Evaluation of Multichannel Annular Denuders for a Newly Developed Ultrafine Particle Sampling System

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Abstract

Laboratory studies evaluated the gas collection efficiencies and particle transmission efficiencies of 1-, 3-, 5-, and 8-channel denuders under simulated atmospheric conditions with the goal of combining an annular denuder–filter pack (AD-FP) system with a newly developed inertial classification. The multichannel denuders having large surface areas were considered for this AD-FP system since the inertial classification system operates at a flow rate of 40 L/min. Gas collection efficiency was measured with phosphorous acid–coated denuders targeted on ammonia gas. Particle transmission efficiency was measured for ammonium sulfate particles. The denuder-coating procedure and the method for extracting the multichannel denuders are described in detail. Gas collection efficiencies for the 1-, 3-, 5-, and 8-channel denuders were 96.3, 98.6, 98.2, and 98.3% (1-ch < 5-ch, 8-ch, and 3-ch), respectively. The extraction efficiency was greater than 96% for all but the 8-channel denuder (92%). The particle transmission efficiencies for the 1-, 3-, 5-, and 8-channel denuders at 40 L/min were 97.3, 97.4, 94.7, and 93.4% (8-ch < 5-ch < 1-ch and 3-ch), respectively. These results indicate that a 1- or 3-channel denuder can be effectively used in this AD-FP system at a flow rate of 40 L/min.

Keywords: Multichannel annular denuder; Gas collection efficiency; Particle loss; Extraction efficiency.

INTRODUCTION

The formation and growth of new aerosol

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particles is a growing interest due to their climatic and health effects. Atmospheric formation from gaseous precursors is a significant source of new aerosol particles into the global atmosphere (Kulmala *et al*., 2004). Several studies suggest that ultrafine particles (< 0.1 μm in diameter; UFPs) disproportionately induce oxidative stress in cells and are more toxic than larger particles of similar composition (Li *et al*., 2003; Nel, 2005; Nel *et al*., 2006). Although UFPs do not contribute large quantities to $PM_{2.5}$ or PM_{10} mass, they dominate the number concentration and most of the surface area (Chow and Watson, 2007). Atmospheric oxidation of gas-phase primary exhaust species can produce low-vapor-pressure compounds that readily condense onto existing particles and produce secondary mass. Particle composition and size can also evolve due to interaction and reaction of gas- and liquid- or solid-phase species at the particle surface or in the bulk solution, as well as through coagulation of existing particles (Moore *et al*., 2007). Therefore, measurement of gaseous compounds participating in the nucleation and growth of particles and determination of the chemical composition of nucleated particles, as well as their other properties, are very important to our understanding of the nucleation process itself.

The use of annular denuders is a promising approach for simultaneously sampling gaseous and particulate-phase aerosols. Since the appearance of the first article on annular denuders (Possanzini *et al*. 1983), this device has been increasingly used for ambient air monitoring. The attractiveness of annular denuders comes from the order of magnitude gain in gas collection efficiency for a given length/flow rate ratio (Ye *et al*., 1991). Particles, which diffuse about 1000 times more slowly than gaseous compounds (Febo *et al.* 1999), are theoretically not trapped on the tube walls but on a filter downstream from the denuder. The combination of an annular denuder and a filter pack system (AD-FP) has been used at flow rates of 10 or 16.7 L/min in a cyclone system with a 2.5 μm cut-point diameter (Possanzini *et al*., 1983; Ye *et al.*, 1991; Baron and Willeke, 2005), to collect both particulate and gaseous phases.

However, to obtain sufficient quantitative accuracy and high time resolution, the sampling flow rate must be relatively high when the method is applied to UFPs or nanoparticles. Recently, a new technique for the inertial classification of UFPs with fibrous filters was developed (Otani *et al*., 2007). With this new technique, pressure drop within the sampling system is lower than existing sampling systems such as low pressure impactor (LPI), and the technique can be used to separate particles smaller than 50 nm or 100 nm. Furthermore, particle masses sufficient for chemical analysis can be collected at a high sampling flow rate (40 L/min), and the composition changes in particles can be measured with high time resolution.

An annular denuder consists of two or more coaxial cylinders so that air is forced to pass through the annular space. An annular denuder can achieve the highest possible efficiency and capacity. The sampling behaviour of a denuder is characterized by the diffusion of gases to the tube walls, which are considered to act as perfect sinks, from a laminar air stream. The condition for a laminar flow is a Reynolds number *(Re)* < 2000 (Possanzini *et al*., 1983). However, the flow regime should be considered for each multichannel denuder to ensure that the flow is laminar at 40 L/min and if the annular denuder technique is to be applied to the newly developed UFPs sampling system. The calculated Reynolds number for a multichannel denuder was all below 2000 at 40 L/min with given geometry of annular denuders in this study. Therefore the diffusion of gases to the tube walls can be considered to act as a perfect sink from a laminar air stream. Although many researchers are currently using multichannel annular denuders, particle losses, gas collection efficiency, the denuder-coating procedure, and the sample extraction method for multichannel denuders have not been studied in detail. Therefore, the particle losses and gas collection efficiencies for multichannel denuders, along with the denuder-coating procedure and extraction method were also studied and introduced here. Furthermore, the adverse health effect of UFPs is much higher than that of fine particles due to their large surface area with higher particle number concentration. Thus, it is very important to evaluate the particle transmission efficiency in denuders to analyze gaseous and particulate atmospheric UFPs in field sampling since the loss of smaller particles (< 0.1 μm) could be greater than that of larger particles (Ye *et al*., 1991; Roussel *et al*., 2004).

In this study, the possibilities of AD-FP system in combination with the newly developed inertial classification system were estimated. Because the AD-FP system was designed to be combined with a cyclone system operated at either 10 or 16.7 L/min, it was necessary to assess the gas collection efficiency and particle loss at 40 L/min within a 1-channel annular denuder. Also, multichannel denuders (3-, 5-, and 8-channel) were considered for the high sampling flow rate since they have much larger surface areas compared to a 1-channel denuder. Ammonia gas was used to measure gas collection efficiency at 40 L/min for 1 h and 6 h, and ammonium sulfate was used to generate particles for particle loss measurement by means of a scanning mobility particle sizer. Both gas collection efficiency and particle loss by diffusion were also theoretically estimated, and the theoretical values were compared with the corresponding experimental values. Our results should assist users in selecting the appropriate number of denuder channels, as well as the amounts of coating and extraction solutions for the channels.

EXPERIMENT

Multichannel Annular Denuders

Annular denuders with the following

Table 1. Dimensions of annular denuders.

	Commercial name	Length (mm)	Surface area (cm^2)	Annular spacing (mm)
1-channel denuder	URG-2000-30B	242	303.5	1
3-channel denuder	URG-2000-30x242-3CSS	242	527.8	1
5-channel denuder	URG-2000-30B5	400	1771.9	1
8-channel denuder	URG-2000-30CF	285	3497.5	1

dimensions (Table 1) were purchased (University Research Glassware, NC, USA).

Experimental Set-up for Determining Gas Collection Efficiency

Ammonia gas was supplied at 0.1–0.2 L/min from a standard ammonia gas cylinder (6.46 ppm, Takachiho Trading, Tokyo, Japan), and the gas was diluted at the glass chamber with purified air supplied from a compressor (Fig. 1).

The mixed air was introduced into the denuder section. The denuder section consisted of two denuders connected in series: the number of channels in the first denuder varied from 1 to 8, and the second denuder was a 1-channel denuder, which was used to estimate the collection efficiency of each denuder. The total flow rate was checked by means of a mass flow meter located in the downstream of the denuders. Neither relative humidity nor temperature was controlled, but both were monitored by means of a thermohygrometer (HI 8564, Hanna Instruments, USA). The temperature was 23°C (± 2.0°C), and the relative humidity was below 25%. The sampling durations were 1 h and 6 h. 6 hour sampling duration was chosen to see whether there is breakthrough or not and for field application.

Experimental Set-up for Particle Loss Measurement

Ammonium sulfate particles were generated by an atomizer (Constant output atomizer, TSI 3076, MN, USA) from an aqueous ammonium sulfate solution (0.003 mol/L). The generated particles were introduced into the glass chamber and diluted with purified air supplied from the compressor at 40 L/min (Fig. 2. a and b). The generated particle size ranged from 0.02 to 0.3 μm.

Particle loss in the annular denuder under test was determined by measuring particle number concentration at the upstream and downstream locations of the test system, and comparing the particle loss with the test system present to the loss with the test system removed. Upstream and downstream concentrations (N_{in} and N_{out}) were measured with a differential mobility analyzer (DMA) to measure the particle size distribution (TSI 3080, USA) and an ultrafine condensation particle counter (UCPC) to measure the particle number distribution (TSI 3025A, USA). Particle loss at 20 L/min was also

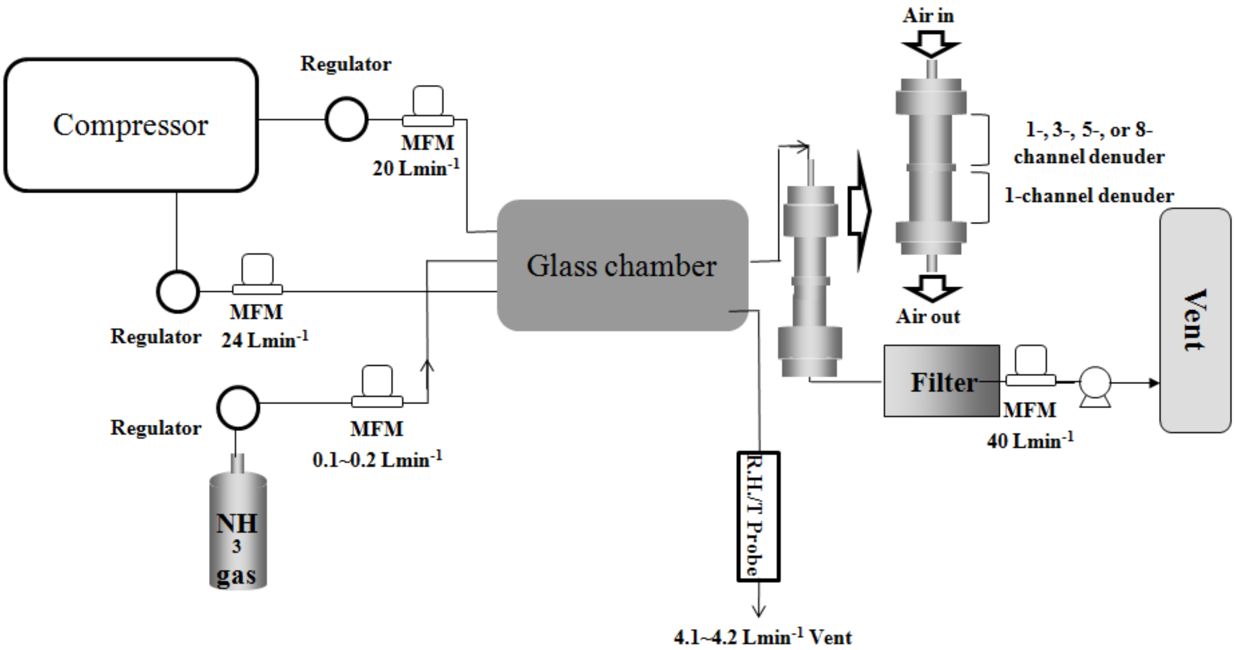


Fig. 1. Experimental set-up for determining gas collection efficiency: MFM, mass flow controller; R.T., relative humidity; T, temperature.

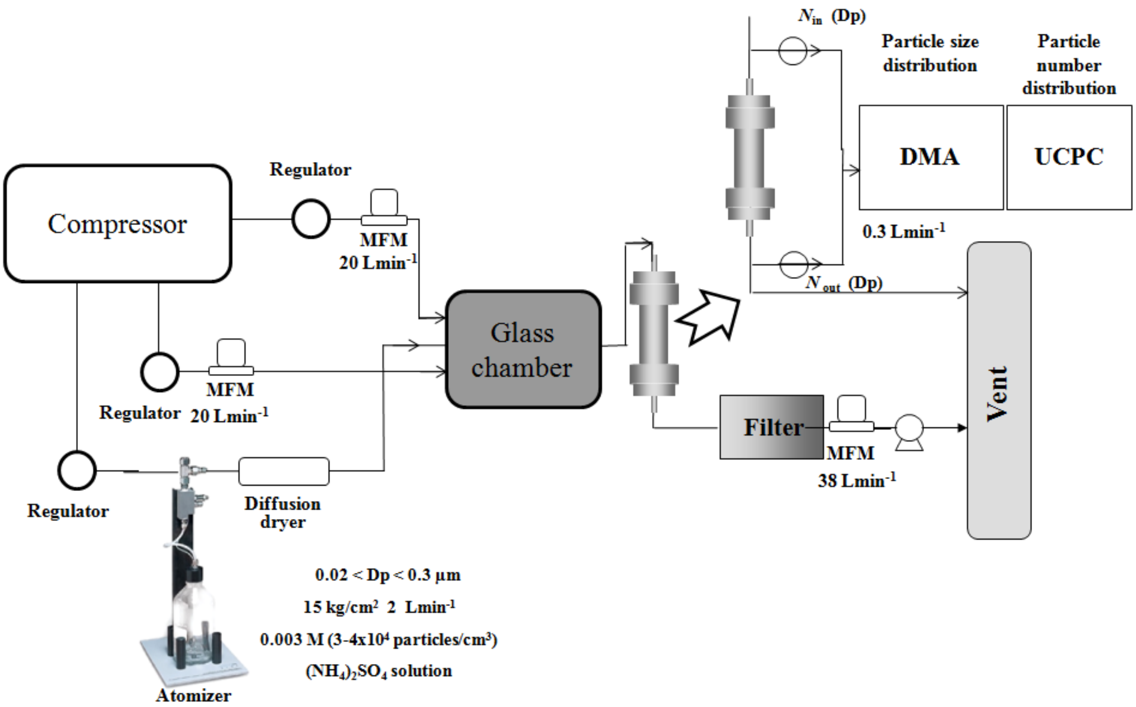
measured to determine the effect of sampling flow rate on electrostatic forces and particle diffusion. The fraction of particle losses (η_L) was estimated from the following equation:

$$\eta_L = 1 - (N_{out}/N_{in}) \quad (1)$$

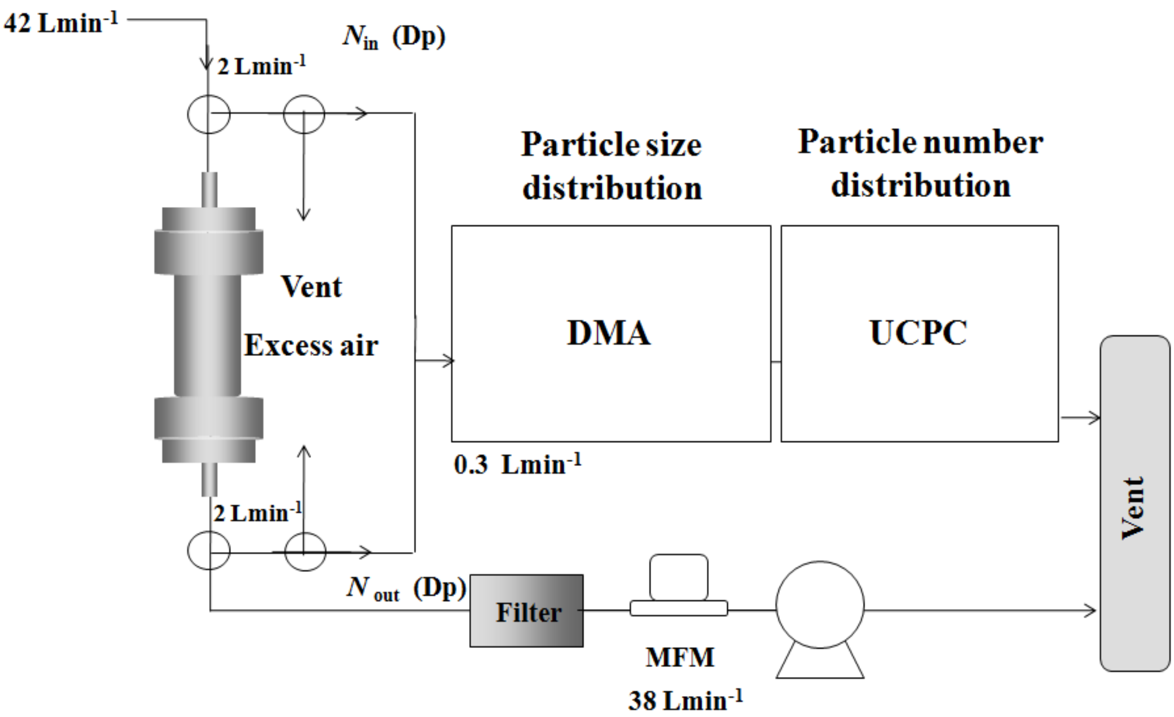
Denuder Cleaning and Coating Procedure

Citric acid has been widely used to coat the walls of denuders used to collect ammonia gas. However, some researchers recently suggested that using citric acid coating for ammonia gas suffers from insufficient strength of the bond between collected ammonia and the coating layer, which causes a release of the collected ammonia both towards the active sites of the denuder glass (Perrino and Gherardi, 1999; McCulloch and Shendrikar, 2000). They studied three coating layers, citric and phosphorous acid and oxalic acid, and found that phosphorous acid proved to be more suitable for ammonia determination. Many studies have used phosphorous acid to collect ammonia gas since its capability was proved (Perrino *et al*., 2002; Yu *et al*., 2006; Edgerton *et al*., 2007).

Denuders were cleaned as follows. Each denuder was cleaned prior to use. First, the denuder was rinsed with methanol for 20 s with manual rotation, as described elsewhere (Winberry *et al*., 1990; Winberry, 1992). Second, the denuder was rinsed with deionised water for 20 s twice with manual rotation. The denuder coating was prepared by dissolving 1% phosphorous acid in a 1:9 water–methanol solution (10 g phosphorous acid, 100 mL deionised water, and 900 mL methanol) as described by Yu *et al*.



(a) Overall experimental set-up.



(b) Experimental set-up in detail.

Fig. 2. Experimental set-up for particle loss measurement (a) with detail (b): UCPC, ultrafine condensation particle counter; DMA, differential mobility analyzer.

(2006). A total of approximately 18 mL of coating solution was applied to the denuder wall for the 1-channel denuder (three 6-mL portions), 30 mL was applied for the 3-channel denuder (three 10-mL portions), 42 mL was applied for the 5-channel denuder (three 14-mL portions), and 60 mL was applied for the 8-channel denuder (three 20-mL portions) to coat the denuder surfaces efficiently. Each coating procedure was carried out for 20 s with manual rotation to distribute the coating solution evenly. The coated denuders were dried with under a flow of ultrapure nitrogen gas (1 L/min) for several minutes; then both ends were capped with clean caps, and the denuders were soon used for gas collection.

Sample Extraction

Sample extraction had to be considered since multichannel denuders were used in this study. The main method used for sample extraction is the U.S. EPA method for an annular denuder with only 1 channel (Winberry *et al*., 1990; Winberry, 1992). Although many researchers are currently using multichannel annular denuders, there have been no detailed studies of sample extraction methods specifically for multichannel annular denuders. When extraction is necessary, the efficiency of the extraction method must be considered. In this study, we used different volumes of extract solution for each multichannel denuder because the denuders have different surface areas. The extraction efficiency of each denuder was calculated to optimize the extraction. Although the U.S. EPA method recommends 10-mL total volume of extract solution for 1-channel denuder, 15 mL total volume of extract solution was applied for 1-, 3-channel denuders for the effectiveness.

The denuders were extracted with deionised water immediately after sampling. The applied extract solution was determined on the basis of pre-experiments. Different volumes (total 30 mL) of extract solution were used for the 5- and 8-channel denuders for the pre-experiments, but the extraction efficiencies were lower than 90% for the 5- and 8-channel denuders. In contrast, extraction efficiencies greater than 95% were obtained for the 1- and 3-channel denuders with a 15-mL total volume of extract solution. Therefore, 15 mL of deionised water was applied for the 1- and 3-channel denuders, and 45 mL of deionised water was applied for the 5- and 8-channel denuders in this study. The total volume of extract solution was divided into three portions to improve the extraction efficiency (three 5-mL portions for the 1- and 3-ch; three 15–mL portions for the 5- and 8-ch). Each extraction was carried out for 1 min with manual rotation.

Sample Analysis

The collected ammonia gas was analyzed by ion chromatography (DX-100 Ion Chromatograph, Dionex, CA, USA). The ion chromatograph was calibrated prior to the analysis by injection of aqueous standards prepared in our laboratory from analytical-grade salts. Concentrations of collected NH_4^+ were calculated from quadratic regression equations for NH_4^+ standards. And the numerical factor 0.944 was used to convert the measured NH_4^+ concentration to the equivalent concentration of ammonia. Therefore the product of the factor and the concentration of the NH_4^+ collected by denuders directly estimated the ammonia concentration ($C_g(NH_3)$) (Winberry *et al*., 1990). The following chemicals were used in this study: ammonium sulfate (99.5% purity, Wako, Osaka, Japan), phosphorous acid (97% purity, Wako), ammonium ion standard solution (NH_4^+ 1000, Wako), sulfuric acid (1 mol/L, Wako), and methanol (99.7% purity, Wako).

RESULTS AND DISCUSSION

Extraction Efficiency

The extracted denuders were extracted once more to measure the extraction efficiency, which was calculated by dividing the amount of gas obtained from the first extraction by the sum of amounts obtained from the first and second extractions. The extraction efficiencies were 96.4, 98.4, 96.7, and 92.0% for the 1-, 3-, 5-, and 8-channel denuders, respectively. The extraction efficiencies of the 1-, 3-, and 5-channel denuders were acceptable, but the efficiency of the 8-channel denuder was somewhat lower than the others, owing to the lack of extraction solution compared to its surface area.

Gas Collection Efficiency

The theoretical gas collection efficiency of a denuder can be determined from the following equations (Possanzini *et al*., 1983):

$$E = 1 - (C/C_0) \quad (2)$$

$$C/C_0 = 0.82 \exp{-22.53\Delta a} \quad (3)$$

$$\Delta a = (\pi DL/4F)(d_1 + d_2)/(d_2 - d_1) \quad (4)$$

where E is the collection efficiency, C_0 is the inlet pollutant concentration, C is the exit pollutant concentration, L is the length of the denuder, d_1 is the inner diameter of the annulus, d_2 is the outer diameter of the annulus, F is the air flow rate, and D is the diffusion coefficient of the pollutant of interest. The diffusion coefficient of ammonia gas (0.227 cm^2/s at 20°C) was used to calculate the theoretical collection efficiency as studied by Andrew (1955). Table 2 shows the gas collection efficiencies. The theoretical collection efficiencies were calculated with Equations 2–4, and the experimental collection efficiencies were calculated with Eq. (2). The theoretical and experimental values are compared in the table. The experimental gas collection efficiencies were nearly equivalent to the theoretical values. The collection efficiency of the 5-channel denuder had a somewhat high standard deviation and this was probably due to the longer denuder length (400 mm) than the others. There was no breakthrough during the 1- and 6-h sampling duration.

Table 2. Gas collection efficiencies and extraction efficiencies for the 1-, 3-, 5-, and 8-ch denuders.

	Experimental collection efficiency (%)	Theoretical collection efficiency (%)	Extraction efficiency (%)
1-ch	96.3 ± 0.5	97.1	96.4 ± 0.4
3-ch	98.6 ± 0.3	99.7	98.4 ± 0.3
5-ch	98.2 ± 1.6	99.9	96.7 ± 0.8
8-ch	98.3 ± 0.2	99.9	92.0 ± 0.7

1. The gas collection efficiencies and the extraction efficiencies are the geometric mean values over 12 replicate tests.
2. Each value is given with the standard deviation obtained over 12 replicate tests.
3. The relative standard deviations were less than 1% and 1.5% for the gas collection efficiency (except for the 5-ch denuder) and the extraction efficiency, respectively.
4. The theoretical gas collection efficiency was calculated with an NH_3 diffusion coefficient (D) of 0.227 cm^2/sec (Andrew, 1955) and at a flow rate of 40 L/min.
5. The concentration of ammonia gas was 7-10 μg/m^3.

Particle Loss

Although the gas collection efficiency is high, particle loss can occur inside of the denuder. Because annular denuders are designed to have a narrow annular space, there could be particle losses, especially for UFPs (< 0.1 μm in diameter) mainly due to the diffusion of particles and electrostatic forces in the annular space. The particle loss by diffusion was evaluated using the equation developed by Hinds (1982). This equation was first established for the diffusion and loss of particles in a rectangular cross section. Ye *et al*. (1991) later adapted it for annular denuders. The fraction of particles lost by diffusion (η_L) in a denuder channel can be estimated from the following equations:

$$\eta L = 1 - 0.910e^{-7.54\mu} - 0.0531e^{-85.7\mu} - 0.0153e^{-249\mu} \quad (5)$$

$$\mu = D(d)\, LW/Qh \quad (6)$$

where $D(d)$ is the diffusion coefficient of a particle diameter, L is the tube length, W is the mean perimeter of the channel under study, Q is the flow rate, and h is the width of the channel. The particle transmission efficiency can be calculated from the following equation:

$$E = (1 - \eta_L)100 \quad (7)$$

The theoretical particle transmission efficiencies and the corresponding experimental values with particle sizes are presented in Table 3 (at 20 L/min) and 3 (at 40 L/min).

The standard deviations seem large; we attributed the large standard deviations mainly due to alterations in the electrostatic surface charge caused by various factors, including changes that occurred during the drying and assembling procedure. Ye *et al.* (1991) reported the electrostatic surface charge problems in an annular denuder previously. Therefore, the large standard deviations, particularly for the 5- and 8-channel denuders, may have been due to the fact that these multichannel denuders have many annulus spaces providing more places to be altered by the electrostatic forces.

As shown in Tables 3 and 4, particle loss increased as the flow rate decreased and the number of channels increased. Because of the vertical position of the denuder and the laminar flow conditions (Re < 2000) in the channel of the denuder, the main mechanisms responsible for the losses of the small particles (< 0.2 μm in diameter) were electrostatic forces and diffusion (Febo *et al.*, 1999). Previous studies (Ye *et al.*, 1991; Roussel *et al.*, 2004) showed that particle loss is negligible for bigger particles (> 0.1 μm in diameter) and significant for smaller particles (< 0.1 μm in diameter). Particle transmission efficiency for all denuder channels at 20 L/min was much lower than the theoretically expected particle transmission efficiency, especially for the particles smaller than 0.04 μm and for the 5- and 8-channel denuders, whereas the particle transmission efficiency at 40 L/min was close to the expected transmission efficiency, especially for the 1- and 3-channel denuders, which indicate that the electrostatic forces were minimized. Particle transmission efficiencies for different numbers of channels at the two flow rates are plotted in Figs. 3. a and b. The figure clearly shows the effect of flow rate and the number of channels on the size-segregated particle losses in the denuders.

Unlike the particles studied by Ye *et al.* (1999) the particles generated from the atomizer in our study could be affected not only by particle diffusion but also by the electrostatic field in the denuder walls because the generated particles were not neutralized before being introduced to the denuder section. The theoretical particle transmission efficiencies presented in Tables 3 and 4 were only considered particle losses by diffusion using Eq. (5) and (6), as described in Section *Particle loss*. Therefore, the particle losses that occurred at 20 L/min were larger than the expected values mainly because of the local electrostatic field in the denuder channels. For particles smaller than 0.04 μm in diameter, electrostatic forces cannot be considered as negligible (Roussel *et al.*, 2004). Madler and Friedlander (2007) studied on transport of nanoparticles, and one of their conclusions was that the surface of nanoparticles is large enough to carry one or more charges which greatly influence their transport in electrical fields.

Furthermore, it was observed that the electrostatic forces could be reduced when the flow rate increased, because the decrease of transmission efficiency was not significant at the high flow rate (40 L/min), whereas the decrease in transmission efficiency was significant at the low flow rate (20 L/min). This lower influence for the highest flow rate is the consequence of the shorter residence time of the particle and

Table 3. Theoretical and experimental particle transmission efficiencies at 20 L/min for each denuder.

	1-ch (%)		3-ch (%)		5-ch (%)		8-ch (%)	
Dp (μm)	Theo	Ex	Theo	Ex	Theo	Ex	Theo	Ex
0.02	97	98 ± 13	96	86 ± 11	93	72 ± 8	89	81 ± 14
0.04	97	98 ± 11	97	89 ± 11	95	80 ± 9	93	91 ± 12
0.08	98	95 ± 6	98	90 ± 7	97	91 ± 9	96	89 ± 10
0.13	98	93 ± 6	98	99 ± 8	98	99 ± 11	97	91 ± 15
0.2	98	97 ± 12	98	97 ± 4	98	95 ± 14	98	96 ± 22
Mean	97.7	94.7 ± 9	97.5	92.7 ± 8	96.5	87.1 ± 10	95.1	88.2 ± 14

1. Each value is given with the standard deviation obtained over 16 replicate tests.

2. Theo, theoretical particle transmission efficiency; Ex, experimental particle transmission efficiency.

Table 4. Theoretical and experimental particle transmission efficiencies at 40 L/min for each denuder.

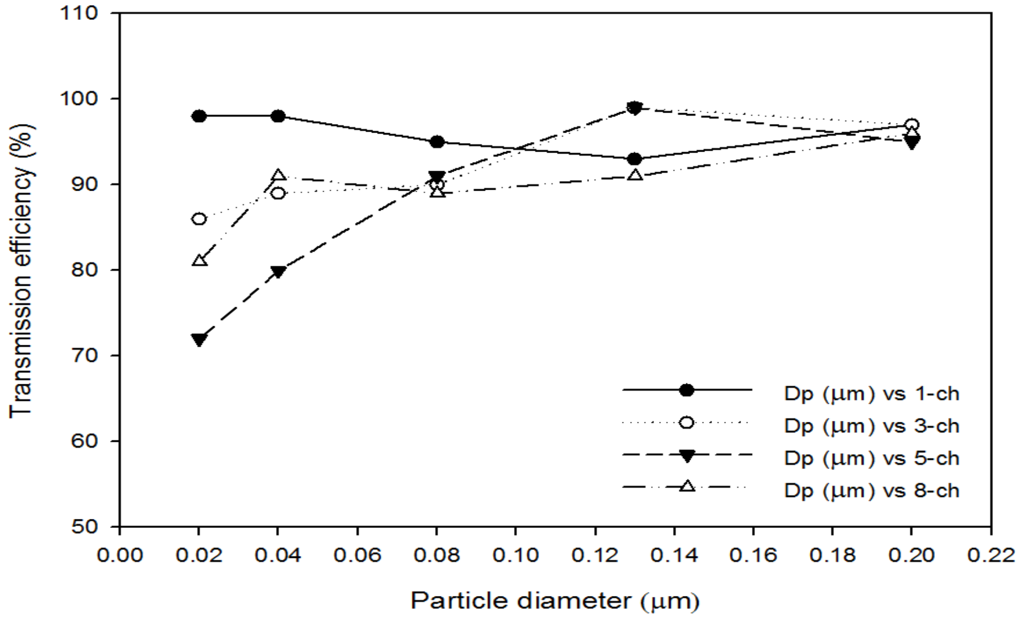
	1-ch (%)		3-ch (%)		5-ch (%)		8-ch (%)	
Dp (μm)	Theo	Ex	Theo	Ex	Theo	Ex	Theo	Ex
0.02	97	96 ± 13	97	98 ± 9	95	91 ± 11	93	94 ± 12
0.04	98	97 ± 6	97	95 ± 7	96	95 ± 7	95	97 ± 13
0.08	98	98 ± 7	98	97 ± 6	97	93 ± 7	97	91 ± 9
0.13	98	98 ± 8	98	98 ± 12	98	99 ± 7	98	94 ± 11
0.2	98	98 ± 8	98	98 ± 12	98	92 ± 12	98	94 ± 13
Mean	97.8	97.3 ± 8	97.7	97.4 ± 9	97.0	94.7 ± 9	96.5	93.4± 12

1. Each value is given with the standard deviation obtained over 16 replicate tests.

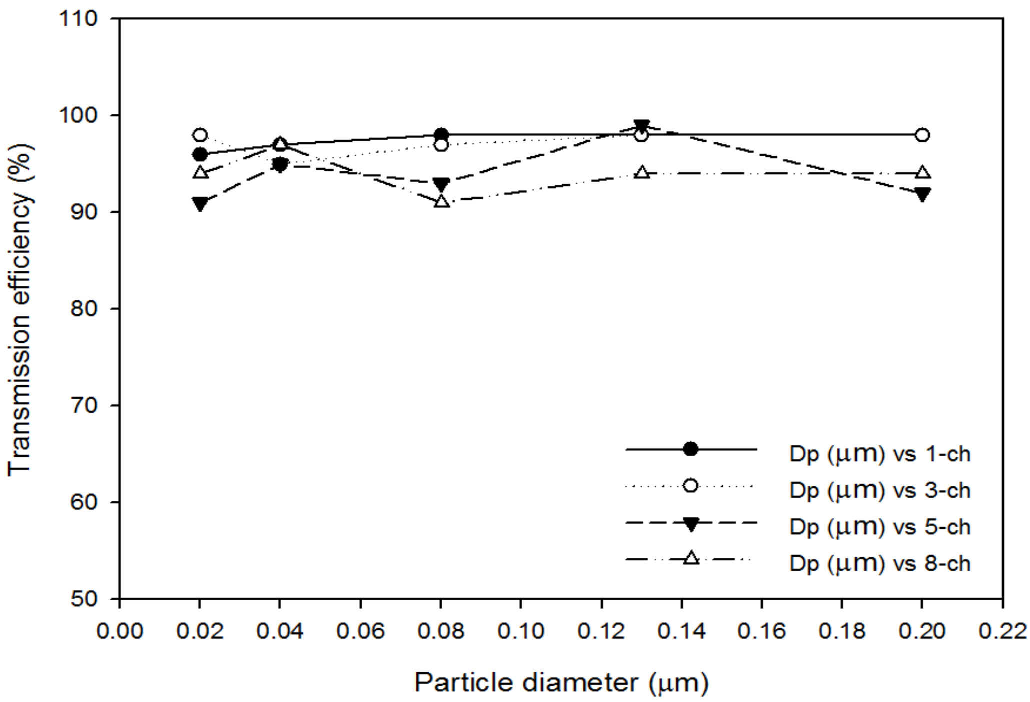
2. Theo, theoretical particle transmission efficiency; Ex, experimental particle transmission efficiency.

minimized electrostatic forces in the denuder channels. As a matter of fact, the residence times at 20 L/min were 0.05, 0.09, 0.28, and 0.54 s for the 1-, 3-, 5-, and 8-channel denuders, respectively whereas the residence times at 40 L/min were 0.03, 0.05, 0.14, and 0.27 s for the 1-, 3-, 5-, and 8-channel denuders, respectively. A similar effect of electrostatic forces and shorter residence time was also observed by Roussel *et al.* (2004) and theoretically considered by Ye *et al.* (1991). The results of particle transmission efficiencies observed in previous studies (Ye *et al.*, 1991; Roussel *et al.*, 2004) were compared with those observed in this study as shown in Table 5. The annular denuders used in the previous studies were manufactured by URG and have the same geometry (1- and 8-channel denuders).

The particle transmission efficiencies at 10 L/min with 1-channel denuder obtained by Ye *et al.* (1991) and at 20 L/min with 1-channel denuder estimated in this study are compared in Table 4. As shown in this table, the particle transmission efficiency of this study was a little lower than that obtained by Ye *et al.* even though our flow rate was



Particle transmission efficiencies at 20 L/min



Particle transmission efficiencies at 40 L/min

Fig. 3. Particle transmission efficiencies for 1-, 3-, 5-, and 8-channel denuders at 20 L/min (a) and 40 L/min (b).

Table 5. Comparison of particle transmission efficiencies obtained in previously reported studies and in this study.

Dp (μm)	1-ch at 20 L/min[a]	1-ch at 10 L/min[b]	8-ch at 40 L/min[a]	8-ch at 34 L/min[c]
0.02	98 ± 13	96	94 ± 12	-
0.04	98 ± 11	98.6	97 ± 13	92 ± 6
0.08	95 ± 6	95.9	91 ± 9	96 ± 3
0.13	93 ± 6	95.9	94 ± 11	95 ± 2
0.2	97 ± 12	-	94 ± 13	94 ± 2
Mean	94.7 ± 9	96.6 ± 2	93.4 ± 12	94.2 ± 3

a: Results from this study.

b: Particle transmission efficiency for an uncoated 1-channel annular denuder for Boltzmann charged particles (Ye *et al*., 1991; the particle transmission efficiency for 0.04 μm particles is the average of the particle loss values for 0.03 and 0.05 μm particles).

c: Particle transmission efficiency for an 8-channel annular denuder coated with XAD-4 for diesel exhausts (Roussel *et al*., 2004).

higher. This result could be explained in terms of differences between generated particles. Ye *et al*. (1991) used monodisperse and neutralized particles or Boltzmann charged particles to estimate the effects of particle diffusion and electrostatic forces separately. However, in the atmosphere, particles are polydisperse, and both neutral and charged particles can be present; hence particle losses could be larger. In contrast, the particle transmission efficiencies at 34 L/min with 8-channel denuder obtained by Roussel *et al*. (2004) and by us at 40 L/min were quite similar. Roussel *et al*. (2004) used diesel vehicle exhaust to estimate the particle transmission efficiency with XAD-4 coated 8-channel denuders. Therefore, they could estimate the particle transmission efficiency under real atmospheric conditions. Because the UFPs and nanoparticles can be lost in denuder systems, it is very important to assess these small particles (< 0.1 μm) in denuders for an AD-FP system. However, it was found that the loss of particles with diameters < 0.1 μm could be minimized when the flow rate is higher; the particle losses in the important 0.02–0.2-diameter range were less than 3% for the 1- and 3-channel denuders at 40 L/min.

CONCLUSIONS

The gas collection efficiencies and particle transmission efficiencies were estimated at 40 L/min using ammonia gas and ammonium sulfate particles (diameters, 0.02–0.2 μm) for 1-, 3-, 5-, and 8-channel denuders to assess the possibility of combining these denuders with a newly developed inertial classification system. Gas collection efficiencies for the 1-, 3-, 5-, and

8-channel denuders were 96.3, 98.6, 98.2, and 98.3% (1-ch < 5-ch, 8-ch, and 3-ch), respectively. The extraction efficiency was over 96% for all denuders except for the 8-channel denuder (92%) with the extraction procedure described herein. The experimental gas collection efficiencies were nearly equivalent to the predicted efficiencies. The particle transmission efficiencies for the 1-, 3-, 5-, and 8-channel denuders at 40 L/min were 97.3, 97.4, 94.7, and 93.4% (8-ch < 5-ch < 1-ch and 3-ch), respectively. Furthermore, it was found that the particle losses can be reduced by increasing the flow rate.

The 3-channel denuder would be the best denuder to use in an AD-FP system combined with the inertial classification system, in terms of the collection efficiency and particle transmission efficiency (for particles with diameters < 0.1 μm). However, the 1-channel denuder could also be used with the new classification system because the 1-channel denuder is competitively priced, coating the denuder wall is easier, and the 1-channel denuder is easy to carry for field sampling. Furthermore, gas collection efficiency for denuder is generally acceptable when it is over 95%.

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