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ABSTRACT. The collection efficiency of liquid impingers was studied experimentally as a function of the sampling flow rate with test particles in the bacterial size range. Three impingers were tested: two All-Glass Impingers (AGI-4 and AGI-30), widely used for bioaerosol sampling, and a newly developed slot impinger. The aerosol particles were generated by a Collison nebulizer, and an Aerosizer was used to measure the particle concentrations and size distributions upstream and downstream of each impinger. The effect of the air pressure drop across the impinger on the Aerosizer performance was investigated, and the particle measurement system was modified and calibrated accordingly. While inertial impaction is the dominant particle removal mechanism in impingers, particle bounce and reaerosolization were also found to have significant effects on the impinger collection characteristics. At relatively high flow rates and low levels of collection fluid (corresponding to the collection fluid level after evaporation of most of the liquid during prolonged impingement), the liquid under the impinger jet was observed to be removed by the air pressure and pushed against the container's walls. Particles, such as bacterial or fungal spores, may thus bounce from the bottom of the collection vessel and escape with the effluent air flow or may impact sideways into the liquid that was previously pushed against the walls. It was found that such particle bounce may significantly reduce the collection efficiency of impingers containing a small amount of liquid. When the impingers were operated at a high level of collection fluid and sufficiently high sampling flow rates, it was observed that the bubbles, rising through the liquid, entrained previously collected particles and created new aerosols by bursting at the liquid–air surface. Such particle reaerosolization was also found to reduce the impinger collection efficiency. AEROSOL SCIENCE AND TECHNOLOGY 26:326–342 (1997)

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INTRODUCTION

Several different physical methods, e.g., impaction, impingement, filtration, and electrostatic precipitation, are in common use to actively sample particles from an air environment. In an impinger, the airborne particles are sampled into a circular or rectangular nozzle in which the particles are accelerated to a high velocity into a liquid (in some devices to approximately sonic speed). As the particles leave the nozzle in a high-velocity jet, they are expected to either impact directly into the liquid or to be taken up by the liquid after impaction onto the bottom surface. The air flow creates a high degree of turbulence and bubble formation as it passes through the liquid.

The first liquid impinger was developed by Greenberg and Smith (1922) and was used as a dust cloud sampler. In the late 1930s, liquid impingement was adapted for microbial aerosol sampling (Miles and Mistra, 1938). Further progress determining the performance of available and newly-developed impingers, made in the 1940s and 1950s, resulted in several design modifications aimed at improving the physical and biological efficiencies of impingers (Rosebury, 1947; Henderson, 1952; Cown et al., 1957; May and Harper, 1957; Shipe et al., 1959; Tyler and Shipe, 1959; Tyler et al., 1959; Wolf et al., 1964). Design modifications of the "Porton" impinger (Tyler and Shipe, 1959) led to the development of its successor, the All-Glass Impinger. The contemporary versions, the AGI-4 and the AGI-30 (Ace Glass Inc., Vineland, NJ), are still widely used for human exposure assessments of air environments contaminated with particulates.

Although liquid impingement is widely used, the performance characteristics of impingers have received considerably less attention in the literature than that of impactors. Interest in evaluating impingement as a particle collection method has been rekindled by today's increasing need to sample biological aerosols. At this time, a liquid collection medium is considered an attractive alternative to a filter or an agar plate, particularly when collecting viable airborne microorganisms or complex toxic materials. Impingers are particularly useful when the dehydration of viable cells should be avoided. Another advantage of liquid collection over collection on an inert or nutrient-containing surface is that liquid suspensions containing high concentrations of airborne microorganisms can be diluted to the level required for the subsequent analysis. The liquid suspension with collected biological materials can also be split into several aliquots to perform separate analyses, e.g., for total and viable microbial counts and chemical or immunological assays.

The performance characteristics of bioaerosol impingers have been evaluated and compared with those of other bioaerosol samplers in several field and laboratory studies published during the past decade: Macher and First (1984), Henningson and Fängmark (1987), Zimmerman et al. (1987), Henningson et al. (1988), Hering (1989), Kang and Frank (1989a,b), Buttner and Stetzenbach (1991), Jensen et al. (1992), Thorne et al. (1992), Nevalainen et al. (1993), Griffiths and DeCosemo (1994); Grinshpun et al. (1994), Henningson and Ahlberg (1994), Juozaitis et al. (1994). Since the early 1960s, some commercially available impingers have been proposed as reference bioaerosol samplers, e.g., the AGI-30 was recommended by Brachman et al. (1964) as a reference sampler. The AGI-30 and other impingers are, however, not yet sufficiently characterized for standardization (Henningson and Ahlberg, 1994).

Analogous to solid-surface impactors, inertia is usually considered the driving mechanism for particle collection in liquid impingers. However, the microbial counts obtained by impingers frequently differ from those obtained with impactors. If the sampled aerosols contain aggregates of bacteria or fungi, impingement may overestimate the actual concentration of airborne biological particles as the microbial clusters may break up during the impingement process. On the other hand, collection of particles by liquid impingement may lead to the

underestimation of the aerosol concentration when particles with sufficient inertia are not efficiently trapped and held by the collection fluid. Muilenberg (1989) attributed this phenomenon to particle hydrophobicity, i.e., hydrophobic particles may not be sufficiently wetted to be retained by the liquid. When operated for half an hour or more, an impinger may lose a significant part of its collection fluid, resulting in a change of the conditions for particle collection. This leads to time dependency of the impinger collection characteristics.

The overall efficiency of an aerosol sampler is defined as the ratio of the particle concentration measured by the sampler to the actual particle concentration in the air. The sampling efficacy of a bioaerosol sampler has physical and microbiological aspects. The physical aspects include the inlet (E_{INLET}) and collection (E_{COLL}) efficiencies (Grinshpun et al., 1994) which define the overall physical sampling efficiency E:

$$E = E_{\text{INLET}} \times E_{\text{COLL}} = (c_{\text{IN}} - c_{\text{OUT}})/c_{\text{IN}} \quad (1)$$

where c_{IN} is the actual concentration of particles of a given size in the vicinity of the sampler (upstream), and c_{OUT} is the concentration of particles penetrating through the sampler without being captured (downstream).

The inlet efficiency is an indicator of the changes in particle concentration that occur when the aerosol moves from the ambient air through the inlet to the collection unit of the sampling device. These changes are caused by inertial motion and gravitational sedimentation during particle aspiration from the air to the sampling orifice, as well as by various particle wall deposition mechanisms in the inlet section of the sampler (Vincent, 1989; Grinshpun et al., 1990; Brockmann, 1993). Thus, the inlet efficiency of an aerosol sampler is the product of its aspiration and transmission efficiencies. Both efficiencies may differ significantly from 100%, especially when sampling large particles nonisokinetically. However, when bioaerosol samplers, such as the All-Glass Impingers, are used to sample bacteria of a typical size range of 0.5–3 μm in diameter, the inertial and gravitational forces are insignificant, i.e., both efficiencies are close to 100%, and thus $E_{\text{INLET}} \approx 100\%$ (Grinshpun et al., 1994). For most impinger sampling of bacteria, Eq. (1) therefore reduces to $E = E_{\text{COLL}}$.

The microbiological aspects of bioaerosol sampling are characterized by the sampler's bioefficiency which deals with the survival and recovery of collected microorganisms. The present study is focused on the process of aerosol impingement, not on what happens after collection. This study, therefore, deals only with the physical sampling efficiency. All experiments were performed under conditions at which the physical efficiency is fully characterized by its collection efficiency component.

Because impactors and impingers utilize inertia as the primary mechanism for collecting airborne particles, their collection efficiencies are expected to be monotonically increasing functions of the particle inertia. Therefore, an increase in flow rate through the sampler should result in a collection efficiency that asymptotically approaches 100% for a given particle size. However, significant deviations from this expectation have been observed in impactors, and have been attributed primarily to particle bounce from the collection surface (Marple and Willeke, 1979; Xu and Willeke, 1993a,b; Xu et al., 1993). As a result, application of a sticky coating to the impaction plates is recommended for many situations (John, 1995). Performance evaluations of impactors, primarily the collection efficiency curves, have been performed either experimentally or theoretically. In the latter case, the investigators have used classical particle impaction models (Marple, 1970; Marple and Willeke, 1976; Marple et al., 1993) or more complex approaches that include particle resuspension, deagglomeration, and other effects (John, 1995).

For impingers, determinations of the collection efficiency have been conducted primarily by experimentation because the

aerosol impacts into a nonstatic liquid, and theoretical predictions are therefore more difficult to perform. Any application of impaction models to a liquid impinger also assumes that the collection efficiency monotonically increases with the sampling flow rate, reaching 100% at sufficiently high flow rates. However, a nonmonotonic dependence of collection efficiency on the sampling flow rate has been observed in our recent experiments with our newly developed slot impinger (Juozaitis et al., 1994; Willeke et al., 1995). This motivated further laboratory studies of aerosol collection in liquid impingers. The principal hypothesis of the present study is that the collection efficiency of aerosol impingers depends on the sampling flow rate in a different and more complex way than that of aerosol impactors as impingement into a liquid is characterized not only by the mechanism of particle inertia.

EXPERIMENTAL METHOD

Test System

The test system for the experiments on aerosol impingement is schematically shown in Fig. 1. This system is a part of the Bioaerosol Sampler Evaluation Facility which has been developed recently and is described in previous papers (Juozaitis et al., 1994; Thompson et al., 1994; Stewart et al., 1995). In the present study, monodisperse test particles were aerosolized from a liquid suspension by use of a three-jet Collison nebulizer (BGI Inc., Waltham, MA) which was operated at a flow rate of $Q_{NEB} = 6$ L/min. The aerosol was then dried and diluted with prefiltered, compressed laboratory air ($Q_{DRY\text{-}NEB} = 40$ L/min) to shrink the droplets to their particulate content before reaching the test chamber through an 80 cm long copper pipe. The particles were charge-neutralized in a 10 mCi ^{85}Kr electrostatic charge neutralizer (Model 3012, TSI Inc., St. Paul, MN) which was built into the copper pipe to prevent electrostatic deposition of particles on the inner surfaces of the test system.

The test impinger sampled aerosol particles from the center of the test chamber at fixed flow rates, Q_{IMPG}, ranging from 2 to about 13 L/min. An aerodynamic particle size spectrometer (Aerosizer, Model API

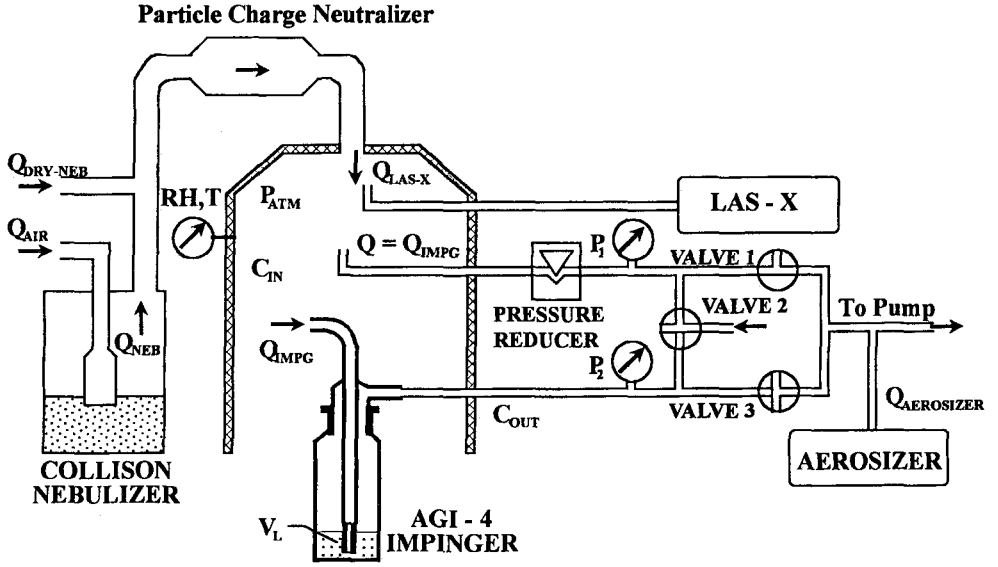

FIGURE 1. Schematic representation of the test system.

Mach II, Amherst Process Instruments, Inc., Hadley, MA) operated at a nominal flow rate of 5.3 L/min was used to measure the aerosol concentrations upstream of the impinger c_{IN} (in the test chamber) and downstream of the impinger c_{OUT}. The particle number concentrations for each of the Aerosizer channels were divided by the logarithmic intervals of the corresponding particle size ranges $\Delta c_{IN}/\Delta \log d_a$ and $\Delta c_{OUT}/\Delta \log d_a$, respectively) and were recorded as a function of the aerodynamic particle diameter d_a. A special multiple valve system (Fig. 1) was designed for these measurements, and the Aerosizer was calibrated accordingly (see next section). To avoid the effect of liquid saturation on the $\Delta c_{OUT}/\Delta \log d_a$ measurement data, dry-air dilution was added downstream of the impinger through valve 2. In addition, a laser aerosol size spectrometer (Model LAS-X, PMS Inc., Boulder, CO), measuring an optical particle diameter range of 0.09–3.0 μm, was used in parallel with the Aerosizer to continuously monitor the aerosol concentration entering the test chamber.

The collection efficiencies of the test impinger were determined with monodisperse particles of sizes that are typical for bacteria. The inlet efficiency for these particle sizes was assumed to be about 100%. Thus, the collection efficiencies were determined from the c_{IN} and c_{OUT} data by Eq. (1). All measurements were repeated at least three times. The air temperature was kept at 20–23°C and the relative humidity (RH) in the chamber at 30–55% during all experiments. The entire setup was placed in a Class II, Type B2, biological safety cabinet (SterilchemGARD, Baker Company, Sanford, ME).

During each test sequence, the particle size distributions were measured by the Aerosizer during 1 min time intervals (up to 5 min total per replication). The volume of collection fluid V_L essentially remained unchanged during this period: the test time was short enough to avoid the effect of liquid evaporation on E_{COLL}, while the testing time was sufficiently long for taking representative samples with the Aerosizer and LAS-X size spectrometers.

Size and Concentration Calibrations of the Aerosizer for Measurements at Reduced Air Pressures

Recent laboratory evalutions of the Mach II model of the Aerosizer and its comparison with other aerosol measurement devices (Grinshpun et al., 1995; Qian et al., 1995) has shown that this instrument is very suitable for the testing of aerosol samplers collecting airborne particles of $d_a \geq 0.5$ μm at normal atmospheric pressure, $P = 1$ atm. The Aerosizer has been used previously to determine the collection efficiencies of bioaerosol impactors (Stewart et al., 1995) by measuring the aerosol concentrations upstream and downstream of the impactors, when both air pressures were approximately 1 atm. However, when sampling into All-Glass Impingers at Q_{IMPG} = 10–12.5 L/min, the air pressure downstream of the impingers, labeled P_2 in Fig 1, may be as low as 0.5 atm. It was found that sampling at reduced pressures affects the performance characteristics of the Aerosizer (and, perhaps, other particle size spectrometers). Therefore, when using the Aerosizer for aerosol measurements upstream and downstream of an impinger, data corrections must be made for the air pressure drop measured across the impinger. Our Bioaerosol Sampler Evaluation Facility was modified so that the pressure drop was the same for upstream and downstream measurements. To achieve this, a pressure reducer was built into the upstream sampling line to match the pressure drop across the impinger, i.e., to set $P_1 = P_2$ at equal flow rates through the impinger and the upstream sampling line.

The changes in recorded particle size and concentration for reduced air pressures at the inlet of the Aerosizer were determined with PSL particles of four typical microbial sizes, d_a = 0.72, 1.09, 3.03, and 5.2 μm. The geometric standard deviation σ_g did not exceed 1.2 for any of the test

aerosols. The aerosol concentrations, measured in the chamber, ranged from ca. 10^{-2} to 10^{2} cm^{-3}. The air pressure drop (ΔP = 1 atm $-P_1$) was varied from 0 to 0.5 atm, simulating the pressure drop across the All-Glass Impinger sampling at 2–13 L/min. The aerosol concentrations at reduced pressure $c_{p<1\,\mathrm{atm}}$ were determined for $P_1 = 0.5$, 0.7, and 0.8 atm and compared with the concentrations measured at $P = 1$ atm, $c_{P=1\,\mathrm{atm}}$. The aerodynamic particle sizes at these same air pressures $(d_a)_{P<1\,\mathrm{atm}}$ were recorded and compared to the calibrated sizes at 1 atm, $(d_a)_{P=1\,\mathrm{atm}}$. Thus, a particle size factor $(d_a)_{P<1\,\mathrm{atm}}/(d_a)_{P=1\,\mathrm{atm}}$ was obtained for each ΔP.

Figure 2 compares the aerosol concentration data at reduced pressure to those measured at atmospheric pressure for PSL particles of $d_a = 1.09$ μm. Linear regression with the correlation coefficient of $r^2 = 0.999$ relates the two concentrations by a constant shift:

$$c_{P=1\,\mathrm{atm}} = b[0] + b[1]c_{P\le 1\,\mathrm{atm}} \qquad (2)$$

where $b[0] = 0$, 0.44, 0.58, and 0.73 atm for $P_1 = 1.0$, 0.8, 0.7, and 0.5, respectively, and $b[1] \approx 1$ for the entire range of the tested air pressures. The data demonstrate that reduction in the air pressure significantly affects the accuracy of the particle concentration measured by the Aerosizer, even at a relatively small ΔP, e.g., the Aerosizer underestimates the concentration by 44% at $\Delta P = 0.2$ atm ($P_1 = 0.8$ atm). This underestimation is caused by the decrease in the air flow rate entering the Aerosizer, which has been observed at reduced pressures. The air flow rate entering the Aerosizer was found to decrease linearly with pressure from 5.3 L/min at 1 atm to 3.9 L/min at 0.8 atm and then to 2.3 L/min at 0.5

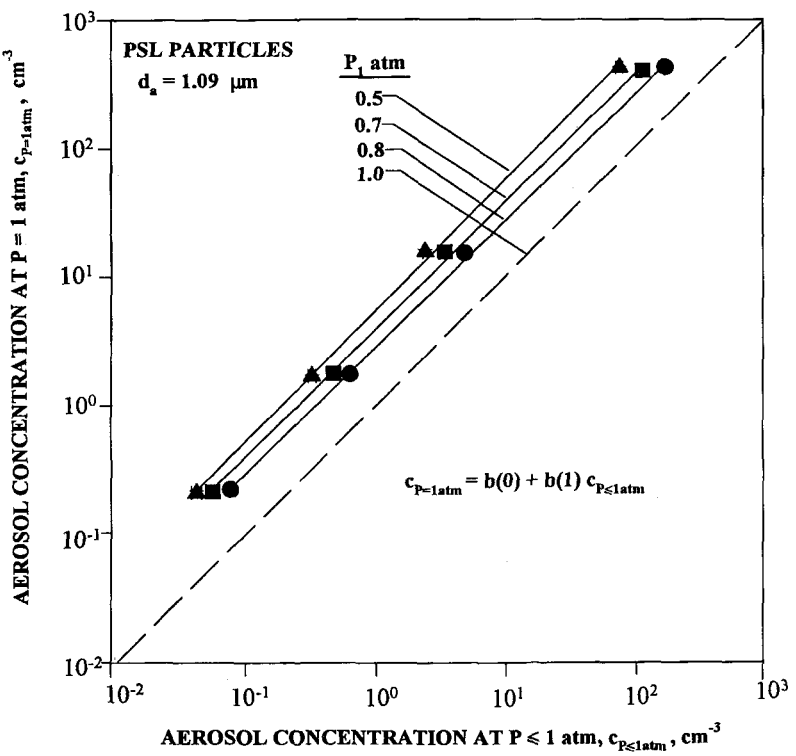

FIGURE 2. Effect of air pressure at the inlet of the Aerosizer on its particle concentration reading.

atm. Thus, the indicated aerosol concentration at $P < 1$ atm is lower than at $P = 1$ atm because the instrument bases its concentration data on its nominal flow rate at normal atmospheric pressure. We believe that the reduced pressure at the impinger outlet and the resulting measurement difficulties are major reasons for the lack of reliable impinger performance data. The measurement system, Fig. 1, is presented in detail as this experimental design can be used in the future for impinger performance evaluations.

Figure 3 shows the particle size factor as a function of the particle aerodynamic diameter at three different pressure drops. It is seen that the particle size is overestimated at reduced air pressure in the Aerosizer inlet. This overestimation increases with ΔP, as also shown by Cheng et al. (1993). We found that this overestimation is a nonmonotonic function of the actual particle size. This reflects the overlapping influence of two particle motion regimes which take place in the tested particle size range: the "slip flow" regime, predominant for submicrometer particles, and the non-Stokesian regime, predominant for supermicrometer particles (Baron and Willeke, 1993). The reduced pressure effects on the particle drag coefficient, caused by these regimes, are of opposite character. The drag force on submicrometer particles decreases at reduced air pressures primarily because of the increase in the mean free path of the gas molecules which, in turn, results in an increase of the slip correction factor. For supermicrometer particles, the influence of the slip correction on particle motion is not so significant, but the influence of non-Stokesian drag becomes increasingly important. As seen in Fig. 3, the opposing effects

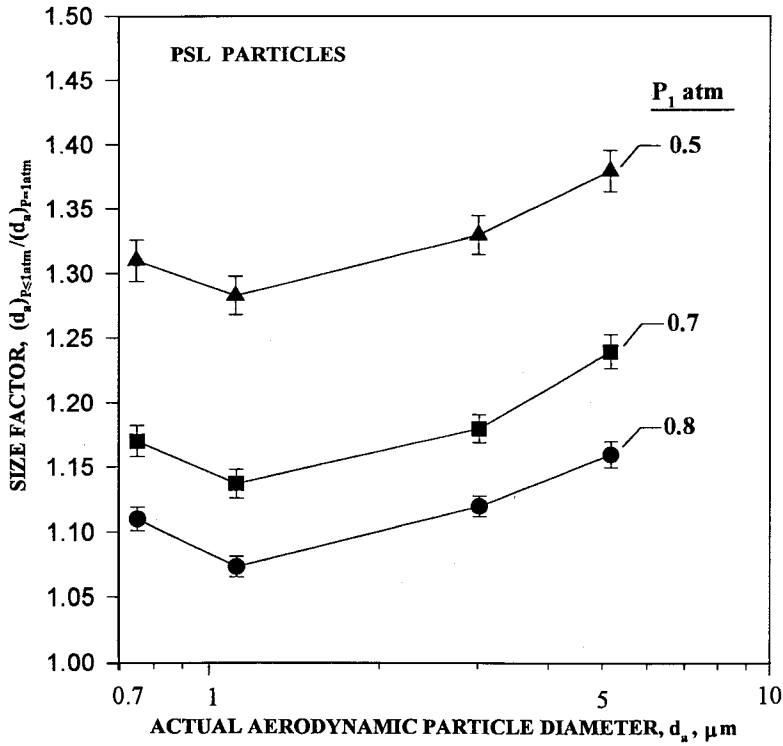

FIGURE 3. Effect of reduced air pressure on the particle size indicated by the Aerosizer.

result in a minimum of the indicated particle size increase at the intermediate size of $d_a \approx 1$ μm. Comparison of the data presented in Figs. 2 and 3 shows that reductions in the air pressure at the Aerosizer inlet result in a less pronounced increase in the particle size than the corresponding decrease in aerosol concentration.

Test Impingers

The impingers used in this study are schematically shown in Fig. 4: two commonly used All-Glass Impingers, the AGI-30 (Fig. 4A) and the AGI-4 (Fig. 4B), and the slot impinger (Fig. 4C). All three impingers have a low cutoff size of about 0.3 μm at $Q_{IMPG} = 12.5$ L/min for the AGIs (Nevalainen et al., 1993) and 10 L/min for the slot impinger (Juozaitis et al., 1994), which makes them suitable for collecting airborne bacteria.

In the AGI-30 and AGI-4, suction was applied to the small exit port at the top of the glass vessel (not shown in Fig 4). Thus, air was drawn through the curved intake tube and down through the impingement nozzle, a capillary of ≈ 20 mm length and ≈ 1.1 mm diameter, which acted as a critical flow orifice. At $Q_{IMPG} = 12.5$ L/min (the recommended flow rate for AGIs), the air pressure drop across the impinger is about 0.5 atm. At this ΔP, the flow in the jet exiting from the nozzle is at sonic velocity, i.e., any increase in suction will not increase the jet velocity further. The distance H_J between the jet orifice and the bottom of the impinger vessel (jet-to-plate distance) was 30 mm for the AGI-30 and 4 mm for the AGI-4. Both types of impinger are usually filled with 20 mL of collection fluid prior to their operation, which results in an initial liquid level of $H_L = 20$ mm (Wolf et al., 1964). Thus, the nozzle of the AGI-30 is 10 mm above the liquid level before flow is established through the impinger (labeled "surface impingement" in Fig. 4A). In contrast, the nozzle exit of the

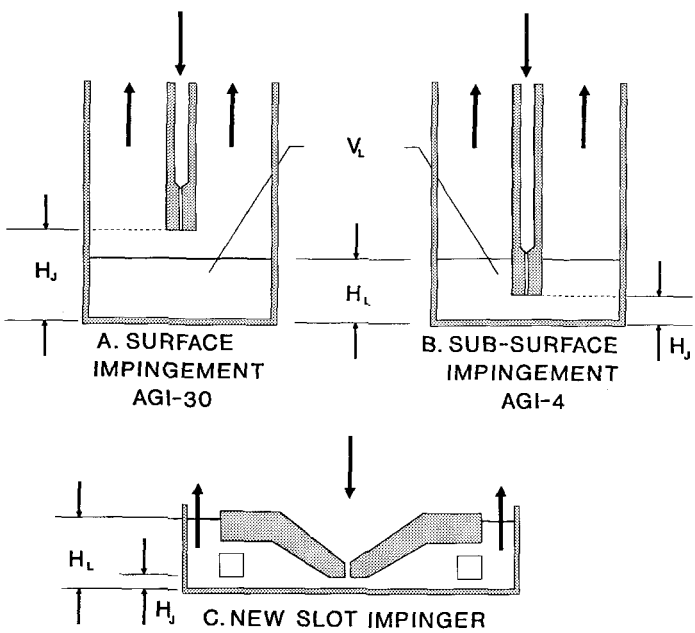

FIGURE 4. Schematic representation of the impingers used in the study. A: AGI-30. B: AGI-4. C: New slot impinger.

AGI-4 is initially 16 mm below the liquid level, which we labeled "subsurface impingement" in Fig. 4B. At the normal operational flow rate, turbulent motion and bubbling of the liquid make the liquid level less definable. The evaporation of the collection fluid during aerosol sampling reduces the liquid volume V_L and thus H_L. To study the loss of liquid on the impinger's collection efficiency, the AGI-4 was tested at two extreme conditions: 1) at the beginning of the sampling period when the liquid level is $H_L = 20\,\text{mm} > H_J = 4$ mm, and 2) at the end of a long sampling period when the level has dropped to $H_L = 1.5$ mm $< H_J = 4$ mm. For these measurements, the sampling flow rate through the impinger was varied from 2 to 13 L/min.

The slot impinger was chosen as an alternate sampler to the All-Glass Impingers. It consists of two parts: a metal inlet and a plastic vessel sealed to the inlet by an O-ring. The bottom part of the inlet unit tapers into a narrow 0.2 mm wide and 13.3 mm long slot. The jet-to-plate distance of this impinger is $H_J = 1$ mm. The slot impinger was filled with 20 mL of the collection fluid (the same V_L as recommended for the All-Glass Impingers), which results in a liquid level of $H_L = 10$ mm, i.e., this sampler operates in the "subsurface" impingement mode. The collection efficiency of this impinger was measured for sampling flow rates of 1–12 L/min. For comparison purposes, experiments were also performed with an empty slot impinger ($H_L = 0$) and a single-stage glass slide impactor. The latter has the same geometry of the inlet and collection units as the slot impinger (Juozaitis et al., 1994). The tests with all impingers were performed either with deionized water or with phosphate buffer solution (KH_2PO_4, 4 g/L + K_2HPO_4, 13.6 g/L) which are sometimes used for bacterial sampling.

Test Aerosols

In the past, impingers were widely used to collect inert particles. At this time, however, they are mainly used for bacterial sampling. For our tests, we choose *Bacillus cereus* spores (ATCC 11778, Difco Laboratories, Detroit, MI) with $d_a \approx 1.1$ μm and inert, monodisperse PSL particles covering most of the bacterial size range ($d_a = 0.55$, 0.72, and 1.09 μm). Both types of test particles are spherical, and are within the particle size measurement range of the Aerosizer and the LAS-X size spectrometers. The PSL particle and bacterial spore concentrations in liquid suspension before nebulization were 10^7–10^8 mL^{-1} (determined with experimental error of $< 20\%$). The PSL spheres are known to be very bouncy when interacting with solid surfaces (John, 1995). We expected the test bacterial spores also to be bouncy because their cells walls are very rigid. In addition, *Bacillus cereus* spores are highly hydrophobic (Wiencek et al., 1990).

RESULTS AND DISCUSSION

The first set of experiments was conducted with four identical AGI-4 impingers and four identical AGI-30 impingers collecting *Bacillus cereus* spores. Each sampler contained $V_L = 20$ mL of deionized water and was initially operated at 10 L/min. At this flow rate, intense bubbling in the liquid was observed. The bubbles burst at the top of the liquid, which caused spontaneous and irregular movement of the liquid surface. All eight samplers demonstrated perfect particle collection at 10 L/min: no airborne spores were detected downstream of the impingers. However, as soon as Q_{IMPG} was increased to levels above 10 L/min, the Aerosizer detected significant concentrations of supermicrometer particles downstream of each impinger. The AGI's collection efficiency was found to decrease from 100% at 10 L/min to 80–90% at the normal operational flow rate of 12.5 L/min. This decrease in E_{COLL} was not very great, but it was significant.

Collection efficiency measurements with the All-Glass Impingers were also performed after 15, 30, 60, and 120 min of

continuous impingement. During this time, the level of the collection fluid dropped from $H_L = 20$ mm to a few millimeters, depending on the relative humidity of the air supplied to the impinger. These tests showed a nonmonotonic behavior of E_{COLL} with time, particularly as the liquid level H_L became very low.

Comparative tests have therefore been conducted with the AGI-4 at a high (H_L = 20 mm) and a low (H_L = 1.5 mm) level of deionized water in the impinger vessel. Figure 5 shows the variation in collection efficiency with flow rate for 1.09 μm PSL particles and 1.1 μm *Bacillus cereus* spores.

At the recommended liquid level of 20 mm and a relatively low flow rate of 2 L/min, more than 50% of the PSL particles and approximately 70% of the *Bacillus cereus* spores were collected by the AGI-4 (Fig. 5A). At higher sampling flow rates of 6–10 L/min, the impinger collection efficiency was above 90% for both the PSL and bacterial particles. A collection efficiency of 100% is theoretically expected for these flow rates when aerosol particles are impacted from the impinger nozzle onto a solid plate (Marple, 1970; Marple and Willeke, 1976, 1979; Marple et al., 1993). Figure 5A shows, however, that an increase in the flow rate to levels above 10 L/min slightly decreased E_{COLL} of the AGI-4. This decrease in overall collection efficiency is attributed to reaerosolization of some particles from the collection fluid. When the flow rate is high enough, particles already collected by the liquid are entrained by rising bubbles, rendered airborne through bursting bubbles and, once airborne, carried upward out of the impinger vessel.

Figure 5B shows that the collection efficiency of the impinger was very different

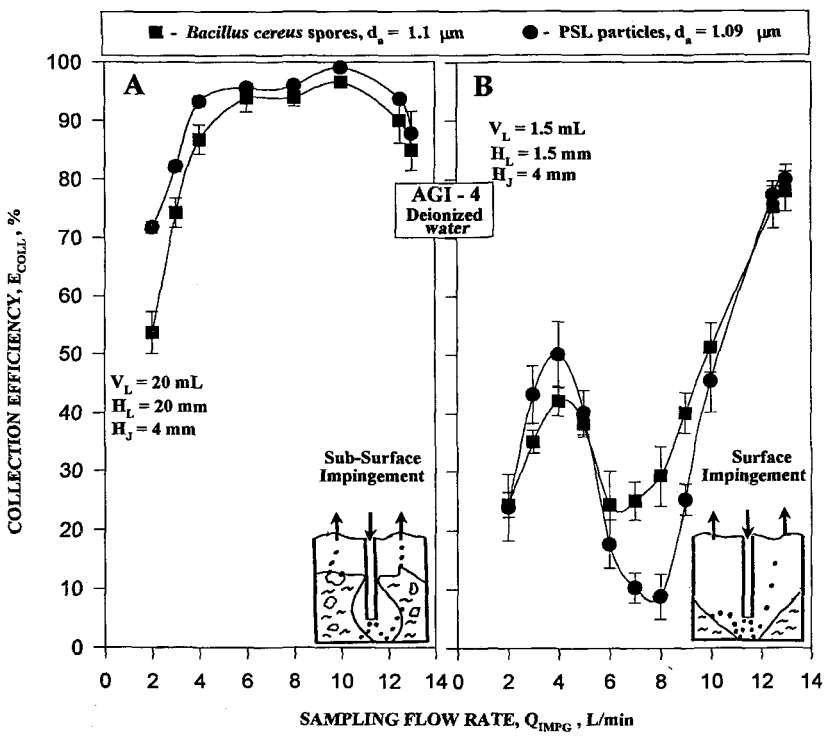

FIGURE 5. Collection efficiencies of the AGI-4 at different levels of collection fluid. A: $H_L = 20$ mm. B: $H_L = 1.5$ mm.

from the monotonic function of the flow rate when the liquid level is low ($H_L = 1.5$ mm). At relatively small sampling flow rates of 2–4 L/min, the collection efficiencies were only about 40% for *Bacillus cereus* spores and 50% for PSL particles. The increasing values in collection efficiency for flow rates increasing from 2 to 4 L/min appear to be in agreement with the values predicted for inertial particle collection on a solid plate. However, a further increase in Q_{IMPG} to 6–8 L/min resulted in a significant decrease in the collection efficiencies for both types of particles. It was observed that, at these flow rates, the shallow liquid layer ($H_L < H_J$) under the AGI-4 capillary was removed by the pressure created by the air jet. Thus, the particles impacted directly onto the glass bottom of the vessel from which they bounced away. The bounced particles may either escape with the effluent air flow or may be impacted into the liquid which is pushed against the vessel's walls by the air jet. In our experiments, the average particle kinetic energy was about 10^{-9} J, which is high enough to allow the elastic latex spheres or spores to rebound from the vessel bottom (this estimate was performed using the model introduced by Xu and Willeke, 1993a). A further increase in the flow rate to values above 8 L/min appears to result in multiple particle bounce from the two opposite surfaces: the vessel bottom and the lower surface of the thick-walled capillary. In this case, the particle kinetic energy for the second bounce is lower, so that the particles may be impacted onto and retained by either the dry or wet surfaces of the vessel. The presence of liquid on the impinger wall and part of the bottom surface increases the retention of these particles. As seen in Fig. 5B, E_{COLL} increases for PSL particles and spores at flow rates above ≈ 8 L/min.

While the data for latex spheres and bacterial spores show the same behavior, the differences in collection efficiency are attributed to their different hydrophobicity and bounce efficiency. In order to test the reaerosolization and bounce hypotheses, further experiments were performed as described below.

Particle Reaerosolization in Impingers

Particle aerosolization was studied by operating the AGI-4 in the aerosol generator mode, i.e., only prefiltered particle-free air entered the impinger filled with 20 mL of a suspension, containing 1.25×10^6 *Bacillus cereus* spores per mL of deionized water. The impinger was tested at two sampling flow rates, 5 and 12.5 L/min, and the measured data were adjusted by the particle concentration and size corrections presented in Figs. 2 and 3. Figure 6 shows that there is significant aerosolization at both flow rates. The mean particle diameter of the particle size distribution obtained at 12.5 L/min ($d_a = 1.08$ μm) is about the same as the aerodynamic size of the test spores; the geometric standard deviation (σ_g) over the measured size range is 1.22. The mean diameter and the geometric standard deviation at 5 L/min are somewhat higher: $d_a = 1.46$ μm and $\sigma_g = 1.37$. The initial sizes of the droplets aerosolized from the bacterial suspension are believed to be higher than the above indicated mean diameters. Most of the droplets' water content is evaporated by the addition of dry air (valve 2 in Fig. 1) prior to particle detection by the Aerosizer.

The experiments were repeated with the AGI-4 filled with filtered, deionized water in order to verify that the peaks of the particle size distributions are due to bacteria in suspension. As seen in Fig. 6, no particles were detected downstream of the impinger in this case. The aerosolization rate for the deionized water should not be very different from the one found for the bacterial suspension at the same flow rate. However, all of the droplets dispersed from the deionized water are fully evaporated before being measured by the Aerosizer. Figure 6 shows that the droplet residues of dispersed deionized water are of sizes below the size measurement threshold of the Aerosizer (< 0.5 μm), i.e., they have not

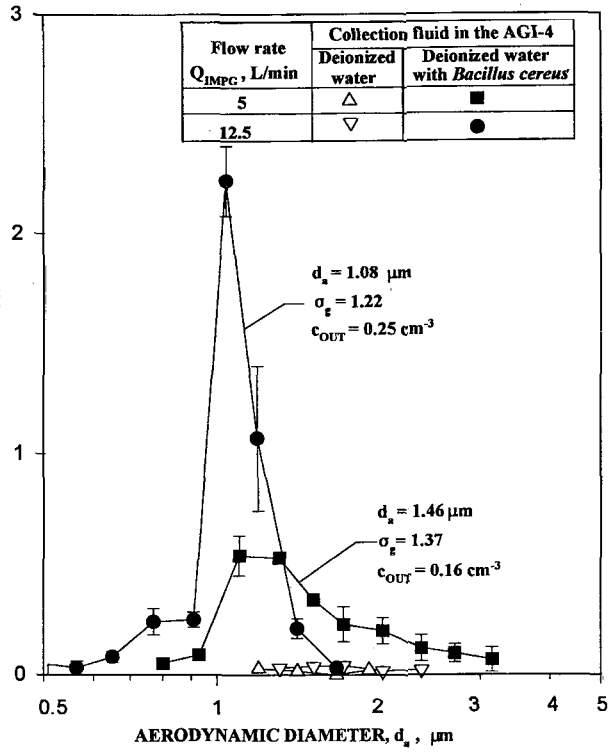

FIGURE 6. Aerosolization of bacteria from the impinger when operated in the aerosol generator mode.

been detected by this instrument. In the case of the dispersed bacterial suspension, the bacteria and accompanying bacterial slime show up as particles with size peaks close to the size of single bacteria.

When impingers are used to collect bacteria, a buffer is frequently used to maintain the viability of the microorganisms. The aerosol dispersed by an impinger depends on the materials dissolved in the liquid, as well as on the impinger design and flow rate and the bacteria already collected. As the liquid is dispersed into droplets and the droplets are shrunk by the addition of dry air, the dissolved material shows up as residue particles. Figure 7 presents the particle size distributions resulting from the dispersion of a widely used phosphate buffer solution. In this experiment, the AGI-4, containing 20 mL of buffer solution without bacteria, is operated at 12.5 L/min of particle-free air. As seen, a bimodal particle size distribution with a principal mode at 1.15 μm results when no drying air is added. When the dry air is introduced downstream of the impinger, the size distribution is also bimodal, but the principal peak is at 0.92 μm. The drying air further evaporates liquid remaining on the particles, dilutes the aerosol concentration, and affects the particle losses in the measurement system.

Particle Bounce in Impingers

Particle bounce in impingers was studied with PSL particles of different sizes using two samplers: the slot impinger (Fig. 4C) filled with 20 mL of phosphate buffer and the glass slide impactor with a grease-coated surface to prevent particle bounce. Both samplers were designed with the same inlet and collection geometries, but using dif-

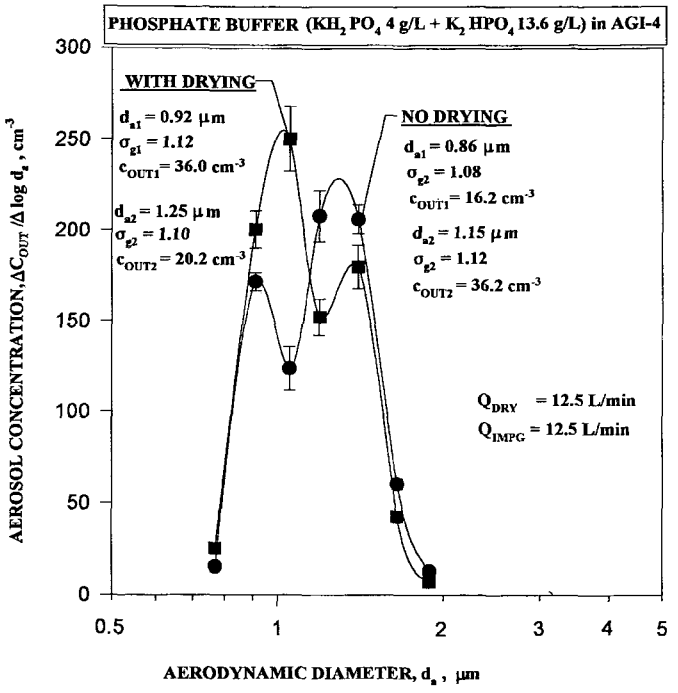

FIGURE 7. Effect of aerosolization from phosphate buffer solution on the particle size distribution downstream of the impinger.

ferent collection media. Figure 8 shows the dependence of collection efficiency on sampling flow rate for the two samples tested with PSL particles of $d_a = 1.09$ μm. At low flow rates, $Q_{IMPG} \leq 4$ L/min, both samplers have similar increases in E_{COLL} with Q_{IMPG}. However, with increasing flow rates, the collection efficiency of the slot impinger decreased and then increased again, similar to the performance of the AGI-4 shown in Fig. 5B. For the same flow rates, E_{COLL} for the impactor with the coated impaction surface remained close to 100%. The difference between the two performance curves, which appeared to be most significant at flow rates of about 6–8 L/min, was due to particle bounce in the tested impinger. This effect was found to be less pronounced for the slot impinger than for the All-Glass Impingers because the air jet velocity in the former was about four times lower than that of the All-Glass Impingers.

If the dip in the performance curve of the slot impinger, seen in Fig. 8, is due to bounce, then one would expect the same or an even greater degradation of the curve when there is no liquid in the impinger. This is shown in Fig. 9 for three different PSL particle sizes. Each of the three graphs shows the performance of the impinger when it is filled with 20 mL phosphate buffer ($H_L = 10$ mm) versus when it is empty ($H_L = 0$). As seen in Fig. 9, the collection efficiency of the impinger with no collection fluid increases with flow rate until Q_{IMPG} reaches its critical value, when the particle inertia is sufficiently high for bounce to occur (Xu and Willeke, 1993a). This critical flow rate decreases with increasing particle size: 8 L/min for $d_a = 0.55$ μm, about 4–6 L/min for $d_a = 0.72$ μm, and 4 L/min for $d_a = 1.09$ μm. When the impinger contained 20 mL of collection fluid, the bounce effect was also found to be most pronounced for the larger particles, which we attribute to the decreasing ratio of the adhesion to kinetic energy with increasing particle size.

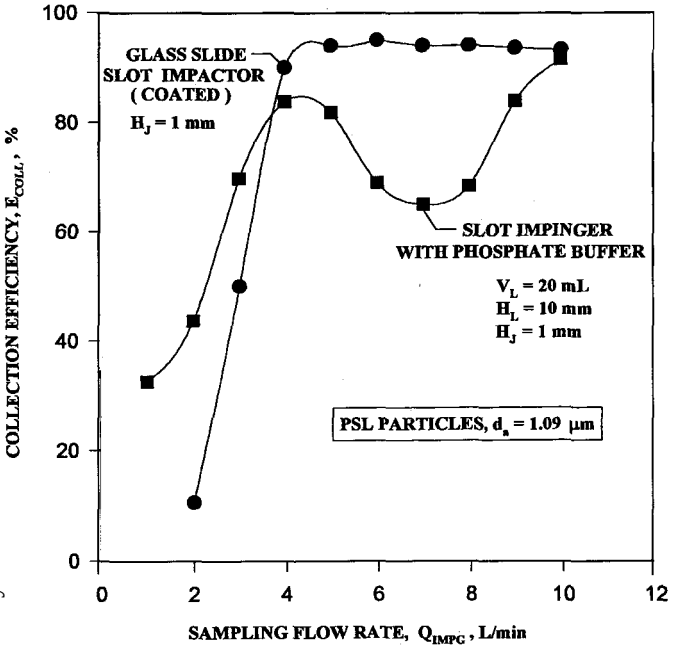

FIGURE 8. Collection efficiencies of the glass slide impactor and the slot impinger with the same inlet (impinger contains phosphate buffer solution) sampling PSL particles of 1.09 μm at different flow rates.

SUMMARY

The primary mechanism for the collection of airborne particles in impingers is inertial impaction. Thus, one would expect the collection efficiency to increase with sampling flow rate and particle size. This study has shown that this is true for a limited range of conditions. For many conditions, however, impinger performance was significantly different from the expected behav-

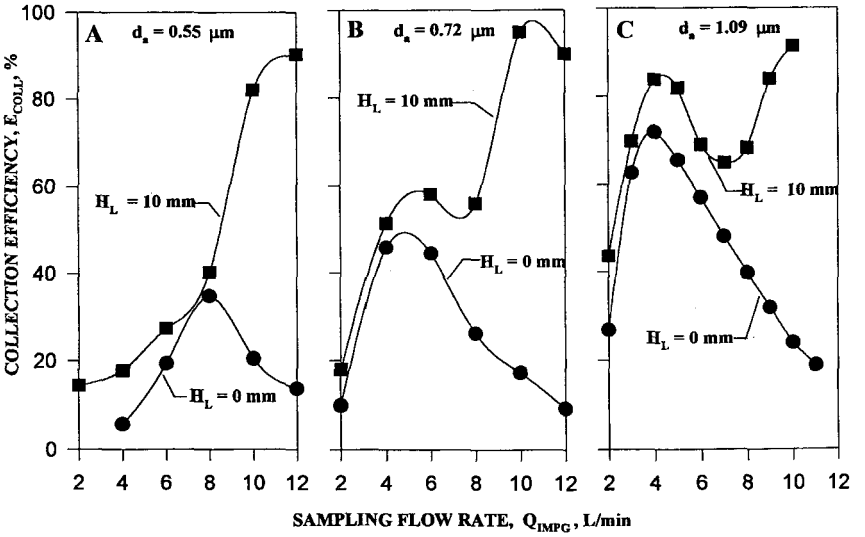

FIGURE 9. Effect of PSL particle bounce on the collection efficiency of the slot impinger when empty versus filled with phosphate buffer. A: PSL $d_a = 0.55$ μm. B: $D_a = 0.72$ μm. C: $d_a = 1.09$ μm.

ior. At low flow rates, the inertia of the particles is low, and therefore the collection efficiency is also low. At high flow rates, the liquid under the impinger jet may be removed by the air pressure and pushed against the walls. This effect is especially pronounced when the level of the collection fluid is low, which usually results from liquid evaporation during the prolonged impingement. If the particles are sufficiently elastic, as is the case with many spores, they may bounce from the bottom of the collection vessel and escape with the effluent air flow or impact laterally into the liquid. Such particle bounce may significantly reduce the collection efficiency of impingers containing only a small amount of liquid. When an impinger operates at a certain level of collection fluid, the air flow turns into bubbles rising through the liquid. At the top of the liquid, the bubbles burst into droplets of supermicrometer size, which may contain some of the smaller particles that were previously collected by the impinger. At high sampling flow rates, the upward air velocity is sufficiently high to entrain these particles out of the impinger. This particle reaerosolization may significantly reduce the impinger's collection efficiency. The hydrophobicity of the sampled microorganisms is expected to affect the fraction of reaerosolized particles.

As seen, the degree of inertial impaction, bounce, and reaerosolization depends on the volume of the collection fluid and the sampling flow rate. Therefore, several different impingement regimes have been identified. Figure 10 schematically illustrates these regimes for the AGI-4 containing high and low levels of liquid. Inefficient particle collection is shown in Fig. 10A and B for low flow rates which impart low inertia to the particles. Figure 10C shows the optimal conditions, i.e., when the collection fluid volume is large enough to minimize bounce and the flow rate is sufficiently high to provide efficient collection, but not high enough to cause significant reaerosolization from the liquid. Figure 10D shows significant particle bounce into air when the liquid volume is reduced (evaporated) to a low

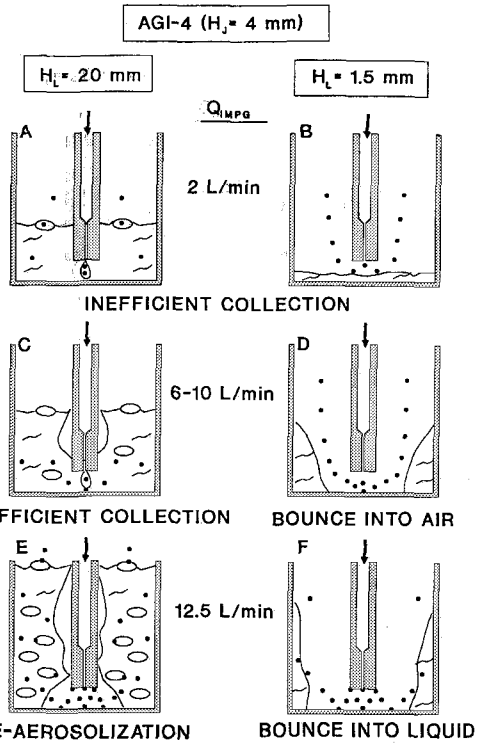

FIGURE 10. Schematic representation of the collection process in an impinger.

level. Figure 10E shows that a fraction of the collected aerosol particles may reaerosolize from the collection fluid when the impinger contains the normal amount of liquid and is operated at the usual (relatively high) flow rate of 12.5 L/min. When the liquid level is low, Fig. 10F, secondary impaction from the bottom of the impingement nozzle bounces the particles into the liquid that is pushed laterally toward the inner walls of the vessel. The processes of the particle bounce and reaerosolization during impingement were found to be similar, but of more complex character when using the AGI-30 versus the AGI-4. We conclude from this study that effects of particle bounce and reaerosolization should also be evaluated for other commercially available impingers and for those under development so that the optimal operating

conditions can be defined for collecting microorganisms or biologically inert particles.

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