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Nanoparticle Penetration through Filter Media and Leakage through Face Seal Interface of N95 Filtering Facepiece Respirators

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National Institute for Occupational Safety and Health recommends the use of particulate respirators for protection against nanoparticles (<100 nm size). Protection afforded by a filtering facepiece particulate respirator is a function of the filter efficiency and the leakage through the face-to-facepiece seal. The combination of particle penetration through filter media and particle leakage through face seal and any component interfaces is considered as total inward leakage (TIL). Although the mechanisms and extent of nanoparticle penetration through filter media have been well documented, information concerning nanoparticle leakage through face seal is lacking. A previous study in our laboratory measured filter penetration and TIL for specific size particles. The results showed higher filter penetration and TIL for 50 nm size particles, i.e. the most penetrating particle size (MPPS) than for 8 and 400 nm size particles. To better understand the significance of particle penetration through filter media and through face seal leakage, this study was expanded to measure filter penetration at sealed condition and TIL with artificially introduced leaks for 20–800 nm particles at 8–40 l minute volumes for four N95 models of filtering facepiece respirators (FFRs) using a breathing manikin. Results showed that the MPPS was ∼45 nm for all four respirator models. Filter penetration for 45 nm size particles was significantly ($P < 0.05$) higher than the values for 400 nm size particles. A consistent increase in filter penetrations for 45 and 400 nm size particles was obtained with increasing breathing minute volumes. Artificial leakage of test aerosols (mode size ∼75 nm) through increasing size holes near the sealing area of FFRs showed higher TIL values for 45 nm size particles at different minute volumes, indicating that the induced leakage allows the test aerosols, regardless of particle size, inside the FFR, while filter penetration determines the TIL for different size particles. TIL values obtained for 45 nm size particles were significantly ($P < 0.05$) higher than the values obtained for 400 nm size particles for all four models. Models with relatively small filter penetration values showed lower TIL values than the models with higher filter penetrations at smaller leak sizes indicating the dependence of TIL values on filter penetration. When the electrostatic charge was removed, the FFRs showed a shift in the MPPS to ∼150 nm with the same test aerosols (mode size ∼75 nm) at different hole sizes and breathing minute volumes, confirming the interaction between filter penetration and face seal leakage processes. The shift in the MPPS from 45 to 150 nm for the charge removed filters indicates that mechanical filters may perform better against nanoparticles than electrostatic filters rated for the same filter efficiency. The results suggest that among the different size particles that enter inside the N95 respirators, relatively high concentration of the MPPS particles in the breathing zone of respirators can be expected in workplaces with high concentration of nanoparticles. Overall, the data obtained in the study suggest that good fitting

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respirators with lower filter penetration values would provide better protection against nanoparticles.

Keywords: face seal leakage; filter penetration; nanoparticles; N95 filtering facepiece respirator; total inward leakage

INTRODUCTION

Recent advances in technology have produced engineered nanoparticles for unique applications. A nanoparticle is defined as a nano-object with all three dimensions in the size range from ~1 to 100 nm (ISO/TS, 2009). Many nanoparticles exhibit distinctive properties different from the common form of the materials. Nanoparticles with novel and dynamic functionality are engineered into multifaceted substances. Several types of nanomaterials are produced in powder form and particles could be released into the air during various processes, including handling, packaging, and disposal. Recent studies have reported the occurrence of high concentration of nanoparticles in some industrial settings (Bello *et al.*, 2009; Johnson *et al.*, 2010). This indicates that workers, if unprotected, could inhale nanoparticles when generating/handling these materials. Inhalation of nanoparticles has been shown to produce adverse effects on pulmonary and systemic functions in exposed animals (Aitken *et al.*, 2004; Byrne and Baugh, 2008; Seaton *et al.*, 2009; Shvedova *et al.*, 2009; Brooks *et al.*, 2010). When engineering and administrative controls are not enough or are unavailable, respiratory protective devices are recommended to reduce the inhalation of harmful particles.

Protection afforded by a particulate respirator is a function of the filter efficiency and any leakage through the face-to-facepiece or component interfaces. National Institute for Occupational Safety and Health (NIOSH) certification of particulate respirators includes filter penetration tests with NaCl aerosols for N-series respirators and dioctyl phthalate aerosols for R- and P-series respirators (NIOSH, 1995). These methods measure particle penetration through filter media of respirators sealed to a test system. Many studies have reported filter penetration values less than the NIOSH maximum allowable levels during testing of filtering facepiece respirators (FFRs) (Qian *et al.*, 1998; Martin and Moyer, 2000; Rengasamy *et al.*, 2007, 2009). On the other hand, some studies have reported higher penetration levels for FFRs when testing with monodisperse aerosols (Martin and Moyer, 2000; Balazy *et al.*, 2006; Eninger *et al.*, 2008). To address the concern for the penetration of smaller size nanoparticles, NIOSH-approved N95 and P100 and European CE-marked FFP1, FFP2, and FFP3 FFRs were tested with <30 nm size particles (Huang *et al.*, 2007; Rengasamy *et al.*, 2008, 2009). Results from these studies showed that nanoparticles <30 nm were efficiently filtered by the FFRs as predicted by the filtration theory with no thermal rebound for particles down to 4 nm.

While filter penetration has been the focus of much research, leakage has received comparatively less attention. Many studies have reported that a respirator wearer must have a good fit that can reduce face seal leakage of particles to achieve higher levels of respiratory protection (Coffey *et al.*, 1998; Zhuang *et al.*, 2003; Reponen *et al.*, 2011). Factors including work activity, breathing rate, and head and body movements influence particle entry through face seal leaks. To address these issues, several studies have measured particle leakage through artificially introduced leaks in the respirator using a manikin or a test subject. Results from a manikin study showed that leakage was strongly dependent on particle size (Hinds and Kraske, 1987). These authors also concluded that the aerosol size distribution inside the respirator will be significantly different from that outside the respirator. Particle leakage was also studied using a human test subject wearing a half-mask respirator with different size and shape holes (Holton *et al.*, 1987). Results showed a decrease in particle leakage with increasing particle diameter from 1.0 to 4.4 µm as well as with decreasing particle size from 0.20 to 0.07 µm, which were attributed to increased inertial losses with increasing particle size and increased diffusional losses with decreasing particles size, respectively.

Nanoparticle leakage through face seal area of respirators, however, has been sparsely reported. Recently, particle penetration through filter media and face seal leakage pathways was studied using a manikin and human test subjects (Grinshpun *et al.*, 2009). Penetration data obtained with human test subjects and manikin were compared for different size particles. Highest penetration values were obtained for particles with a geometric mean diameter (aerodynamic diameter) of ~78 nm size through the face seal leakage of subjects tested with N95 FFRs and surgical masks. Results showed that the number of particles passing through face seal leakage far exceeded the number of particles penetrating through the filter media. The authors also confirmed that head and body movement had a pronounced effect on the relative contribution of the two penetration pathways. Similarly, protection factor (PF), a ratio of particle concentration outside to inside of a respirator, measured for N95 FFRs worn by test subjects was less for particles ~78 nm than those values obtained for smaller as well as larger size particles indicating higher leakage through respirator sealing area (Lee *et al.*, 2008; Reponen *et al.*, 2011).

This paper is a continuation of previous work in our laboratory, which measured the total inward leakage (TIL) for a wide size range of particles for N95 model FFRs with artificial leaks using a manikin (Rengasamy and Eimer, 2011). The results showed higher TIL values for 50 nm most penetrating particle size (MPPS) particles than for 400 nm size particles suggesting that higher concentration of nanoparticles relative to smaller and larger size particles could occur inside the respirators in nanoparticle workplaces. To gain insight into particle penetration through filter media and leakage through face seal interface, the study was expanded to include TIL measurements for four N95 model FFRs with different filter penetration characteristics. Filter penetration and TIL values for 20 to 800 nm range NaCl particles were measured using FFRs sealed to a manikin with different leak hole sizes and breathing flow rates. Filter penetration and TIL values obtained for the most penetrating particles at different leak sizes and minute volumes were compared with those values obtained for 400 nm size particles. The influence of filter penetration and leakage on TIL values for nanoparticles at different flow rates and leak sizes is also discussed.

MATERIALS AND METHODS

Filtering facepieces

Four models of NIOSH-approved N95 FFRs were purchased commercially. The four models were chosen to span a wide range of filtration performance. FFR models were assigned labels A, B, C, and D based on initial penetration values at the MPPS using a 3160 Fractional Efficiency Tester (TSI 3160; TSI inc., Shoreview, MN, USA) as described below. The manufacturers and models (in parentheses) of the N95 FFRs are 3M (1870), San Huei United Company (1895N), Gerson (1730), and Willson (1105N). None of the N95 models tested in the study had exhalation valves.

Particle size distribution

Polydisperse NaCl aerosol was employed to measure the TIL of particles through a respirator sealed to a manikin placed inside a test chamber (48 × 48 × 48 cm^3). Polydisperse aerosols were generated using a 1.5% NaCl solution with a constant output atomizer (TSI 3076) and passed through a dryer, a ^{85}Kr charge neutralizer (TSI 3077A) and then into the test chamber. An aerosol sample from the test chamber was analyzed for number concentration for different size particles (20–1000 nm range) over 240 s and repeated after a 30-s interval, using a scanning mobility particle sizer (SMPS; TSI 3080) in the scanning mode with an ultrafine condensation particle counter (TSI 3776). From the SMPS scans, the count median diameter (CMD) and mode size (peak) of the test aerosols were obtained (n = 2).

Monodisperse aerosol penetration test method

Prior to the TIL measurements, the MPPS for each FFR was tested against different size monodisperse NaCl particles using a TSI 3160 as described previously (Rengasamy *et al.*, 2007). Initial percentage penetration levels for 10 different monodisperse aerosols (centered at 20, 30, 40, 50, 60, 80, 100, 200, 300, and 400 nm) were measured for each sample at a constant flow rate 30 l min^{-1}. Average penetrations were based on three test samples (n = 3) for each N95 model.

Filter penetration and face seal leakage measurements using a manikin set-up

To evaluate the role of filter penetration and face seal leakage on TIL, a manikin set-up was employed as described previously with minor changes (Fig. 1) (Rengasamy and Eimer, 2011). An FFR was sealed to a manikin head form using a silicone sealant and placed inside a test chamber (48 × 48 × 48 cm^3). The head form was connected to a breathing simulator (Breathing Simulator Series 1101; Hans Rudolph, Inc., Shawnee, KS, USA) through an isolation chamber consisting of a rubber bladder inside a glass container. The isolation chamber was used to prevent particles created by the breathing simulator pump from getting inside the breathing zone of the respirator. The breathing simulator allowed for several parameters to be varied including tidal volume and breathing rate which provided simulation of more realistic breathing conditions compared to a previous study (Rengasamy and Eimer, 2011). Four breathing rates were chosen to span the breathing range of a human adult: 8, 20, 30, and 40 l minute volumes with 0.5, 1.0, 1.25, and 1.5 l tidal volumes, respectively. Aerosol was supplied from a constant

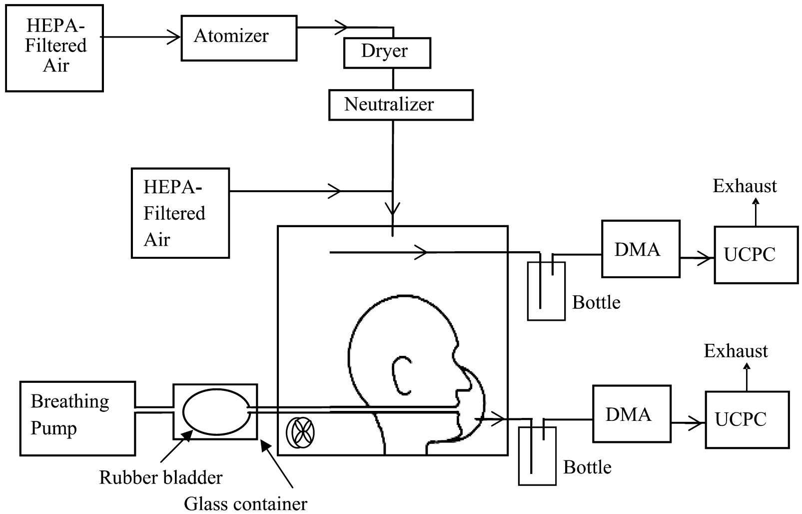

Fig. 1. Flow diagram of the experimental set-up used for nanoparticle leakage under simulated breathing conditions.

output atomizer (TSI 3076), using a 1.5% NaCl solution, dried and then passed through a charge neutralizer (TSI 3077A). The aerosol was then diluted with 50 l min^{-1} of dry air passed through high-efficiency particulate air filter (HEPA) and fed into the test chamber. Excess aerosol exited through a hole (1.3 cm diameter) in the back of the test chamber. A small fan was installed at the bottom of the chamber to ensure an even distribution of particles throughout.

The test FFR was equipped with one sample probe to measure aerosol concentration inside the FFR and two 'artificial leak' probes to simulate face seal leakage (Fig. 2) as described previously (Rengasamy and Eimer, 2011). The leak probes were filled with non-hardening putty and the increasing leak sizes were obtained by carefully pushing hypodermic needles (16- and 13-gage needles with inner diameters of 1.18 and 1.80 mm, respectively) through the putty to provide consistent leak channels through the needles (~2.56 cm long). Care was taken to ensure that the needle was kept open after inserting into the putty. The tip of the needle was positioned approximately closer to the inner surface of the respirator. The largest leak size (~1 cm long) of the study was obtained by removing all the putty from the leak probes providing a 3 mm hole. It should be noted that the length of the smaller leak sizes and the largest leak size was different. The length, diameter, and position of the leak probes as well as the sample probe may influence the data collected in the experiments. Further studies are needed to assess any potential effects of the above factors on particle concentration inside the FFR sealed to a manikin. For each test, two similar size leaks, one on each

Fig. 2. Typical FFR with a sample probe and one of two leak probes.

side, were introduced simultaneously. Samples from inside and outside of the FFR were withdrawn and analyzed by two SMPS systems simultaneously as described previously (Rengasamy and Eimer, 2011). The two SMPS systems scanned the particles in the 20 to 800 nm size range three times each 3 min. The samples were obtained during manikin breathing at 8, 20, 30, and 40 l minute volumes with tidal volumes of 0.5, 1.0, 1.25, and 1.5 l, respectively. These tidal volumes corresponded to 16, 20, 24, and 26.7 breaths/min, respectively. Both filter penetration and TIL values were calculated as the ratio of particle concentration inside (C_{in}) of the FFR to the outside concentration (C_{out}) and multiplied by 100%.

Filter penetration and TIL measurements with isopropanol-treated FFRs

To better understand the interaction between filtration and leakage processes, filter penetration and TIL were measured using N95 FFRs (N95-B and N95-D models) that had their electrostatic charge removed. Electrostatic charge on filter media can be removed by liquid isopropanol (IPA) immersion as described previously (Chen and Huang, 1998; Rengasamy *et al.*, 2009). The FFRs were dipped into IPA for 10 min, removed and then allowed to dry overnight. Removal of electrostatic charge from the filter media of FFRs has been found to shift the MPPS to larger (>100 nm) sizes (Chen and Huang, 1998; Martin and Moyer, 2000; Rengasamy *et al.*, 2009). The FFRs were then tested for filter penetration and TIL as described above for FFRs with no treatment. Four FFR samples of N95-B and N95-D models were used for this study ($n = 4$).

Data analysis

The data were analyzed using the SigmaStat® (Jandel Corporation) computer program. Filter penetration values for 45 and 400 nm size particles for the four models at three different flow rates were analyzed using a two-way analysis of variance (ANOVA). A three-way ANOVA was conducted to evaluate the significance of TIL data for 45 and 400 nm size particles at the three flow rates and leak sizes. All pairwise multiple comparisons were performed using the Tukey test.

RESULTS

Particle size distribution

For filter penetration and leakage measurements with a manikin in the chamber, aerosol particles were generated using a 1.5% NaCl solution and particle size distribution was measured using the SMPS system when the manikin was not breathing. The particle size distribution of NaCl test aerosols showed a CMD of ~82 nm with a mode size (peak) at ~75 nm (Fig. 3).

MPPS for the respirators

Filter penetration for monodisperse aerosols in the 20–400 nm range was measured at 30 l min^{-1} constant flow rate using the TSI 3160. Figure 4 shows filter penetration curves for the four N95 model respirators against 10 different monodisperse aerosols at sealed conditions. All four N95 model FFRs showed an MPPS in the 40–60 nm size range as expected for FFRs containing electrostatic filter media (Rengasamy *et al.*, 2007). Particle penetration was several times higher for the particles at the MPPS than for the 400 nm size range for all models. The filter penetrations at the MPPS for models N95-A, N95-B, N95-C, and N95-D were 0.11, 0.27, 0.61, and 1.47%, respectively (Table 1, top row). Model N95-A and N95-B showed smaller filter penetration values than the other models.

Filter penetration and TIL measurements under breathing conditions

Filter penetration of sealed FFRs at different flow rates was measured using a manikin test system. Filter penetrations for N95-B and N95-D increased with increasing breathing flow rate from 8 to 40 l min^{-1} as shown in Fig. 5. The MPPS for the filters for all four FFR model was ~45 nm at 40 l min^{-1} breathing flow (Fig. 6). The penetration values for 45 nm particles were 1.48, 2.8, 2.95, and 4.34% for models N95-A, N95-B, N95-C, and N95-D, respectively, at 30 l min^{-1} (Tables 1 and 2). These values were several times higher than the filter penetrations measured using monodisperse aerosols with the TSI 3160. The filter penetrations for 45 nm size particles were larger than the values for

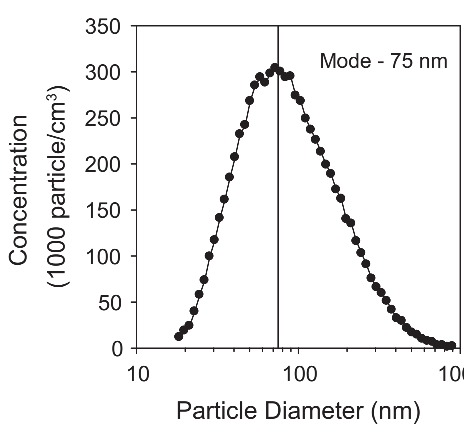

Fig. 3. Typical particle size distribution curve for aerosols generated using 1.5% NaCl solution.

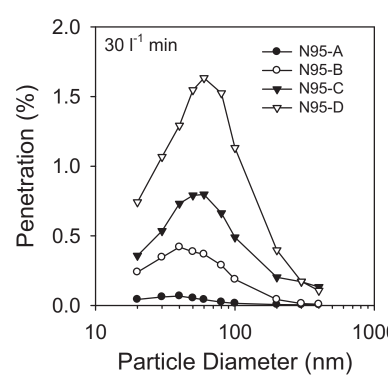

Fig. 4. Typical filter penetration levels for monodisperse aerosols for the four N95 FFR models at 30 l min^{-1} constant flow rate measured using the TSI 3160.

Table 1. Filter penetration for the four N95 FFR models. Average of three samples ($n = 3$) and error represents 1 SD.

Penetration test method	Filter penetration (%)			
	N95-A	N95-B	N95-C	N95-D
TSI 3160 (MPPS) (30 l min^{-1})	0.11 ± 0.08	0.27 ± 0.10	0.61 ± 0.21	1.47 ± 0.22
Manikin head form (MPPS) (30 l min^{-1})	1.48 ± 0.78	2.80 ± 1.06	2.95 ± 0.30	4.34 ± 0.91

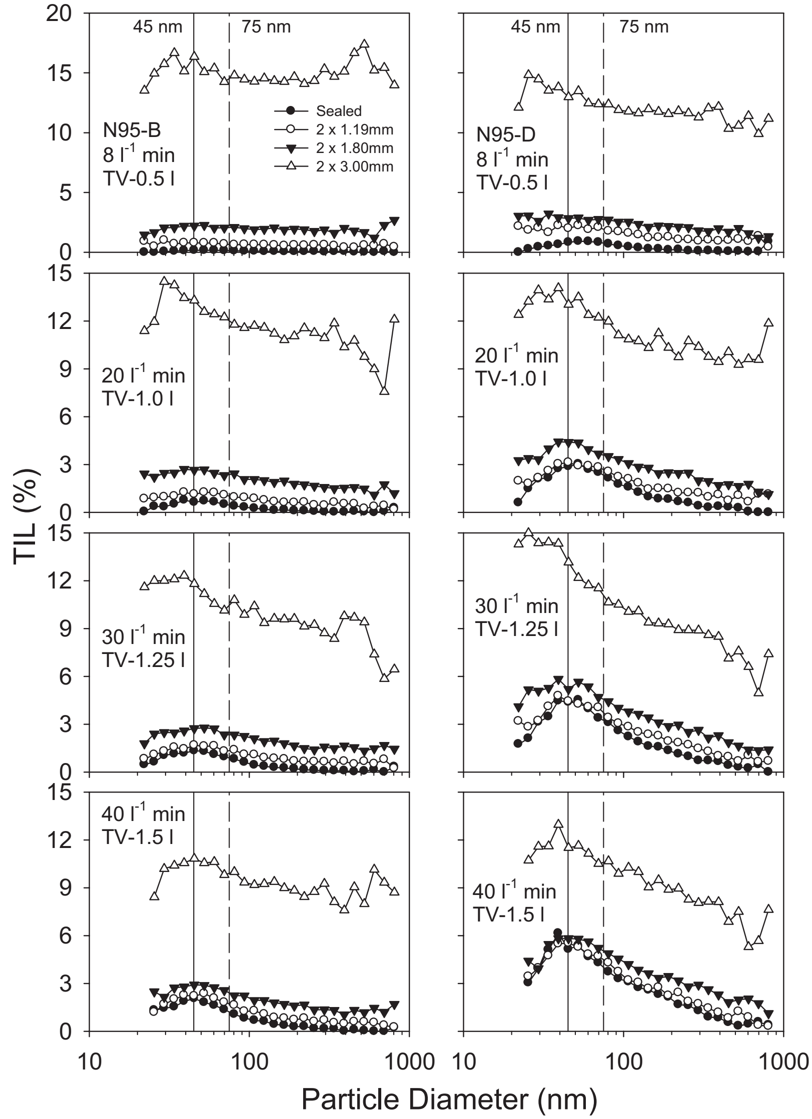

Fig. 5. Typical TIL values for the N95-B (left) and the N95-D (right) FFR models at 8 l min^{-1} (top row), 20 l min^{-1} (second row), 30 l min^{-1} (third row), and 40 l min^{-1} (bottom row) breathing minute volume. The symbols indicate sealed (solid circle), 2 × 1.19 mm leaks (open circle), 2 × 1.80 mm leaks (solid triangle), and 2 × 3.00 mm leaks (open triangle). The solid vertical line corresponds to the approximate average MPPS (45 nm) of the four N95 FFR models. The dashed vertical line corresponds to the mode of the challenge aerosol (75 nm) (TV, tidal volume).

400 nm size particles at different breathing flow rates using the manikin. The difference in filter penetration between 45 and 400 nm particles was found to be statistically significant ($P < 0.05$) for all four model FFRs (Table 3). Only N95-D model FFRs showed a significant difference ($P < 0.05$) in filter penetrations for 45 and 400 nm size particles with minute volume.

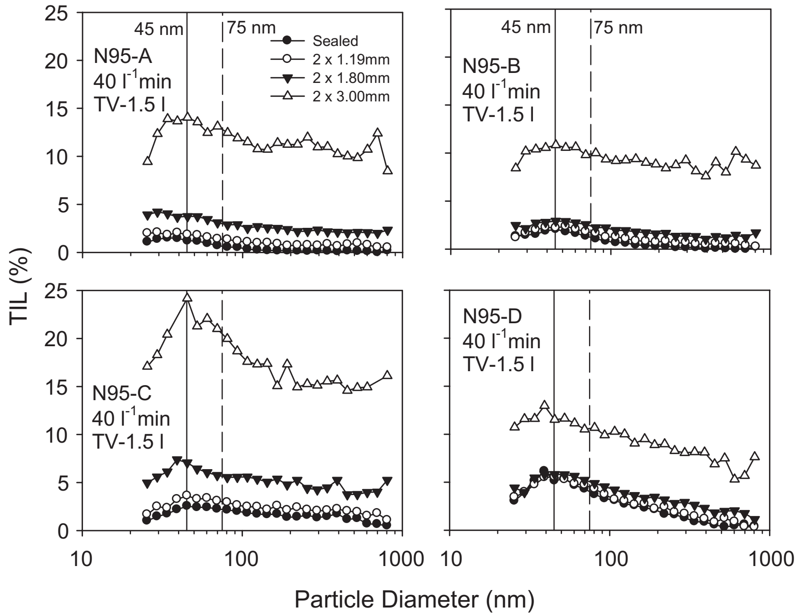

Fig. 6. Typical TIL values, for the four N95 FFR models, at three leak sizes at 40 l min^{-1} breathing minute volume. The symbols indicate sealed (solid circle), 2 × 1.19 mm leaks (open circle), 2 × 1.80 mm leaks (solid triangle), and 2 × 3.00 mm leaks (open triangle). The solid vertical line corresponds to the approximate average MPPS (45 nm) of the four N95 FFR models. The dashed vertical line corresponds to the mode of challenge aerosol (75 nm) (TV, tidal volume).

Table 2. Comparison of filter penetration and TIL at four different breathing minute volumes and three leak sizes. Average of four samples (n = 4) and error represents 1 SD.

Leak size (mm)	Flow rate (l min^{-1})	TIL (%)							
		N95-A		N95-B		N95-C		N95-D	
		Particle size (nm)							
		45	400	45	400	45	400	45	400
Sealed	8	0.65 ± 0.43	0.51 ± 0.44	1.04 ± 1.06	0.97 ± 1.92	2.20 ± 1.03	1.64 ± 0.99	1.30 ± 0.67	0.69 ± 0.75
	20	1.02 ± 0.46	0.41 ± 0.26	2.25 ± 1.20	1.24 ± 0.90	2.36 ± 0.26	1.13 ± 0.44	2.97 ± 0.68	0.65 ± 0.46
	30	1.48 ± 0.78	0.50 ± 0.36	2.80 ± 1.06	1.20 ± 0.76	2.95 ± 0.30	1.09 ± 0.51	4.34 ± 0.91	0.88 ± 0.43
	40	1.72 ± 0.66	0.47 ± 0.38	3.32 ± 0.89	1.13 ± 0.77	3.21 ± 0.93	1.19 ± 0.55	6.76 ± 4.24	1.55 ± 1.59
2 × 1.18	8	1.71 ± 0.45	1.33 ± 0.56	3.74 ± 2.78	3.51 ± 2.90	3.11 ± 1.18	2.62 ± 1.15	2.11 ± 0.35	1.07 ± 0.20
	20	2.26 ± 0.60	1.32 ± 0.49	3.24 ± 1.66	2.19 ± 1.23	3.82 ± 1.17	2.14 ± 0.86	3.54 ± 0.96	1.22 ± 0.61
	30	2.83 ± 0.92	1.45 ± 0.52	4.16 ± 2.46	2.45 ± 1.81	4.34 ± 1.19	2.08 ± 0.85	4.65 ± 0.83	1.30 ± 0.36
	40	2.65 ± 0.63	1.24 ± 0.40	3.88 ± 1.33	1.45 ± 0.66	4.07 ± 0.47	2.25 ± 0.77	7.14 ± 3.91	2.24 ± 1.81
2 × 1.80	8	5.60 ± 1.32	4.13 ± 1.09	5.42 ± 2.34	4.88 ± 2.11	6.40 ± 1.62	5.19 ± 1.85	3.60 ± 0.59	2.67 ± 0.62
	20	4.91 ± 1.00	3.06 ± 0.85	5.08 ± 1.71	3.50 ± 1.51	8.10 ± 3.57	4.78 ± 2.06	4.95 ± 1.12	2.52 ± 0.84
	30	5.06 ± 1.25	2.93 ± 0.46	5.45 ± 1.83	3.63 ± 1.49	8.93 ± 4.77	4.43 ± 0.246	5.67 ± 1.18	2.17 ± 0.56
	40	4.64 ± 0.64	2.66 ± 0.48	5.05 ± 1.43	2.78 ± 1.31	6.99 ± 2.64	4.30 ± 1.80	7.78 ± 4.35	3.04 ± 1.86
2 × 3.00	8	25.5 ± 3.88	19.9 ± 3.99	20.7 ± 3.84	19.8 ± 3.87	31.9 ± 12.6	25.5 ± 9.29	22.1 ± 10.1	19.8 ± 8.16
	20	19.6 ± 3.46	12.9 ± 1.42	18.1 ± 3.70	14.7 ± 3.09	30.4 ± 9.42	19.6 ± 5.07	18.2 ± 7.34	14.0 ± 6.63
	30	19.4 ± 4.63	12.9 ± 1.85	16.2 ± 3.37	12.2 ± 1.75	29.1 ± 12.8	16.8 ± 5.84	15.1 ± 4.00	11.1 ± 4.09
	40	17.3 ± 2.15	10.8 ± 1.02	13.6 ± 3.36	10.1 ± 3.39	22.7 ± 6.19	15.6 ± 2.88	15.1 ± 4.05	10.6 ± 2.44

Figure 5 shows increasing TIL values with increasing leak size and minute volumes for two models (N95-B and N95-D), while Fig. 6 shows the TIL values for all four N95 models (A, B, C, and D) at the highest minute volume, 40 l. Particles that penetrated through the filter media as well as

Table 3. Two-way ANOVA for the filter penetration data for the four N95 model FFRs. Particle size and breathing minute volumes are the independent variables.

	P-values (two-way ANOVA)			
	45 versus 400 nm			
	N95-A	N95-B	N95-C	N95-D
Particle size	<0.001*	0.009*	<0.001*	<0.001*
Minute volume	0.167	0.836	0.619	0.008*
Particle size × minute volume	0.164	0.379	0.168	0.078

*Statistically significant (*P*-value <0.05).

Table 4. Three-way ANOVA for TIL data for the four N95 model FFRs. Particle size, breathing minute volume, and leak size are the independent variables.

	P-values (three-way ANOVA)			
	45 versus 400 nm			
	N95-A	N95-B	N95-C	N95-D
Particle size	<0.001*	<0.001*	<0.001*	<0.001*
Minute volume	<0.001*	<0.001*	0.260	0.526
Leak size	<0.001*	<0.001*	<0.001*	<0.001*
Particle size × minute volume	0.688	0.238	0.587	0.222
Particle size × leak size	<0.001*	0.418	0.003*	0.961
Minute volume × leak size	<0.001*	<0.001*	0.128	<0.001*

*Statistically significant (*P*-value <0.05).

leaked through artificial holes showed an MPPS in the ∼45 nm range at all test conditions for all four N95 models. It should be noted that the MPPS at the largest leak size tested in the study was at a smaller size than 45 nm, which may be due to the turbulence of aerosols inside the respirator. In general, N95-A and N95-B models showed lower TIL values for different size particles than the other two models (Fig. 6). Similar results were obtained at other flow rates. TIL values for 45 nm size particles were larger than the values for 400 nm size particles at many flow rates and leak sizes tested in the study (Table 2). The mean TIL values for 45 and 400 nm size particles were significantly different ($P < 0.05$) for all four models (Table 4). All four models showed statistically significant difference ($P < 0.05$) for 45 and 400 nm size particles with different leak sizes. Statistical significance ($P < 0.05$) for different minute volumes was only found for two FFR models (N95-A and N95-B). Minute volume and leak size were also found to have statistically significant interaction ($P < 0.05$) for three N95 models (N95-A, N95-B, and N95-D).

Filter penetration and TIL with IPA-treated FFR

Filter penetration and TIL values for the charge removed (discharged) N95-B and N95-D models were measured at the same test conditions described for the FFR models with no treatment. Filter penetration results for discharged N95-B and N95-D models at sealed condition showed a shift in the MPPS from ∼45 to ∼150 nm size ranges at different flow rates (Fig. 7). The results indicated the removal of electrostatic charges from filter media as shown previously (Rengasamy *et al.*, 2009). Filter penetration values for 150 nm size particles were larger than the test aerosol mode size (75 nm). In general, TIL value for the MPPS (150 nm) increased with increasing minute volume. A small increase in TIL value for 75 and 150 nm size particles was obtained with smaller leak sizes (Fig. 7). Similar TIL values for different size particles in the 20–800 nm range was obtained at smaller minute volumes (8 and 20 l) and with the largest leak size tested in the study.

DISCUSSION

Four different N95 model FFRs sealed to a manikin test system when challenged with polydisperse NaCl aerosol particles (CMD ∼82 nm and mode size 75 nm) at different breathing minute volumes showed an MPPS in the ∼45 nm range. Penetrations for 45 nm size particles (MPPS) were significantly ($P < 0.05$) higher than the values for 400 nm size particles for all N95 model FFRs consistent with the previous report (Rengasamy and Eimer, 2011).

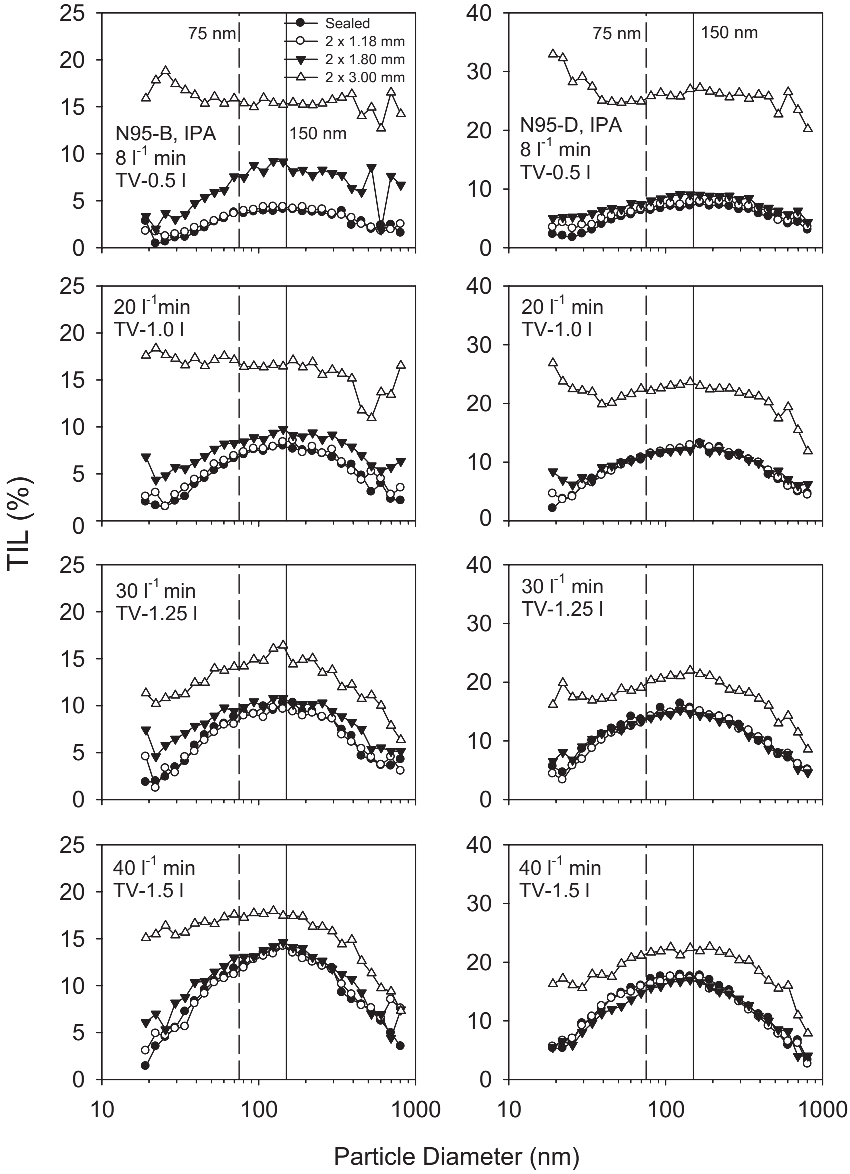

Fig. 7. Typical TIL values for the N95-B (left) and the N95-D (right) treated with IPA at 8 l min^{-1} (top row), 20 l min^{-1} (second row), 30 l min^{-1} (third row), and 40 l min^{-1} (bottom row) breathing minute volume. The symbols indicate sealed (solid circle), 2 × 1.19 mm leaks (open circle), 2 × 1.80 mm leaks (solid triangle), and 2 × 3.00 mm leaks (open triangle). The solid vertical line corresponds to the approximate average MPPS (150 nm) of the four N95 FFR models. The dashed vertical line corresponds to the mode of challenge aerosol (75 nm) (TV, tidal volume).

Filter penetrations for the MPPS ranged from 1.48 to 4.34% for FFR models N95-A to N95-D using the manikin system and 0.11–1.47% using the TSI 3160 at 30 l min^{-1}. The discrepancy may partly be explained by the difference in the manikin and TSI 3160 test systems and test conditions [sealing an FFR to a manikin or a flat plate, breathing (cyclic) minute volume or constant flow, scanning 20–800 nm aerosols or monodisperse aerosols] employed in the two methods. A breathing minute volume of 30 l min^{-1} is known to increase the filter penetration of nanoparticles for N95 FFRs compared to constant flow rates (Haruta *et al.*, 2008). Increasing the minute volume from 8 to 40 l increased the penetrations for 45 as well as 400 nm size particles for all models. When artificial leaks were introduced, the MPPS remained at ~45 nm and the TIL value increased with increasing leak size and minute volume for all four N95 model FFRs. TIL value for 45 nm was higher than the value obtained for 400 nm size particles at all test conditions. A three-way ANOVA of the data showed significant difference ($P < 0.05$) for the TIL values of 45 and 400 nm size particles for all respirator models. The mean TIL values for 45 and 400 nm size particles among the leak sizes were also significantly different ($P < 0.05$). Only N95-A and N95-B showed significant differences in the TIL for 45 and 400 nm size particles with different minute volume.

Interestingly, the results from the study provided a better understanding of the contribution of filter penetration and face seal leakage of nanoparticles to the TIL. N95 FFRs with electrostatic charge tested in the study showed an MPPS of ~45 nm size as reported previously (Hinds, 1999). During TIL measurement, aerosols (NaCl, mode ~75 nm) in the test chamber entered through the leak and peaked at ~45 nm size (MPPS). The shift in the aerosol mode (peak) size can be explained by the higher penetration level for the MPPS particles through the filter medium as well as by the efficient capture of larger size (>100 nm) particles by the filter medium during manikin breathing. The results showed that artificial leaks only allowed the test aerosols (NaCl, mode size ~75 nm) to enter inside the FFR to increase the concentration, while filter penetration determined the TIL values for different size particles. The increase in TIL value for the MPPS with increasing breathing minute volumes and leak sizes can partly be attributed to the cyclic air flow conditions. The results showed that penetration through filter media and leakage through face seal pathways interdependently increased the TIL value for various size particles with increasing leak sizes and minute volumes.

The importance of filter penetration in the TIL of the MPPS particles can also be seen from the results obtained in parallel experiments with electrostatic charge removed (discharged) N95 FFRs. Filter penetration and TIL values were measured with discharged N95 FFRs models sealed to a breathing manikin head. The discharged N95 FFRs challenged with test aerosols (mode size ~75 nm) showed a shift in the MPPS to ~150 nm size range at sealed conditions. The data are consistent with results obtained in previous studies that employed IPA to remove electrostatic charges from filter media (Chen and Huang, 1998; Martin and Moyer, 2000; Rengasamy *et al.*, 2009). Artificial leaks increased the TIL values for the MPPS (150 nm) and other size particles at different breathing flow rates confirming that leakage facilitates the test aerosols to enter inside the FFR, while filter penetration determines the TIL values for different size particles. Overall, the TIL data obtained with the discharged N95 FFRs confirmed the interdependency of filter penetration and face seal leakage processes.

The results obtained in the study address the issue of respiratory protection against nanoparticles. Many commonly used N95 and P100 FFRs are electrostatic and have an MPPS at ~50 nm. The filtration characteristics of electrostatic respirators with an MPPS <100 nm range produce higher filter penetration and TIL for nanoparticles. At the same time, charge removed or mechanical respirators show an MPPS at ~150 nm. The use of mechanical respirators in nanoparticle workplaces is expected to show lower concentration of nanoparticles inside the respirator than larger size particles (150 nm). This indicates that mechanical respirators would be advantageous over electrostatic respirators of the same rated filter efficiency for protection against nanoparticles in workplaces. It should be noted that charged particles are captured by weak image forces of mechanical filters rather than by the coulombic forces of electrostatic filters. Nevertheless, high-efficiency mechanical filters designed with lower resistance may offer better protection for nanoparticles.

The increase in TIL values with increasing leak sizes at different flow rates tested in the study indicated that face seal leak is a critical component of respiratory protection as described previously (Han and Lee, 2005; Grinshpun *et al.*, 2009). Data from one study showed that particle penetration through face seal leaks was between 3 and 5% for particles in the 0.1–1 μm range, while filter penetrations were <1% for the same size range of particles indicating that more nanoparticles get into the breathing zone through leakage than by filter penetration (Grinshpun *et al.*, 2009). Similar findings were obtained in another study that measured leakage for a Korean half-mask, and FFRs including 'top class', 'first class', and 'second class' with filter efficiency levels ≥99.0, ≥94.0, and ≥80.0%, respectively (Han and Lee, 2005). Maximal and minimal leakage values as well as the maximal/minimal ratio values by respirator brand and type were measured for human test subjects. The largest minimal leakages measured were 0.18, 0.39, 0.6, and 0.89% for half-mask, top class, first class, and second class respirators, respectively. These values were found to be very small when compared to Korean compliance standards. The authors explained that if filter penetration had been large, then minimal leakage would have been large (Han and Lee, 2005). This would have resulted in a small maximal to minimal penetration ratio. However, the maximal to minimal ratios were between 20 and 292 for the different types of respirators and were attributed to leakage through face seal pathways but not through filter penetration (Han and Lee, 2005). The above studies, however, failed to address the contribution of filter penetration to TIL of particles.

Data from our study showed a direct relationship between filter penetration and TIL values. Filter penetrations for 45 nm size (MPPS) particles were relatively less for model N95-A and N95-B than the values obtained for the other two models (N95-C and N95-D) with sealed condition at different flow rates (Table 2). In general, N95-A and N95-B showed relatively small TIL values for 45 nm size particles than the values obtained for the other two models (N95-C and N95-D) at smaller leak sizes at all flow rates. Overall, TIL values obtained for the four N95 model FFRs were dependent on the filter penetrations measured at sealed condition. The data obtained in the study agree with previously published results (Han and Lee, 2005). In that study, TIL for Korean half-masks and different class FFRs with test subjects were measured. Among the FFRs, average TIL values were 5.0% for top class FFRs with lower filter penetration values ($<1.0\%$) and were approximately two times lower than the TIL values for FFRs certified with higher filter penetrations (first class $<6.0\%$ and second class $<20.0\%$). Their results indicated that FFRs with lower filter penetrations produce lower TIL values. The contribution of particle penetration through filter media to TIL is supported by the findings from a previous study (Liu *et al.*, 1993). In that study, a theoretical expression for PF (an inverse function of TIL) based on filter penetration, leakage, and flow rates through filter media and leak holes was derived and compared with experimental results. For experiments, a relatively less penetrating 10 nm size monodisperse NaCl particles was used to measure particle leakage using a manikin. Leakage was measured with controlled leak holes at steady flow rates using three respirators with different filter penetrations at three different flow rates. PF was found to be inversely proportional to filter penetration, leak size, and flow rate under steady flow conditions. A good agreement between the theoretical and experimental results was obtained. TIL results obtained for the four models of N95 FFRs in our study are consistent with the above findings showing filter penetration dependence of TIL. Cyclic flow condition used in the study is likely to increase filter penetration and TIL for nanoparticles at flow rates such as 30 l min^{-1} (Haruta *et al.*, 2008).

Leak size and flow rate can significantly impact the TIL expected of an FFR with relatively small filter penetration value. In general, results from our study showed that TIL values for all four N95 models increased with increasing leak sizes and flow rates. In the case of N95-A, FFR with a small filter penetration value, TIL for 45 nm (MPPS) increased with breathing flow rates from 8 to 40 l min^{-1} at sealed condition and with the smallest leak sizes tested in the study. However, the TIL for 45 nm particles was higher at 8 l min^{-1} than at higher flow rates at the largest leak size tested in study (Table 2 and Fig. 5). TIL values for 45 nm size particles obtained at 8 l min^{-1} flow rate at the second leak size were nine, five, three, and three times larger than the filter penetration values for N95-A, N95-B, N95-C, and N95-D models, respectively, at sealed conditions. Penetrations at the largest leak size were 39, 21, 15, and 17 times greater for the same models, respectively. This may be related to filter resistance, penetration, geometry of leak sizes, location of sample, and leak probes and flow through the cylindrical leak holes. Further studies are needed to assess the effects of the above factors on TIL values. The results obtained in the study indicate that a respirator with a relatively low filter penetration at sealed condition may experience higher magnitude of increase in TIL values at a relatively low breathing flow rate such as 8 l min^{-1} employed in the study. This is consistent with the conclusion that a HEPA respirator may provide less protection than a dust–mist respirator when there is a substantial lack of face seal fit and the breathing rate is low (Chen and Willeke, 1992). In that study, a HEPA respirator with a large leak size at 5 l min^{-1} flow allowed more aerosols through the leak than a dust–mist respirator with relatively low filtration efficiency. Thus, a low filter penetration N95-A model FFR similar to a HEPA respirator may provide lower protection to the wearer when a significantly large size leak is created during work activities at a low breathing flow rate.

The artificial static leaks employed in our study using the manikin are different from the variable leak sizes created during work activities of the respirator wearer. Several studies have reported that artificial leaks introduced in the manikin test system are static and differ from the dynamic leaks created during the maneuvers of a worker (Krishnan *et al.*, 1994; Myers *et al.*, 1995; Janssen and Weber, 2005). Unlike a manikin, the leaks created by workers body and head movements may be intermittent and particle deposition in human airways during breathing can influence the measurement of filter penetration and face seal leakage. The artificial leaks introduced in the manikin study are comparable to those leaks that occur in human test subjects (Lee *et al.*, 2008; Grinshpun *et al.*, 2009; Reponen *et al.*, 2011). The smaller leak sizes (2 × 1.19 and 2 × 1.8 mm) used in our study produced TIL values generally <5% for nanoparticles (<100 nm) at 30 and 40 l breathing minute volumes. These values are closer to the mean inspiratory flow rates reported for test subjects performing fit test exercises (Grinshpun *et al.*, 2009). The TIL value 5% obtained in our study corresponds to a PF of 20 (100/5). The largest leak size (2 × 3 mm) produced TIL values > 10 equivalent to PF values <10 (100/10) for nanoparticles. The TIL values obtained in our study are comparable to the PF values measured for test subjects in previous studies (Lee *et al.*, 2008; Reponen *et al.*, 2011). Four N95 models tested in the above studies showed PF values <30 (50th percentile) for particles <120-nm size particles. This indicates that the leak sizes employed in our study are closer to the leak sizes that occur in human beings at controlled experimental conditions. Further studies are needed to validate the comparability of the TIL data obtained with a breathing manikin and the PF measured for test subjects. Nevertheless, filter penetration and TIL data obtained at different test conditions consistently showed an MPPS at ∼45 nm. Moreover, filter penetrations and TIL values for 45 nm size particles were higher than the values obtained for 400 nm size particles at all test conditions. The data obtained indicate that filter penetration and face seal leakage processes can be studied using a manikin test system in a reproducible manner.

The data obtained in the study suggest that the TIL for the MPPS would be higher relative to smaller and larger size particles in the breathing zone of respirators in workplaces where high concentration of nanoparticles exists. However, it should be noted that the overall concentration of particles inside the respirator is expected to be less than the concentration outside. The results obtained in the study showed that filter penetration had a direct relationship to the TIL values for the different N95 model FFRs. In general, N95 models (N95-A and N95-B) that had the smallest filter penetration values for the MPPS also showed smallest TIL values for the same size particles, while the models (N95-C and N95-D) with higher filter penetrations had higher TIL values. The results suggest that FFRs with relatively low filter penetrations for the MPPS are likely to provide higher protection for nanoparticles. Penetration through leakage increases the TIL values for nanoparticle with increasing leak sizes indicating that lower TIL values for nanoparticles can be obtained at smaller leakages. The filter penetration and leakage data obtained in the study are congruent to the results obtained for test subjects (Lee *et al.*, 2008; Grinshpun *et al.*, 2009; Reponen *et al.*, 2011). These authors showed lower PF for particles <120 nm than those for larger size particles. The above studies also showed that good fitting respirators are needed to achieve higher PF values. Taken together, the data obtained in our laboratory suggest that good fitting respirators with lower filter penetration values would provide better protection against nanoparticles.

CONCLUSIONS

Four N95 model FFR sealed to a manikin showed an MPPS at ∼45 nm. Filter penetration for 45 nm size particles was significantly ($P < 0.05$) higher than the values for 400 nm size particles at 8–40 l min^{-1} flow rates. A consistent increase in filter penetrations for 45 and 400 nm size particles was obtained with increasing flow rate. Leakage of test aerosols with a mode size of ∼75 nm through artificial holes increased the TIL for ∼45 nm (MPPS) as well as other size particles at different flow rates indicating that leakage allows the test aerosols inside the FFR, while filter penetration determines the TIL for different size particles. TIL values for 45 nm size particles were significantly ($P < 0.05$) higher than the values obtained for 400 nm size particles for all four N95 models. N95 models with relatively small filter penetrations showed lower TIL values than the models with higher filter penetration values. The results indicated that TIL for nanoparticles was dependent on filter penetration in addition to leakage at different flow rates and leak sizes. N95 FFR with their electrostatic charge removed showed a shift in the MPPS to ∼150 nm when TIL was measured with the same test aerosols (mode size ∼75 nm) at different leak sizes and flow rates confirming the interaction between filter penetration and face seal leakage processes. The shift in the MPPS from 45 to 150 nm for the charge removed filters indicates that mechanical filters may perform better against nanoparticles than electrostatic filters rated for the same filter efficiency. The data indicate that among the different size particles that enter inside the N95 respirators, relatively high concentration of the MPPS particles inside the breathing zone of N95 respirators can be expected in workplaces handling high concentration of nanoparticles. Overall, the data obtained in the study suggest that good fitting respirators with lower filter penetration values would provide better protection against nanoparticles.

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