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EVALUATION OF PHYSICAL CAPTURE EFFICIENCY AND DISINFECTION CAPABILITY OF A NOVEL IODINATED FILTER MEDIUM

**Shanna Ratnesar, Chang-Yu Wu, Joe Wander, Dale Lundgren, Sam
Farrah, Prinda Wanakule, Matthew Blackburn and Mei-Fang Lan**

**Department of Environmental Engineering Sciences
University of Florida
412 Black Hall
Gainesville, FL 32611**

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26 Shanna Ratnesar¹, Chang-Yu Wu^{2*}, Joe Wander³, Dale Lundgren², Sam Farrah⁴,
27 Prinda Wanakule⁵, Matthew Blackburn⁶ and Mei-Fang Lan⁷
28

29 ¹Advanced Engineering Sciences, ITT Corp

30 ²University of Florida, Department of Environmental Engineering Sciences

31 ³Air Force Research Laboratory, Tyndall Air Force Base

32 ⁴University of Florida Department of Microbiology and Cell Sciences

33 ⁵University of Florida Department of Agricultural and Biological Engineering

34 ⁶University of Florida Department of Chemical Engineering

35 ⁷University of California – Davis, Department of Psychiatry and Behavioral Sciences
36

37
38 *To whom correspondence should be addressed:

39 University of Florida

40 Department of Environmental Engineering Sciences

41 406 AP Black Hall

42 PO Box 116450

43 Gainesville, FL 32611-6450

44 TEL: 352-392-0845

45 FAX: 352-392-3076

46 Email: cywu@ufl.edu

1 **Abstract**

2 A novel filter medium has been developed that combines the use of filtration and
3 iodine disinfection to provide protection against airborne pathogens. The physical
4 capture efficiency and biological disinfection capability of this iodinated resin medium
5 were evaluated. Significant physical capture efficiency (> 97%) was observed for both
6 the iodine-treated and untreated media tested, and there was no significant difference in
7 capture efficiency between them. The efficiency was greater than 99% in many cases.
8 The pressure drag was less than 10% of the glass fiber HEPA filter (0.0054 in
9 H₂O/(in/min) vs. 0.065 in H₂O/(in/min)). Biological disinfection by the medium was
10 evaluated using *Micrococcus luteus* and *Escherichia coli* vegetative bacterial cells. High
11 biological deactivation efficiency was observed (99.997%). Viable penetration through
12 the biocidal filters was observed in only 2 of 10 experiments. A near-contact mechanism
13 in which iodine is displaced from the triiodide complex is proposed to explain the higher
14 biological removal efficiency compared to the physical capture efficiency exhibited by
15 the iodinated filters. The results show that an antimicrobially augmented filter medium
16 can provide effective protection against airborne pathogens with a significantly lower
17 pressure drop than that imposed by conventional high-efficiency filtration systems.

18
19 **Keywords:** Iodine, Filtration, Bioaerosol, Disinfection, Pressure Drag

1 **Introduction**

2 As signaled by the anthrax incident in 2001, the emerging threat of bioterrorism is
3 a great concern for national security. Among the various pathways for an attack,
4 aerosolization is considered to be the most effective form to disperse biological agents to
5 the widest area in the shortest time (Kortepeter and Parker 1999). The pandemic Severe
6 Acute Respiratory Syndrome (SARS) and avian flu also clearly indicate the grave impact
7 of bioaerosols on public health. There are many technologies that can effectively remove
8 bioaerosols from the air. For example, High Efficiency Particle Air (HEPA) filters are
9 commonly used to physically remove allergens (such as pollen) from the air. The capture
10 efficiency of filters depend on several factors including the size of the challenging
11 aerosols, the filter fibers, the velocity of airflow through the filter, and the presence or
12 absence of electric charge on the fibers or particles (Hinds 1999). For respiratory
13 protection of military personnel or emergency workers, bioaerosol removal technologies
14 must meet additional requirements to be applicable. They must not exert a high demand
15 for consumable materials or dissipate large amounts of energy. They should be simple to
16 install and operate, versatile, and able to function in a wide range of conditions. They
17 should also be able to handle and not be compromised by materials commonly present in
18 battlefields or workplaces (*e.g.*, dust).

19 Another important factor in bioaerosol protection is the viability of collected
20 microorganisms. A significant fraction of airborne microorganisms remain viable after
21 collection on filtration devices. There is a great potential for microbial growth on filters
22 under favorable conditions, because of this remaining viability. Proper nutrition and
23 moisture conditions may allow microbial growth and then subsequent re-entrainment

1 from filter media. For example, molds are able to grow on fibrous media if provided with
2 70–80% relative humidity and atmospheric dust (Maus et al. 2000). Bacterial and mold
3 spores collected on air filter media can survive over prolonged periods of time and pose a
4 potential for microbial growth, especially when humidity is high ($RH > 70\%$) and filters
5 are not exposed to airflow. Abundant production and gradual release of spores into the
6 clean-air stream of the filters is likely to occur (Maus et al. 2000). Conventional
7 filtration, therefore, may not offer the best solution for respiratory protection according to
8 the reasons described above. Large pressure drop can lead to breathing difficulties and
9 subsequently reduced mobility, agility and stamina of the protected person. Air leakage
10 also increases with filter pressure drop. The inability of conventional media to disinfect
11 collected microorganisms also needs to be overcome.

12 The halogens iodine and chlorine are antimicrobial agents of great importance.
13 Halogen disinfection is a form of chemical sterilization in which oxidation of cell
14 constituents and halogenation of cell proteins occurs (Prescott et al. 2002). Iodine has
15 been used as a disinfectant for potable and on-site water treatment, and is known for its
16 stable chemical storage characteristics (Brion and Silverstein 1999). Iodine in the
17 oxidation state of zero (I_2) is not highly soluble in water but may be introduced by heat
18 vaporization, crystal dissolution, oxidation of iodide (I^{-1}) ion, and release from iodine-
19 containing resins or from the direct addition of high-strength iodine/alcohol solutions or
20 triiodide (I^{-3}) ions (Black et al. 1968). Iodine has the ability to bind to quaternary
21 ammonium anion exchange resins as tri- or penta-iodide complexes (Berg et al. 1964;
22 Brion and Silverstein 1999; Chang 1958; Taylor et al. 1970). These complexes are a

1 demand type disinfectant, releasing iodine only when needed, thus allowing for a longer
2 lifetime of product and minimizing casual exposure to iodine.

3 The use of filtration in combination with an iodinated resin has previously been
4 adopted for water disinfection aboard spacecraft. Such a resin consists of polyiodide
5 anions bound to quaternary ammonium fixed positive charges on a poly(styrene–
6 divinylbenzene) copolymer anion exchange resin (Marchin et al. 1997). A stable ionic
7 bond is formed between the iodine complex and the resin polymer, controlling the release
8 of free iodine for disinfection of microorganisms. The bound polyiodide anions release I₂
9 into water when they come in contact with suspended microorganisms, which results in
10 “devitalization” of the microorganism due to oxidation that hinders its cellular functions.

11 The use of such an iodinated resin product in combination with filtration has also
12 been proposed for the removal of bioaerosols, although there is only limited research
13 reported about the use of this product to disinfect air. According to a patent by Messier
14 (2000), such a resin can achieve high removal efficiency of microorganisms in air under
15 various conditions. Further evaluation of the iodinated resin product, however, is needed
16 to characterize its removal. The objective of this study was, hence, to appraise the use of
17 iodine in the iodine-exchanged resin for air disinfection. This was accomplished by
18 assessing the biological removal efficiency in comparison with its physical removal
19 efficiency. Its pressure drag was compared to that of a glass fiber filter. The effects of
20 flow rate and filter thickness were investigated. Two types of vegetative cells were tested
21 in addition to inorganic aerosols.

22

23

1 **Experimental Methodology**

2 The experiments were carried out in two phases. In Phase I, the physical removal
3 efficiency was evaluated using ammonium fluorescein aerosols. In Phase II, two types of
4 vegetative cells were challenged to assess the filter's biological removal efficiency. The
5 same experimental system was used in both phases.

6

7 ***Experimental Set-up***

8 Figure 1 shows the schematic of the experimental set-up. Aerosols were
9 generated using a six-jet Collison nebulizer (Model CN25, BGI Inc.) at a flow rate of
10 10 Lpm for Phase I experiments. Filtered compressed air passing through a diffusion
11 dryer was used as dilution air and was introduced into the dilution dryer chamber at twice
12 the amount of the aerosol flow to evaporate the water content of the droplets (May 1972).
13 An excess airflow point was used to control the flow going through the target filter at the
14 designated level (13, 15, and 21 Lpm) while maintaining the flow rate of 28.3 Lpm
15 required for the cascade impactor. For the bioaerosol experiments, 7 Lpm of air was used
16 for the nebulizer, and dilution air was 13 Lpm. Additional air was introduced
17 downstream of the dryer to maintain 15 Lpm passing through the filter. Relative
18 humidity and temperature downstream of the dilution dryer were monitored (HX 94,
19 Relative Humidity/Temperature Transmitters, Omega Engineering). The aerosols were
20 then passed through the test filter (or empty filter holder in the control runs). A
21 Magnehelic gage reading 0–10 in H₂O was employed to evaluate the pressure drop across
22 the resin/iodine filters. A reading was taken every minute for each run. Pressure drop is
23 due to the resistance to airflow across a filter. Penetrating aerosols leaving the test filters

1 were captured and classified by particle size on a six-stage Andersen viable impactor
2 (Model #10-820). All the flow rates were controlled by pre-calibrated rotameters.

3

4 ***Test Particles***

5 In Phase I, ammonium fluorescein particles were employed to evaluate the
6 physical capture efficiency of the filters due to the lower detection limit of fluorescence
7 compared to gravimetric measurements. A 6.75-g/L fluorescein solution in 0.1 N NH₄OH
8 was aerosolized by the Collison nebulizer. This concentration was chosen to allow for the
9 production of larger particles and enhanced detection. The mass median diameter of the
10 dry fluorescein particles was calculated to be ~ 0.27 μm, based on the following equation
11 (Hinds 1999):

$$12 \quad d_a = d_d (F_v)^{1/3} \quad (1)$$

13 where d_d is the mass median diameter of the atomized droplet (~ 3 μm; (May 1972) and
14 F_v is the volume fraction of fluorescein in the solution.

15 In Phase II, microorganisms for bioaerosol challenges were selected based on
16 several factors. Nonpathogenic representatives of possible biological weaponry agents
17 were employed. A commonly known airborne pathogen is *Yersinia pestis*, the organism
18 that causes bubonic plague (Morris and Sandana 2005). As a representative for this
19 organism, *Escherichia coli* was utilized. *E. coli* is a Gram-negative rod-shaped bacterium
20 that ranges in size from 2 to 3 μm in length and 0.25 to 1 μm in diameter. The strain used
21 was obtained from the Water Reclamation Facility in the Department of Environmental
22 Engineering Sciences at the University of Florida. The samples obtained were inoculated
23 and maintained on Difco tryptic soy agar and grown at 33°C prior to sampling.

1 *Micrococcus luteus* is another frequently used representative bioaerosol (Agranovski et
2 al. 2003; Li and Lin 2001; Wake et al. 1997). *M. luteus* cells are Gram-positive, non-
3 motile, nonsporulating, round bacteria normally found in clusters or tetrads. The
4 individual cells are 0.9 to 1.8 μm in diameter (Wake et al. 1997). *M. luteus* samples were
5 obtained from the University of Florida, Department of Microbiology and Cell Sciences.
6 Cells were inoculated on standard nutrient agar (Difco 0001) and maintained at 33 °C
7 prior to sampling. Prior studies (Crook et al. 1998; Wake et al. 1997) have shown that
8 Ringer's solution can successfully maintain the viability of stressed bacteria used in
9 aerosol studies. Hence, bioaerosol suspensions for use in the Collison nebulizer were
10 created by washing bacterial cells off agar slants using 1 mL of 25% Ringer's Solution
11 (Fisher, S77939). The slants were agitated for 20 seconds using a standard vortex, and
12 varying amounts of each sample were aseptically pipetted out of the slant test tubes and
13 into Ringer's solution contained in the nebulizer reservoir for each experiment.

14

15 ***Test Filter***

16 Filter media were provided by Triosyn Corp. through the U.S. Air Force Research
17 Laboratory. The iodinated resin is produced by thermally fusing pure iodine crystals with
18 a quaternary anion exchange polymer under high pressure. Iodine-treated and untreated
19 filters of 1 mm thickness were tested for physical capture efficiency and to evaluate
20 whether differences in morphology due to the iodine treatment would affect capture
21 efficiency. Heavier filters of 2 mm thickness were used to evaluate the effect of depth.
22 The bioaerosol experiments were conducted using medium-depth filters with an
23 approximate thickness of about 1.5 mm.

1 The filter media tested were 47 mm in diameter (area 17.35 cm²). A common
2 respirator cartridge has a cross-section area of approximately 100 cm² and a nominal
3 breathing rate is about 85 Lpm (Di Ionno and Messier 2004). Accordingly, the flow rate
4 used for testing the 47-mm filter was scaled down to 15 Lpm to produce a similar face
5 velocity. Two other flow rates, 13 and 21 Lpm, were used to evaluate the effects of flow
6 velocity. Bioaerosol challenges were conducted only at 15 Lpm air flow rate across filter
7 surfaces.

8

9 ***Experimental Procedure***

10 Phase I

11 Each experiment was run for 15 minutes. This amount of time was shown to be
12 sufficient to deliver a measurable amount of fluorescent particles for evaluation, while
13 not causing a mound effect due to accumulation of particles under each impactor jet,
14 which might alter the collection characteristics of the Andersen impactor. After
15 collection, the individual stages containing fluorescein particles were rehydrated in
16 aqueous 0.1 N NH₄OH solution. They were then treated with 20 mL of methylene
17 chloride to dissolve the grease coating on each plate and sonicated for 10 minutes.
18 Twenty mL of 0.1 N NH₄OH was added to each sample after sonication and poured into a
19 test tube. Due to the immiscibility of methylene chloride and ammonium hydroxide, a
20 fluorescein–ammonium hydroxide solution separated to the top of each sample while the
21 methylene chloride, containing the grease, sank to the bottom. The top layer of each
22 sample was then pipetted into a quartz cuvette for analysis (Vanderpool et al. 1987).

1 Mass concentrations of fluorescein solution from each stage were measured using
2 a Sequoia–Turner 112 Digital Filter Fluorometer (G.K. Turner Associates). A calibration
3 of concentration vs. fluorometer reading was performed using samples of known
4 fluorescein concentration in 0.1 N NH₄OH for fluorometer range settings of 1X, 3X, 10X,
5 and 30X. Fluorometer readings less than 30 and higher than 100 were considered to be
6 less reliable and therefore were not used in this study. Accuracy was maintained by
7 selecting range settings that best suited the concentration of the solution being analyzed.

8 Several factors may skew the results of fluorometric analysis, including
9 fluorescence given off by reagents, filter substances reacting with chemicals, or flaws
10 within different cuvettes used in analysis. For this reason, several background tests for
11 the various filter media were conducted. The results indicate that there was no
12 interference with fluorescence analysis by the glass fiber and untreated filters. Variations
13 in the optical properties of each cuvette were also shown to have negligible effect on data
14 reproducibility. A maximum standard deviation of only 0.8% was observed at 500 µg/L
15 based on 36 fluorometric readings of different cuvettes oriented at different directions
16 and at differing concentrations. However, a negative interference was observed for the
17 iodine-treated filters. The interference may be attributed to chemical reactions occurring
18 between the iodine and fluorescein in the rehydrated filter solution, and it can result in a
19 lower apparent concentration. A concentration-based interference curve was therefore
20 established to adjust the values measured.

21
22
23

1 Phase II

2 To measure the inlet concentration of bioaerosols entering the test system vs.
3 those captured by filtration, two impactors were used in parallel. One impactor contained
4 no filter upstream, whilst the other contained an iodine-treated filter upstream. The inlet
5 concentrations were measured for the first and last five minutes of each experiment using
6 Petri dishes on all six stages of the impactor. Due to the very low outlet concentration or
7 penetrating concentration, the bioaerosol was collected on only one Petri dish on stage six
8 (last stage). Every 20 minutes throughout the 2 h run, the outlet Petri dishes were
9 changed out to prevent desiccation of the agar surface. After each plate was removed
10 from the impactors, it was labeled and placed in an incubator at 33 °C for 24–36 h. It was
11 noted that growth of colonies on the surface of the impaction zone was in the same
12 pattern as in the Phase I experiments. The optical count of the number of positive holes in
13 each sample was corrected for the number of colony-forming units (cfu) that impacted
14 onto an agar surface following the positive hole method (Thermo Electron Corp., 2003).

15

16 *Statistical Analysis of Data*

17 Three controls (with no filters upstream) and three experimental runs (with filters
18 upstream) were conducted separately for each flow rate tested. The least squares method
19 was used to correlate the data (Vining 1998). This method considers the data points
20 assessed during the experiments and finds the best straight-line equation to represent all
21 combinations of the data. The x -axis represents the control mass fraction measured for
22 each stage for this analysis, and the y -axis represents the difference between the control
23 mass fraction and the penetration mass fraction for each stage. The efficiencies reported

1 were the slopes calculated based on the least squares methods, and the error ranges
2 reported were the standard errors for the y estimates.

4 ***Filter Morphology Analysis***

5 Microscopic images of the filters before and after sampling were taken using
6 scanning electron microscopy (SEM) to evaluate any differences in morphology due to
7 treatment or use of the filters. Elemental analysis of objects magnified with SEM was
8 performed using energy dispersive X-ray (EDX) analysis.

10 **Results and Discussions**

11 ***Morphology Analysis of Filter Media***

12 Iodine-untreated filters were analyzed via SEM prior to experimentation (Figures
13 2a and 2b). A dense woven structure of long fibers of different thickness was observed
14 by visual analysis. Iodine-treated filters were also analyzed via SEM prior to
15 experimentation (Figures 2c and 2d). Small dark flecks were observed around the outer
16 perimeter of the filter surfaces. EDX analysis was used upon magnification to determine
17 the elemental composition of fibers (Figure 2c) and black specs (Figure 2d) seen in the
18 SEM photographs. The EDX analysis of the small flecks indicated the presence of iodine
19 (Figure 3a). Similar results were observed for the EDX taken of the filter fibers,
20 demonstrating the presence of iodine on the fiber surface.

22 ***Phase I – Physical Capture***

1 The mass size distribution of particles produced in the system reaching the point
2 of filtration was determined during control runs using no filter upstream of the impactor.
3 Figure 4 shows the size distribution at 15 Lpm as an example. The shape of the
4 distribution of other flow rates had a similar pattern, while the total mass increased as the
5 flow rate increased. The majority of the fluorescent particles were collected on the
6 downstream filter stage ($<0.65 \mu\text{m}$) and 5th and 6th stages ($2.1\text{--}1.1$ and $1.1\text{--}0.65 \mu\text{m}$,
7 respectively) for each flow rate. The data for stages 1 to 4, however, were not used due
8 to the low concentrations of detectable particles. The mean corresponding total mass
9 concentrations were 1.33×10^7 , 2.14×10^7 , and $4.03 \times 10^7 \mu\text{g}/\text{m}^3$ for 13, 15, and 21 Lpm,
10 respectively.

11 Table 1 summarizes the physical capture efficiency at each stage of the impactor
12 for iodine-treated and untreated filters at different flow rates. Figure 4b shows, as an
13 example, the mass size distribution downstream from an iodinated resin filter. As shown,
14 significant capture (greater than 97%) was observed for both the iodine-treated and
15 untreated filters tested for stages 5 and 6 and for the downstream filter (DF). Filters were
16 visually analyzed after sampling and rehydration. It was observed that some amount of
17 fluorescein was not captured by the upstream iodinated resin filter. No fluorescence was
18 observed on the downstream filters when similar experiments were performed using glass
19 fiber filters upstream, indicating that the glass fiber filters have higher removal efficiency
20 for fluorescence particles than the iodinated resin filters do. Rehydrated solutions in test
21 tubes were also visually analyzed prior to pipetting into cuvettes. No visible fluorescence
22 was observed when glass fiber filters were located upstream, whereas visible
23 fluorescence could be seen from the iodinated resin samples.

1 Removal efficiency greater than 99.8% was observed for the 1.1–2.1 μm particles
2 for all flow rates tested. The efficiency decreased to less than 99% for particles smaller
3 than 0.65 μm and to 96.84% for the iodine-treated filters tested at 15 Lpm. Impaction is
4 the most important mechanism for the larger particles (1.1–2.1 μm). Hence, increasing
5 the flow velocity (21 Lpm vs. 13 Lpm) resulted in higher efficiency. Conversely, the
6 dominant capture mechanism for smaller particles (less than 0.65 μm) is diffusion. A
7 higher flow rate resulted in shorter retention time for diffusion and consequently lower
8 collection efficiency. The highest capture efficiency was generally observed in the
9 1.1–2.1 μm range, and efficiency decreased as the particle size decreased. Both treated
10 and untreated filters appeared to perform similarly based on the data, which showed no
11 significant difference. The thicker filters (2 mm) appeared to perform better than the
12 regular iodine-treated filters at 15 Lpm. This was expected, due to the increased
13 possibility for impaction and longer retention time leading to greater diffusion of particles
14 to fiber surfaces. However, the improvement was not huge because the thin filters
15 (1 mm) already had exceptional capture efficiency.

16 Pressure drop across the filters was recorded. The system was tested with and
17 without the use of the aerosolized particles to determine how particle accumulation on the
18 filter affected pressure drop. A pressure drop of 0.2 in H_2O was observed when no filter
19 was inserted into the filter holder, which was attributed to the wire mesh backing that was
20 placed behind the filter for support. The initial pressure drop when the filter (1 mm
21 thickness) was placed was 1.8 in H_2O at 15 Lpm and 2.3 in H_2O for 21 Lpm. It then
22 increased as the particles flowed through the testing filters and were subsequently
23 captured.

1 Pressure drag (S) is a measure of the filter's aerodynamic resistance to air flow
2 and can be calculated by dividing pressure drop across a filter (Δp) by filtration velocity
3 (U) as (Noll 1999):

$$4 \quad S = \frac{\Delta p}{U} \quad (2)$$

5 It is worthwhile noting that the initial pressure drag of the iodinated filters was
6 significantly less than the associated pressure drag of the glass fiber filters tested,
7 0.0054 in H₂O/(in/min) vs. 0.065 in H₂O/(in/min). Lower filter drag is associated with
8 less-labored respiration, which is beneficial when the mobility of the protected person is
9 critical. Another useful criterion for comparing different types of filters is filter quality,
10 q_F (Hinds 1999),

$$11 \quad q_F = \frac{\ln(1/P)}{\Delta p} \quad (3)$$

12 where P is the aerosol penetration through the filter. The iodinated filter's filter quality
13 was 9.2 kPa⁻¹, which is higher than the glass fiber's value, 1.8. It again supports that the
14 iodinated filter exhibits better quality.

15

16 ***Phase II — Biological Disinfection***

17 Two types of bacteria were evaluated for the bioaerosol challenges, *E. coli* and *M.*
18 *luteus*. The size distributions of colony-forming units detected at the inlet are shown in
19 Figure 5. As shown, the majority of the bioaerosol particles generated were detected in
20 the 1.1–2.1 μm range, which agrees with the nominal sizes reported in the literature. The
21 overall disinfection efficiency was 99.997 ± 0.004% for *M. luteus* and 99.998 ± 0.005%
22 for *E. coli*. It should be noted that penetration was detected in only two of 10

1 experiments. These results indicate that the iodinated resin filters cause close to a 5-log
2 average removal of bioaerosols when tested using the methods discussed in this study.
3 The similar efficiencies for both species also suggest that the mechanism works for both
4 Gram-positive and -negative species.

5 In addition, disinfection removal efficiency was approximately 2 logs higher than
6 the physical efficiency (99.8% physical capture efficiency at 15 Lpm, Table 1). This
7 removal efficiency increase may be explained by reaction with gaseous iodine present in
8 the filtration system. However, measurements of I₂ vapor downstream applications of
9 similar iodinated polymer materials showed low iodine vapor concentration (< 0.2
10 mg/m³; OSHA TLV is 1 mg/m³) (Di Ionno and Messier 2004; Di Ionno et al. 2001).

11 Another possible explanation is near-contact transfer of I₂ as the bacteria pass near the I₃⁻
12 complex. Figure 6 displays the concept. Bacteria are almost universally anionic at their
13 surface. When they fly in proximity to the I₃⁻ complex on the polymer surface, the charge
14 on the microbe surface displaces the I⁻ ion and captures the I₂ molecule. The I₂ molecule
15 then reacts with an iodlatable group on the microbe. Thus, disinfection can occur near
16 but without direct contact with filter surface. Confirmation of this hypothesis, however,
17 requires further investigation.

18 The initial pressure drag of the 1.5-mm thickness filter was 0.006 in
19 H₂O/(in/min), which is still less than 10% of that of a glass fiber HEPA filter. The filter
20 quality based on the biological removal was 19.9 kPa⁻¹. The high biological disinfection
21 efficiency, low pressure drag and high filter quality together demonstrate that the
22 iodinated filter offers a superior alternative to conventional HEPA filtration for removing
23 biological agents.

1 Combinations of SEM with EDX were used to evaluate filters after experimental
2 runs with *M. luteus* suspended in 25% Ringer's solution. Figures 2e and 2f display their
3 images. Particles of various size and shape were observed in contact with the filter
4 fibers. The filter fibers appear to be coated in a white substance. Also notable is the
5 absence of the large, dark specs observed in the filters prior to experimentation. The
6 EDX analysis (Figure 3b) verified that the particles were primarily composed of sodium
7 and chlorine, indicative of the Ringer's solution used in the experiment. The coating is
8 expected to have no influence on the viability of penetrating microorganisms. It is,
9 however, not clear what impact—if any—the coating had on the viability of those cells
10 collected on the filter. It should be noted that the Ringer's solution is necessary to keep
11 the microorganisms alive during the experiments, since pure water lyses the cells. Thus,
12 it is unlikely that the undesired presence of the materials can be avoided in the testing.

13 The viability of those cells captured on the filter is also of interest. Unfortunately,
14 this could not be verified because the retrieval of those collected microorganisms requires
15 the use of water. Iodine released into the water can easily disinfect those cells as reported
16 in numerous studies (Berg et al. 1964; Black et al. 1968; Marchin et al. 1997), thus
17 yielding no viable cells for verification. Nevertheless, from the low level of penetration,
18 it can be expected that the possibility of survival on the filter is rather low.

19 It is well known that many microorganisms aggregate. There are concerns
20 whether aggregation would provide shielding protection to those hiding in the core. This
21 is likely not to be an issue for vegetative cells in the iodinated resin filter system. Large
22 aggregates of vegetative cells are at least a few micrometers, and hence they will be very
23 efficiently captured by the physical mechanism. Gas-phase or captured iodine can also

1 penetrate the aggregates to reach the bacteria inside. Thus, aggregated vegetative cells
2 pose no greater threat when such a filtration system is employed. The same statement,
3 however, may not be extended to virus particles. Virus aggregates may still well be in
4 the submicrometer range, where the collection efficiency is lower.

5
6 **Conclusions**

7 In this study, the physical capture and biological disinfection efficiency of a novel
8 biocidal filter medium were evaluated. Significant capture (greater than 97%) by the
9 filters was observed for a wide particle size range. In most cases the efficiency was
10 greater than 99%. Efficiency was the highest for larger particles (1.1–2.1 μm), and it
11 increased slightly as flow velocity increased, since impaction is the main collection
12 mechanism for this size range. Efficiency was the lowest for smaller particles (less than
13 0.65 μm , collected in the downstream filter), and it decreased as flow velocity increased
14 since diffusion is more important for smaller particles. No discernible difference in the
15 physical capture efficiency between the iodine-treated and untreated filters suggests no
16 mechanical impact of the treatment. The iodinated resin filters were not as efficient at
17 physical removal of aerosols as the glass fiber HEPA filter; nonetheless, the pressure drag
18 was 90% less and the filter quality for biological removal was higher. Enhanced capture
19 efficiency can be easily achieved by using a thicker filter while still being in the
20 acceptable range of pressure drop.

21 Two types of microorganisms were used in the bioaerosol challenge experiments,
22 *M. luteus* and *E. coli*, which were dominantly in the 1.1–2.1 μm aerodynamic range.
23 There was no difference in removal efficiency between these two species representing
24 Gram-positive and -negative bacteria; the average efficiency was approximately

1 99.997%, or close to 5-log removal of bioaerosols. Indeed, only two of 10 experiments
2 performed had detectable penetration through the iodine-treated filters. This efficiency
3 showed much greater biological removal than was physically established. This may be
4 indicative of the presence of gaseous iodine within the filtration system or downstream of
5 the filters or, as we propose, of a direct mechanism involving capture of I₂ from the
6 medium. Whether the bacterial cells collected on the filter were viable could not be
7 verified due to the well known disinfection capability of iodine in water. Nonetheless,
8 the low penetration fraction of viable bioaerosols implies an extremely low survival
9 fraction, if any. The high biological disinfection capacity combined with the low
10 pressure drag and high filter quality demonstrates the novel filter medium to be a superior
11 alternative to conventional filtration for the removal of micrometer bioaerosols. Its
12 application to much smaller virus aggregates, however, needs to be investigated. The
13 effective lifetime should also be determined.

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15
16

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17
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Table 1 Physical Capture Efficiency of Iodinated Resin Filters per Stage for 1 mm thick filters

Sample, Flow rate	Stage# (Size range, μm)				Total (2.1-0.03)
	5 (2.1-1.1)	6 (1.1-0.65)	DF (0.65-0.03)	Total (2.1-0.03)	
Treated @ 13 Lpm	99.86 \pm 0.0111	99.96 \pm 0.0292	99.22 \pm 0.167	99.34 \pm 0.118	
Untreated @ 13 Lpm	99.91 \pm 0.00447	99.79 \pm 0.0263	99.12 \pm 0.184	99.62 \pm 0.830	
Treated @ 15 Lpm	99.81 \pm 0.00509	99.21 \pm 0.0464	96.84 \pm 0.0634	97.32 \pm 0.381	
Untreated @ 15 Lpm	99.89 \pm 0.0328	99.87 \pm 0.0234	99.38 \pm 0.101	99.43 \pm 0.0826	
Treated @ 21 Lpm	99.99 \pm 0.000308	99.51 \pm 0.0199	98.85 \pm 0.149	99.01 \pm 0.0877	
Untreated @ 21 Lpm	99.93 \pm 0.0320	99.53 \pm 0.0308	98.95 \pm 0.263	99.07 \pm 0.191	
Thick* Treated @ 15 Lpm	99.94 \pm 0.00276	99.97 \pm 0.00191	99.50 \pm 0.126	99.56 \pm 0.0904	

* 2 mm

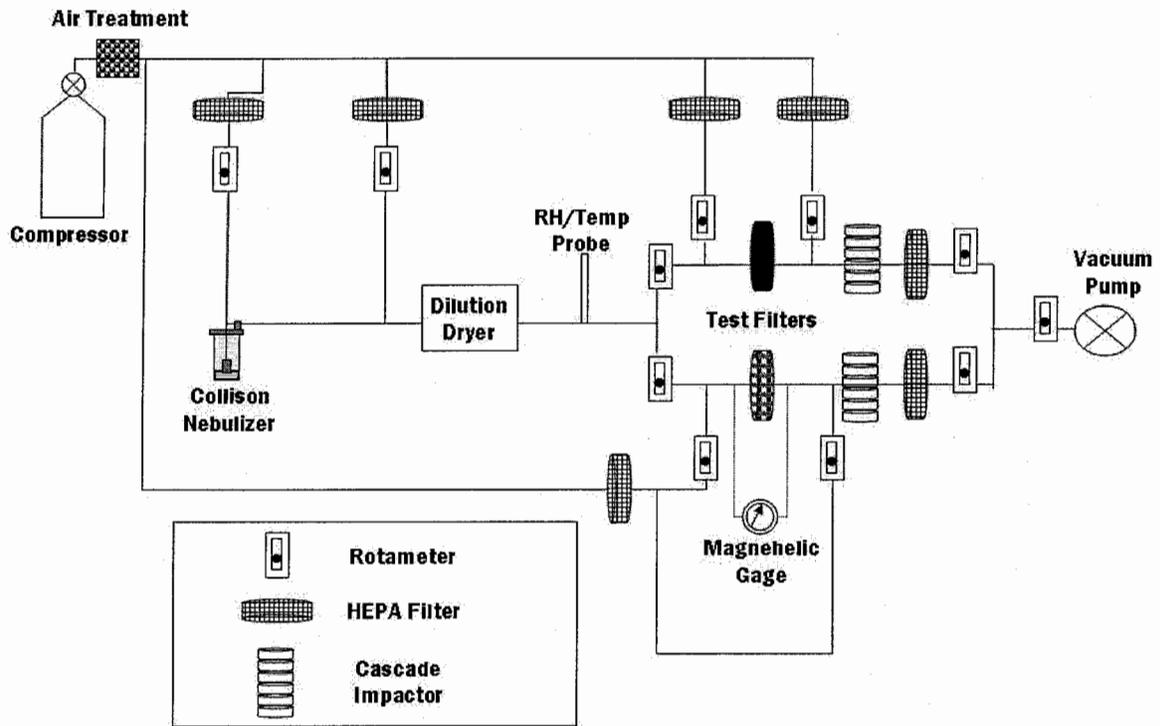
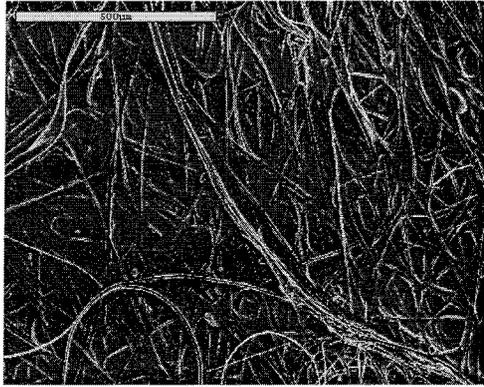
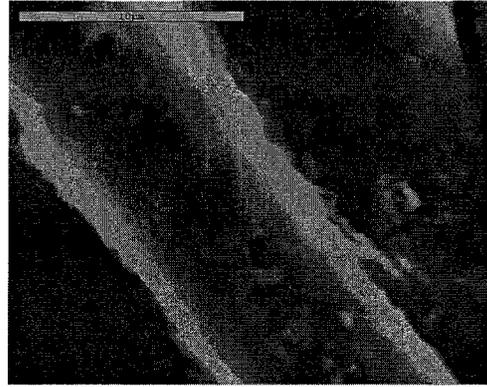


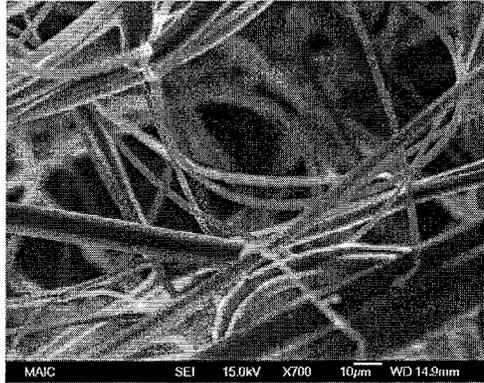
Figure 1 Experimental Set-Up



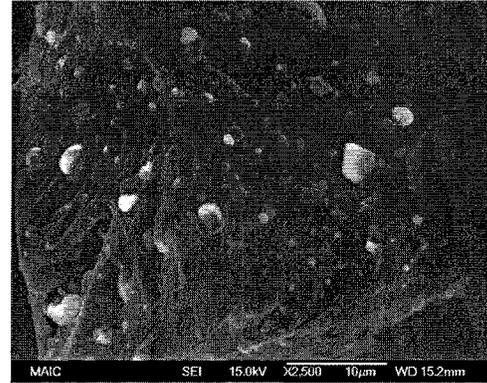
(a)



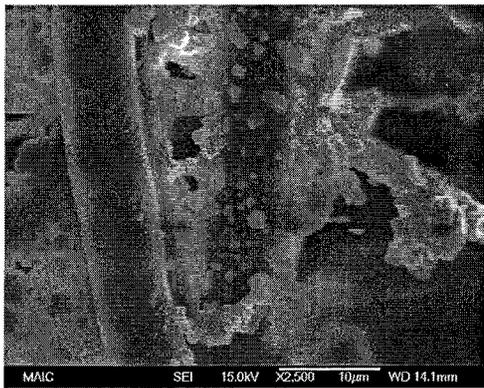
(b)



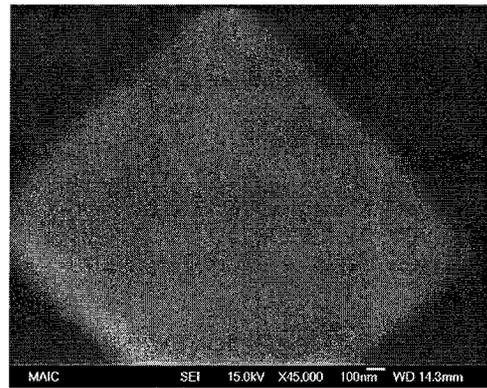
(c)



(d)

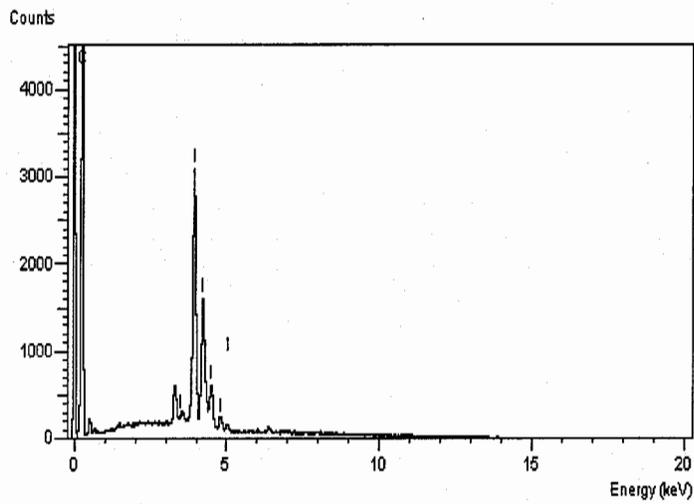


(e)

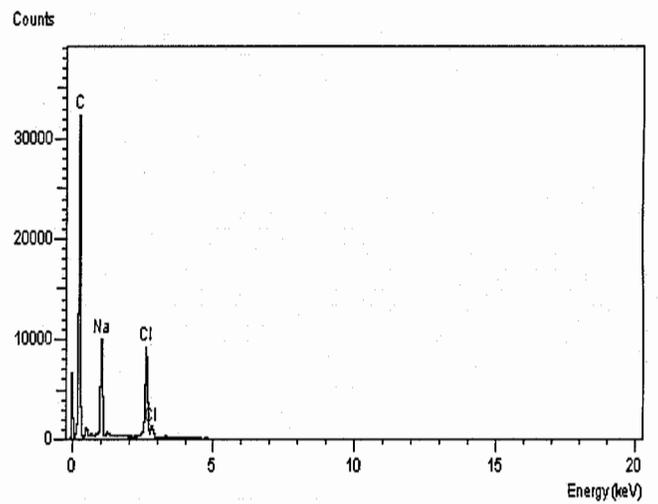


(f)

Figure 2 SEM images of untreated filters (a) 5100X, (b) 54000X; SEM images of fresh iodine treated filters: (c) fibers at 700x, and (d) enlarged fleck at 2500X; SEM images of used filters: (e) 2500X, and (f) enlarged particle at 45000X



(a)



(b)

Figure 3 EDX spectrum: (a) enlarged fleck on fresh iodine-treated filter (Figure 2d), (b) enlarged particle on used filter (Figure 7(f))

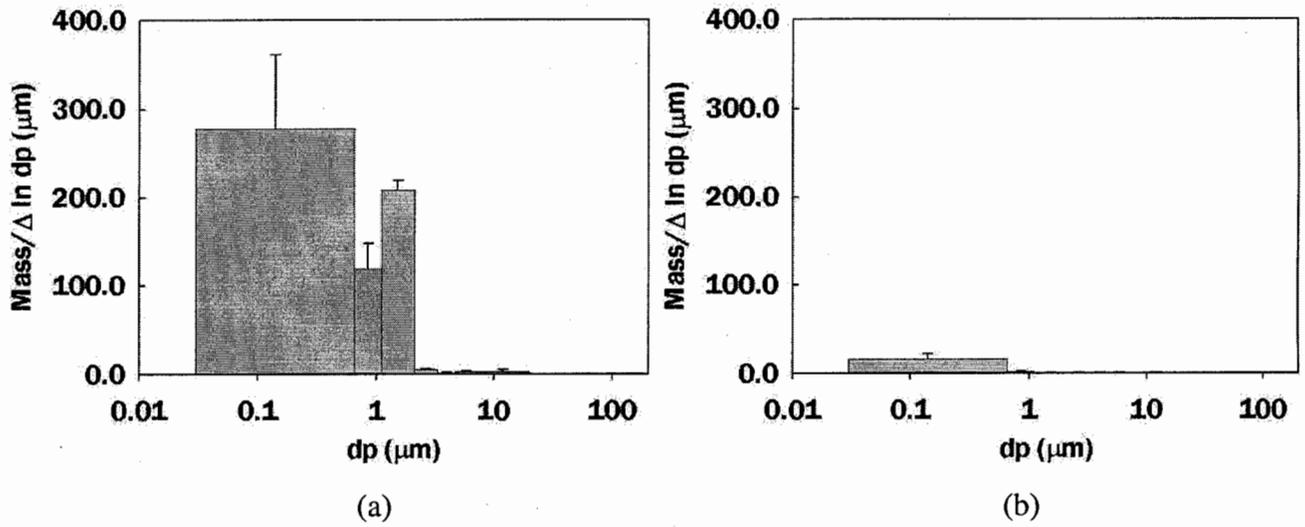


Figure 4 Mass size distribution of ammonium fluorescein particles for Phase 1 at 15 Lpm: (a) control experiments, (b) with an iodinated filter upstream

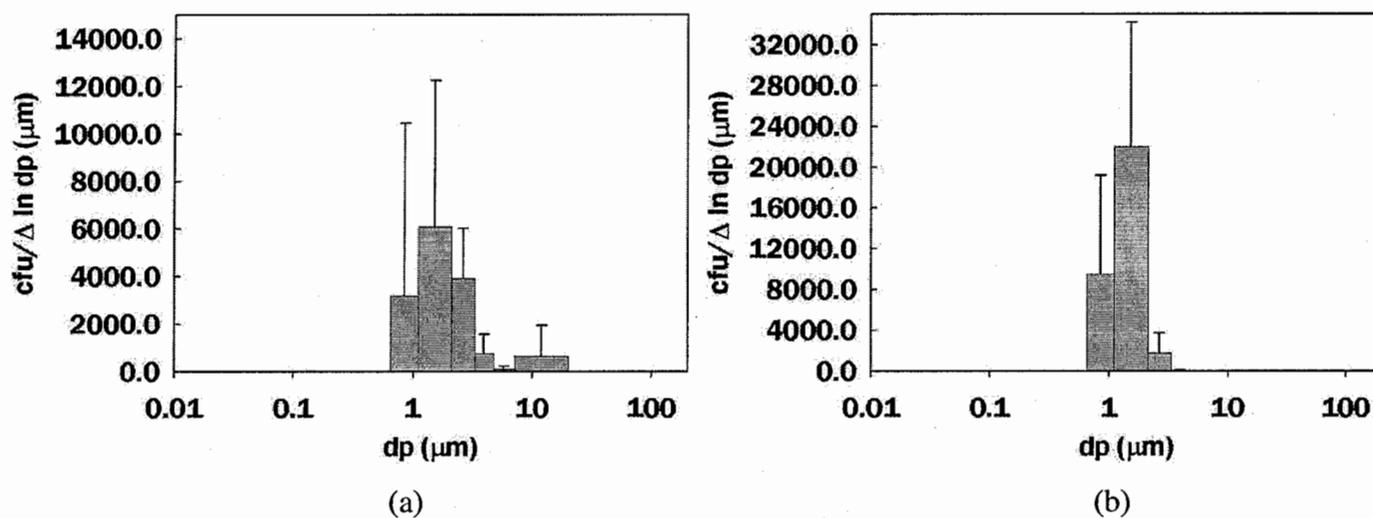


Figure 5 Size distribution of bioaerosols generated at 15 Lpm for (a) *M. luteus*, and (b) *E. coli*

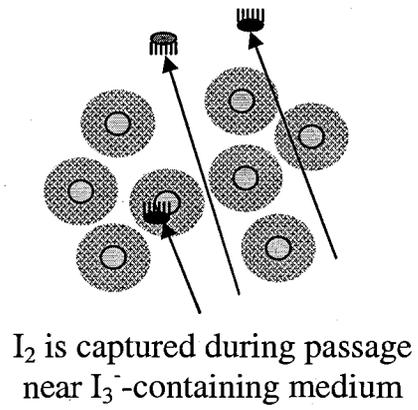
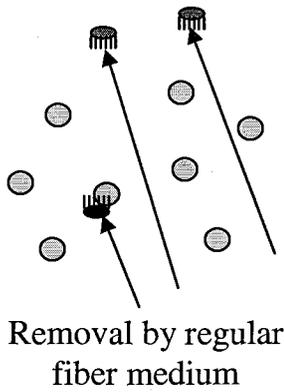
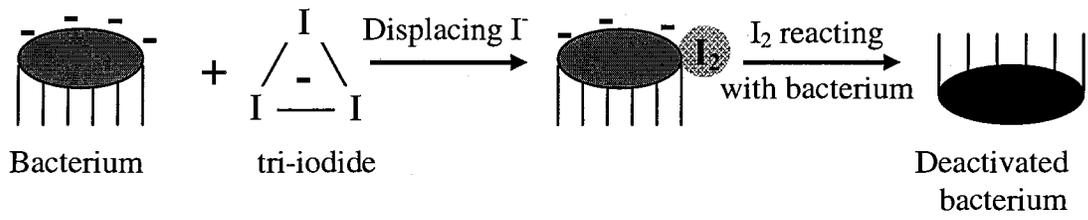


Figure 6 Conceptual schematic of disinfection near the iodinated polymer

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