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Aerosolization of Particles from a Bubbling Liquid: Characteristics and Generator Development

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ABSTRACT. A new aerosol generator is introduced in which particles suspended in a liquid are aerosolized by gentle bubble bursting. Tangential injection of dry air to the bubbling surface dries the airborne droplets immediately after aerosolization so that they rapidly shrink in size and are carried out from the generator by inward and upward swirling air motion. The new generator has been evaluated with monodisperse PSL particles in the size range of 0.73–5.1 μm and with a saline solution using a time-of-flight aerodynamic particle size spectrometer (Aerosizer). It was found that, in contrast to pneumatic nebulization (e.g., with a Collison nebulizer), the new generator's output in undesirable liquid droplets is very small, while its output in dry PSL particles is high. When using the new aerosol generator, a minimum number of the nebulized droplets is returned to the liquid pool, thus optimizing the number of particles available as test aerosols. The aerosol concentration was found to be constant and stable for at least 30 min with the prototype generator tested. It is shown that the relative humidity of the effluent flow can be regulated. For microorganisms aerosolized by this generator, the shear stress is expected to be considerably lower than in conventional aerosol generators. © *1997 American Association for Aerosol Research.* AEROSOL SCIENCE AND TECHNOLOGY 26:175–190 (1997)

INTRODUCTION

The size and concentration calibration of aerosol instruments is usually performed with inert airborne particles that are spherical, such as polystyrene latex (PSL), or close-to-spherical, such as sodium chloride (Chen, 1993). In generating such aerosol particles, it is usually not critical which generation method is used as long as the desired particle size and concentration remain constant for the duration of the instrument calibration. However, when evaluating or calibrating samples used for collecting airborne microorganisms and aeroallergens, such as bacteria, fungal spores, viruses, or other materials of biological origin, the survivability of the organisms during the generation process is also of concern. Thus, the motivation for our research was to find a generation method that imparts the least stress to viable microorganisms dispersed by a liquid.

We have developed such a method for the generation of bioaerosols by dispersing the particles from a bubbling liquid. In

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evaluating this new method, we compared it to existing aerosol particle generation methods utilizing conventionally used PSL and sodium chloride (NaCl) particles. When evaluating our new method, we found that it also had advantages for the generation of conventionally used calibration aerosol particles. Our evaluation of the new generation method with conventional aerosol particles is the subject of this paper.

One of the frequently used ways to generate an aerosol is by compressed-air atomization of particle suspensions or solutions (John, 1993). This generally produces a broad spectrum of primary droplets (May, 1973). Most of these droplets are impacted onto a surface in the nebulizer so that all the larger droplets are recirculated into the liquid, and only a small mass fraction of the aerosol leaves the nebulizer. The large surface area of the nebulized cloud and the high percentage of recirculated droplets leads to a high degree of liquid evaporation in such a nebulizer. Thus, the amount of liquid lost by evaporation may exceed 50%, while less than 50% of the liquid leaves the nebulizer as droplets (Dennis, 1990). This process of pneumatic nebulization produces a droplet cloud with the majority of particles in the 0.1–0.5 μm size range. The particle concentration in this size range is generally much higher than the droplet concentration in the more desirable larger size range. If an optical single particle counter is calibrated or used in the evaluation system, the high concentration of small particles may lead to coincidence problems in the view volume of the counter (Willeke and Liu, 1976). The latter can be reduced, however, by passing the aerosol cloud through a virtual impaction stage (Pilacinski et al., 1990).

When bacteria are dispersed by the pneumatic nebulization technique, their high acceleration in the dispersing nozzle and their exposure to high-velocity air streams subject them to high shear forces that may affect their viability (Griffiths and DeCosemo, 1994). Thus, there is a need for an aerosol generation technique that imparts less stress on the organisms. We have achieved low-stress dispersion of bacteria and other biological materials by bubbling air through a liquid suspension of these materials. The bubbling of air or other gases through a bulk liquid is a common phenomenon, and is found in many natural and man-made processes (Baron and Willeke, 1986; Szewczyk et al., 1992; Juozaitis et al., 1994; Huang et al., 1994). The rising bubbles entrain the particles in the suspension and disperse most of them in small film droplets that result from the breakup of the bubble surfaces. Additional larger droplets result from the jet that propels upward when the depression at the top of the liquid fills in upon the bursting of each bubble.

Tomaides and Whitby (1976) have shown that each bubble of 0.5–6 mm diameter produces many small film droplets and a few larger jet droplets. Wangwongwatana et al. (1990) found that the geometric mean diameter of the droplets generated from 2.6-mm bubbles was about 2.0 μm with a geometric standard deviation of about 2. Blanchard and Syzdek (1982) have shown that bubbles of 1.7-mm diameter produce 10–20 film droplets with their sizes ranging from < 2 μm to over 30 μm diameter (half the droplets are < 10 μm). As the bubbles rise through the water, they collect particles on their surfaces by interception. This results in an enrichment of the particle concentration in the bubble surfaces relative to the bulk concentration in the liquid reservoir. For bubbles rising a distance of less than 2 cm through the bacterial suspension, they found a bacterial enrichment factor in the droplets between 10 and 20, i.e., entrainment of the bacteria by the bubbling surface significantly increased the concentration of bacteria in the dispersed liquid relative to the bulk liquid in the vessel. From these previous findings, we concluded that a bubbling-type generator can produce aerosol particles of the same size range as that of conventional pneumatic nebulizers. While future studies will deal with the issue of microbial stress reduction in bacterial dispersions, this paper shows the dispersion of conventional test particles by the new generation technique, and compares the data with test particle dispersion by a conventional pneumatic nebulization technique.

NEW TWO-FLOW AND SINGLE-FLOW BUBBLING GENERATORS

Two-Flow Bubbling Generator

The new two-flow bubbling generator shown in Fig. 1a consists of three major components: 1) a vessel with a cover (made of nalgen polypropylene for easy sterilization by autoclaving), 2) an air dispersion unit that creates bubbles in the liquid (made of a medium-porosity fritted disk), and 3) two glass tubes for the dry air supply that evaporates the liquid from the effluent droplets. The passage of clean, dry air through the medium-porosity fritted disk, Q_{bubl}, produces fine bubbles of about 2 mm, which rise to the top surface of the vessel's liquid while scavenging particles contained in the liquid. Once at the top, liquid in the bubbles drains downward, thus thinning the bubble surfaces until they burst into small droplets. When the top surface of an air bubble bursts, "film droplets" are created. Part of the film pulls back into the bulk liquid and moves toward the center of the liquid depression formed by the bubble. This creates an upward moving jet in the center of the liquid depression. The jet shoots up at high velocity and breaks up into several droplets. These "jet droplets" tend to be larger than film droplets (Blanchard and Syzdek, 1975; Baron and Willeke, 1986). The effluent film droplets and jet droplets may contain either no particles or one or more particles per droplet, depending on the particle concentration in the liquid and the operational characteristics of the bubbling liquid generator.

Immediately upon bursting of the bubbles, the resulting droplets are dried and carried upward in an inward swirl by two clean, dry air streams at the overall flow rate of Q_{dry}. The air streams are produced by airflow through two tubes of 1 mm diameter, aligned tangentially with the vessel's inner surface. The Reynolds number of the airflow in the tubes exceeds 15,000. Thus, a turbulent vortex is produced in the air space above the liquid. The swirl velocity increases toward the center of the vessel due to conservation of the flow's angular momentum (Dietrich, 1990). This inward motion of the swirl decreases particle losses to the inner surfaces of the vessel. Since the new generator was specifically designed for the dispersion of solid particles, the focus of this paper is on the aerosolization of solid particles. The new technique can also be used for the aerosolization of dissolved solid particles, which is not evaluated in this paper.

Single-Flow Bubble Generator

Figure 1b presents an alternate to the new bubbling generator. It is a single-flow generator in which the bubbling air flow and the drying air flow are combined into one flow, $Q_{\mathrm{bubl+dry}}$. The air dispersion unit, consisting of a medium-porosity fritted cylinder, is partially submerged in the liquid. Due to the increased resistance to air flow through the liquid, only about 1/10th of the total air flow passes through the liquid. This minor air flow produces the droplets. The majority of the air flow passes through the tiny holes in the upper portion of the fritted cylinder, producing small scale turbulence in the air.

The vessel was fabricated from two 25-mm-diameter plastic sampling cassettes. Such cassettes are in common use for industrial hygiene sampling. The hole in the inlet section of the lower cassette was enlarged to fit the fritted cylinder. A second cassette with an enlarged outlet port was connected to it.

The major advantage of the single-flow bubbling generator is its simplicity and small size. However, it offers very limited opportunity for optimizing the ratio of Q_{dry} to Q_{bubl}. This is a disadvantage if compared with the versatility of the two-flow bubbling generator.

CONVENTIONAL (COLLISON) JET NEBULIZER

The Collison jet nebulizer has been used in many studies for dispersing microbial suspensions (e.g., Jensen et al., 1992; Stewart et al., 1995). It is also in prominent use for the dispersion of inert particles (May, 1973; Marple and Rubow, 1980; Gussman, 1984). The performance of the new aerosol generators was, therefore, compared to that of the Collison nebulizer.

The process of particle dispersion in the Collison jet nebulizer, schematically shown in Fig. 2, is similar to that of many other commercially available jet nebulizers (Sterk et al., 1984; Nerbrink et al., 1994): high-pressure air is pushed through a nozzle from which it exits as a high-velocity air jet. This air jet creates a low-pressure region which causes the liquid suspension to rise up a feed tube and be nebulized into droplets. The process of nebulization is complex: the air jet entrains the liquid as a cylindrical rod or sheet until it becomes unstable and disintegrates into droplets (Lefebre, 1989). The resulting droplet size distribution is, therefore, very wide. Most of these droplets are large, which causes their impaction onto the internal wall of the vessel from where they are drained back into the liquid reservoir. The smaller, unimpacted droplets follow the upward air flow and are emitted through the outlet port. At that point, drying air may be added if the goal is to primarily generate solid residues of the droplets.

In a jet nebulizer, the mean droplet size depends on the physical properties of the operating fluid, and is inversely proportional to the square root of the pressure

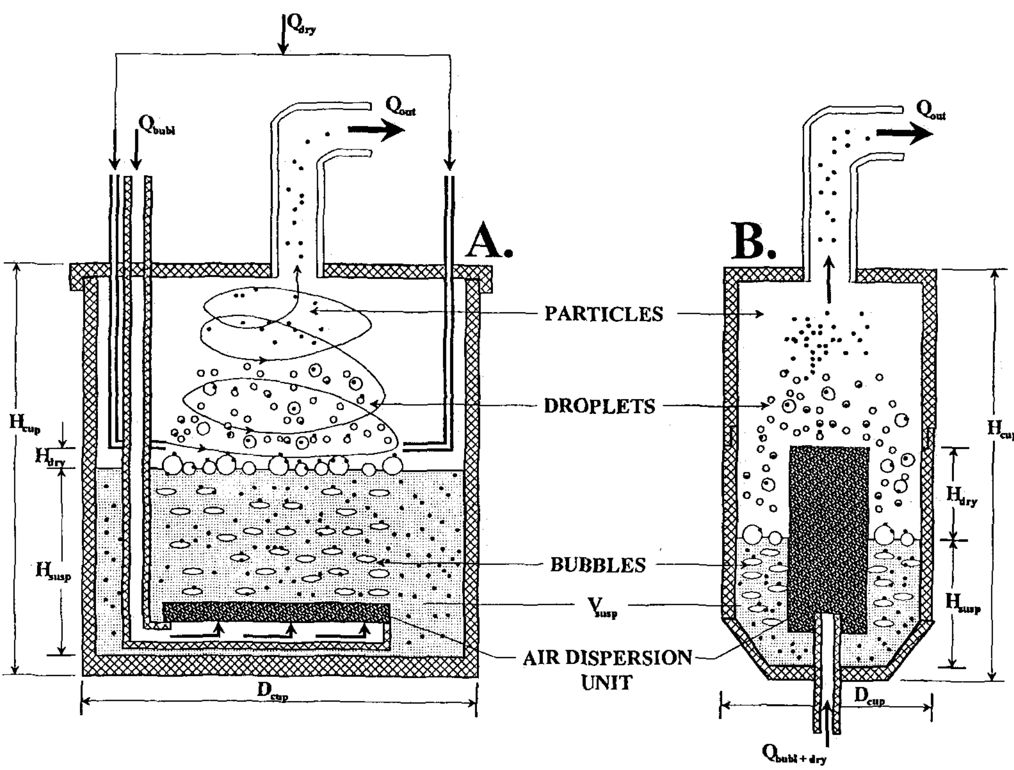

FIGURE 1. New bubbling aerosol generators. (a) Two-flow. (b) Single-flow. (Dimensions of the drawing are not to scale.)

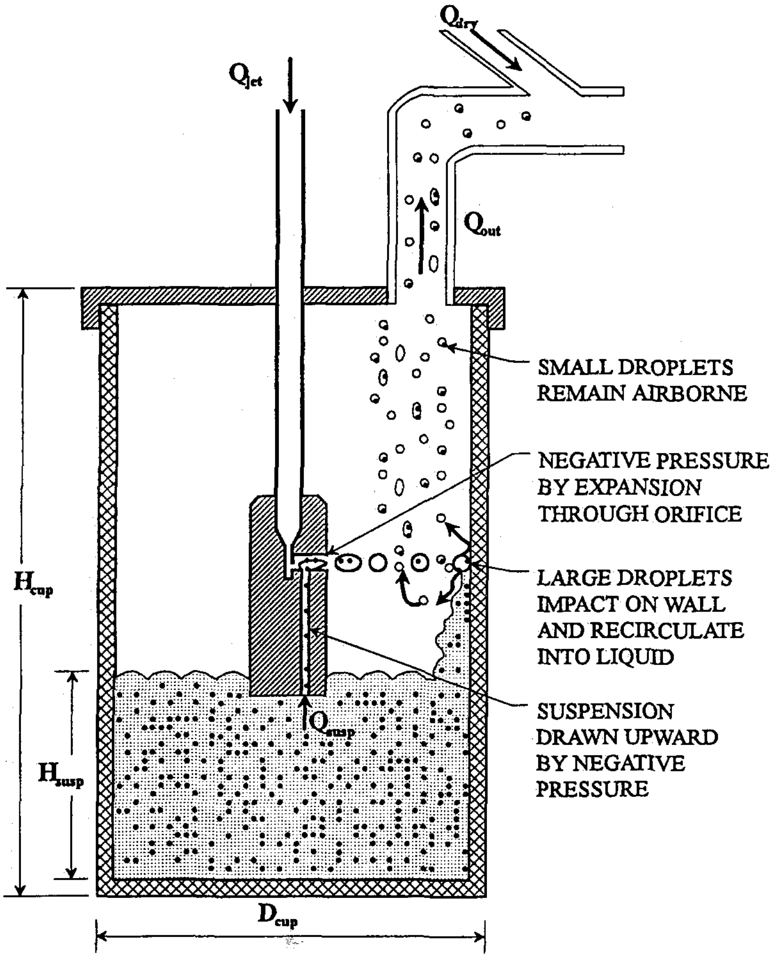

FIGURE 2. Schematic representation of the dispersion process in the Collison nebulizer. (Dimensions of the drawing are not to scale.)

drop across the nozzle (Dietrich, 1990). Variation of the pressure drop changes the ratio of the liquid flow rate from the suspension, Q_{susp}, to the jet flow rate, Q_{jet}.

EXPERIMENTAL PROCEDURE

Mass Balance

In the Collison nebulizer, the rate of aerosolization from the liquid suspension is very high, but only a small portion of the aerosolized liquid leaves the device as sufficiently small droplets. In contrast, the two-flow bubbling aerosol generator was designed to dry the droplets immediately upon generation and carry away the dry droplet residues. We have performed mass balance calculations that compare the rates of liquid suspension leaving each generator as vapor, liquid droplets, or solid particles. The concentration of water vapor, c_{wv} (in $\text{g} \cdot \text{cm}^{-3}$), that either enters the nebulizer, $c_{wv,\text{in}}$, or leaves the nebulizer, $c_{wv,\text{out}}$, is given by the Antoine equation (Reid et al., 1977):

$$c_{wv} = \left(\frac{\mathrm{RH}}{100}\right) \times 363.8 \exp\left(-\frac{4943}{273.15 + T_a}\right), \quad (1)$$

where RH is the relative humidity of the air (in percent) and T_a is the temperature of the air entering or leaving the aerosol generator (in °C). With this equation, we can calculate the rate of mass gain or loss of the nebulizer due to the water vapor in the air flow, m_{wv}:

$$\dot{m}_{wv} = c_{wv} Q, \quad (2)$$

where Q is the air flow rate into or out of the liquid suspension. The rate of liquid mass leaving the nebulizer suspension, $\dot{m}_{\mathrm{out}}$, can thus be calculated from the measured rate of change in nebulizer weight, $\Delta \dot{m}_{\mathrm{susp}}$, and the calculated rate of water vapor mass entering the nebulizer, $\dot{m}_{mw,\mathrm{in}}$:

$$\dot{m}_{\mathrm{out}} = \Delta \dot{m}_{\mathrm{susp}} + \dot{m}_{wv,\mathrm{in}}. \quad (3)$$

This output is made up of three components (Stapleton and Finlay, 1995):

$$\dot{m}_{\mathrm{out}} = \dot{m}_{p,\mathrm{out}} + \dot{m}_{wv,\mathrm{out}} + \dot{m}_{l,\mathrm{out}}, \quad (4)$$

where $\dot{m}_{p,\mathrm{out}}$ is the rate of solid particle mass leaving the aerosol generator, $\dot{m}_{wv,\mathrm{out}}$ is the mass of water vapor in the effluent flow, and $\dot{m}_{l,\mathrm{out}}$ is the rate of liquid mass leaving the generator as droplets. The value of $\dot{m}_{p,\mathrm{out}}$ was found by converting the number concentrations measured by the aerosol size spectrometer to overall particulate mass. The values of $\dot{m}_{wv,\mathrm{out}}$ were calculated by use of Eqs. (1) and (2). Equations (3) and (4) were used to calculate $\dot{m}_{l,\mathrm{out}}$.

The amount of water leaving the generator, $\dot{V}_{\mathrm{water,out}}$, depends on the water content of the incoming air (volume $= \dot{m}_{wv,\mathrm{in}}/\rho_{\mathrm{water}}$, where ρ_{water} is the water density). The major portion of the water output volume is the loss in suspension volume, $\Delta \dot{V}_{\mathrm{water}}$, due to water evaporation and aerosolization of the liquid as droplets. Thus, the total liquid output is

$$\dot{V}_{\mathrm{water,out}} = \Delta \dot{V}_{\mathrm{water}} + \frac{\dot{m}_{wv,\mathrm{in}}}{\rho_{\mathrm{water}}}. \quad (5)$$

Experimental Setup

The experimental setup is schematically shown in Fig. 3. Liquid suspensions of the test particles were dispersed in random order by one of the three aerosol generators: the Collison nebulizer, the two-flow bubbling generator, or the single-flow bubbling generator. The dispersions from the Collison nebulizer were diluted and dried with compressed laboratory air, Q_{dry}. In the two-flow bubbling generator, Q_{dry} was introduced directly into the nebulizer, at a short distance, H_{dry}, above the stationary liquid suspension level, H_{susp} (see Fig. 1). In the single-flow bubbling generator, most of the air flow through the fritted cylinder is intended to dry the effluent droplets in the space where they originate. Additional dry air can be added above the outlet port.

Before entering the sampling chamber (550 cm^3), the combined generation and drying air flow, Q_{total}, passes through a 10-mCi ^{85}Kr particle change neutralizer (model 3012; TSI Inc., St. Paul, MN); see Fig. 3. This reduces the level of electrostatic charges on the test particles, and thus their electrostatic attraction to the walls of the test system.

The size distribution measurements of the dry aerosol particles were performed with an Aerosizer, an aerodynamic size spectrometer that sizes particles aerodynamically down to about 0.5 μm (API Mach II, Amherst Process Instruments, Inc., Hadley, MA). Liquid sprays produced by conventional nebulizers are polydisperse and usually have a high number concentration. When performing droplet size distribution measurements by sampling the liquid droplet aerosol into a dynamic size spectrometer, the droplets may change their sizes within milliseconds due to liquid evaporation from their surfaces. Thus, the experimental data may depend upon the distance from the nebulizer to the mea-

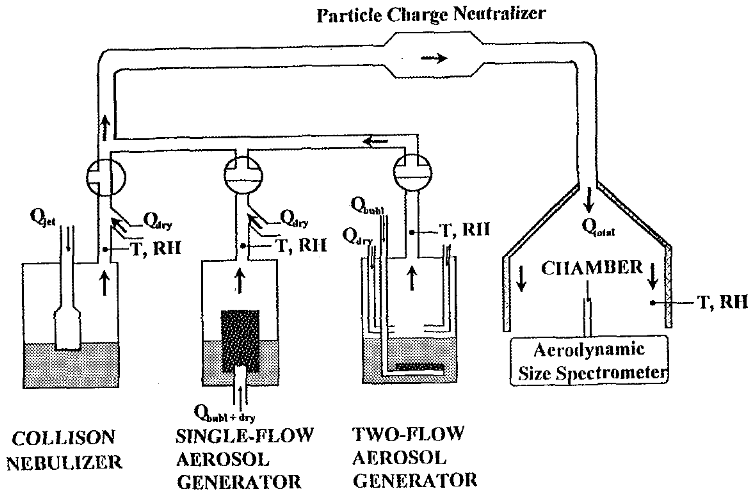

FIGURE 3. Experimental setup.

surement device and upon the specific measurement technique used (Gebhart, 1994). To avoid these uncertainties, we have exposed all liquid dispersions to drying air, and have compared the size distributions of the particles in their dry state.

The temperature and relative humidity were measured by certified traceable digital hygrometer/thermometers (Fisher Scientific, Pittsburgh, PA) on the outlet side of the aerosol generators and in the test chamber. The sensors were protected from droplet and particle impaction by small plastic shields. Since a large amount of liquid is evaporated during droplet generation in the Collison nebulizer, we assume that the relative humidity in the outlet of this nebulizer is over 99%, as estimated by Ferron and Gebhart (1988) for isotonic solutions. All three aerosol generators were weighed before and after operation (model P1210; Mettler Instrument Corp., Hightstown, NJ).

Suspension Preparation

The liquid dispersion experiments were performed with PSL particles suspended in deionized water at three concentration levels, c_{susp}, of 10^6, 10^7, and 10^8 particles $\cdot$ mL^{-1}. The physical particle diameters were $d_p = 0.73$, 1.6, 2.96, and 5.1 μm. The initial PSL particle concentration of the commercially available liquid suspension (Bangs Laboratories, Inc., Carmel, IN) is

$$c_{susp} = 1.828 \times \frac{10^{11}}{d_p^3}, \qquad (6)$$

where c_{susp} is in PSL particles $\cdot$ mL^{-1} and d_p is in μm. The initial PSL particle concentration was diluted with deionized water to the desired concentration by serial dilutions with 1 ± 0.02 mL and 10 ± 0.06 mL serological pipets (Fisher Scientific, Pittsburgh, PA). The exact particle concentration in each suspension was determined by placing the suspension into a brightline hemocytometer with a 0.1-mm-deep chamber (Hausser Scientific Partnership, Horsham, PA) and counting the particles under a bright-field, phase-contrast microscope (Labophot-2, Nikon Corp., Tokyo, Japan). In order to have a fast method for checking the particle concentration, the transmittance at 940 nm was measured with a spectrophotometer for each PSL particle concentration (Spectronic 21D, Milton Roy Co., Rochester, NY). The dependence of transmittance on PSL particle concentration was approximated by linear regression. Differences between measured and calculated concentration values were $\leq 20\%$. Whenever a particle concentration needed to be adjusted upward or downward, an aliquot of a higher concentration PSL suspension or of deionized water was added and the transmittance was remeasured.

RESULTS AND DISCUSSION

Table 1 presents major performance characteristics of the Collison nebulizer, and the single-flow and two-flow bubbling aerosol generators. All tests were performed with PSL particles of $d_p = 1.6$ μm at $c_{susp} = 10^8$ mL^{-1}. This table shows the input and output quantities of these devices as determined through our experiments and calculations using Eqs. (1)–(4). The data on the rate of decrease in mass of liquid suspension, $\Delta\dot{m}_{susp}$, represent the results of weighing the aerosol generators before and after one hour of operation. The values of the rate of overall particle mass output, $m_{p,out}$, are derived from the particle output concentration measurements with the Aerosizer. As seen, about half the output mass of the Collison nebulizer is water vapor, $\dot{m}_{wv,out} \approx 6.20$ g$\cdot$h^{-1}, and about half is water droplets, $\dot{m}_{l,out} \approx 5.51$ g$\cdot$h^{-1}. The overall mass of the PSL particles generated by the Collison nebulizer, $\dot{m}_{p,out} \approx 0.0065$ g$\cdot$h^{-1}, is about three orders of magnitude lower than the overall liquid mass generated as droplets. The output of the single-flow bubbling generator is similar: more than 60% of the output mass consists of liquid droplets. In contrast to these two generators, almost all of the output of the two-flow bubbling aerosol generator is water vapor for low or high Q_{dry} values. For an input relative humidity in the two-flow generator, RH$_{in}$, of $25.0 \pm 2.5\%$, and $Q_{bubl} = 1.5$ L$\cdot$min^{-1}, a low Q_{dry} of 3.0 L$\cdot$min^{-1} results in an output relative humidity, RH$_{out}$, of 97%. It is seen that even at such a high RH$_{out}$, there is minimal or no output in liquid droplets (shown as ~ 0 g$\cdot$h^{-1} in Table 1). An important advantage of the two-flow generator is, therefore, its generation of particles with no or minimum water content in them. At low Q_{dry}, however, we could not achieve a sufficiently high output concentration of these dry particles. The large droplets generated from the bursting bubbles were not dried sufficiently and, therefore, fell back into the suspension. To produce a high number concentration of aerosolized PSL particles, the two-flow generator was operated at a high dry air flow rate, $Q_{dry} = 15$ L$\cdot$min^{-1}. This resulted in a sufficiently high output of dry PSL particles with no liquid droplets present in the effluent.

Figure 4 shows the rate of total liquid output relative to the effluent air flow, $\dot{V}_{water,out}/Q_{out}$, for all three aerosol genera-

TABLE 1. Input and Output of the Collison Nebulizer and the Two New Bubbling Aerosol Generators[a]

Nebulizer	$c_{wv,in}$ (g$\cdot$cm^{-3})	$c_{wv,out}$ (g$\cdot$cm^{-3})	$m_{wv,in}$ (g$\cdot$h^{-1})	$m_{wv,out}$ (g$\cdot$h^{-1})	Δm_{susp} (g$\cdot$h^{-1})	m_{out} (g$\cdot$h^{-1})	$m_{p,out}$ (g$\cdot$h^{-1})	$m_{l,out}$ (g$\cdot$h^{-1})
Collison[b]	0.0000043	0.000017	1.57	6.20	10.14	11.71	0.0065	5.51
			±0.22	±0.11	±0.44	±0.52	±0.0023	±0.64
Single-flow[c]	0.0000042	0.000016	0.13	0.48	1.22	1.33	0.0009	0.85
			±0.02	±0.06	±0.28	±0.28	±0.0002	±0.29
Two-flow[d]	0.0000043	0.000017	1.15	4.32	3.40	4.55	0.00008	~0
(at low Q_{dry})			±0.19	±0.48	±0.41	±0.63	±0.00001	
Two-flow[e]	0.0000042	0.000013	4.22	12.75	8.66	12.88	0.0024	~0
(at high Q_{dry})			±0.67	±2.43	±0.19	±0.70	±0.0006	

[a] PSL particles, $d_a = 1.6$ μm; $c_{susp} = 10^8$ mL^{-1}.

[b] $Q_{jet} = Q_{out} = 6$ L$\cdot$min^{-1}, RH$_{in} = 25.1 \pm 2.6\%$, $T_{in} = 20.6 \pm 0.5$°C, RH$_{out} = 99.4\%$, $T_{out} = 18.7 \pm 1.5$°C.

[c] $Q_{bubl+dry} = Q_{out} = 0.5$ L$\cdot$min^{-1}, RH$_{in} = 25.2 \pm 2.3\%$, $T_{in} = 20.6 \pm 0.5$°C, RH$_{out} > 97\%$, $T_{out} = 20.4 \pm 0.8$°C.

[d] $Q_{bubl} = 1.5$ L$\cdot$min^{-1}, $Q_{dry} = 3.0$ L$\cdot$min^{-1}, $Q_{out} = 4.5$ L$\cdot$min^{-1}, RH$_{in} = 25.0 \pm 2.5\%$, $T_{in} = 20.6 \pm 0.5$°C, RH$_{out} > 97\%$, $T_{out} = 20.1 \pm 0.9$°C.

[e] $Q_{bubl} = 1.5$ L$\cdot$min^{-1}, $Q_{dry} = 15$ L$\cdot$min^{-1}, $Q_{out} = 16.5$ L$\cdot$min^{-1}, RH$_{in} = 24.9 \pm 2.6\%$, $T_{in} = 20.6 \pm 0.5$°C, RH$_{out} = 75.1 \pm 8.2\%$, $T_{out} = 18.4 \pm 1.7$°C.

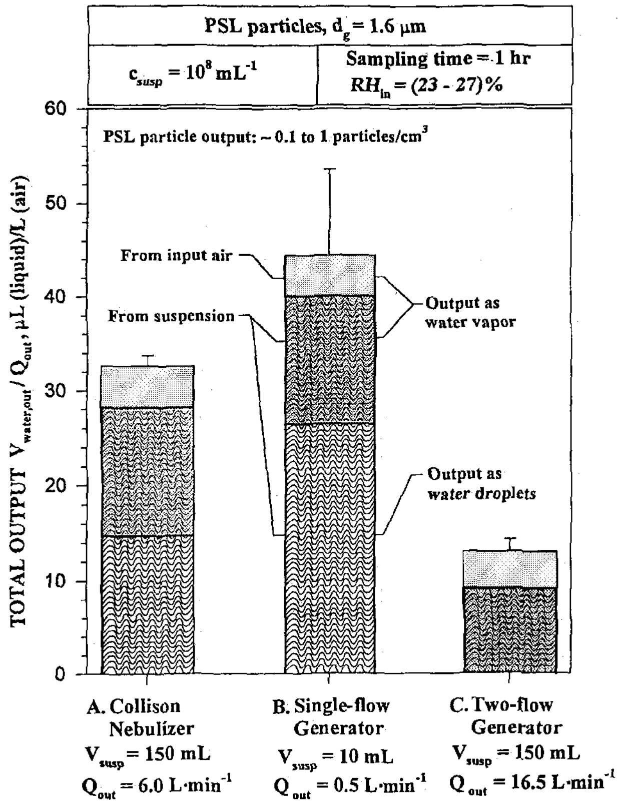

FIGURE 4. Rate of total water volume output in three generators. (a) Collison nebulizer: H_{cup} = 130 mm, H_{susp} = 80 mm, D_{cup} = 57 mm. (b) Single-flow bubbling aerosol generator: H_{cup} = 120 mm, H_{susp} = 23 mm, H_{dry} = 15 mm, D_{cup} = 22 mm. (c) Two-flow bubbling aerosol generator: H_{cup} = 110 mm, H_{susp} = 58 mm, H_{dry} = 2 mm, D_{cup} = 65 mm.

tors. This diagram results from our calculations using Table 1 (the data for the two-flow generator correspond to the "high" Q_{dry} of 15 L·min^{-1}). $\dot{V}_{water,out}$ [Eq. (5)] is the rate of total liquid volume leaving the generator vessel during its operation, and Q_{out} is the effluent air flow rate. The output flow rate of the two-flow bubbling generator was calculated as the sum of the bubbling flow rate through the fritted disk, Q_{bubl}, and the drying air flow rate, Q_{dry}, applied at distance H_{dry} = 2 mm above the liquid surface.

In Fig. 4, each generator's liquid output in water droplets is differentiated from its output as water vapor. Each output in water vapor consists of the water vapor in the incoming air and liquid evaporated from the suspension. As seen, the output in water vapor is about the same for each of the three generators. The main difference is in the output of water droplets from the suspension. While it is below the detectable limit (~ 0 μL·L^{-1}) for the two-flow generator, it is quite high for the other two generators. The PSL particle output was in the range of 0.1–1 particles/cm^3 for all three generators. At the conditions indicated in Fig. 4, the two-flow generator had the highest dry PSL particle generation relative to its effluent air flow rate. Its output in water droplets was negligibly small, and thus its total output consisted only of dry particles and water vapor (Fig. 4c). With an input of Q_{bubl} = 1.5 L·min^{-1} and Q_{dry} = 15 L·min^{-1} at RH$_{in}$ = 24.9 ± 2.6%, the output relative humidity was RH$_{out}$ = 75.1 ± 8.2%, which resulted in a sufficiently high production rate of dry PSL particles.

The total liquid output from the Collison nebulizer was measured to be 32.5 ± 1.6 μL of water per liter of air passed through the suspension. This is in the same range as measured by May (1973) and predicted by Mercer et al. (1968). The relative humidity on the input side was the same for all three generators, $RH_{in} = 25.1 \pm 2.6\%$. The air jet formed by the Collison nebulizer nozzle had a flow rate of $Q_{jet} = 6.0$ L·min^{-1}. On the output side, the air flow rate was the same ($Q_{out} = Q_{jet}$), but the relative humidity was found to be about 100%. Thus, the total output of the Collison nebulizer shown in Fig. 4a is due to the water vapor gain in the effluent air flow and the dispersion of liquid suspension as airborne droplets.

The single-flow bubbling aerosol generator yields the highest total output among the three tested devices, 44.3 ± 9.0 μL·L^{-1}. The output air flow rate is also equal to the input flow rate ($Q_{out} = Q_{bubl+dry}$). A fraction of the input flow is used to aerosolize the liquid, while the remainder is intended to dry the droplets. However, the fritted cylinder, immersed in the liquid suspension, also draws liquid upward into its pore structure from where it is evaporated into the effluent air. Thus, the droplets are not completely dried in the prototype tested, and the concentration of liquid droplets in the effluent is high because the same drying of the liquid droplets dispersed from the suspension reduces their size sufficiently to be carried away by the effluent air flow. Therefore, the total water volume output of the single-flow aerosol generator is relatively high.

In addition to its ability to generate dry test aerosols, the lower total liquid volume output in the two-flow bubbling aerosol generator is also an advantage over the Collison nebulizer. Given the same amount of liquid (150 mL) in these two devices, the new device can be used for a longer period of time. The Collison nebulizer is known to concentrate the particles in suspension with time due to significant recirculation of the aerosolized liquid (May, 1973). The new bubbling aerosol generator avoids this by immediately moving the film and jet droplets formed at the top of the liquid upward. As the two-flow bubbling aerosol generator has shown greater versatility, and is therefore expected to have greater utility, only this device is further evaluated in the remainder of this paper.

Figure 5 shows the measured rates of decrease in the initial liquid suspension volume $\Delta V_{susp}/Q_{out}$ ($\Delta V_{susp} \approx \Delta V_{water,out}$) as a function of the dry air flow rate, Q_{dry}, for the two-flow bubbling generator operated at $Q_{bubl} = 1.5$ L·min and $V_{susp} = 150$ mL and tested with PSL particles of $d_p = 1.6$ μm. As seen, the relative humidity in the effluent air flow depends on the flow rate of drying air injected just above the top surface of the liquid suspension. The RH_{out} values were determined experimentally. We have found that at $Q_{dry} \sim 0$–4 L·min^{-1}, the air in the generator outlet was essentially saturated and the suspension loss was maximal and approximately constant. At $Q_{dry} > 4$ L·min^{-1}, the measured suspension loss decreased as the relative humidity in the outlet port of the generator decreased with increasing levels of Q_{dry}. As it was difficult to accurately measure RH_{out} at $Q_{dry} \leq 4$ L·min^{-1}, we attempted to calculate this relative humidity from the adiabatic mixing of two air streams. Using the psychometric chart (*ASHRAE Handbook*, 1985), a flow of $Q_{bubl} = 1.5$ L·min^{-1} at RH ~ 100% mixed with a flow of $Q_{dry} = 3$ L·min^{-1} at $RH = 25.0 \pm 2.5\%$ results in an effluent RH of ~ 55% at $T = 20.1 \pm 0.9$°C. However, Fig. 3 shows that the measured RH for these conditions is close to 100%. We believe that droplet evaporation still occurs at these conditions. At higher levels of Q_{dry}, however, all droplets have either dropped back into the liquid suspension or have evaporated to their PSL or residue size. Thus, the relative humidity in the effluent air flow decreases for dry air flow rates above 4 L·min^{-1}. The drying of droplets occurs very fast; e.g., a droplet with a mean diameter of 2.0 μm (a typical droplet size, as shown by Wangwongwatana et al., 1990) will evaporate in about 0.1 s at RH = 55% and $T = 20$°C (Ferron and Soderholm, 1990).

The two-flow generator has been evaluated for the dispersion of PSL particle suspensions into air. The concentrations of the

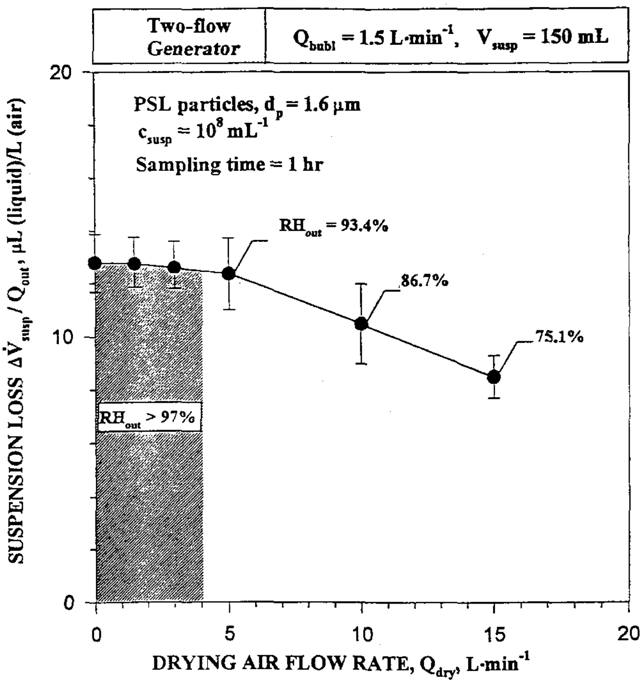

FIGURE 5. Liquid suspension loss at different drying air flow rates in the two-flow bubbling generator. $H_{dry} = 2$ mm.

suspensions being dispersed ranged from 10^6 to 10^8 particles per mL of water and the PSL particle size from $d_p = 0.73$ to 5.1 μm. The resulting aerosol concentration, N_{out}, was measured as a function of the aerodynamic diameter, d_a. Figure 6 displays the measured particle size distributions for two selected PSL particle sizes and three suspension concentrations at three drying air flow rates: $Q_{dry} = 0$, 10, and 15 L·min^{-1}. We found that the size distributions for each particle size were similar for each suspension concentration, c_{susp}, but differed in concentration by a factor 10 corresponding to the decade differences in particle concentration in the liquid. For 2.96 μm PSL particles, Fig. 6 shows a bimodal particle size distribution. The principal mode is at the PSL particle size, and the secondary mode peaks are between $d_a = 0.5$ and 1 μm. It appears that the secondary peaks were caused by residue particles from the surfactant. Particles of 2.96-μm diameter have a much larger surface than 0.73-μm diameter particles; therefore, more surfactant was present in the 2.96-μm suspension. Also, the 2.96-μm PSL particle suspension used was a few years older than the 0.73-μm PSL particle suspension. Thus, the residues from the 2.96-μm PSL particle suspension were within the measurement range of the Aerosizer, while those of the 0.73-μm PSL suspension were below the Aerosizer's lower limit of detection.

Figure 6 also shows that the monodispersity of the PSL particle peak increases with increasing drying air, i.e., with decreasing relative humidity. When a bubble breaks, some droplets contain PSL particles. At high drying air flow rates, the droplets are dried quickly by the inward and upward vortex flow of dry air, which results in removing all liquid from the droplets containing PSL particles. When drying is not sufficient, some of the water surrounding the PSL particles does not evaporate, and a less monodisperse size distribution of droplets is recorded. When Q_{dry} is low, most droplets fall back into the suspension because they are not immediately reduced

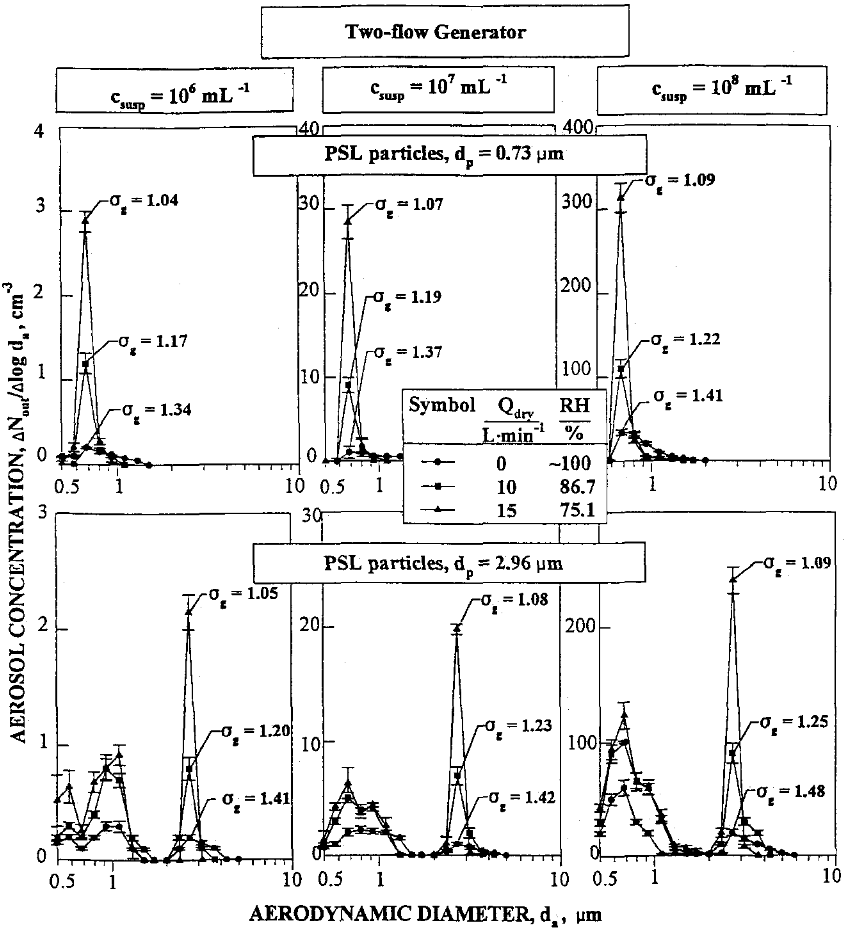

FIGURE 6. Effect of drying air flow rate on PSL particles size and concentration. $H_{dry} = 2$ mm, $V_{susp} = 150$ mL, $Q_{out} = 16.5$ L·min^{-1}, $Q_{bubl} = 1.5$ L·min^{-1}.

in size by drying air. Thus, the geometric standard deviation, σ_g, is broad and the PSL concentration is low for low Q_{dry}. With increasing dry air flow rate, the PSL particle peak becomes higher and more monodisperse.

For the measurements displayed in Fig. 6, we chose PSL particle concentrations in the liquid that avoid the formation of doublets and higher multiplets. Even at the highest PSL concentration of 10^8 mL^{-1}, the data (right-hand figures) for the high drying flow rate show essentially no doublet formation.

Figure 7 shows the peak PSL particle aerosol concentration as a function of the aerodynamic particle size for three suspension concentrations when the two-flow generator is operated at $Q_{bubl} = 1.5$ L·min^{-1}, $Q_{dry} = 15$ L·min^{-1}, and $V_{susp} = 150$ mL. It is seen that each ten-fold increase in PSL particle concentration in the liquid suspension results in a ten-fold increase in the airborne concentration. For each level of c_{susp}, the aerosol concentration of PSL particles is approximately the same for particles ranging in physical size from 0.73 to 1.60 μm ($d_p = 0.73$ μm converts to $d_a =$

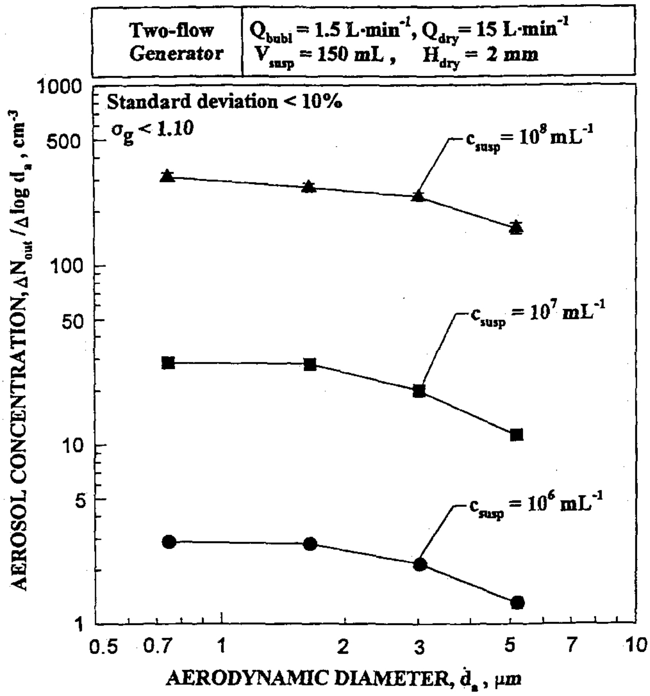

FIGURE 7. Effect of PSL particle size and concentration in the liquid suspension on their aerosolization.

0.75 μm for PSL particles with a density of 1.05 $g \cdot cm^{-3}$). The aerosol concentration decreases to about half of that value when the PSL particle size is further increased to 5.1 μm. The higher inertia particles of 5.1 μm have many more opportunities for getting lost to the inner surfaces of the ducting and valving of the measurement system. Therefore, we attribute this decrease primarily to the particle wall losses in the measurement system rather than to losses inside the generator itself. The latter is expected to be minimal due to the effective inward swirl of the air motion above the liquid surface of the two-flow generator. The standard deviation of all aerosol concentration measurements for each point in Fig. 7 was less than 10%. Aerosol concentration versus time measurements have shown that the aerosol concentration varied by less than 10% during the test period of 30 min. If this generator is to be used over much longer periods of time, without replenishing the PSL content in the liquid suspension, the concentration of PSL particles in the effluent air stream will decrease. The decrease with time will depend on the volume of liquid in the two-flow generator and on the air flow rate through the liquid.

The measured geometric standard deviation of the PSL particle peak shown in Fig. 6 never exceeded 1.10, which indicates the ability of the two-flow generator to emit monodisperse PSL particles at $Q_{dry} = 15$ L $\cdot$ min^{-1} and $H_{dry} = 2$ mm. The aerosol size spectrometer indicates a higher σ_g than that of the PSL particles themselves because of the instrument's limit of σ_g resolution. Figure 8 shows the effect of drying height H_{dry} on the resulting aerosol concentration. As seen, the aerosol concentration decreases by a factor of about 2.5 when the injection point of dry air is increased from 2 to 15 mm above the top of the liquid suspension. This increase in H_{dry} results in a reduction of the amount of drying air reaching the droplets formed at the bubbling surface. Thus, a smaller number of droplets is shrunk to the size which allows their upward and outward motion

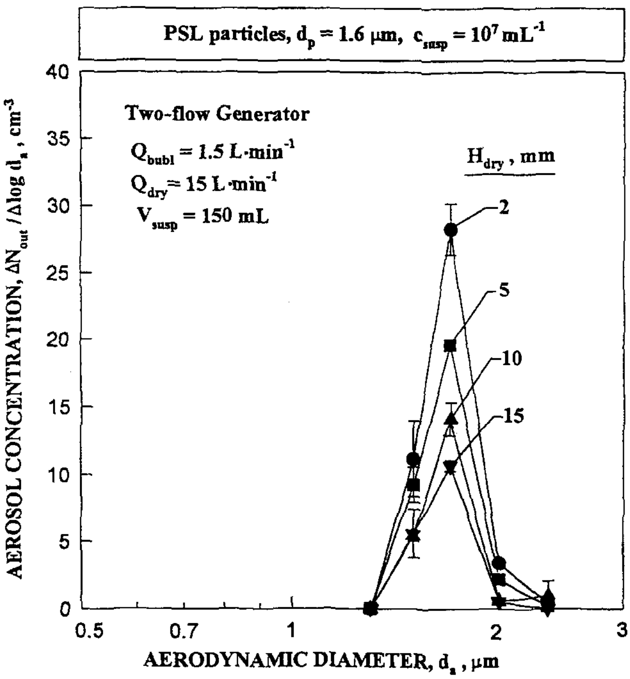

FIGURE 8. Effect of drying distance H_{dry} on aerosol generation in the two-flow bubbling aerosol generator. H_{dry} is defined in Fig. 1(a).

with the swirling air. Distance H_{dry} increases naturally during the operation of the generator when the liquid suspension height, H_{susp}, decreases (Fig. 1a). As the suspension losses in the two-flow aerosol generator are low, H_{susp} decreases with time very slowly, which explains the high stability of the aerosol concentration (standard deviation $< 10\%$) for at least 30 min.

PSL particle suspensions are typically stabilized against coagulation by the addition of an anionic surfactant. When a liquid suspension of PSL particles with the surfactant is aerosolized by the two-flow bubbling aerosol generator, the drying air shrinks the droplets that do not contain any PSL particle to the size that corresponds to the mass of the residual surfactant. This is seen in Fig. 9 which presents the size distribution of 5.1-μm PSL particles generated from a liquid suspension of PSL particles containing the surfactant versus one with the surfactant removed. Removal of the water-soluble surfactant was achieved by centrifuging the PSL suspension for 2 min, long enough for the particles to settle to the bottom of the vial. The surfactant liquid was then removed and the PSL particles were resuspended in deionized water. As seen in Fig. 9, surfactant residues (circular data points) are present in the range of 1–4 μm when the original PSL suspension is aerosolized. Once the surfactant is removed before aerosolization, the PSL particle peak remains, but the lower size peak disappears (square data points).

Also, the bubbling process in the new device is expected to be more gentle for particles in the suspension than the process of suspension recirculation at high pressure in the conventional jet nebulizers. This advantage is important for the generation of viable bioaerosol particles such as sensitive bacteria.

CONCLUSIONS

Particulate materials suspended or dissolved in a liquid are conventionally aerosolized by pneumatic nebulization, e.g.,

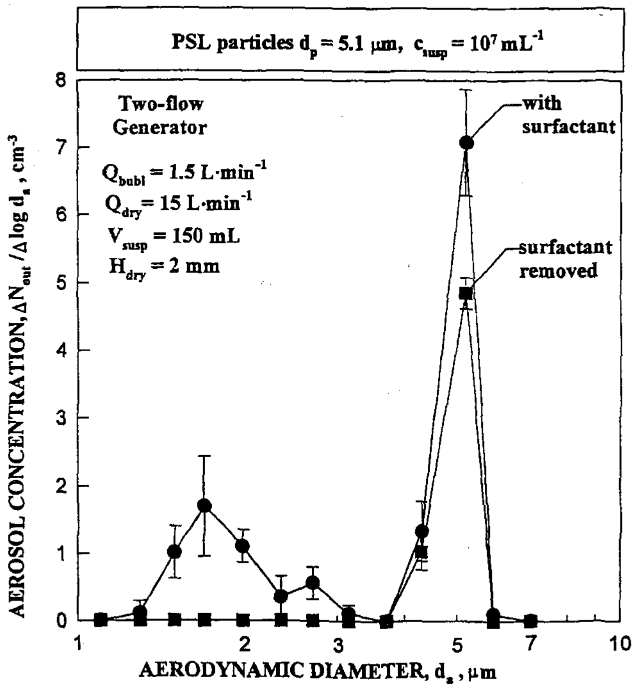

FIGURE 9. Aerosolization of PSL particles before and after surfactant removal from the PSL particles.

with a Collison nebulizer. This study has shown that the test aerosol particles can also be aerosolized by bubbling air through a liquid suspension of the particles, and then drying and swirling away the droplets produced by the bursting bubbles. Two new generator designs were tested: a single-flow and a two-flow version. The new two-flow bubbling aerosol generator was found to give the better performance. It was found that, in contrast to pneumatic nebulization by, e.g., a Collison nebulizer, the output in liquid droplets was negligibly small in the new two-flow generator, while the output in dry PSL particles was high. An advantage is seen in the new technique in that there is less recirculation of liquid. The aerosol concentration was constant for at least 30 min with the prototype generator tested. The relative humidity in the outlet port of this generator can be varied by adjusting the flow rate of the drying air. If used for the aerosolization of bacteria or other biological materials, the organisms are expected to be subjected to much less shear stress during aerosolization. This will be shown in a forthcoming study.

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