the removal of huge or coagulated particles ($PM_{1.4}$-TSP) along with a pre-cut inertial filter using webbed SUS fibers for the removal of fine particles ($PM_{0.5}$-$PM_{1.4}$), the present $PM_{0.1}$ inlet for the personal sampler was practical for the chemical analysis of collected particles. This sampler was proven effective even under the limitations of a small-capacity portable battery pump, which was rated at less than the minimum change for separation performance. The novel $PM_{0.1}$ personal sampler is compact and lightweight (under 1 kg including a portable battery pump), which is important for the practical application of a personal sampler.

Keywords: $PM_{0.1}$; Nanoparticles; Personal exposure; Inertial filter.

INTRODUCTION

During the assessment of the health effects of airborne particulates, it is necessary to determine both the concentration and composition of the particles in the breathing zone with regards to aerodynamic particle size, which affects the regional deposition of particles inhaled into the human respiratory system. This is particularly important for ambient nanoparticles (< 100 nm), since they can contain a large portion of hazardous chemicals from anthropogenic sources and can penetrate deeply inside lungs, eventually reach the alveolar region. Moreover, their chemical compositions will be more quickly dispersed throughout the human body (Hinds, 1999; Bolch *et al.*, 2001; Warheit, 2004; Hussain *et al.*, 2011). Exposure to nanoparticles has been associated with pulmonary inflammation, immune changes, and a contribution to undesirable cardiovascular effects (Donaldson *et al.*, 2002; Granum and Lovik, 2002; Borm and Kreyling, 2004). Moreover, $PM_{0.1}$ in environmetns ilfluenced by human activities, e.g., powder production in a factory, burning of agricultural crop waste, and cigarette smoking, is being reported in ever-increasing concentrations (Phillips and Bentley, 2001; Behera *et al.*, 2004; Davidson *et al.*, 2005; Herner *et al.*, 2005; Morawska *et al.*, 2008; Ngo *et al.*, 2010). In order to evaluate health influences and risks, therefore, the monitoring of environmental nanoparticles is crucially important.

The evaluation of nanoparticle exposure has been concerned not only on nanoparticles from daily human activities and environments, but also on nanomaterials that are an inherent part of nanotechnological developments (Kuhlbusch *et al.*, 2011). Although the number of personal exposure studies on fine particles has continually increased (Du *et al.*, 2010; Borgini *et al.*, 2011; Lim *et al.*, 2012; Jahn *et al.*, 2013), relatively few studies have focused on monitoring the personal exposure to fine particles in the nano-size range via a portable personal sampler (Young *et al.*, 2013). Therefore, the development of a portable personal sampler that could be used to evaluate nanoparticle exposure would be indispensable in any discussion on the health risks and infuluences posed by nanoparticles.

Various types of portable personal samplers equipped

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with a battery pump have been used for the evaluation of the personal exposure in workplaces and in living environments. Few of these personal samplers, however, have been applicable to the collection of nanoparticles. This has been due to the difficulty posed by the large degree of pressure drop that is needed for the separation of nanoparticles when using conventional methods that employ a low-pressure impactor. In order to overcome this difficulty, the authors developed a personal sampler based on the "inertial filter" technology (Furuuchi *et al.*, 2010). However, because of the difficulty posed by a pressure drop through the inertial filter under the limited capacity of a portable battery pump, the best cutoff size that could achieved was ~140 nm with a 6 L/min of a sampling flow rate, which was insufficient for a characterization as "nanoparticles". Although an impactor type of personal sampler was recently devised with a cutoff size of 100 nm (Tsai *et al.*, 2012), its sampling flow rate (2.0 L/min), was not always sufficient for the chemical analysis of particles that could be collected in working (6–8hours) and living environments (12–24 hours). Hence, a cutoff size of 100 nm must be achieved for a practical samplng air-flow rate that should approximate 4-6 L/min, or more. Another difficulty frequently encountered in the practical application comes from the existence of huge and coagulated particles, which are typically observed in the handling of fine powder in workplaces and in the vicinity of roadside environments. The loading of these particles on the inertial filter for nanoparticle separation increases the pressure drop and also accelerates the rate of bouncing problems encountered with coarse particles. Hence, given the wide range of concentration and size distribution of particles, it is very important to overcome these problems if the practical application of a personal sampler is to be effective.

In this study, the $PM_{0.1}$ sampler for the evaluation of the personal exposure to nanoparticles was designed based on a novel approach that uses a layered mesh inertial filter while targeting the application to practical environments including roadsides and highly contaminated workplaces. Separation performances were evaluated for the $PM_{0.1}$ sampler consisting of the layered mesh filter and other pre-separators for the removal of coarse particles. The influence of particle loading on the pressure drop and separation performance, which is important for practical applications, was also evaluated.

INERTIAL FILTERS AND $PM_{0.1}$ PERSONAL SAMPLER

Layered Mesh Inertial Filter for the $PM_{0.1}$

Fig. 1 shows the structure of the layered mesh inertial filter used for the $PM_{0.1}$ separation. It consists of commercially available layered square mesh copper TEM grids (Glider, G600HSS) sandwiched by manufactured copper spacers with a circular hole (ϕ 1.9 mm, t = 30 µm) stacked in a circular nozzle (ϕ 3 mm, 9 mm nozzle length) with a bell shaped inlet through an aluminum cartridge. The geometry of the original inertial filter used webbed stainless steel fibers (Otani *et al.*, 2007; Eryu *et al.*, 2009; Furuuchi *et al.*, 2010). This new inertial filter was made up of layered TEM grids that provide a uniform structure of fibers aligned perpendicular to the flow direction along the nozzle, which maximizes the inertial effect on particles and provides less pressure drop with no loss in separation performance. The uniformity of the layered-mesh structure projected in the flow direction is a key point in the preparation of the layered-mesh inertial filter since the aerosol particles may penetrate directly through the opening between mesh wires because of a large inertial effect (Eryu *et al.*, 2009). Hence, wire mesh screens must be aligned tangentially uniform by shifting each TEM grid for 15 degree in order to maximize the coverage of the nozzle cross-section by the mesh wires. The advantages of the layered mesh inertial filter cannot be obtained by the original structure of randomly orientated SUS fibers packed in a circular nozzle since it is difficult to make the structure of packed fibers uniform over the cross-section and depth of a nozzle that has a diameter of less than 2 millimeters. The analysis of chemical components such as PAHs can be done for particles collected on TEM grids by the extraction, e.g., by immersing TEM grids in a solution for the extraction. The specifications of the TEM grids are listed in Table 1. Five TEM grids and spacers were used for each filter based on the preliminary experiments and numerical analysis (Eryu *et al.*, 2009; Takebayashi, 2012).

Pre-cut Inertial Filter for $PM_{0.5}$

In order to prevent clogging and bouncing of coarse particles on the layered mesh $PM_{0.1}$ inertial filter, a pre-cut inertial filter consisting of webbed SUS fibers (d_f = 9.8 µm) packed in a ϕ 4.75 mm circular nozzle (5.5 mm length)

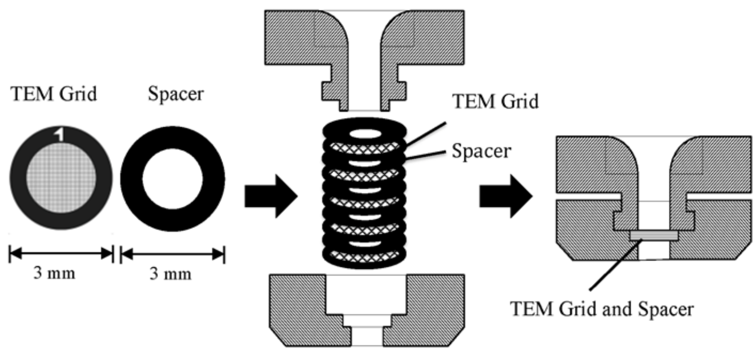

Fig. 1. Schematic of the main inertial filter.

Table 1. Specification of TEM grids used for the layered mesh inertial filter.

Grid type	Code	Material	Mesh (lines/inch)	Pitch (μm)	Bar width (μm)	Hole width (μm)	Thickness (μm)	Q (L/min)	Cutoff size (nm)
square mesh	Glider G600HSS	Cu	600	42	5	37	8	5	100

through a metal cartridge was used upstream from the layered mesh inertial filter. This type of inertial filter had a relatively large dust-loading capacity and provides less pressure drop than that of the impactor. The pre-cut inertial filter had a geometry that was similar to the original one but with a different diameter for the nozzle and SUS-fiber loading to decrease the cutoff size from 700 to 450 nm. This was intended to reduce the amount of particles penetrating to the layered mesh inertial filter to help maintain performance. The specifications of the pre-cut inertial filter are shown in Table 2.

Pre-cut Impactors

The pre-cut inertial filter was expected to have a larger capacity for particle loading and fewer re-suspended particles compared with the impaction plate of an impactor. However, the dust loading capacity was suspected to be insufficient for the measurement in environments highly contaminated by the huge and coagulated particles that are typically observed in fine powder handling processes and road-side environments. In order to avoid penetration by these particles into the pre-cut and layered mesh inertial filters, therefore, a commercially available two-stage pre-cut impactors (SHIBATA, ATPS-20H) were used for the removal of particles in the micron size range. Cutoff sizes were estimated to be 5.6 and 1.4 μm at 5 L/min for the 1st and 2nd stages, respectively, of the pre-cut impactors, as estimated using an equation for inertial separation (Hinds, 2009), where the cutoff sizes were originally designed to be 10 and 2.5 μm at 1.5 L/min. The pre-cut impactors are important for practical application in workplaces that are highly contaminated by coagulated particles in order to maintain the separation performance of the inertial filters and to minimize the pressure drop due to particle loading.

$PM_{0.1}$ Inlet for a Personal Sampler

Fig. 2 shows the geometry of the $PM_{0.1}$ personal sampler inlet, which consisted of two different types of inertial filters located downstream from the two-stage pre-cut impactors and was followed by a backup filter on a thin stainless filter holder. The surface of the impaction plate for the 1st stage of the pre-cut impactor was covered by silicon grease (Dow Corning, 03253589) to a uniform thickness of approximately 0.2 mm while a glass fiber filter 10 mm in diameter (Pallflex, T60A20) was attached to the impaction plate of the 2nd stage. The outlet of the $PM_{0.1}$ personal sampler was connected to a portable battery pump (Hario Sci., HSP-5000) using a flexible resin tube. The weight of the $PM_{0.1}$ personal sampler was 112 g for the sampler inlet (6.5 cm maximum width and 11.4 cm height) and 700 g for the portable pump (85 mm width, 60 mm depth and 155 mm height), which makes it easy to handle in the field.

EXPERIMENTS

Separation Performances of Inertial Filters and Pre-cut Impactors

The separation performance of the inertial filters was evaluated using the an experimental setup shown in Fig. 3, which consisted of an evaporation-condensation type of aerosol generator, a nitrogen gas generator for the carrier gas supply, HEPA filters, mass flow controllers, a neutralizer (^{241}Am), a differential mobility analyzer (DMA), a test inertial filter in a holder, a digital manometer and measuring instruments for particle number concentration. The performance was evaluated following an established procedure (Furuuchi *et al.*, 2010). $ZnCl_2$ powder was dosed on an alumina boat in a tubular image furnace, then $ZnCl_2$ was heated to 190–320°C followed by cooling to room temperature in order to obtain the $ZnCl_2$ particles. After classifying the generated particles by DMA, the particles were used for the test aerosol (~20–520 nm in aerodynamic diameter, geometric standard deviation σ_g = 1.06–1.30). The mono-dispersed $ZnCl_2$ particles were diluted with air through a HEPA filter and supplied to the inertial filter placed in a holder.

The collection efficiency was determined based on the number concentration measured by a laser aerosol spectrometer (TSI, LAS model 3340), a condensation particle counter (TSI, CPC model 3785), and a scanning mobility particle sizer (TSI, SMPS model 3080). The pressure drop through the inertial filter was monitored using a digital manometer (EXTECH, HD 750). The mobility equivalent diameters of the $ZnCl_2$ particles were converted to aerodynamic diameters using a measured density (1508 kg/m^3 averaged for 40 nm to 350 nm) of generated particles via an aerosol particle mass analyzer (KANOMAX, APM model 3600).

The performance of pre-cut impactors was evaluated using the configuration shown in Fig. 4. A condensation

Table 2. Specification of the pre-cut inertial filter.

Inertial Filter	d_f (μm)	Fiber material	Type	L_n (mm)	D_n (mm)	Q (L/min)	Fiber loadings (mg)	Fiber volume fraction (-)	Cutoff size (nm)
Pre-cut inertial filter	9.8	SUS-316L	web	5.5	4.75	5	10	0.0133	450

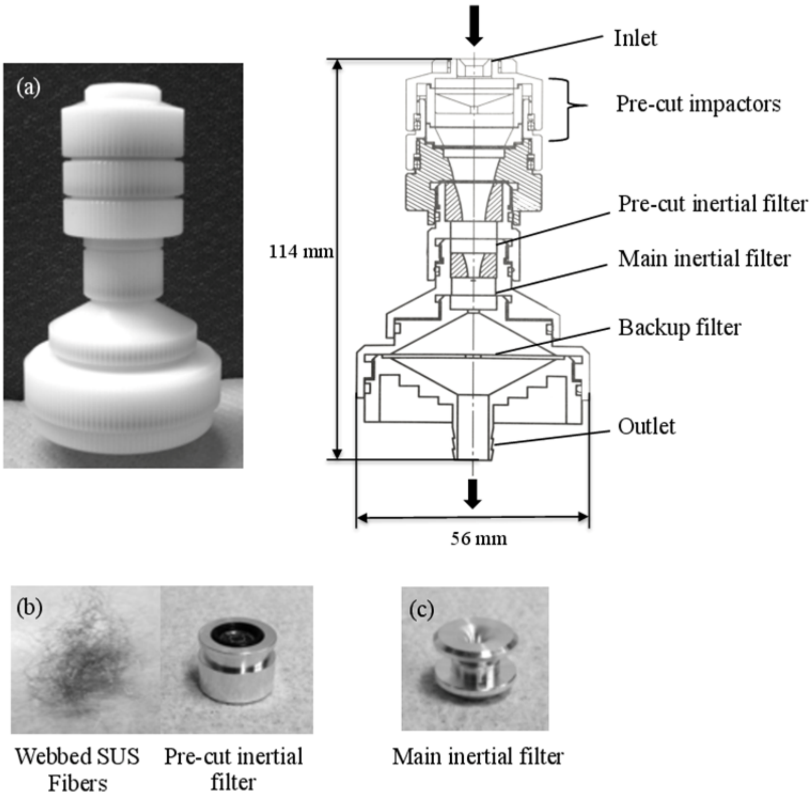

Fig. 2. $PM_{0.1}$ personal sampler inlet and inertial filters used: (a) an outside picture and structure of $PM_{0.1}$ personal sampler inlet, (b) the pre-cut inertial filter and stainless steel (SUS) fibers used, and (c) the main inertial filter (layered mesh geometry).

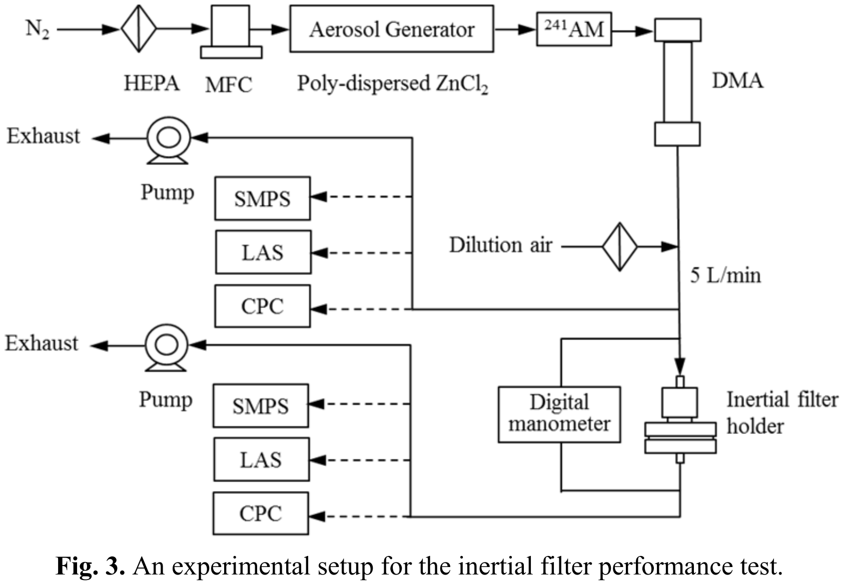

Fig. 3. An experimental setup for the inertial filter performance test.

aerosol generator (TOPAS, SLG 270) was used to obtain a high number concentration of mono-dispersed NaCl coarse particles (~540–2840 nm in aerodynamic diameter, σ_g = 1.22–1.29), which are electrically neutral and correspond to the heating temperature between 220 and 280°C. Generated mono-dispersed NaCl particles were diluted by mixing with filtered air via a HEPA filter, then them to the pre-cut impactor filters. The collection efficiency of the pre-cut impactor filters was determined based on the number concentration measured by an aerosol particle sizer (TSI, APS model 3321). A pressure drop through the inertial filter was also monitored using a digital manometer (EXTECH, HD 750).

Effect of Surface Coating of the Inertial Filters

The influence of the surface treatment of the inertial filter

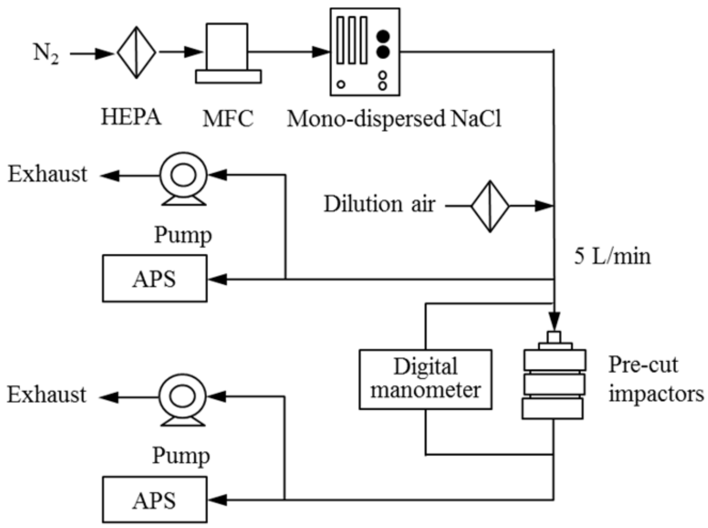

Fig. 4. An experimental setup for the pre-cut impactors performance test.

fibers to reduce the bouncing effect of coarse particles was also investigated. Fiber surfaces of the pre-cut and the main inertial filters were coated by glue, or, by dropping 1 wt% water solution of water soluble glue (Tombo, HCA-122) onto the pre-cut and main inertial filters, which held them on the $PM_{0.1}$ inlet, followed by drying via flowing a HEPA filtered air through each inertial filter for 1 hour. Based on observation using an optical microscope, there was no remaining water glue solution or dried glue at any of the corners or edges of the mesh grids, which may have influenced the flow and particle motion.

Influence of Particle Loading on Pressure Drop

The influences of particle loading on the pressure drop and separation performance of the $PM_{0.1}$ inlet were investigated for different size ranges of particles: coarse particles on the order of microns that may be predominant in some workplaces or roadsides, and fine particles that are the main fraction of smoke particles including cigarette smoke and automobile exhaust particles, etc. As coarse loading test dust, JIS No. 5, which is a mineral dust of 85 ± 5% as the coarse particles (> 5 µm) in mass, was used. As fine loading test particles, incense smoke particles, which ranged concentrations between 100–200 nm, were used. The JIS No.5 dust was dispersed by an ejector (Sympatec, RODOS type) to a mixing box then introduced to the $PM_{0.1}$ inlet. Incense smoke particles were diluted by filtered air through a HEPA filter then introduced to the $PM_{0.1}$ inlet. In order to obtain various particle loadings on the filters, the sampling was adjusted between 60 and 120 min for the JIS No. 5 dust and between 5 and 10 min for the incense smoke particles. The pressure drop was measured using a digital manometer (EXTECH, HD 750) before and after sampling.

Validation of the $PM_{0.1}$ Personal Sampler

For the validation of measurement by the $PM_{0.1}$ personal sampler, the concentration and size distribution of ambient aerosol particles were compared between the $PM_{0.1}$ personal sampler and the Nanosampler (NS, KANOMAX, Model 3180; Furuuchi *et al.*, 2010) after the same period of aerosol sampling. The validation was conducted on a balcony of the 6th floor in a 7-story building at Kanazawa University on the Kakuma campus, Kanazawa. Binder-less quartz fibrous filters (Pallflex, 2500QAT- UP) were used for the validation. They were weighed after the conditioning at 20°C and 50% RH in a weighing chamber for 48 hours both before and after the sampling.

RESULTS AND DISCUSSION

Separation Performance of the Inertial Filters

Fig. 5 shows the collection efficiency curves for the pre-cut and main inertial filter along with a combination of those filters and pre-cut impactors measured at an airflow rate of 5 L/min. The cutoff size of the pre-cut filter was estimated at ~450 nm with a pressure drop of 0.6 kPa. The cutoff size of the main filter could be adjusted by ~100 nm by changing the filtration velocity, or, the size of a spacer hole, with an acceptable steepness of the efficiency curve at 4.6 kPa of pressure drop. The dashed curve in Fig. 5 denotes a prediction based on the filtration theory along with a numerical simulation for a fiber with a square cross-section (Hinds, 1999; Otani *et al.*, 2007; Eryu *et al.*, 2009), where the fiber volume fraction α was adjusted to fit a d_{p50} = 100 nm (α = 0.21). Although there was good consistency in the separation tendency between measured and predicted efficiencies, the measured collection efficiency for coarse particles larger than ~200 nm was slightly lower than that from the prediction. This may have been the influence of bouncing or a re-suspension on the TEM grid mesh fibers when dealing with this size range of particles. Because of Brownian diffusion, the collection efficiency for particles in the 10–20 nm range increased both in the pre-cut inertial filter and in the main inertial filter. This increase may be greater for particles for smaller than 10 nm, but from the point view of particle mass, it may not be so important. The pre-cut inertial filter just has only a slight influence on the main filter performance. Values of the total pressure

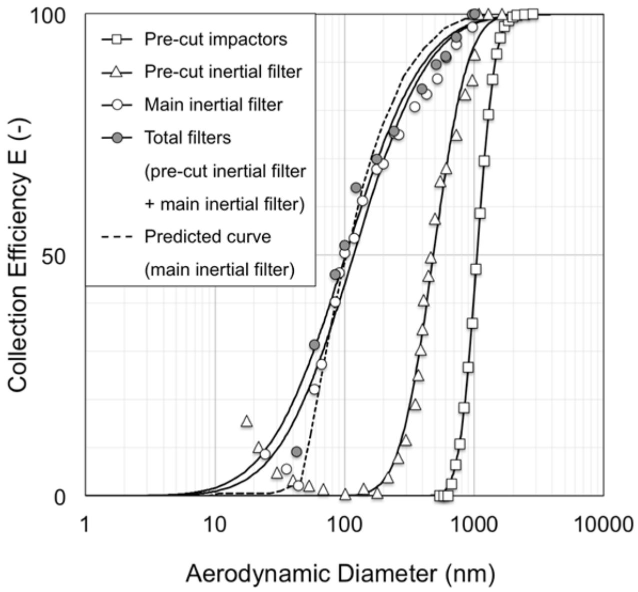

Fig. 5. Collection efficiency curves for the pre-cut impactors, the pre-cut inertial filter and the main inertial filter and the combination of the pre-cut and main inertial filters.

drop of 5.2 and 7.7 kPa for tandem inertial filters and tandem inertial filters + pre-cut impactors + a backup filter, respectively, are low enough to be powered by a portable battery pump (the maximum allowable pressure drop is 15 kPa at 5 L/min). This creates a large allowance for an increased pressure drop due to particle loading and tubing.

As shown in Fig. 6, the collection efficiency of the main inertial filter was clearly improved for coarse particles larger than ~200 nm by glue coating and almost reached the predicted value, or, the maximum performance, as denoted by the dashed curve. Fig. 7 shows the total collection efficiency curves for the glue-coated pre- and main inertial filters. The increase in collection efficiency was negligibly small for the pre-inertial filter so that improvements in the performance corresponded mostly to the main inertial filter, as shown in Fig. 7. The pressure drop through the glued inertial filters increased 10–20%, corresponding to a total pressure drop through the $PM_{0.1}$ inlet of 8.10 kPa, which was still much lower than the allowable value (15 kPa). Hence, the coating by water-soluble glue can be a tool that can be used to improve the separation performance of coarse particles, although the background for the chemical analysis of particles collected on TEM grids should be carefully evaluated.

Influence of Particle Loading on Pressure Drop and Separation Performance

Total pressure drops through the pre-cut impactors, the inertial filters and the backup filter of the $PM_{0.1}$ personal sampler was measured along with that of the total $PM_{0.1}$ personal sampler in relation to loaded masses of JIS No.5 test dust and incense particles. The total pressure drop was increased by dust loading up to the maximum allowable pressure, or, to 15 kPa of the portable battery pump. There was a predominant increase in the pressure drop in the main inertial filter, particularly for the incense particles, while the changes in the impactors, the pre-filter, and the backup filter were not so important. Depending on the size and characteristics of the particles, the maximum amount of particles collected on the backup filter, which can be used not only for mass evaluation but also for various chemical analyses, ranged between 0.1–0.3 mg when using the present battery pump. This amount is sufficient for the analysis of chemicals such as carbon components and polycyclic aromatic hydrocarbons (PAHs), and it can be increased by using a pump with a larger capacity.

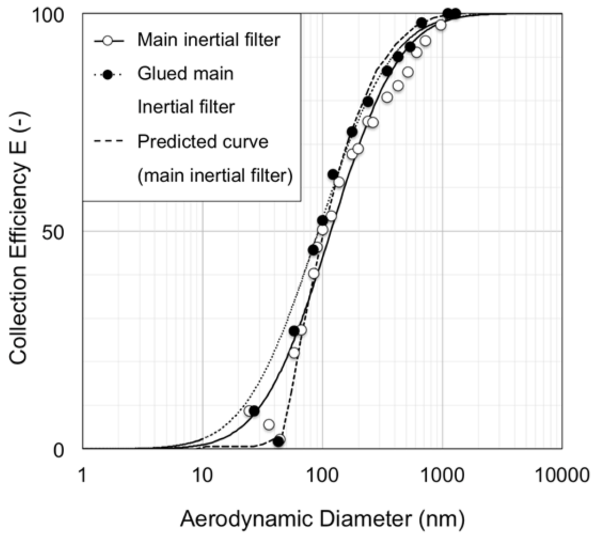

Fig. 6. Effect of glue coating on TEM grids on the collection efficiency of the main inertial filter.

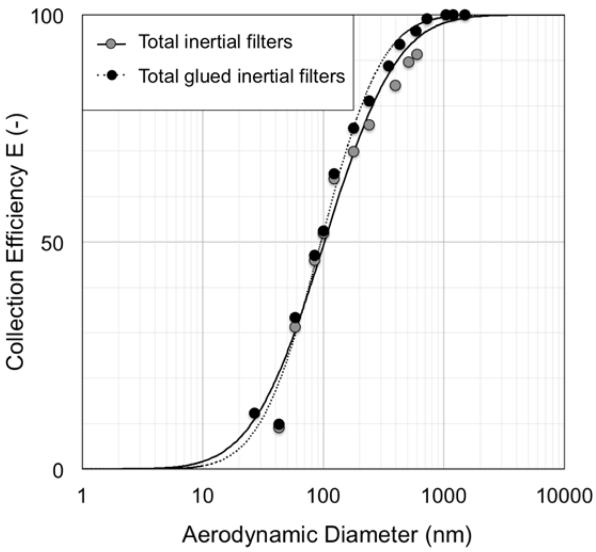

Fig. 7. Effect of glue coating on the total collection efficiency of the pre-cut and main inertial filters.

The separation performance of the main inertial filter was evaluated when loaded with 0.1 mg of incense particles, and a collection efficiency curve is shown in Fig. 8. The cutoff size was decreased to ~94 nm, or, ~6% that of the non-loaded case. This may be a practical level for many field measurements.

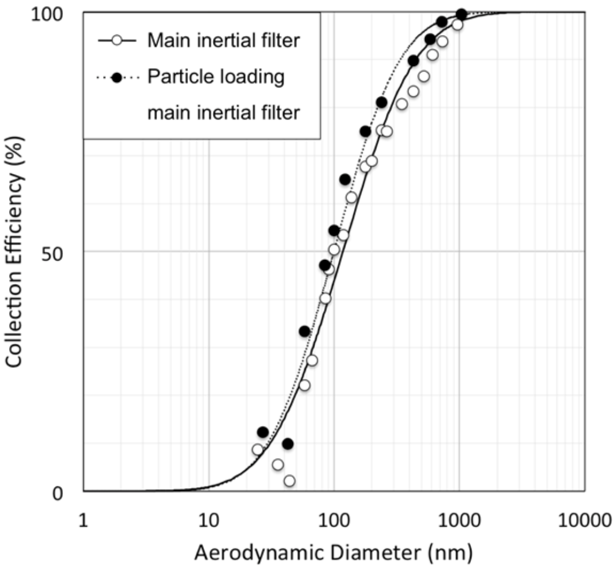

Fig. 8. Comparison of collection efficiency of the main inertial filter before and after the particle loading of 0.1 mg.

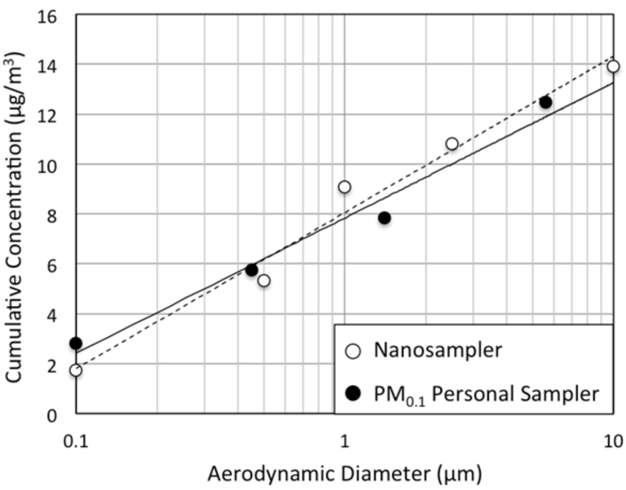

Fig. 9. Comparison of aerosol particle cumulative concentrations obtained by $PM_{0.1}$ personal sampler with Nanosampler for ambient aerosol sampling.

Comparison of the $PM_{0.1}$ Personal Sampler with a Nanosampler

Fig. 9 shows the cumulative concentration of size-fractionated particles collected by the $PM_{0.1}$ personal sampler with pre-cut impactors compared with those collected using a Nanosampler (NS) (Kanomax, Model 3180) (Furuuchi *et al.*, 2011). The similarities in the concentration and size distribution between those from the $PM_{0.1}$ personal sampler and NS were reasonable.

CONCLUSIONS

For practical applications in environments that include sampling from roadsides and in highly contaminated workplaces, the $PM_{0.1}$ sampler was successfully devised by improving a prototype of the personal sampler for the evaluation of personal exposure to nanoparticles (Furuuchi *et al.*, 2010). The inertial filter with a layered mesh geometry demonstrated a separation performance with a cutoff size of 100 nm and a small pressure drop of ~5 kPa. Through the combination of a layered mesh inertial filter for the $PM_{0.1}$ and pre-cut impactors for the removal of huge or coagulated particles ($PM_{1.4}$-TSP) along with a pre-cut inertial filter using webbed SUS fibers for the removal of fine particles ($PM_{0.5}$–$PM_{1.4}$), the present $PM_{0.1}$ inlet for the personal sampler was practical for the chemical analysis of collected particles. This sampler was proven effective even under the limitations of a small-capacity portable battery pump, which was rated at less than the minimum change for separation performance. The devised $PM_{0.1}$ personal sampler is compact and lightweight (under 1 kg including a portable battery pump), which is important for the practicality of a personal sampler. The devised $PM_{0.1}$ personal sampler has been used to evaluate the exposure to nanoparticles in various environments and results will be reported in the near future.

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