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Design and development of an electrostatic sampler for bioaerosols with high concentration rate

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Abstract

Integration of bioaerosol sampling methods with modern analysis techniques, such as the polymerase chain reaction as well as our ability to detect low concentrations of airborne agents require samplers that are able not only to efficiently collect the biological particles, but also to concentrate them in small amounts of fluids. In this research, we began development of a novel bioaerosol sampler, where a combination of electrostatic collection mechanism with superhydrophobic ("Lotus leaf" type) collection surface allows for efficient particle collection, removal and concentration in water droplets as small as 5 μL. This new sampling concept allowed achieving very high sample concentration rates (up to 1 million and higher) and could be applied to detect low concentrations of bioaerosols in various environments.

The prototype electrostatic precipitator with superhydrophobic surface had a shape of a half-pipe, where a top plate served as the ground electrode, while the collecting surface was 3.2 mm wide rectangular electrode coated with a superhydrophobic substance and positioned in a groove in the flat bottom surface. Airborne particles drawn into the sampler were positively charged and then by the action of an electrostatic field deposited onto the negatively charged electrode. The sampler was positioned at a ∼1° inclination angle to the horizontal, and the injected water droplets rolled off of electrode's surface removing deposited particles. Sampler's performance has been analyzed with polystyrene latex particles of five aerodynamic diameters (0.5, 1.2, 1.9, 3.2, and 5.1 μm), collecting droplet volumes ranging from 5 to 60 μL, and sampling flow rates of 2, 5, and 10 L/min. It was determined that vast majority of particles deposited onto the electrode are removed by the first rolling droplet, which for 3.2 μm particle and 20 μL droplet translated into a concentration rate of 3×10^5. By narrowing the electrode to 2.1 mm and lowering the droplet volume to 5 μL we achieved the concentration rate as high as 1.2×10^6. These concentration rates were sustained for sampling times as long as 60 min. This novel sampling concept demonstrates a great potential for sampling and detecting airborne microorganisms in low concentration environments.

Keywords: Bioaerosol sampling; Electrostatic precipitation; Concentration rate; Collection efficiency; Superhydrophobic surface

1. Introduction

Exposure to airborne biological agents, especially pathogenic or allergenic microorganisms, may cause a wide range of respiratory and other health disorders in occupational and general populations. Such exposures are increasingly recognized as a cause of preventable airborne infections and hypersensitivity diseases (World Health Organization,

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1990). Although relatively little data exist on the presence of cells or cell material of fungi and bacteria in fine particle samples (Womiloju, Miller, Mayer, & Brook, 2003), some studies indicate that perhaps as many as 10% of urban and rural fine aerosols are biological in nature (Monn, 2001). Douwes, Thorne, Pearce, and Heederik (2003) concluded that many bioaerosol species that may cause health effects are currently not yet identified and more research is needed to establish better tools for assessing and controlling our exposure to indoor and outdoor bioaerosols. In addition, improved exposure assessment and protection of populations and resources at risk from biowarfare agents requires advanced air sampling devices that feature high collection efficiency and can detect low agent concentrations.

Currently, bioaerosols are commonly collected using techniques that require active sampling, such as impaction, impingement, or deposition on filters. Recently, there has been an increased interest in collection of microorganisms using electrostatic precipitation due to its lower power requirements compared to inertial techniques while still allowing efficient particle removal from the air. In electrostatic precipitators, airborne particles are electrically charged and then removed from the air stream by an electrostatic field. Removal of non-biological aerosol particles by electrostatic precipitators has been widely studied from the theoretical and practical points of view (Cardello, Volckens, Tolocka, Wiener, & Buckley, 2002; Lu & Hungsung, 1998; Rose & Wood, 1956; Zhuang, Kim, Lee, & Biswas, 2000), owing to its widespread practical applications. These devices provide efficient particle capture while causing minimal impedance to the gas flow. A successful application of electrostatic precipitation for collection and enumeration of viable airborne microorganisms has also been described (Mainelis, Adhikari, et al., 2002; Mainelis, Willeke, Adhikari, Reponen, & Grinshpun, 2002; Yao, Mainelis, & An, 2005).

In addition to collection efficiency, another metric with which liquid-based bioaerosol samplers are being characterized, especially recently, is the "concentration rate". It is defined as a ratio of particle concentration in collection liquid versus the airborne particle concentration per time unit. It can also be expressed as a ratio of sampling flow rate (L/min) of a sampler divided by the volume of the collection fluid (L) and multiplied by the collection efficiency of the device. High concentration rates reduce the sampling time needed to detect airborne particles and enable detection of lower particle concentrations (Haglund, 2003). High concentration rates also compensate for average or even low collection efficiencies of samplers. However, concentration rates of 1 million or higher are obtainable in few research samplers (Seo, 2007). Traditional liquid samplers exhibit relatively low sample concentration rates even when assuming 100% collection efficiency (any reduction in collection efficiency reduces the concentration rate by the same factor). For example, a Biosampler (SKC Inc., Eighty Four, PA) operating at 12.5 L/min and sampling into 5 mL of liquid would have a concentration rate of up to 2500. InnovaTek, Inc. (Richland, WA) introduced the BioGuardian air sampler which operates form 100 to 1000 L/min and collects sample into 10–15 mL of liquid, which results in a maximum concentration rate of 100,000. The SpinCon air sampler by Specter Industries, Inc. (Kansas City, MO) samples at 450 L/min and concentrates sample into 10 mL of liquid (maximum concentration rate of 45,000). The BioCapture 650 (MesoSystems Technology, Inc., Albuquerque, NM) is a portable sampler that uses a sampling flow rate of 200 L/min and collects particles into 5 mL of liquid (maximum concentration rate 40,000). The Lawrence Livermore National Laboratory (LLNL, Livermore, CA) has developed a stationary Autonomous Pathogen Detection System (APDS) that is capable of continuous and fully autonomous monitoring for multiple biowarfare agents. The APDS operates at flow rates up to 3750 L/min and can achieve concentration rates as high as 750,000 when collecting 3 μm PSL particles into 4 mL of liquid (Mainelis, Masquelier, Makarewicz, & Dzenitis, 2005). However, its size, power, and cost requirements would make it difficult to apply in most occupational and residential environments. Among the electrostatic precipitators, a briefcase-sized sampler developed by the Savannah River Technology Center samples at an airflow rate of 300 L/min and collects particles into 20 mL of liquid (Carlson, DeGange, Cable-Dunlap, & Halverson, 2004). Coyle and Bindra (2004) presented an electrostatic sampler having similar collection characteristics and flow rates. These samplers have concentration rates of about 15,000–20,000.

As could be seen, the concentration rates of majority of bioaerosol samplers, especially the compact ones, are still in the order of tens of thousands even when assuming 100% collection efficiency. Since the increase in sampler's concentration rate enables detection of lower airborne microorganism concentrations, even if a sampler features lower collection efficiency, it is important to develop samplers that feature high concentration rates. Another desirable sampler's feature is low power consumption. Since compared to inertia-based techniques, electrostatic precipitators require less energy, development of an electrostatic sampler capable of high concentration rates would be especially beneficial.

In this manuscript we present a concept and characteristics of a novel bioaerosol sampler, where a combination of electrostatic collection mechanism and a collection surface coated with material, water droplet contact angle of which exceeds 150° (superhydrophobic surface), allows achieving very high concentration rates.

In nature, superhydrophobic surface properties allow certain plants ("Lotus leaf" type) to be cleaned from dust pollution by a simple rain shower (Barthlott & Neinhuis, 1997). Superhydrophobic nature of the leaf's surface makes water droplets form spheres with very little adhesion to the surface which then roll off very easily even at small inclinations. Microscopic examination of such surfaces revealed the presence of microstructured surface as well as coating by water-repellent crystals (Ma & Hill, 2006). When a water droplet rolls over a particle deposited on such a surface, the particle is wetted, adheres to the droplet and is removed from the surface. Thus, this "self-cleaning" property of Lotus leaf was incorporated into our new sampler. Here, the airborne particles are electrostatically deposited onto a superhydrophobic surface from where they are removed and collected by small rolling water droplets (from 5 to 60 μL) for subsequent analysis. In this electrostatic precipitator with superhydrophobic surface (EPSS) the airborne particles can be collected for a desired period of time and the deposited particles can then be removed at the end of the sampling period thus accruing all the collected particles in a small liquid droplet. Since the deposit is suspended in liquid, the presence of biological particles can be determined by using the traditional culturing and microscopy techniques as well as the modern molecular analysis tools, such as polymerase chain reaction (PCR).

The main goal of this manuscript is to present the concept of this novel bioaerosol collector, analyze its performance as a function of collecting droplet size and quantity, sampling flow rate, and other parameters, and to investigate the feasibility of the proposed bioaerosol sampling concept for monitoring bioaerosols present in low concentrations. Since the main goal of this stage of investigation was to determine the optimal sampling parameters of the EPSS, the experiments have been performed with polystyrene latex (PSL) particles of sizes simulating common bioaerosol particles. Later stages of the research will investigate sampler's performance with several airborne biological agents.

2. Design of the new electrostatic precipitator with superhydrophobic surface (EPSS)

The EPSS designed and evaluated in this study is shown in Fig. 1. The device has a shape of a closed half cylinder and consists of two separate components: the round top part containing the ground electrode and the flat bottom part containing the collection electrode. Fig. 1a presents a 3D view of the entire EPSS, while Fig. 1b shows a 3D view of the bottom plate. The collection electrode is a thin copper strip coated with a superhydrophobic substance. When the collection electrode is placed inside the bottom plate, the top of the electrode is slightly below the surface of the plate forming a groove for improved guidance of the collecting droplet (Fig. 1b). Two different collection electrode configurations were tested in the study: the majority of the experiments were performed with an electrode 3.2 mm wide and positioned 0.5 mm below the plate surface. Some experiments were performed with an electrode that had the width of 2.1 mm and was 0.3 mm below the plate. In both configurations the length of the electrode was 254 mm. During the sampler's operation, the charged airborne particles are pulled into the sampler (charging conditions are described below) and the configuration of ground and collection electrode focuses the electrostatic field lines so that particles are subjected to an electrostatic field and deposited onto the collection electrode covered with the superhydrophobic substance. After a sampling period, liquid droplets of a desired volume are sequentially injected at the top of the collection plate using a micropipette. Due to the inclination of the collection chamber (1–30°) and high liquid contact angle, the liquid droplets roll down picking up deposited particles and are collected in a vial located downstream of the collection chamber.

Although the collection electrode captures most of the particles, a fraction of the particles may be lost to the ground electrode and non-conductive parts of the sampler due to static charges. To minimize such losses, a large portion of the bottom plate is covered by an inlayed conductive material (copper). The particles not captured by the EPSS are conveyed out of the device via exhaust.

3. Experimental setup and testing methodology

Fig. 2 shows the schematic of the experimental setup used to evaluate the performance of the electrostatic collector with superhydrophobic surface. The entire experimental setup was housed inside a Class II Biosafety cabinet (NUAIRE Inc., Plymouth, MN) so that airborne particles not collected by the device are properly eliminated.

A Collison nebulizer (BGI Inc., Waltham, MA) was used to aerosolize the green fluorescent PSL particles (Duke Scientific, Palo Alto, CA) from a liquid suspension at a flow rate, Q_A (4 L/min) and the test aerosol was dried and diluted with HEPA-filtered air flow, Q_D (36 L/min). The 40 L/min aerosol stream was passed through a 2-mCi Po-210 charge neutralizer to reduce aerosolization-related particle charges to Boltzmann equilibrium. The electrically

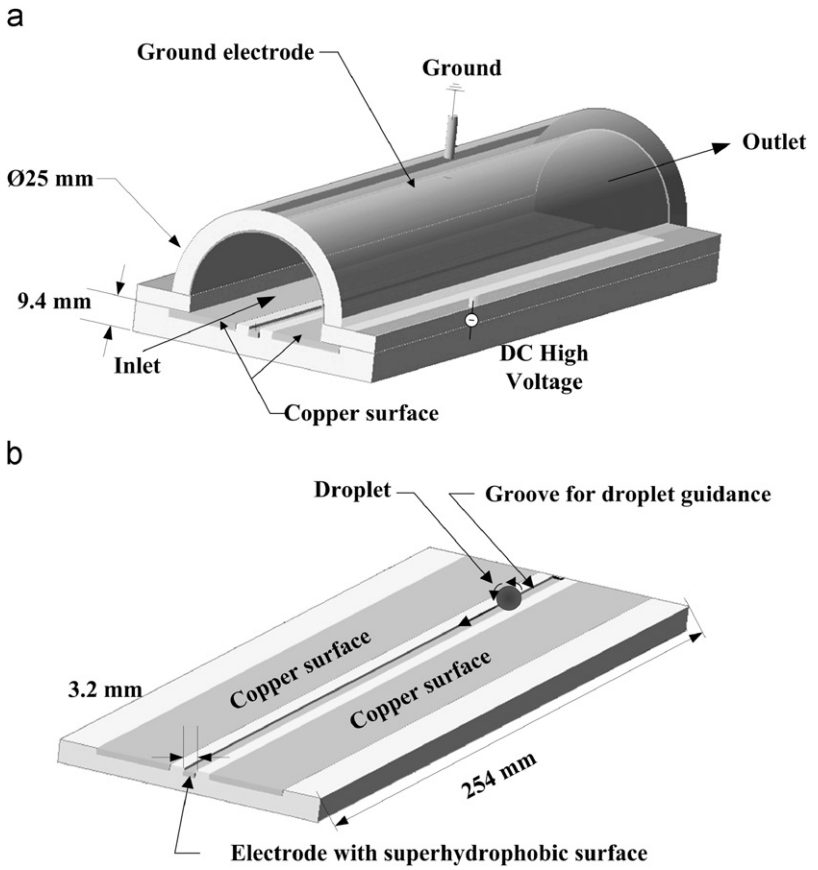

Fig. 1. The schematic representation of the electrostatic precipitator with superhydrophobic surface (EPSS). (a) Overall view and (b) bottom plate.

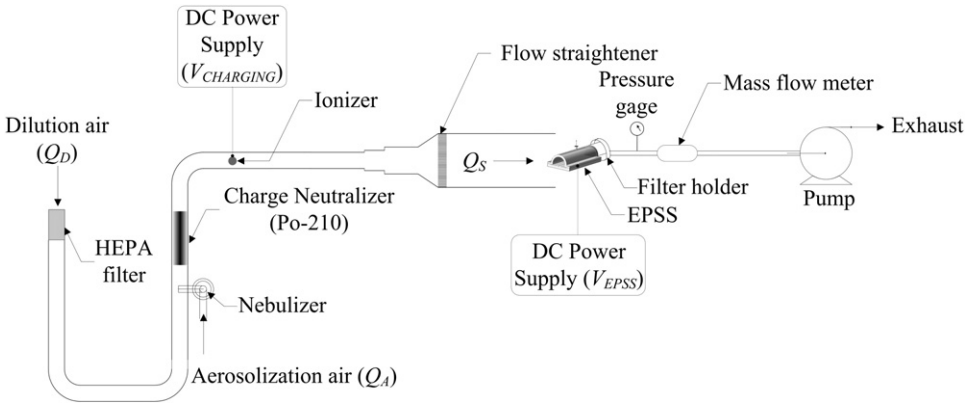

Fig. 2. The schematic diagram of experimental setup.

neutralized particles then passed through a 0.035 m duct housing a vertically oriented ionizer (AS 150, Wein Products Inc., Los Angeles, CA) which imparted positive charge on the particles under controlled voltage and current settings. The electrically charged aerosol passed through a flow straightener and entered the testing chamber where it was collected by the EPSS at a flow rate Q_S.

During the tests, the EPSS was placed at a distance of 2-duct diameters downstream of the flow straightener and, depending on the test, collected particles from 10 to 60 min. A 47 mm glass fiber after-filter (Type A/E, Pall Inc., East Hills, NY) was used to collect the particles not deposited inside the EPSS, thereby allowing to determine the concentration of particles retained inside the EPSS.

For each test, the collecting electrode was coated with superhydrophobic spray HIREC-1450 (NTT Corporation Inc., Japan) and left to dry at 60 °C for at least 1 h. The coating procedure was repeated twice to achieve a uniform coating. One stable DC power supply (BK Precision, Yorba Linda, CA) provided power to the ionizer (12 V/50 mA), while another stable DC high voltage power supply (Bertan Associates, Inc., Valhalla, NY) provided negative voltage (−7 kV) to the precipitator to collect the positively charged particles. The operating values for these two voltages were established in the preliminary experiments as yielding the most efficient deposition of particles inside the EPSS and were not varied throughout the experiments. The effect of charging conditions on particle deposition inside the EPSS will be addressed in subsequent research.

Fluorescent PSL particles of five sizes, i.e., 0.5, 1.2, 1.9, 3.2, and 5.1 μm in aerodynamic diameter (d_a) were used to characterize the sampler's performance at three collection flow rates, $Q_S = 2$, 5, and 10 L/min, which were monitored using a mass flow meter (TSI Inc., Shoreview, MN). The particle sizes were selected to represent the most common size range of airborne bacteria and fungi.

After each test, the particles deposited on the superhydrophobic surface were removed by three sequential water droplets, which were collected in separate vials. The performance of the sampler was tested with water droplets of 5, 20, 40, and 60 μL. The mass concentration of PSL particles removed by each droplet, collected by the after-filter, as well as the concentration of particles deposited on the ground electrode and elsewhere inside the sampler were quantified using a fluorometer (Sequoia-Turner Corporation, Mountain View, CA).

Each sequential hydrosol sample (first, second, and third water droplets) collected in a vial was evaporated using a heat gun (Master-Carr, Inc., Robbinsville, NJ) and then 4 mL ethyl acetate (EMD Chemicals Inc., Gibbstown, NJ) was added to the vial and set aside for 20 min to dissolve the PSL particles. An after-filter containing particles that escaped the EPSS sampler was soaked in 25 mL of ethyl acetate in a glass container for 4 h to elute the fluorescein dye from the PSL particles. The mass of PSL particles remaining on the collection electrode (not removed by three rolling droplets) as well as the mass of PSL particles deposited on other components of the collector was quantified by extracting them using a defined quantity of ethyl acetate and analyzing using a fluorometer.

The concentration of aerosolized PSL particles was such as to ensure that fluorometer reading of each sample was approximately 10-fold of the background fluorescence of ethyl acetate and the measurements were adjusted for background readings. In addition, the concentrations of all analyzed samples were within the linearity range of the fluorometer.

A fraction of particles deposited in any individual part of the system (water droplet(s), collection electrode, ground electrode, bottom plate, or after-filter), η_i, can be defined as a ratio of the relative concentration of particles in the individual part, C_i, to the total relative concentration of the aerosol entering the system, C_{TOTAL}, and can be expressed as

$$\eta_i = \frac{C_i}{C_{TOTAL}} = \frac{C_i}{\sum_i C_i} \tag{1}$$

The C_{TOTAL} is the sum of aerosol particle concentration in all individual parts of the system and is easily obtainable once the individual concentrations are determined. For all testing conditions the sampling was isoaxial and isokinetic or near-isokinetic and, according to formulas provided by Hinds (1999), the inlet efficiency for 3.2 μm particles was 96% and greater. In addition, the C_{TOTAL} for 3.2 μm particles was compared against particle concentration in the test chamber (upstream of the sampler) measured by a filter in an isokinetic probe and the two concentrations were found to agree within 4%. Thus, to minimize the numbers of measurements, the C_{TOTAL} was used as a reference value for calculating various performance parameters of the sampler.

The relative aerosol concentration, C_i, for each part is calculated from

$$C_i = \frac{I_i \cdot V_i}{Q_S \cdot t} \tag{2}$$

Here, I_i is the concentration of fluorescein eluded in ethyl acetate (fluorometer reading) for a component i, V_i the volume of solution used to elute the tracer for a component i, Q_S the air sampling flow rate, and t the sampling time.

The efficiency with which the particles deposited on the collection electrode were removed by each sequential water droplet, η_{Rj}, can be expressed as

$$\eta_{Rj} = \frac{C_{WDj}}{C_{Electrode} + \sum_j C_{WDj}} = \frac{C_{WDj}}{C_{Electrode+WDs}} \tag{3}$$

where C_{WDj} is the PSL particle concentration in each sequential water droplet ($j = 1$, 2, or 3), $C_{Electrode}$ is the concentration of particles remaining on the electrode (not removed by three water droplets), and $C_{Electrode+WDs}$ is the concentration of particles deposited on the electrode during the sampling. The $C_{Electrode}$ was determined by washing the electrode with a defined amount of ethyl acetate and then determining the concentration of particles using Eq. (2). The concentration of particles in each water droplet, C_{WDj}, relative to the mass of all particles entering the sampler also determines the overall collection efficiency of the sampler based on the j-th water droplet, η_{Ej}:

$$\eta_{Ej} = \frac{C_{WDj}}{\sum_i C_i} \tag{4}$$

The η_{Ej} value incorporates the efficiency with which the particles are deposited on the collecting electrode and extracted from it into j-th water droplet.

In addition to the collection efficiency, another metric that describes the performance of an aerosol collector is the concentration rate, which represents the ratio of particle concentration in the liquid versus the concentration of particles in the air over a period of time. The concentration rate R_{Cj} with units of min^{-1}, based on the j-th droplet could be expressed as follows:

$$R_{Cj} = \frac{Q_S}{V_{WDj}} \times \eta_{Ej} \tag{5}$$

where V_{WDj} is the volume of the j-th liquid droplet.

The primary goal of this project was to analyze the particle deposition patterns inside the EPSS, the ability of the rolling droplets to remove the particles deposited onto the collection electrode and to assess the feasibility of the proposed sampling method for detection of low bioaerosol concentrations. Overall, an investigation of the new sampler's ability to collect and concentrate particles in liquid droplet(s) was conducted over a range of particle sizes (0.5–5 μm), water droplet sizes (5–60 μL), number of water droplets (1–3), sampler's inclination angles (1–30°), and sampling time period (10–60 min) at different sampling flow rates (2, 5, and 10 L/min).

3.1. Data reproducibility

It is important to determine not only the particle deposition on the electrode and other components of the EPSS, but also the reproducibility of results from one set of experiments to another, including the uncertainty (precision) of the results. For each set of repeated experiments one can determine a coefficient of variation (COV) as follows:

$$COV = \frac{\sqrt{\frac{1}{N}\sum_{k=1}^{N}(x_k - \bar{x})^2}}{\bar{x}} \tag{6}$$

where N is the number of repeats, x_k is a value for a repeat k, and $\bar{x}$ is the value of the mean. When n sets of the same experiments are performed, one can calculate the mean value of the COVs, $\overline{COV}$, as well as the standard deviation of the COVs, σ_{cov}, from several sets of experiments as

$$\overline{COV} = \frac{1}{n}\sum_{i=1}^{n} COV_i$$

$$\sigma_{COV} = \sqrt{\frac{1}{n}\sum_{i=1}^{n}(COV_i - \overline{COV})^2} \tag{7}$$

The inherant reproducibility of the measurement data could be expressed by calculating the relative precision (RP), which is defined as $RP = \sigma_{COV}/\overline{COV}$ (McFarland et al., 1999). The RP value indicates the ability of a particular

experimental setup and methods to maintain precision from one set of experiments to another. The RP accounts for the variability in random error, but does not account for any systematic error and it changes from one set of experiments to another.

4. Results and discussion

Collection efficiency of electrostatic precipitators is often expressed as a function of charging voltage and current and/or collection voltage at different particle sizes or flow rates. However, since the primary focus of this project was to examine particle deposition and removal processes inside the EPSS as well as its ability to concentrate the collected particles in a small droplet, the current and voltage values were kept constant throughout the experiments.

Fig. 3 shows the deposition fractions inside the EPSS for different PSL particle sizes ($d_a = 0.5$, 1.2, and 3.2 μm) and different water droplet sizes (10, 20, 40, and 60 μL) when sampling at a flow rate of 10 L/min and at 1° inclination angle. In these figures, the η_i is the deposition fraction for each of these components: cumulative for three water droplets, remaining on collecting electrode (particles not removed by the three droplets), ground electrode, bottom plate, and after-filter. It could be seen that for each particle size, the distribution of deposition fractions is similar across water droplet sizes, although larger droplets seem to accrue a larger fraction of deposited particles. The fraction of particles initially deposited on the collecting electrode (particles in water droplets plus those remaining on the electrode), $C_{\text{Electrode+WD}s}$, increased with increasing particle size. For each particle size, the $C_{\text{Electrode+WD}s}$ averaged over droplet sizes was approximately 58.5 ± 4.1%, 72.5 ± 3.8%, and 88.0 ± 3.7%, respectively.

In Stokes regime, the electrical mobility of a charged particle is directly proportional to the amount of charge on the particle and inversely proportional to the particle diameter (Hinds, 1999). Since the amount of charge acquired by a particle in the field charging regime is proportional to d_p^2 (Hinds, 1999), the electrical mobility is directly proportional to the particle diameter when charging conditions remain the same. Since the charging conditions were constant and the collection efficiency is a direct function of electrical mobility, this explains why higher particle fraction was deposited on the collection electrode as the particle diameter increased.

The EPSS uses electrostatic forces to remove charged particles from the aerosol stream onto the collection electrode, while uncharged particles or particles carrying low charge may be retained on other components of the EPSS (ground electrode or bottom surface) due to inertial or gravitational effects. Given sufficient amount of electrical charge on the airborne particles, deposition by electrostatic forces is expected to be the dominant mechanism. The extent of particle losses inside the EPSS (particles deposited on surfaces other than the collection electrode) decreased from 23–32% range for 0.5 μm particles to 4–9% range for 3.2 μm particles. The fraction of particles collected on the after-filter

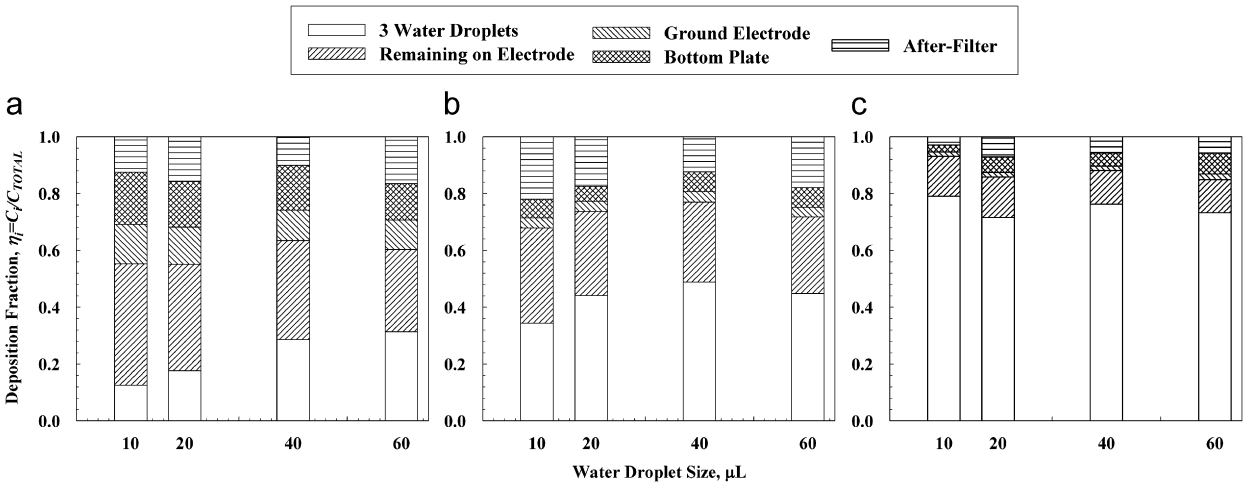

Fig. 3. Deposition fraction for each component (three water droplets, collection electrode, ground electrode, bottom surface, and after-filter) as a function of water droplet size (10, 20, 40, and 60 μL) when collecting PSL of (a) 0.5 μm, (b) 1.2 μm, and (c) 3.2 μm at 10 L/min flow rate and at the 12 V/50 mA charging condition and 7 kV collection voltage. The maximum standard deviation is 0.04 from three repeats.

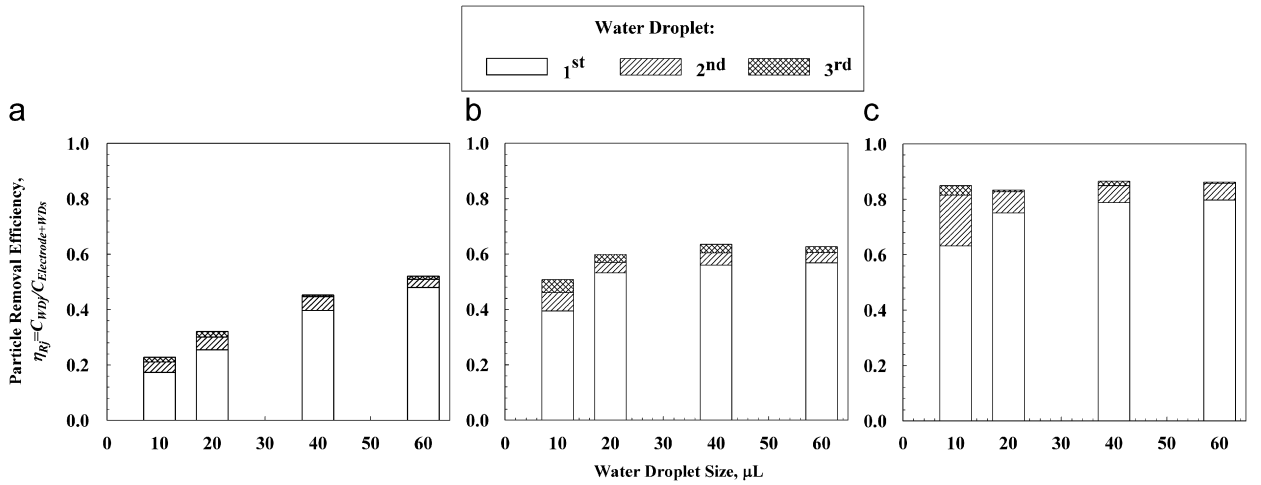

Fig. 4. Efficiency of particle removal from the collection electrode by each sequential water droplet as a function of water droplet size (10, 20, 40, and 60 μL) when collecting PSL of (a) 0.5 μm, (b) 1.2 μm, and (c) 3.2 μm at 10 L/min flow rate and at the 12 V/50 mA charging condition and 7 kV collection voltage. The maximum standard deviation is 0.043 from three repeats.

decreased from 13.7% for 0.5 μm particles to 5.4% for 3.2 μm particles. These results are again related to the electrical mobility of the particles.

The experiments conducted above indicated that the particles can be effectively deposited on the collection electrode. However, for a successful application of this sampling method it was important to determine what fraction of particles deposited on the superhydrophobic surface can be removed by each sequential rolling droplet. These data as a function of particle and droplet size are shown in Fig. 4 for 1° inclination angle of the EPSS. The most important result observed here is that the vast majority of particles that can be removed from the electrode is removed by the first rolling water droplet. The second droplet removed only a few percent of the deposited particles, while the third droplet removed 1–2% in most cases. This result is important because accumulation of most of the particles in a single droplet allows achieving higher concentration rates. If two droplets were needed, the concentration rate would be cut in half. It could also be seen that the removal efficiency, η_{Rj}, increases as a function of droplet volume, especially for the smallest, 0.5 μm PSL particles. For the 1.2 and 3.2 μm particles, the η_{Rj} of the first droplet is 40% and 65%, respectively, for the 10 μL droplet. For larger water droplets the removal efficiency increases to 53–57% for 1.2 μm particles and to 75–80% for 3.2 μm particles. This observation is most likely due to the fact that smaller particles are generally more difficult to be removed from surfaces (Hinds, 1999).

Fig. 5 shows the 3.2 μm PSL particle removal efficiency, η_{Rj}, by 40 μL water droplet from the superhydrophobic surface as a function of the inclination angle (1°, 5°, 10°, 20°, and 30°) at 10 L/min sampling flow rate. It could be seen from this figure that the particle removal efficiency decreases as the inclination angle increases to 20° and higher. The average particle removal efficiency for the first water droplet at 1–10° angle was 78.3 ± 0.8%, but it has decreased to about 65.3 ± 2.0% for 20° angle and 56.9 ± 2.9% for 30° angle. Since at higher angles the droplet tends to roll off quicker under the effect of gravity, the observed decrease is likely due to the shorter contact time of the droplet and the surface. Thus, to ensure a more efficient removal of particles deposited on the superhydrophobic surface of the new sampler, the inclination angle should be 10° or less.

Since the earlier experiments indicated that the vast majority of collected particles is removed by the first water droplet, the overall collection efficiency and the concentration rate of the sampler were also calculated (Eqs. (4) and (5)) based on the values obtained with the first water droplet. The value of this overall collection efficiency, η_E, is a product of the efficiency with which the particles are deposited on the collecting electrode and the efficiency of particle extraction from the electrode by the first water droplet. The overall collection efficiency, η_E, and concentration rate, R_C, are presented in Figs. 6 and 7, respectively, for two water droplet volumes (20 μL versus 40 μL) as a function of particle size ($d_a = 0.5$, 1.2, 1.9, 3.2, and 5.1 μm) and different sampling flow rates (2, 5, and 10 L/min). The particle charging and collection voltage settings were kept the same as in previous experiments. In these figures, the symbols indicate

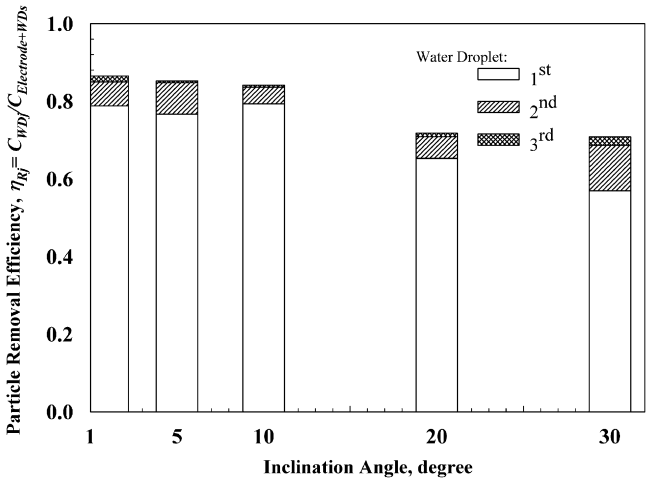

Fig. 5. Efficiency of particle removal from the collection electrode by each sequential 40 μL water droplet as function of the sampler's inclination angle (1°, 5°, 10°, 20°, and 30°) at 10 L/min flow rate and at the 12 V/50 mA charging condition and 7 kV collection voltage. The maximum standard deviation is 0.014 from three repeats.

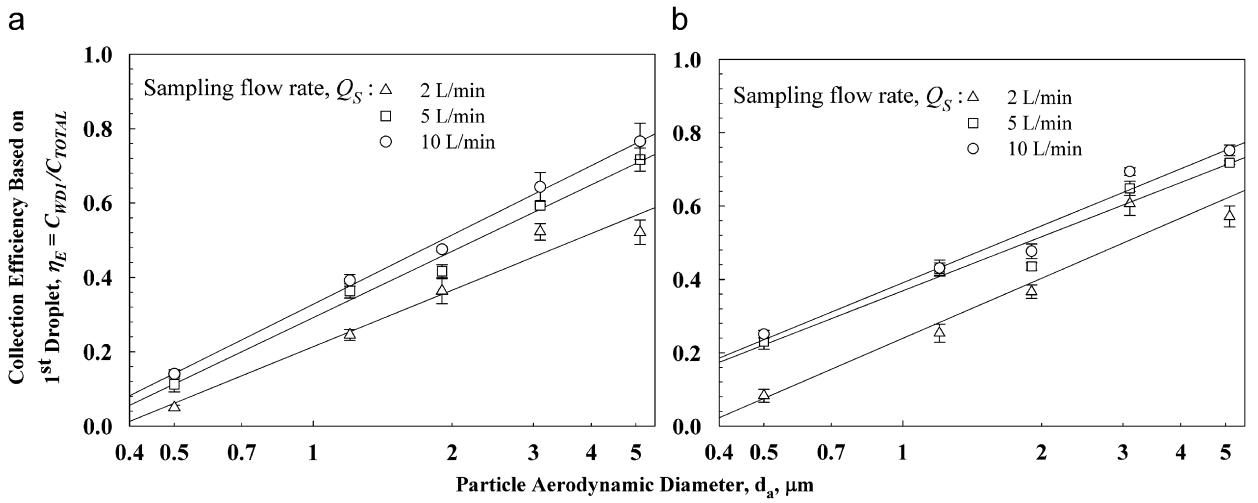

Fig. 6. Sampler's collection efficiency based on the first water droplet (particles accumulated in the first droplet versus particles entering the sampler) as a function of particle size (0.5, 1.2, 1.9, 3.2, and 5.1 μm) at 2, 5, and 10 L/min flow rates and at the 12 V/50 mA charging condition and 7 kV collection voltage for two water droplet sizes: (a) 20 μL and (b) 40 μL. The error bars represent the standard deviations from three repeats. The minimum R^2 values of the trendlines are 0.96 (a) and 0.93 (b).

average values and standard deviations from triplicate measurements, while the solid lines are linear regression lines. As could be seen from Fig. 6, the collection efficiency clearly increases for larger particles. This is a result of a more efficient removal of larger particles from the deposition electrode as was shown in Fig. 4. Another result to note from Fig. 6 is an increase in collection efficiency with increasing sampling flow rate. Although particle residence time inside the EPSS decreases at higher flowrates, our analysis of the deposition pattern inside the EPSS showed the reduction of losses inside the EPSS (particles on surfaces other than collection electrode) at increasing flow rates. At lower flow rates, gravitational settling may contribute to considerable losses which are minimized as the flow rate is increased (McFarland, Mohan, Ramakrishna, Rea, & Thompson, 2002). In addition, any losses due to image charge effects are also reduced as the flow rate increases and residence time decreases.

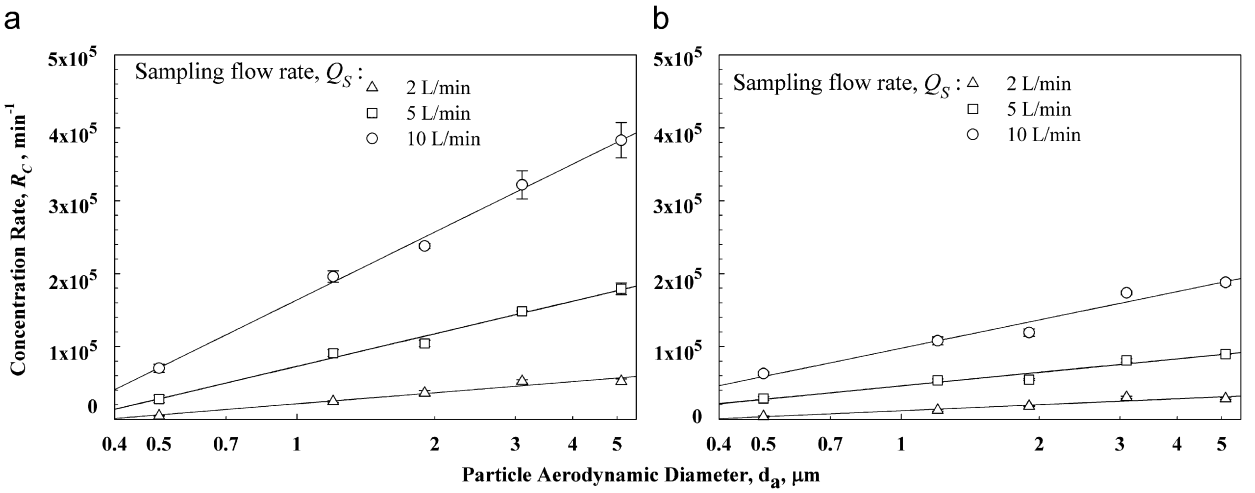

Fig. 7. Sample concentration rates based on the first water droplet, as a function of particle size (0.5, 1.2, 1.9, 3.2, and 5.1 µm) at 2, 5, and 10 L/min flow rates and at the 12 V/50 mA charging condition and 7 kV collection voltage for two droplet sizes: (a) 20 µL and (b) 40 µL. The error bars represent the standard deviations from three repeats. The minimum R^2 values of the trendlines are 0.96 (a) and 0.93 (b).

According to Lighthart (1997), about 90% of ambient bacterial aerosols are larger than 2.1 µm and more than 75% of such particles are larger than 3.3 µm. Based on Fig. 6, the collection efficiencies using 20 and 40 µL water droplets for particles 3.2 µm and larger ranged from 42–72% at 5 L/min and 47–77% at 10 L/min to 36–62% at 2 L/min. Using these collection efficiency data, we calculated the concentration rates for 20 and 40 µL droplet volumes and each sampling flow rate which are presented in Fig. 7. As could be seen, for 20 µL droplet and the sampling flow rate of 10 L/min the concentration rate for 1.9 µm particles is 240,000 and reaches 382,000 for 5.1 µm particles. These values are much higher than concentration rates achieved by other compact samplers as summarized in the Introduction. When the droplet volume increases to 40 µL, the concentration rate averaged over all particles sizes decreases by approximately a factor of 1.7 compared to 20 µL droplet (Fig. 7) mostly due to the dilution of the sample in larger droplet volume. On the other hand, reduction of the droplet volume to 10 µL would increase the concentration rate to approximately 600,000.

Another factor affecting the concentration rate (R_C) may be the width of the collection electrode relative to the droplet size. Thus, to possibly improve the concentration rate even further by using a smaller droplet (5 µL), the width of the collection electrode was reduced by a third from its original size: from 3.2 to 2.1 mm. The width of the new electrode corresponds to the diameter of 5 µL water droplet which ensures that the droplet covers most of the width of the collection electrode. The results in Fig. 8 achieved with 3.2 µm particles indicate that the concentration rate increases when a narrower electrode is used (data for 10 and 20 µL droplets). More importantly, a narrower electrode (2.1 mm) allows application of a tiny droplet of 5 µL with which an unprecedented concentration rate of 1.2 million is achieved when sampling 3.2 µm PSL particles at a flow rate of 10 L/min. Thus, the EPSS can efficiently operate at a sampling flow rate of 10 L/min with about 30–500 times higher concentration rate as compared to other liquid samplers (e.g., Biosampler, BioGuardian, BioCapture, etc.).

A number of current analytical methods use at least 50 µL of liquid for the sample analysis and may not yet benefit from the use of droplets as small as 5 µL. However, as analytical technology advances towards "laboratories-on-a-chip", a particle collector capable of concentrating particles in small amounts of liquid would be needed for successful integration of collection and detection capabilities.

All the data presented above were achieved with the sampling time of 10 min. In many bioaerosol sampling applications longer sampling times are needed. Fig. 9a shows the fraction of 3.2 µm PSL particles deposited on the electrode, $C_{\text{Electrode+WD}s}$, relative to all other components on the sampler, C_{TOTAL}. Fig. 9b shows the collection efficiency, η_E, for sampling times of 10, 30, and 60 min. After 10 min sampling, about 80% of particles are deposited on the electrode, with the rest of the particles deposited elsewhere in the EPSS. As the sampling continues, the unipolarly charged particles deposited not on the collection electrode begin to repel each other, and the electrode potential becomes dominant force

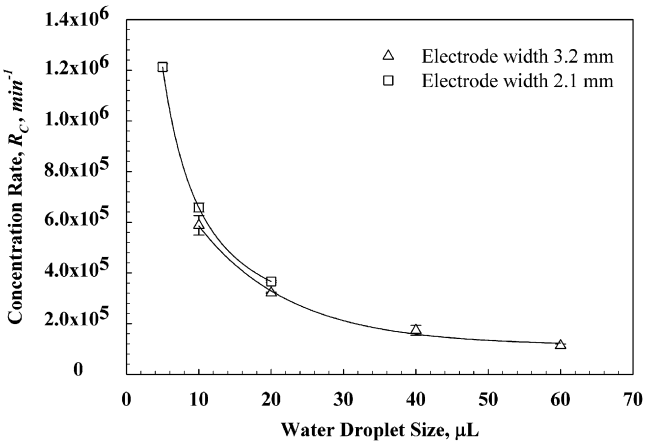

Fig. 8. Comparison of sample concentration rates based on the first water droplet at two different collection electrode widths (3.2 versus 2.1 mm) as a function of droplet size (5, 10, 20, 40, and 60 µL) at 10 L/min flow rate and at the 12 V/50 mA charging condition and 7 kV collection voltage with 3.2 µm PSL. The error bars represent the standard deviations from three repeats.

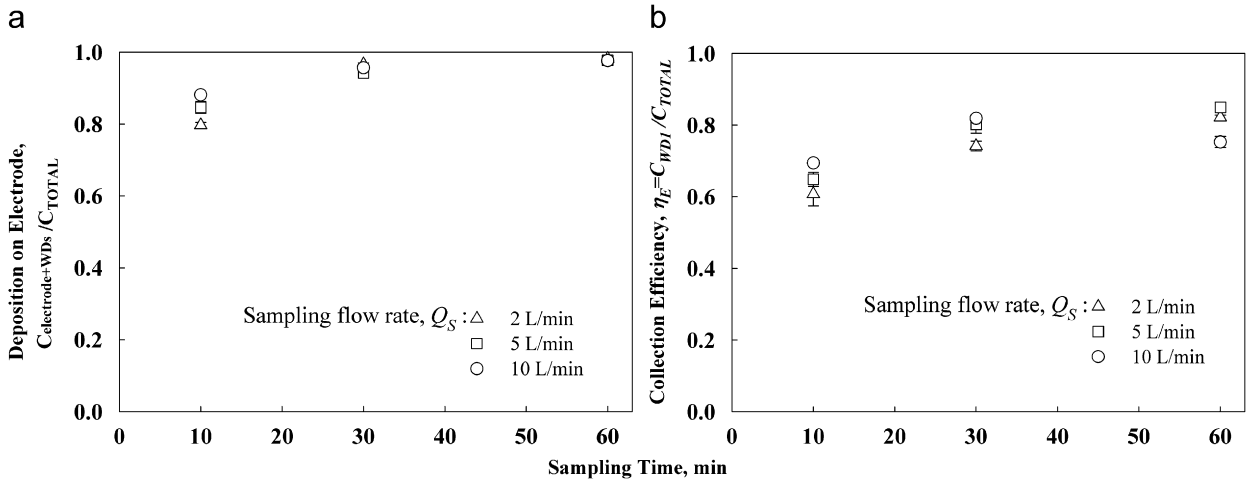

Fig. 9. Sampler's performance as a function of sampling time (10, 30, 60 min) at 2, 5, and 10 L/min flow rates and at the 12 V/50 mA charging condition and 7 kV collection voltage with 3.2 µm PSL: (a) fraction of particles deposited on collection electrode and (b) collection efficiency based on first 40 µL water droplet. The error bars represent the standard deviations from three repeats.

for particle collection and thus the fraction of particles deposited on the electrode increases. After 30 and 60 min of sampling, the deposition on the electrode increased on an average by 14% and 16%, respectively, for all sampling flow rates. As far as the collection efficiency based on the first water droplet (Fig. 9b), it increased with longer sampling time and exhibited trends similar to those observed in Fig. 9a. However, after 60 min of sampling the collection efficiency at 10 L/min decreased by 10% as compared to 30 min sampling time. Most likely because the 40 µL water droplet has become saturated and its ability to remove the particles deposited on the electrode after 60 min sampling has decreased. The decrease may not be observed in experiments with lower particle concentrations, such as encountered in ambient environment.

The main application of this sampling technology would be for collection of airborne microorganisms. Using the optimum sampling parameters determined in this research, we are currently experimenting with bacterial cells. The data indicate that the new sampling methodology allows achieving collection efficiencies of 50–60% and concentration rates as high as 1.2 million.

Table 1
Overall relative precision of COVs for each particle deposition fraction in the electrostatic precipitator for experiments performed with 40 μL water droplet at 2, 5, and 10 L/min sampling flow rates and at the 12 V/50 mA charging condition and 7 kV collection voltage with 3.2 μm PSL.

Sampling flow rates	Deposition fraction	COV (%)		Mean COV (%)	Standard deviation of COV values (%)	Ratio of standard deviation to the mean COV, RP
		Set 1	Set 2			
2 L/min	After-filter	37.3	12.5	24.9	17.5	0.70
	Ground electrode	8.8	14.1	11.5	3.8	0.33
	Bottom surface	17.4	6.4	11.9	7.8	0.65
	Collection electrode	19.7	14.7	17.2	3.5	0.21
	1st WD	5.4	3.7	4.5	1.2	0.26
	2nd WD	26.3	39.6	33.0	9.4	0.29
	3rd WD	10.3	13.5	11.9	2.3	0.19
5 L/min	After-filter	17.6	26.9	22.3	6.6	0.30
	Ground electrode	9.0	42.2	25.6	23.5	0.92
	Bottom surface	10.0	4.3	7.1	4.0	0.56
	Collection electrode	16.8	11.5	14.2	3.8	0.26
	1st WD	3.0	3.4	3.2	0.3	0.08
	2nd WD	34.6	12.8	23.7	15.4	0.65
	3rd WD	35.1	23.9	29.5	7.9	0.27
10 L/min	After-filter	8.4	19.1	13.7	7.5	0.55
	Ground electrode	7.7	5.9	6.8	1.3	0.19
	Bottom surface	5.5	15.3	10.4	6.9	0.66
	Collection electrode	15.9	15.3	15.6	0.4	0.03
	1st WD	1.5	2.4	1.9	0.6	0.33
	2nd WD	17.1	19.8	18.4	1.9	0.10
	3rd WD	44.9	52.7	48.8	5.5	0.11
				Average of all values		0.36

5. Discussion of data reproducibility

The reproducibility of the data was examined using the methodology presented in "Testing Methodology" section and by performing two separate sets of tests in triplicate with 40 μL water droplet at 2, 5, and 10 L/min sampling flow rates with 3.2 μm PSL particles. The results are presented in Table 1. Charging conditions were kept constant at 12 V/50 mA for the charging ionizer and the sampling was performed with 7 kV sampling voltage. For each set of tests, we calculated a COV for each particle deposition fraction (particles removed by the first, second, and third water droplets, remaining on the electrode or other surfaces, and collected on the after-filter), average COV as well as the RP.

As could be seen from Table 1, the value of the RP averaged for all deposition fractions was RP = 0.36. This value represents the inherent reproducibility of our measurement methodology. The achieved RP value of 0.36 implies that for a mean COV of 10% the standard deviation among COVs would be 3.6%; for the COV of 20%, the standard deviation would be 7.2%. These values indicate that our experiments were highly repeatable.

6. Conclusions

A novel bioaerosol sampler with a capacity to achieve concentration rates of over 1 million and to accrue collected particles in liquid volumes as small as 5 μL has been successfully designed and tested. The electrostatic precipitator with superhydrophobic surface (EPSS) achieves this performance by using electrostatic precipitation and special conditioning of the collection electrode. Inspired by the Lotus leaf, application of the superhydrophobic coating on the collection surface provides for highly efficient transfer of deposited particles into a liquid sample. It was found that a vast majority of the deposited particles were removed by the first applied droplet, and that much fewer particles were removed by subsequent droplets. This feature is very important for achieving high sample concentration rates. We found that the high concentration rates were maintained during sampling times as long as 60 min. At very low

bioaerosol concentration as well as for routine bio-defense monitoring applications, sampling times exceeding 1 h might be necessary. These challenges for the proposed sampling technology will be addressed in later research.

The novel bioaerosol sampling methodology presented here provides very high concentration rates with low power expenditure, which is desirable for wide-spread application of the method. In addition, in bioaerosol detection applications, where the agents are generally present in small concentrations, the EPSS would provide a highly concentrated sample as a small liquid volume which can significantly enhance our ability to detect the biological agents. Also, use of liquid sampling medium allows for sample analysis by several methods, including the polymerase chain reaction and similar ones. In this part of the project, the use of polystyrene particles allowed us to determine the optimal parameters for the sampler's operation. In future projects, these parameters will be used to analyze the sampler's performance when collecting low concentrations of airborne bacteria and fungi.

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