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Aerosol Science 36 (2005) 1400–1408

Journal of Aerosol Science

www.elsevier.com/locate/jaerosci

Counting and particle transmission efficiency of the aerodynamic particle sizer

John Volckens[a,*], Thomas M. Peters[b]

[a] U.S. Environmental Protection Agency, National Exposure Research Laboratory, Research Triangle Park, NC 27711, USA
[b] Department of Environmental and Occupational Health, University of Iowa, Iowa City, IA, USA

Received 29 January 2005; received in revised form 14 March 2005; accepted 25 March 2005

Abstract

The aerodynamic particle sizer (APS) measures the size distributions of particles with aerodynamic diameter between 0.5 and 20 μm in real time. To provide accurate size distributions, the APS must measure both particle size and concentration correctly. The objective of this study was to characterize the counting efficiency of the APS as a function of particle size (0.8–10 μm), particle type (liquid or solid), and APS model number (3310 vs. 3321). For solid particles, counting efficiencies ranged between 85% and 99%. For liquid droplets, counting efficiencies progressively declined from 75% at 0.8-μm drops to 25% for 10-μm drops. Fluorometric wash tests indicated that transmission losses occur when larger droplets impact on the instrument's inner nozzle. However, transmission losses did not account entirely for the reduced droplet counting efficiencies, indicating that additional losses may have occurred downstream of the inner nozzle. Between instrument comparisons revealed that although multiple APSs report similar number concentrations, small deviations in particle sizing can produce substantial errors when number concentrations are converted to mass concentrations.

Keywords: Aerodynamic particle sizer; Time-of-flight; Counting efficiency; Particle size

* Corresponding author. Present address: Department of Environmental and Radiological Health Sciences, Campus Delivery 1681, Colorado State University, Ft. Collins, CO 80523. Tel.: +1 970 491 6341; fax: +1 970 491 2940.

E-mail address: john.volckens@colostate.edu (J. Volckens).

0021-8502/$ - see front matter
doi:10.1016/j.jaerosci.2005.03.009

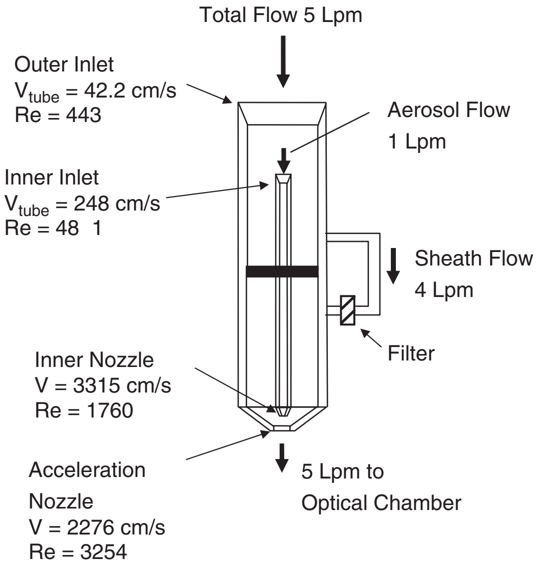

Fig. 1. Diagram of the aerosol flow path in the APS.

1. Introduction

The aerodynamic particle sizer (APS) (model 3321, TSI Inc., Shoreview, MN) measures aerosol size distributions from 0.5 to 20 μm by determining the time-of-flight of individual particles in an accelerating flow field. To provide accurate size distributions, the APS must determine both particle size and number concentration correctly. Generally, sizing accuracy has not been a problem with the APS, except in the case where large droplets having low viscosity and surface tension may deform during acceleration through the sensing zone (Baron, 1986; Bartley, Martinez, Baron, Secker, & Hirst, 2000; Chen, Cheng, & Yeh, 1990). Counting accuracy, however, has been an issue with this instrument (Armendariz & Leith, 2002; Kinney & Pui, 1995; Peters & Leith, 2003).

A simplified version of the aerosol flow path in the APS is shown in Fig. 1. Aerosol enters the outer inlet at a flow of 5 Lpm and is divided into two flows: 1 Lpm of sample flow enters the inner inlet, while the remaining 4 Lpm of sheath flow is passed through a high efficiency filter. Aerosol traverses the inner tube, exits the inner nozzle, and recombines with the cleaned, particle-free sheath flow (4 Lpm), and the summed 5 Lpm passes through an acceleration nozzle and then into the sensing zone. In the sensing zone, particles are sized according to their transit time through two overlapping laser-diode beams. Flow dynamics at the inner nozzle and acceleration nozzle are in the turbulent-transition regime with Re numbers of 1700–3200, respectively.

The overall counting efficiency (η) for a particle counting instrument is a product of aspiration (η_a), transmission (η_t), and detection (η_d) efficiencies, which can be represented mathematically as (adapted from Brockmann, 2001)

$$\eta_{\text{overall}} = \eta_a \eta_t \eta_d. \tag{1}$$

Each of these efficiencies is a function of particle diameter. Factors external to the APS affect particle aspiration, such as windspeed and direction (Chen, Barber, & Zhang, 1998). Separate aspiration efficiencies exist for the outer inlet and the inner inlet (see Fig. 1 for details). Whereas outer inlet aspiration has not been well characterized, Kinney and Pui (1995) found inner inlet aspiration to be affected by super-isokinetic sampling conditions: air velocity is approximately 40 cm/s within the outer inlet and approximately 250 cm/s within the inner inlet (Fig. 1). Transmission efficiency includes any losses that occur when particles deposit upon the interior walls of the inner inlet/nozzle assembly. The aspiration and transmission characteristics of the instrument have remained constant because inlet and nozzle geometries and flow rates have not changed since the first APS (model 33) was introduced in 1980 (Wilson & Liu, 1980). However, the detection efficiency of the APS, dependent upon optics and signal processing circuitry, may have changed as newer APS models were introduced: model 3310 in 1987; model 3320 in 1997; and model 3321 in 2001 (Blackford, Hanson, Pui, Kinney, & Ananth, 1988; Caldow, Quant, Holm, & Hairston, 1997; Stein et al., 2002).

Several studies have investigated the overall counting efficiency of the APS. Remiarz, Agarwal, and Johnson (1982) compared the response of an APS 33 to the output of the vibrating orifice aerosol generator (VOAG); in this case, the VOAG was considered a primary standard for particle number concentration. They found that the overall counting efficiency of the APS model 33 for liquid dioctyl phthalate droplets ranged from 100% for particles between 1.5 and 4 μm to 75% for 10-μm particles. Blackford et al. (1988) used the same method with corn oil to determine the overall efficiency of the APS 3310: 95% between 1 and 3 μm; 80% for 4-μm particles; and 36% for 10-μm particles. In both experiments, the APS sampled from the downward flowing airstream of the VOAG drying column.

Kinney and Pui (1995) physically removed the inlet section from an APS and replaced the optical detection region with a filter. They used a fluorometric-detection technique to determine aspiration and transmission efficiency of the inner inlet/nozzle assembly as a function of monodisperse particle size. They too sampled from the downward flowing airstream of the VOAG drying column. They found that the aspiration efficiency of the inner inlet and nozzle is very sensitive to the degree of super-isokinetic sampling, and that transmission efficiency is reduced due to inertial impaction of droplets on the interior walls of the inner inlet. When Kinney and Pui added aspiration and transmission efficiencies together, they obtained similar counting efficiencies as reported by Blackford.

Other researchers have measured the overall counting efficiency of the APS by comparison to a cascade impactor. For the older APS 3320, Armendariz and Leith (2002) reported overall counting efficiencies for oil mist droplets that increased from 30% for 0.5-μm particles to 100% for 0.9-μm particles, then decreased to 60% for 5-μm particles. For the newer APS 3321, Peters and Leith (2003) reported overall counting efficiencies for oil mist droplets from 40% for 0.8-μm particles to 60% for 4-μm particles. In contrast to the earlier studies, the APS and the cascade impactor both sampled at 5 Lpm each from a chamber. However, cascade impactors have their own inherent biases (e.g., aspiration losses, non-ideal stage cuts, inter-stage losses). Therefore, whether the counting errors for the new APS model 3321 are the result of biases from particle aspiration, particle transmission, optical measurement, or impactor error remains somewhat unclear.

In this study, overall counting efficiency ($\eta_a \eta_t \eta_d$) was determined by comparing an APS 3321 measurement to a filter catch of monodisperse, fluorescently tagged particles. Fluorometry avoids the biases associated with cascade impaction and offers a low limit of detection with good precision. The primary objective was to determine counting efficiency for the APS 3321 as a function of particle size (0.8–10 μm) and particle type (liquid or solid). A secondary objective was to compare the performance of several APS models concurrently (one model 3310 vs. three model 3321s).

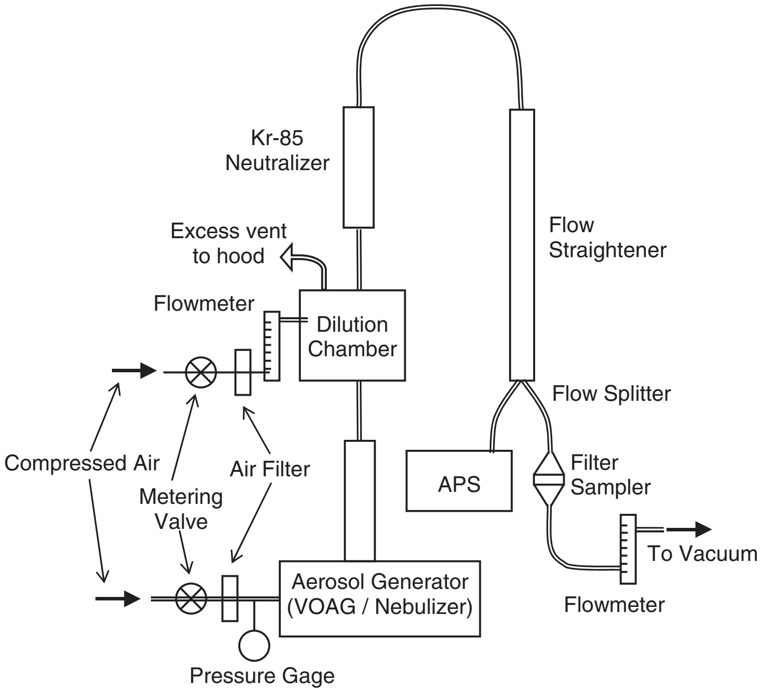

Fig. 2. Experimental setup for the APS counting efficiency tests.

2. Methods

2.1. Counting efficiency of the APS 3321

Counting efficiency was determined by comparing the response of the APS 3321 (s/n 1270) to the fluorometric material collected on a filter. A schematic of the experimental setup is shown in Fig. 2. Monodisperse liquid droplets were created using serial dilutions of an ethanol-based solution containing oleic acid tagged with uranine (90:10 mixture) and aerosolized with a vibrating orifice generator (VOAG 3450 TSI Inc., Shoreview, MN). Two types of solid, monodisperse aerosols were generated: (1) fluorescent, polystyrene latex spheres (Duke Scientific, Palo Alto, CA) nebulized from suspension in distilled, deionized water, and (2) ammonium fluorescein particles created with the VOAG (Vanderpool & Rubow, 1988).

After exiting the generator, aerosol was mixed in a dilution chamber with filtered, compressed air, and then passed through a Kr-85 charge neutralizer (Model 3012, TSI Inc., Shoreview, MN). Next, the aerosol passed through a 180° bend, followed by a straightening section to make the velocity and particle concentration profile uniform. A sharp-edged flow splitter directed the aerosol into either the APS or a filter sampler. The filter holder was fitted with an inlet identical to the outer inlet of the APS, and both samplers were operated at a flow rate of 5 Lpm to ensure equivalent outer inlet aspiration efficiencies. As a result, outer inlet aspiration efficiency was not evaluated here; inner inlet aspiration and nozzle transmission efficiency were included in counting efficiency measurements. Preliminary tests were conducted with

Table 1
Experimental variables for counting efficiency tests

Particle type	Particle composition	Generation method	Extraction solvent	Aerodynamic diameters (μm)	Models tested
Liquid	Uranine and oleic acid	VOAG[b]	0.1 N NaOH	0.8, 1.4, 3.4, 5.7, 10.5	3321
Solid	Fluorescent PSL[a]	Nebulizer	Xylene	0.8, 1.0, 3.0, 5.1	3321, 3310
Solid	Ammonium Fluorescein	VOAG	0.1 N NaOH	9.4	3321

[a]Polystyrene latex (PSL).
[b]Vibrating orifice aerosol generator (VOAG).

total filters drawing 5-Lpm airflow on each leg of the flow splitter. These tests demonstrated an equivalent split for an aerosol composed of 10-μm drops. Sampling times were varied from 2 to 20 min to obtain a sufficient amount of fluorometric material on the filter for detection. Table 1 provides the relevant particle generation variables. Three tests were conducted for each particle size.

Immediately after sampling, filters were solvent extracted by sonication to disperse the fluorescent material freely throughout the extraction solution. The PSL spheres were extracted with 10 mL of xylene (Fisher Scientific, Fairlawn, NJ) (Glenny, Bernard, & Brinkley, 1993); the oleic acid droplets were extracted with 10 mL of 0.1 N sodium hydroxide (Fisher Scientific, Fairlawn, NJ) (Tolocka, Tseng, & Wiener, 2001); and the ammonium fluorescein particles were extracted with 10 mL of 0.1 N ammonium hydroxide (Fisher Scientific, Fairlawn, NJ) (Vanderpool & Rubow, 1988). The inner inlet/nozzle assembly of the APS was then removed from the instrument, and particle transmission losses were determined using a fluorometric wash-off technique (Tolocka et al., 2001). First the exterior of the inlet was wiped clean and then separate washes were conducted on the inner inlet, the nozzle tip, and the remainder of the interior of the inner tube. Filter and wash extracts were quantified with a digital filter fluorometer (Turner Quantech model FM109515, Barnstead/Thermolyne Inc., Dubuque, IA). Particle number concentration was calculated by converting particle mass, as determined by fluorescence spectroscopy, to number knowing the density and physical diameter of each particle size. Physical diameters of VOAG generated aerosols were confirmed by collecting particles on Nyebar coated glass slides and sizing the droplets with a Nikon Microphot-FXA light microscope (Nikon Instruments Inc., Marietta, GA) using digital sizing software (ImageJ, NIH, Washington, DC). The droplet distortion correction by Baron, Deye, Martinez, and Jones (2004) was applied to all results to account for deformation of liquid droplets in the acceleration nozzle.

2.2. Model comparison

A second experiment compared the response of four APS instruments simultaneously measuring a series of monodisperse PSL particles. This experiment followed a similar setup to that in Fig. 2, except that a 4-way flow splitter was used to divide the aerosol between samplers. Three APS model 3321 (s/n 1270, 1277, 1282) and one model 3310 (s/n 7634) sampled nebulized solutions of solid, monodisperse PSL spheres. Three tests were conducted for each PSL particle size listed in Table 1.

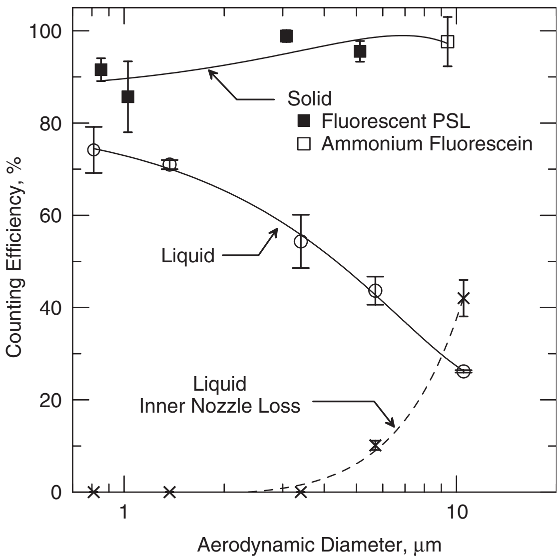

Fig. 3. Counting efficiency of the APS 3321 and liquid losses within the inner nozzle vs. aerodynamic particle diameter.

3. Results

3.1. Counting efficiency of the APS 3321

Fig. 3 presents counting efficiency of the APS 3321 as a function of particle size for both solid and liquid particles. Counting efficiencies for solid particles ranged from 85% to 99%. No fluorometric material was recovered in the inner inlet, the interior of the inner tube, or inner nozzle for tests conducted with solid aerosols. Counting efficiency for liquid droplets progressively decreased from 74% at 0.8 um to 26% at 10 um. Fig. 3 also presents droplet transmission losses in the tip of the inner nozzle as a percentage of particles sampled by the instrument. These losses were evident for droplets larger than 5 µm, and increased substantially with droplet size. For the 10-µm droplets, 3% deposited in the inner inlet, 4% deposited within the inner tube, 31% deposited at the tip of the inner nozzle and 26% were counted correctly by the detector. For the 5-µm drops, 0.5% deposited within the inner tube, 9% deposited at the tip of the nozzle, and 44% were counted correctly by the detector. A negligible amount of fluorometric material was detected in the inner inlet, the interior of the inner tube, and the inner nozzle for the 1 and 3 µm drops.

3.2. Model comparison

Fig. 4 shows number and mass concentrations of PSL spheres measured by four APS instruments sampling simultaneously. Although paired *t*-tests indicated that particle number concentrations measured by each instrument were statistically different ($p_{avg} = 0.06$, except when counting 3-µm particles where $p_{avg} = 0.29$), these differences were small, as the average coefficient of variation between instruments was 3.8% across all particle sizes. Measured mass concentrations, however, were both substantially and significantly different between the instruments ($p_{avg} = 0.007$). The coefficient of variation in mass

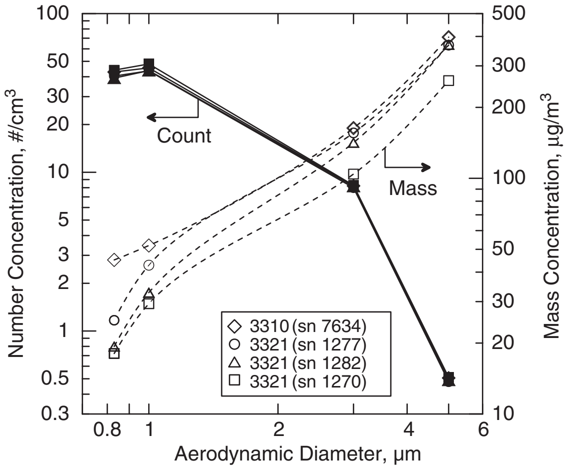

Fig. 4. Average measurements of number (left axis) and mass (right axis) concentration for PSL spheres by four APS instruments operating simultaneously. Error bars representing standard deviations of concentrations measured between tests for each instrument are within the size of the data points.

concentrations measured between the four instruments was 46.5, 26.0, 18.7, and 17.4% for particle sizes of 0.83, 1, 3, and 5 µm, respectively. No trends in counting and sizing were detected between model types 3310 and 3321.

4. Discussion

The overall counting efficiency of the APS 3321 was near 100% for the solid particles, but substantially lower for the liquid droplets. Transmission losses in the inner inlet/nozzle assembly were negligible for solid particles, but increased with particle size for liquid droplets; this trend coincided with a decrease in droplet counting efficiency. These observations indicate that solid particles probably bounce off the surfaces of the inner nozzle and re-entrain into the flow. Most of the lost droplet counts can be attributed to transmission losses within the inner nozzle (Fig. 1). Analysis of the inner inlet/nozzle wash off data revealed that the majority of droplets deposited at the 60° beveled tip near the nozzle exit, just before joining the sheath flow. However, these transmission losses do not account for all of the missing droplet counts. The fate of the remaining 10-µm droplets, 36% by comparison to the filter catch, is unknown. The percentages of 'unaccounted' droplets for the 0.8, 1, 3, and 5 µm sizes are 25%, 29%, 45%, and 46%, respectively.

Several loss mechanisms may contribute to these 'unaccounted' droplets: aspiration losses, transmission losses, detector error, and experimental error. Outer-inlet aspiration losses may be ruled out because both the APS and filter used identical outer inlets operating at 5 Lpm. Likewise, low aspiration efficiency of the inner inlet in the APS is unlikely to account for this phenomenon because the losses are so great: 45% of the 5-µm particles remain unaccounted compared to the near 100% inner inlet aspiration efficiency reported by Kinney and Pui (1995) for the APS 3310.

However, droplets might have deposited onto the interior surface of the acceleration nozzle, prior to entering the detection region (Fig. 1). Particle losses in the acceleration nozzle were not measured as part of this work because of the difficulties associated with removing it from the instrument. Particle Stokes numbers (Stk), defined as the ratio of the stop distance to the nozzle jet width, are substantially higher in the inner nozzle than in the acceleration nozzle, indicating that particle deposition due to impaction is more probable within the inner nozzle. However, shear forces resulting from the mixing of sheath and sample flows in the acceleration nozzle might have been sufficient to cause particles to diverge from the main flow and reach the walls of the acceleration nozzle.

Two types of detector errors are possible: (1) particles entering the light beam do not scatter sufficient light to create a signal, and (2) particles in the sensing region miss the laser beam and scatter no light whatsoever. The former error is processed by the APS software as an Event Type I error (TSI, 2004) and contributed, on average, to less than 5% of the total counts measured during any test runs reported here, regardless of particle type and size. The latter error would be related to the ratio of the laser beam volume to the particle flow volume in the sensing zone, which is unknown.

Small discrepancies in particle sizing lead to large discrepancies in particle mass concentrations, which are calculated from the third moment distribution of particle diameter, d_p^3. For example, when sizing 1-μm PSL spheres, the count modes for the four instruments were 1.04, 1.11, 1.19, and 1.11 μm. If we assume that two instruments counted similar number concentrations, a 15% discrepancy in sizing the diameter would produce a 50% difference in reported mass concentration. Such discrepancies are easily seen in the mass distribution data plotted in Fig. 4. For the work conducted here, errors in particle sizing could have contributed to particle counting inefficiencies, since counting efficiency was determined indirectly from a mass measurement. However, we do not believe that to be the case, since the APS (s/n 1270) was factory calibrated prior to testing and measured particle diameters, from which mass was determined, were independently verified using light microscopy.

5. Conclusions

Several conclusions may be taken from this work:

- Solid particles tend to bounce off interior surfaces of the inner nozzle and, therefore, are counted more efficiently than liquid droplets that tend to stick upon contact.
- Liquid drops deposit at the beveled tip of the inner nozzle, a trend that increases with droplet size.
- Our work could not account for all of the ‘missing’ droplet counts; thus, another loss mechanism must be present for droplets.
- Counting and processing circuitry are capable of near 100% efficiency for solid, monodisperse particles; this observation is true for the APS 3310 and the APS 3321.
- Small sizing errors produce substantial mass biases with monodisperse particle distributions.

Future work should address the uncertainties in droplet counting efficiency of the APS.

Acknowledgements

The authors wish to thank Katherine Coutros, Leonard Stockburger, and Robert Vanderpool for loaning their APS instruments, and David Leith and Maryanne Boundy for providing logistical support for this work.

Disclaimer

The United States Environmental Protection Agency through its Office of Research and Development funded and managed the research described here. It has been subjected to Agency review and approved for publication as an EPA document.

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