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Particle Count Statistics Applied to the Penetration of a Filter Challenged with Nanoparticles

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Abstract

Statistical confidence in a single measure of filter penetration (P) is dependent on the low number of particle counts made downstream of the filter. This paper discusses methods for determining an upper confidence limit (UCL) for a single measure of penetration. The magnitude of the UCL was then compared to the P value, UCL ≤ 2P, as a penetration acceptance criterion (PAC). This statistical method was applied to penetration trials involving an N95 filtering facepiece respirator challenged with sodium chloride and four engineered nanoparticles: titanium dioxide, iron oxide, silicon dioxide and single-walled carbon nanotubes. Ten trials were performed for each particle type with the aim of determining the most penetrating particle size (MPPS) and the maximum penetration, P_{max}. The PAC was applied to the size channel containing the MPPS. With those P values that met the PAC for a given set of trials, an average P_{max} and MPPS was computed together with corresponding standard deviations. Because the size distribution of the silicon dioxide aerosol was shifted towards larger particles relative to the MPPS, none of the ten trials satisfied the PAC for that aerosol. The remaining four particle types resulted in at least 4 trials meeting the criterion. MPPS values ranged from 35 – 53 nm with average P_{max} values varying from 4.0% for titanium dioxide to 7.0% for iron oxide. The use of the penetration acceptance criterion is suggested for determining the reliability of penetration measurements obtained to determine filter P_{max} and MPPS.

INTRODUCTION

Particle collection efficiency by a filtration device, E, is determined as the percent difference between particle counts obtained upstream of a collection or separation device, C_u, relative to those obtained downstream, C_d. For both filters and aerosol samplers it is often more important to refer to the percent of particles that penetrate, P, the device, which can be calculated by 100 – E, or directly by $C_d/C_U(100)$. Methods used to determine the efficiency of a device to capture particles over a range of particle sizes have involved the production of a series of monodisperse aerosols that covered the size range of interest. This method was used, for example, to determine the collection efficiency of aerosol samplers (Harper et al., 1998; Mark and Vincent, 1986) and respirator filters (Lee and Liu, 1982; Qian et al., 1998; Stevens and Moyer, 1989). With the advent of particle counters capable of size discrimination into multiple size bins, such as the scanning mobility particle sizer (SMPS), filter and sampler efficiency studies were modified to utilize particle counter/sizers since they could more easily develop a complete efficiency curve from the administration of a

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single polydisperse aerosol (Liden, 1993; Maynard and Kenny, 1995; Balazy et al., 2006). In some cases the two approaches were used in the same study. For example, Rengasamy et al. (2009) utilized the dispersion of monodisperse aerosols and an SMPS to measure the collection efficiency of filtering facepiece respirators.

Although modern particle counters can readily provide values for C_u and C_d, a reliable estimate of P is related to the magnitude of the particles available to count downstream of the device. To demonstrate with a simple example; if $C_u = 100\ cm^{-3}$ and $C_d = 1\ cm^{-3}$ is counted downstream, then a P = 1% results. However, if, during the same experiment one additional particle cm^{-3} is counted downstream, then a P = 2% results and thus doubles the measured penetration of the device. Therefore, when C_d is small any changes in the value of C_d can have a large impact on P.

The question then arises as to the statistical confidence applied to P values obtained from experiments with low C_d values. An obvious solution would be to increase C_u to provide a greater likelihood of increasing C_d and therefore minimizing the effect that varying C_d values have on an estimate of P. However, when using the method in which a polydisperse aerosol is applied to a filter, then, given its (typically) log-normal distribution, there will necessarily be particle size ranges with low counts in the tails of the distribution relative to those in the middle of the distribution. As demonstrated in the previous example, low counts in the tail areas will increase uncertainty in the estimate of P in those size ranges. If the uncertainty in P for any particle size can be estimated then another solution is to discard size bins that do not meet a criterion that stipulates the maximum acceptable level of uncertainty in P. To address the need to quantify this uncertainty, techniques used to evaluate the statistical confidence in estimates of particle penetration will be described. Furthermore, the use of one of these methods to determine uncertainty in a P value will be applied when determining the maximum P and most penetrating particle size (MPPS) of a filtering facepiece respirator (FFR) challenged with engineered nanoparticles.

Count Statistics

Statistical techniques used to evaluate counts made by electronic particle counters, as well as manual counts made when viewing particles captured on a filter with a microscope, are based on the assumption that the counts follow a Poisson distribution (Herdan et al., 1960). As described in most introductory statistics texts, that distribution is one of several that characterize the distribution of a discrete random variable that constitutes a relatively rare event over time, area, volume, or other metric (e.g. Navidi, 2008). The mean, λ, of a Poisson "process" is therefore a rate expressed as the ratio of a non-negative integer and a value for the unit of some space or time applied to the denominator. If that space is a volume, as when counting particles in air, λ can be considered the mean count concentration and the discrete variable is the count of particles, n.

Referring to the case of using a particle counter that counts n particles per unit volume, V, then $\lambda = n/V$, and the probability distribution of the parameter of λV can be expressed as (Ott, 1995; Van Slooten, 1986):

$$P(n)=\frac{e^{-\lambda V}(\lambda V)^n}{n!} \quad \text{Eq. 1}$$

This probability distribution equation is useful for cases such as those considered by Van Slooten (1986) in which it is important to understand the probabilities of encountering various particle counts in a cleanroom environment given a historical or expected value for those counts = λV. However, most modern particle counters do not provide an integer count of particles, n, but rather the count concentration, n/V. Furthermore, in most cases where these instruments are employed, λ is not known and will vary between air volumes sampled. The issue, then, is not the probability of obtaining a certain integer count of particles in a measured volume but, rather, the statistical confidence in the measurement of the count concentration. In that case, λ is not a constant as given in Eq. 1, but represents a random variable, $\hat{\lambda}$, for which a confidence interval (CI) can be applied to express the uncertainty in its measurement (Van Slooten, 1986).

As explained by Johnson and Kotz (2005), it is not possible to construct a CI for a variable with a discrete Poisson distribution with an exactly specified coverage probability of say 95%. However, a technique to develop CIs based on the relationship between the Poisson distribution and the χ^2 distribution, first developed by Garwood (1936), is widely accepted as the most "exact" estimate for those limits (Johnson et al., 2005; Ulm, 1990). As alluded to above, the rate expressed by $\hat{\lambda}$ in the case of particle counting with an SMPS can be defined as a randomly varying particle count concentration, C. In this case, applying the method developed by Garwood results in a lower confidence limit ($\mathrm{LCL_C}$) and upper confidence limit ($\mathrm{UCL_C}$) of C expressed as:

$$\mathrm{LCL_C}=\left(\chi^2_{\alpha/2,2\mathrm{C}}\right)\Big/2 \quad \text{Eq. 2}$$

$$\mathrm{UCL_C}=\left(\chi^2_{1-\alpha/2,2(\mathrm{C}+1)}\right)\Big/2 \quad \text{Eq. 3}$$

In the past, these limits had to be calculated with the use of χ^2 probability tables such as those provided by Box et al. (1978), but can now be determined with the use of a spreadsheet containing a function that produces the table value, such as the CHIINV() function in Microsoft Excel.

Because of difficulty in the past calculating the limits given in Eqs. 2 and 3, approximations for results obtained when applying those equations were made knowing that the Poisson distribution closely approximates the normal distribution for large n (Johnson et al., 2005). The development of approximations for the CI of a Poisson parameter was strongly influenced by occupational epidemiologists interested in the probability distribution of the standardized mortality ratio (SMR). The SMR is assumed to follow a Poisson distribution and is computed by dividing the observed number of deaths by the expected number of deaths (Ulm, 1990; Ury and Wiggins, 1985; Vandenbroucke, 1982). These methods form their basis under the assumption that variability in the rate is caused by variability in the observed deaths (numerator) where the expected deaths (denominator) can be considered constant, which is analogous to a fluctuating number of particles in a specified volume. Furthermore, as explained byBox et al. (1978), a square-root transformation of a Poisson variable not only stabilizes its variance, but results in a constant value for the variance of 0.25. In the case of determining a 95% CI for a large, normally distributed sample, this would involve adding and subtracting $1.96\sqrt{0.25}$, which is essentially equivalent to adding or subtracting unity. Therefore, CIs about a measure of particle concentration, n/V, can be estimated from the following (Vandenbroucke, 1982):

$$\frac{\left(\sqrt{\mathrm{n}}\pm 1\right)^2}{\mathrm{V}}. \quad \text{Eq. 4}$$

Further modifications to this approach are reviewed by Ulm (1990). An expansion of the squared term in the numerator of Eq. 4 gives $n\pm 2\sqrt{n}+1$, which is similar to the estimator suggested by Ury and Wiggins (1985). The simplest approximation found in the literature, again derived from research associated with the SMR (Bland, 1995) as well applications in medicine (Dobson et al., 1991), has the following form

$$\frac{\mathrm{n} \pm 1.96\sqrt{\mathrm{n}}}{\mathrm{V}}, \quad \text{Eq. 5}$$

which is sometimes shown with 2 replacing 1.96 (Wang and Winters, 2004).

The magnitude of n needed to apply the approximation given in Eq. 5 varies by source from 5 to 100 (Johnson et al., 2005; Montgomery and Runger, 2003; Van Slooten, 1986; Wang and Winters, 2004). This CI follows from the convenient property of a Poisson variable where the variance is equivalent to the mean (Johnson et al., 2005). Furthermore, the CI of a Poisson variable can be constructed around a single measurement in contrast to the development of a CI for a normally-distributed variable which requires a sample of n>1 observations. It should be noted that the CI given in Eq. 5 is sometimes shown in the literature with only the numerator portion displayed (e.g. Wang and Winters, 2004). This form of the equation suggests the development of confidence limits around variations in integer counts, n. However, the parameter which is bounded by the limits is λ, which, for airborne particle counts, is a concentration, which requires that V be placed in the denominator. Or, a more useful form of the equation that uses the nomenclature described here is: $\mathrm{C} \pm 1.96\sqrt{\mathrm{C}}$.

Penetration Confidence Interval

Given that $C_d \leq C_U$ when testing particle filters, then P is a proportion with possible values bounded by 0 and 1. Furthermore, to count a particle downstream of a filter from a total of upstream counts, n_{up}, can be considered a "success" as versus the "failure" of not counting it, which is characteristic of a variable with a binomial distribution. Like the Poisson distribution, the binomial distribution can be approximated by the normal distribution when, in this case, a large number of upstream counts are used to calculate P. For a binomially distributed variable with large n, a 95% UCL for P, UCL_P, can be calculated by (Johnson et al., 2005):

$$\mathrm{UCL_P}=\mathrm{P}+1.96\sqrt{\frac{\mathrm{P}(1-\mathrm{P})}{\mathrm{n_{up}}}}. \quad \text{Eq. 6}$$

A UCL is given Eq. 6 rather than an expression for the entire CI as given in previous equations because a statistical analysis of P should be more concerned with the magnitude of the UCL than that of the LCL of a CI from the practical standpoint that high penetration results in greater aerosol exposure to the wearer of the FFR.

A different approach was taken by Leith and First (1976) who developed a standard error for P based on both n_{up} and downstream counts, n_{down}, which resulted in the following equation for computing UCL_P:

$$\mathrm{UCL_P}=\mathrm{P}+1.96\mathrm{P}\sqrt{\frac{1}{\mathrm{n_{up}}}+\frac{1}{\mathrm{n_{down}}}}. \quad \text{Eq. 7}$$

Additional UCLs established for P are provided in standards set for evaluating the efficiency of industrial high-efficiency particulate air (HEPA) filters (ASHRAE, 2007; BS EN, 2009; IEST, 2007). Wang and Winters (2004) provide numeric examples for applying the recommended practice developed by the IEST (2007). The IEST describes a UCL_P based on Eqs. 2 and 3 for $n \le 50$, and Eq. 5 for $n > 50$ (using the nomenclature defined here) as the ratio of the downstream count upper limit, UCL_{Cd} , divided by the upstream count lower limit, LCL_{Cu}.

$$\mathrm{UCL_P} = \frac{\mathrm{UCL_{Cd}}}{\mathrm{LCL_{Cu}}} \quad \text{Eq. 8}$$

The IEST method is, therefore, not based on assuming that P is a binomial variable but, rather, creates a "worst-case" UCL based on a combined use of the confidence limits given in Eqs. 2 and 3 which derive from the counts having a Poisson distribution. Additional consideration for particle count variation is also applied to Eq. 8 in the IEST recommended practice (2007) for the case where the upstream counts are diluted prior to measurement, referred to as the "correlation ratio".

A British standard for filter efficiency (BS EN, 2009) describes a method for determining an LCL about efficiency, E, given that this is the worst-case scenario from that perspective. However, the approach for doing so is equivalent in all other ways to that of the IEST method for calculating a UCL for P with the exception that values derived from Eqs. 2 and 3 are provided for $n \le 100$, and Eq. 5 is used for $n > 100$.

A United States (US) standard for testing filter removal efficiencies (ASHRAE, 2007) takes a much different approach from those previously discussed. A CI for P is not based on the inherent variability of individual counts but is computed after a number of trials, k, from which an average P, $\overline{P}$, can be determined and a UCL established as for a small, normally-distributed sample:

$$\mathrm{UCL}_{\overline{P}} = \overline{P} + t\frac{\delta}{\sqrt{k}} \quad \text{Eq. 9}$$

where δ is the pooled standard deviation of the correlation ratio and the observed penetration, and t is the Student-t value based on k-1 degrees of freedom.

The use of Eqs. 6 – 8 for computing UCL_P are compared in Figure 1. The curves shown were created by assessing C_d from 1 to 1000 and changing associated C_u values to maintain a constant $P = 0.01$. Figure 1 demonstrates that the IEST method results in the most conservative estimates of UCL_P. Whereas, assuming P is a proportion with a binomial distribution and using Eq. 6 to calculate a UCL results in the lowest upper limits. Interestingly, the use of Eq. 7 developed by Leith and First (1976) results in limits almost identical to those computed from Eq. 6. Noting that, for small P, the term 1 - P in Eq. 6 tends to unity and the term $1/n_{up}$ in Eq. 7 tends to 0, it can be shown that those two equations produce almost equivalent CI values under the condition of small P.

Hypothetical Example

A hypothetical example that demonstrates the application of UCLs for individual P values determined over a range of particle size channels sampled by a SMPS can be provided by first investigating a best-case scenario. First assume P values, which change from near 0 for very small particles to a peak value (the most penetrating particle size, MPPS) and back to near 0 for large particles, can be modeled as having a lognormal distribution relative to particle diameters with a geometric mean (GM) equal to the MPPS and some geometric standard deviation (GSD). Then the ideal situation for measuring that P-curve will occur when the maximum number of particles can simultaneously be applied to all size channels as will occur if the test aerosol also has a lognormal distribution and with a GM and GSD equivalent to that of the P-curve.

To demonstrate, assume the upstream aerosol has a GM = 100 nm with a GSD = 1.80 and a maximum count of 5000 cm^{-3}. Furthermore, assume the P-curve can be modeled as having a log-normal distribution with GM = 100 nm and GSD = 1.80. Figure 2(a) displays UCL_P values for this situation produced by application of Eq. 8 over a range of particle sizes from 10 – 1000 nm for a device with a maximum P, P_{max}, of 5% such as an N95 FFR. As shown in Figure 2(a), the UCL_P developed from Eq. 8 greatly expands in the size ranges corresponding to the tails of the aerosol distribution where counts are low. As also shown in Figure 2(a), the transition in UCL_P values developed from Eq. 8 is erratic where particle diameters are associated with low concentrations. This is due to the application of the CHIINV() function which truncates the concentration to the nearest integer. This is useful in cases where particle sizer software applies correction factors to integer counts per volume resulting in real-number concentration values, but results in a saw-tooth behavior of estimates of UCL_P between particle size channels.

If the polydisperse aerosol produced to test the device has a GM that is not equal to the MPPS, and/or the GSD is different from that presumed for the P-curve, then larger UCL_P values will be expected where counts are low. Figure 2(b) shows the case where all conditions are equal to those used to create Figure 2(a) except that the aerosol GM = 200 nm. Here, the UCL_P values have expanded greatly on the left side of the P-curve because very few particles are available in that region and contracted on the right side because of the larger counts in that region.

Method Application

With an overall goal of determining the maximum penetration, P_{max}, of FFRs, the statistical methods reviewed above provide a solution to the concern that the possible occurrence of low counts in the size bins where this is likely to occur will greatly reduce overall confidence in the result. This problem is important when challenging respirators with an aerosol for which the geometric mean (GM) diameter cannot be easily controlled. For example the concentration of a salt solution can be adjusted to change the GM diameter to produce an aerosol with a GM near the most penetrating particle size (MPPS) (Hinds, 1999). However, if other aerosols are produced, say from dry generating devices or from a liquid suspension of a powder, then the GM diameter may not be readily adjusted. To evaluate the uncertainty in an estimate of P resulting from low particle counts in cases where the GM is not close to the MPPS, Eq. 8 was utilized to calculate the UCL of P_{max} and then the constraint, $\mathrm{UCL} \leq 2P$, was applied as a penetration acceptance criterion (PAC) for including a P_{max} value from a particular trial when determining an average P_{max}, P_{max}, from all trials conducted for a particular dust type.

Table 1 shows upstream counts needed to obtain various levels of uncertainty for respirators designed to achieve a 5%, 1% and 0.03% penetration corresponding to N95, N99, and N100 respirators, respectively. For example, from Table 1 the number of upstream counts needed to achieve the PAC for an N95 FFR is 700. Furthermore, if the actual penetration at the MPPS is less than 5%, then higher counts would be needed to meet the same criteria. The high upstream counts needed for devices with P = 0.03% or 1% necessitates the use of a diluter prior to measurement with a particle counter, many of which produce count errors above 10,000 cm^{-3}. In that case, the additional uncertainty associated with estimating a correlation (dilution) ratio should also be considered (Wang and Winters 2004).

METHODS

Filter Penetration System

Particle penetration of a single N95 FFR type (Aero Technologies, 3M Corp., St. Paul, MN) was assessed in a laboratory-fabricated respirator sampling enclosure (Figure 3). The relative humidity and temperature in the enclosure was maintained at or below 40% and 25°C, respectively. The FFR was secured in a polycarbonate casing with two sections. The top section had an opening that had been cut to fit the curved outer dimensions of the FFR to be tested less the 8-mm band of material that normally contacts skin. The FFR was pushed through this hole and sealed on the back side along its edge with tape. After applying the FFR to the upper section, the two sections were sandwiched together and connected via bolts in each corner. When the bolts were tightened, the upper section effectively sealed the FFR to the uncut bottom piece of plastic while maintaining its shape. The bottom section had a large threaded hole behind the middle of the FFR which allowed it to be threaded to a 2.54-cm steel pipe protruding through the enclosure. The other end of the pipe was connected via a T-fitting to a vacuum pump which pulled 85 l min^{-1} of air (42 CFR 80, 1995) as monitored by a calibrated Venturi flow meter. A 0.64-cm diameter stainless-steel tubes was used to sample upstream air directly below the FFR casing. A tube of similar length and diameter was enclosed within the 2.54-cm steel pipe so that sampling end was within the cup space of the FFR to sample downstream air. The downstream ends of both sample lines entered a two-way valve to switch air entering an SMPS. Initial tests were conducted without an FFR in line to ensure that particle counts measured with either line were similar.

Four different nanoparticle powders were used to produce the challenge aerosols: 21-nm primary particle size titanium dioxide (TiO_2, Nanostructured and Amorphous Materials, Inc., Houston, TX), 20-nm primary particle size silicon dioxide (SiO_2, Evonik Degussa Corporation, Parsippany, NJ), 20-50 nm primary particle size iron oxide (Fe_2O_3, Nanostructured and Amorphous Materials, Inc., Houston, TX), and single walled carbon nanotubes (SWCNT, Nanostructured and Amorphous Materials, Inc.) with an outer diameter of 1-2 nm and a length of approximately 1.5 μm.

All powders were prepared as 6.7 mg/mL suspensions in water conditioned by reverse osmosis and ultra-filtration. Immediately after adding the powder to the water, the solution was sonicated by high frequency probe (model 550, Fisher Scientific, Pittsburgh, PA) for 10 min. The solution was then added to the reservoir of a six-jet Collison nebulizer (BGI Inc., Waltham, MA). The nebulizer was operated at 20 psi from a HEPA-filtered compressed air source. Sodium chloride (NaCl) was also used as challenge aerosol as a standard reference for comparison with the other aerosols. When producing a NaCl aerosol, a 0.10 mg/mL solution was prepared in the same water.

Given that relatively low upstream counts can produce statistically relevant penetration values when testing N95 FFRs (Table 1), we diluted the upstream aerosol to less than 10,000 cm^{-3} prior to entering the sealed box containing the FFR so as not to overload the CPC and avoid the additional uncertainty induced by diluting high upstream counts prior to their measurement. This dilution was accomplished with the use of a variable venturi concentration selector (Ortho Dial-n-Spray, The Scotts Company LLC, Marysville, OH) designed to regulate pesticides sprayed through a garden hose, but that enabled accurate control of suction from a 19 L drum into which the aerosol was injected (Figure 3). The diluted aerosol was charge-neutralized prior to entering the enclosure.

Filter Penetration Testing

Ten FFRs were tested for each particle type. Upstream and downstream aerosol counts were measured using a scanning mobility particle sizer (SMPS) that consisted of an electrostatic classifier with a “long” differential mobility analyzer (Model 3936, TSI Inc., Shoreview, MN) and a water-based condensation particle counter (CPC, Model 3785, TSI Inc., Shoreview, MN). The SMPS was set to take consecutive measurement every four minutes that allowed for a 60 s delay between 180 s scans.

For each test, three upstream samples were initially taken followed by three downstream samples and then another three upstream samples. This sampling scheme was performed to compensate for temporal changes in aerosol concentration during a test. Previous sample runs were conducted to ensure that there were no residual particles remaining when switching between upstream to downstream samples. The six upstream samples and the three downstream samples in each size bin were averaged. Penetration was then calculated for each bin by dividing the average downstream count by the average upstream count.

Data analysis for a powder type initially consisted of locating the size bin containing P_{max} for each of the ten trials and computing a UCL_P for each P_{max} value. The mid-diameter associated with the size bin containing P_{max} was noted as the MPPS for that trial. If the penetration acceptance criterion (PAC), $UCL_P \leq 2P_{max}$, was met, the P_{max} value was retained and combined with all other P_{max} values from the other nine trials that met the PAC. Given the remaining P_{max} values, an average, $\bar{P}_{max}$, and standard deviation were calculated, and from which $UCL_{\bar{P}}$ was obtained by application of Eq. 9. An average of all MPPS values from size bins that met the PAC was also computed.

RESULTS

Plots are given in Figures 4 and 5 of the particle count distribution as well as the penetration and associated UCL_P values obtained during one of the ten trials that best represented the average condition for a particle type. These values obtained from a NaCl trial are plotted in Figure 4 where it can be seen that upstream counts peaked near 30 nm whereas penetrations peaked near 50 nm. This offset resulted in low UCL_P values for P values associated with particle channels below 50 nm and widely expanding UCL_P values above 50 nm. Regardless, the ratio of UCL_P to P was below 2 for the size bin containing P_{max} for all ten trials and therefore met the PAC for those trials (Table 2).

Results comparing P, UCL_P, and counts for the four nanoparticles tested are shown in Figure 5. Only SWCNT particles produced a count distribution that peaked near the MPPS to obtain reliable penetration results for that powder type for all ten trials. The TiO_2 and SiO_2 powders produced distributions with a GM near 110 nm so that particle numbers in the size bins near the MPPS were low. As a consequence, only four TiO_2 trials and no SiO_2 met the PAC. The Fe_2O_3 count distribution revealed a pronounced bimodal quality (Figure 5d) with a dip in counts between the two peaks near the size bins containing the MPPS, which resulted in only six of the ten trials meeting the PAC. The MPPS for these four particle types varied between 35 and 53 nm (Table 2).

The maximum penetration, $\bar{P}_{max}$, averaged from all trials that met the acceptance criterion and associated $UCL_{\bar{P}}$, for each particle type except SiO_2 are given in Table 2. The $\bar{P}_{max}$ for NaCl was very close to 5% as expected since this particle type is accepted for use in testing N95 respirators (42 CFR 80). The penetration for TiO_2 was likewise below 5%, however those for SWCNT particles and Fe_2O_3 exceeded 5%. A two-sided t-test for $\alpha = 0.05$ conducted to compare the $\bar{P}_{max}$ values obtained for the two particle types which met the PAC for all ten trials, NaCl and TiO_2, resulted in no significant difference ($p = 0.091$).

DISCUSSION

This paper combines two objectives, (1) the development of a statistical method for evaluating the uncertainty of a calculation of particle penetration through a filter relative to the magnitude of the upstream and downstream counts used to make that calculation, and (2) the use of that method when evaluating the penetration of engineered nanoparticles through an N95 filter. After a thorough review of the scientific literature concerning filter performance, no article could be found in which Poisson count statistics in the manner described here were used to generate a measure of uncertainty in filter penetration values. Although not in the scientific literature, as mentioned above, the statistical techniques required obtain a confidence limit about a measure of penetration are incorporated into the methods for filter testing established by two standard setting agencies (IEST 2007, BS EN 2009). However, these methods do not include an acceptance criterion as suggested here, which is therefore a novel concept that cannot be compared to similar criterion. The criterion of $UCL_P \le 2P$ is admittedly broad as it implies that the true P may be as high as twice the measured P, but was adopted here to compensate for an inability to adjust the GM of the nanoparticle aerosols to a value closer to the MPPS. If this method were to be applied to filter testing using a salt solution, for example, then it would be reasonable to make the criterion more conservative, say $UCL_P \le 1.25P$.

The second statistical technique used in this study, described in ASHRAE 52.2 (2007), which involves the development of a confidence interval about P values measured from multiple trials (Eq. 9), has been used in some form by many other researchers (e.g. Balazy et al., 2006; Cho et al., 2010; Kim et al., 2007; Rengasamy et al., 2009). The resulting error bars about penetration (or efficiency) values shown in plots in these papers indicate the precision in the repetitive measurements made for each particle size. However, in addition to stipulating a method for determining a confidence interval about a set of P values (as partially shown in Eq. 9), the ASHRAE method also applies a criterion that forces an increase in the number of trials performed above at least n = 3 so that $UCL_P - \bar{P} \le f\,\bar{P}$ where f is a fraction of value 0.07, 0.15, or 0.20 applied to particle size ranges in three groups of instrument size channels from smallest to largest diameters as measured by an optical particle counter between 0.3 – 10 μm. This method therefore applies an acceptance criterion to increase the precision of the estimate of $\bar{P}$ by increasing the number of trials. Similar methods to ensure an adequate number of samples based on a predetermined desired level of precision have been used, for example, in studies involving counting bacterial cells (Montagna, 1982), and is the basis for establishing the minimum number of fields to view when performing a count of asbestos fibers according to NIOSH Method 7400 (NIOSH, 2003).

The use of an acceptance criterion based on Poisson count statistics when evaluating the penetration of various nanoparticle types resulted in the exclusion of data from one particle type, SiO_2. The nanoparticles that resulted from a nebulized suspension of the bulk powder apparently agglomerated to the extent that there were insufficient counts in the size channels bounding the MPPS. At least four of the ten trials performed when aerosolizing the other three nanopowders (SWCNT, Fe_2O_3, TiO_2) resulted in counts high enough to meet the criterion. Extensive research was performed by Schmoll et al. (2009) on factors affecting the size distribution and concentration of aerosols produced by nebulizing a powder suspension. They initially discovered that contaminant particles are created from the nebulization of processed water alone with a peak near 20 nm, presumably by the creation of salts from solutes that cannot be removed from the water. The addition of a nanopowder then resulted in a bimodal aerosol distribution retaining a peak at 20 nm and a second one consisting of agglomerated primary nanoparticles typically near 110 nm. Furthermore, the suspension concentration affected the size distribution where a high concentration overwhelmed the production of contaminant particles derived from the water used to make the suspension thus eliminating the first peak. Increasing the suspension concentration also increased the concentration of the aerosol in the test chamber and also increased the GM size presumably by additional agglomeration. The SiO_2 and TiO_2 distributions shown in Figure 5 demonstrate this effect where the suspension used, 6.7 mg mL^{-1} was sufficient to eliminate the 20-nm peak. However, an equally high Fe_2O_3 suspension concentration did not eliminate that peak which can be seen in the first mode of the distribution resulting from that aerosol. Schmoll et al. (2009) also demonstrated that sonicating the suspension from 5 to 20 minutes did not achieve a desired reduction in the GM of the resulting aerosol over that when the suspension was not sonicated. Therefore, attempts to reduce the GM of these aerosols closer to that of the MPPS were not successful for the production system used in this study. Therefore, attempts to study the filtration performance of respirators with engineered nanoparticles is hindered by the need to produce high counts which increases agglomeration resulting in non-ideal size distributions relative to the P-curve of respirators.

The average maximum penetrations for SWCNT and Fe_2O_3 exceeded 5% whereas those for NaCl particles were slightly below 5%. Rengasamy et al. (2009) found 20-nm and 30-nm NaCl particles to have a slightly higher penetration than that of similarly sized silver nanoparticles. However, they also found a significant interaction between nanoparticle type and N95 manufacturer, which suggested that penetrations differed by respirator manufacturer. In this study, we found rather large differences in maximum penetration for one FFR type challenged with the same aerosol. For example, the $\bar{P}_{max}$ for SWCNT was 6.7% (Table 2) but the range in P_{max} values for the ten trials was 3.4 – 9.9%. $\bar{P}_{max}$ for Fe_2O_3 was also above 5%. We did not perform 20 trials as stipulated in 42 CFR 84, but comparisons with that method are of limited use here as it is based on a pass-fail criteria rather than the evaluation of an average penetration. Further study is needed to determine whether there are morphological or other characteristics of these engineered nanoparticles that would cause them to break through the filter mesh of an N95 FFR with higher penetration than the expected 5%.

The MPPS values obtained in this study ranged between 35 – 53 nm. For comparison, the study by Rengasamy et al. (2009) resulted in an MPPS of 40 nm for NaCl compared to a value of 53 nm from this study. These MPPS values are small compared to the conventional expectation that respirators have an MPPS near 300 nm. As explained by Stevens and Moyer (1989) that MPPS value was based on theoretical calculations of fiber efficiency and, furthermore, is an aerodynamic diameter. In studies which utilized an SMPS (Balazy et al., 2006; Rengasamy et al., 2009), the MPPS is expressed as a mobility diameter which will have a smaller diameter for materials with a density > 1 g cm^{-1}. For example, Stevens and Moyer (1989) measured the MPPS for four different electret "dust and mist" respirator when challenged with NaCl at 85 L min^{-1} resulting in values between 40 – 100 nm count median mobility diameter. In addition to the diameter type , the MPPS is also influenced by the electrostatic qualities of the electret filters commonly used in modern N95 FFRs, such as the one used in this study, as well as the charge state of the particles and filtration velocity . Other particle penetration studies utilizing electret filters have shown that the highest penetration will be for the condition when using a discharged filter capturing uncharged particles, and the lowest penetration when capturing charged particles with a charged filter ((Romay et al., 1998)). Kanaoka et al. (1987) showed that the MPPS will vary from a high value near 300 nm mobility diameter when capturing charged particles and a low value < 60 nm when capturing uncharged particles. The use of an electret filter in combination with a statically discharged aerosol in this study resulted in a combination of low penetration and low MPPS.

CONCLUSIONS

A survey of the literature resulted in a method for calculating an upper confidence limit of a filter penetration value based on Poisson count statistics. This method can be adopted to establish a level of uncertainty for establishing the validity of a penetration value based on upstream and downstream particle counts in a given size bin. The probability of having valid penetration values in all size bins reported by a multi-channel particle counter such as an SMPS will be maximized if the median diameter of the aerosol used to test the filter is equivalent to the most penetrating particle size (MPPS) of the filter. A second statistical analysis based on multiple trials is also suggested to determine an upper confidence limit of the average of penetration values obtained for each size bin. When these statistical methods were applied to the penetration of sodium chloride particles through an N95 respirator a relatively low level of uncertainty was obtained when predicting the maximum penetration and MPPS because the median sodium chloride particle diameter was close to the MPPS. However, there was higher uncertainty when determining the penetration of some nanoparticle types due to an inability to prevent the agglomeration of the particles that resulted in count distributions with a median diameter much larger than the MPPS. Maximum penetration values for nanoparticles were within 2% of the allowed level of 5%. No significant difference was found between the average penetration obtained from one of the engineered nanoparticles, SWCNT, and that of sodium chloride.

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NOMENCLATURE

$\mathbf{C}$	particle count concentration (cm^{-3})
$\mathbf{C_d}$	count concentration downstream of a device (cm^{-3})
$\mathbf{C_u}$	count concentration upstream of a device (cm^{-3})
δ	pooled standard deviation of multiple P values (%)
$\mathbf{E}$	efficiency (%)
$\mathbf{k}$	sample size of multiple trials
λ	mean count concentration (cm^{-3})
$\hat{\lambda}$	measured estimate of λ (cm^{-3})
$\mathbf{LCL_C}$	lower confidence limit of a single measure of C (cm^{-3})
$\mathbf{LCL_{Cu}}$	lower confidence limit of a single measure of C_u (cm^{-3})
$\mathbf{n}$	non-negative integer particle count
$\mathbf{n_{down}}$	non-negative integer particle count downstream of a device
$\mathbf{n_{up}}$	non-negative integer particle count upstream of a device
$\mathbf{P}$	penetration (%)
$\bar{\mathbf{P}}$	Mean penetration of series of trials (%)
$\mathbf{P_{max}}$	maximum penetration from all size bins measured during a single trial
$\bar{\mathbf{P}}_{\mathbf{max}}$	mean of maximum penetration values from multiple trials

UCL_{C}	upper confidence limit of a single measure of C (cm^{-3})
UCL_{Cd}	upper confidence limit of a single measure of C_d (cm^{-3})
UCL_{P}	upper confidence limit of a single calculation of P (%)
$\mathrm{UCL}_{\bar{P}}$	upper confidence limit of $\bar{P}$ (%)
V	air volume (cm^3)

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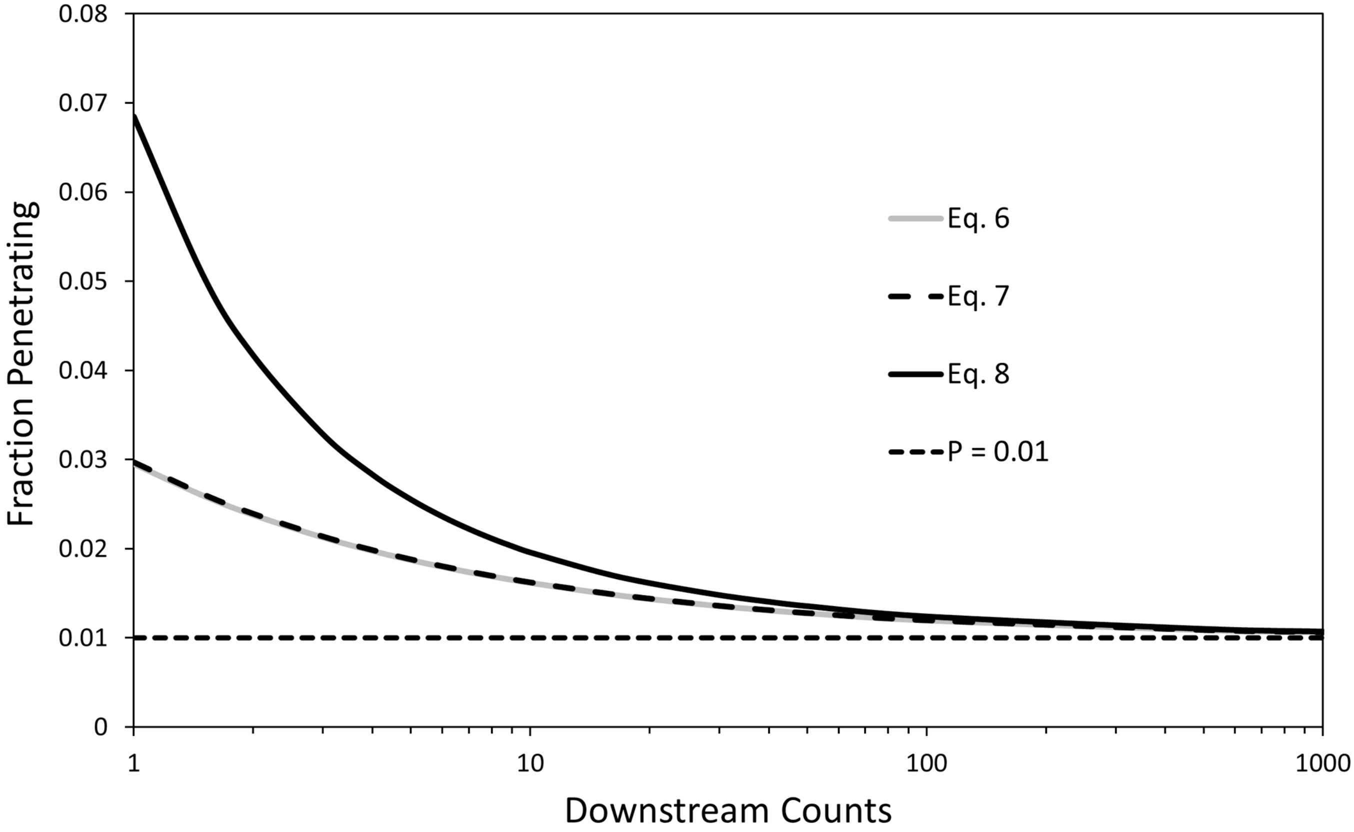


Figure 1.
95% upper confidence intervals for methods designated by their text equation number.

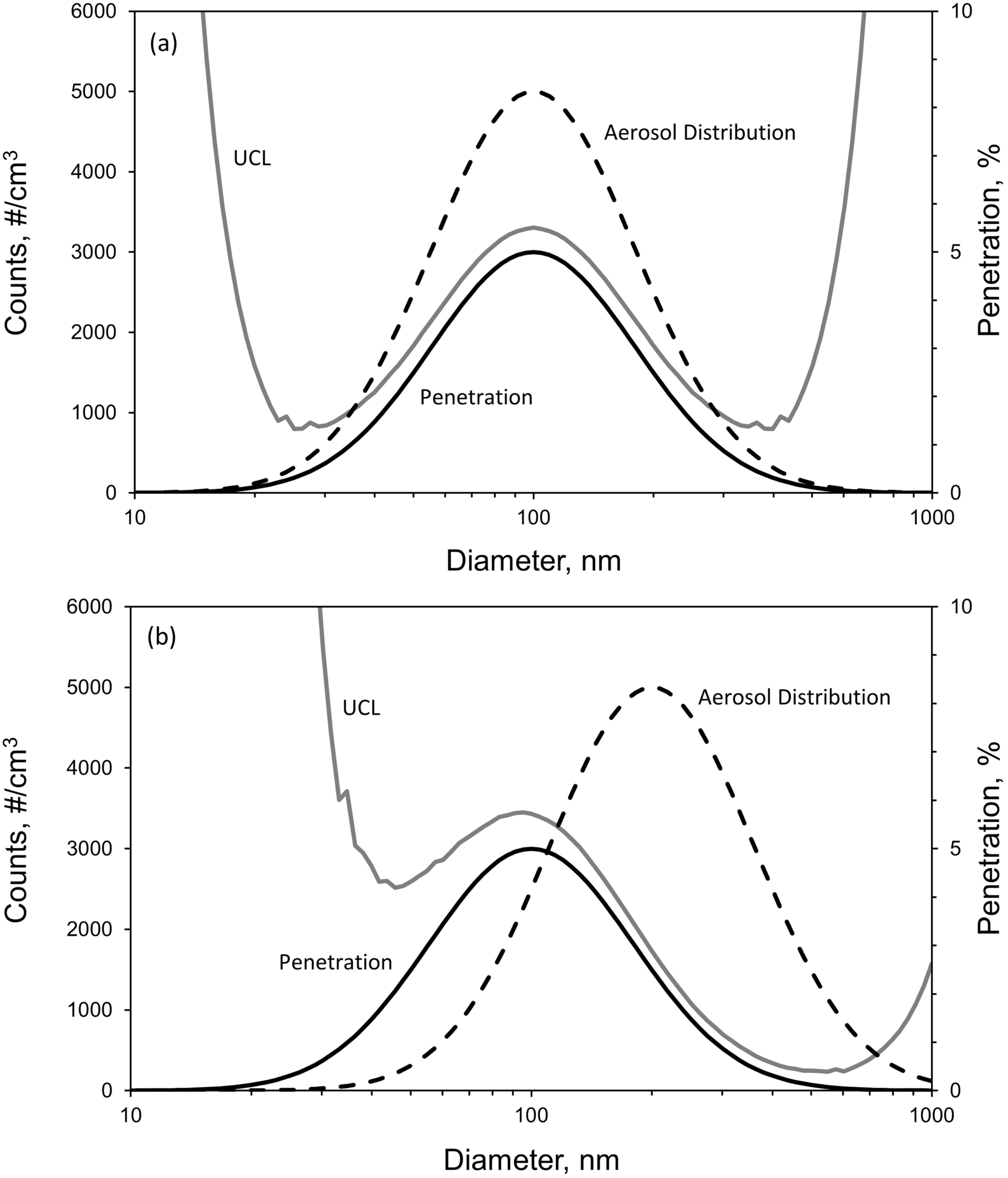

(a)
UCL
Aerosol Distribution
Penetration
Counts, #/cm3
Penetration, %
Diameter, nm
0
1000
2000
3000
4000
5000
6000
0
5
10
10
100
1000
(b)
UCL
Aerosol Distribution
Penetration
Counts, #/cm3
Penetration, %
Diameter, nm
0
1000
2000
3000
4000
5000
6000
0
5
10
10
100
1000

Figure 2.

Hypothetical upper 95% CIs developed from two methods when (a) the GM of the aerosol coincides with the MPPS of the filter, and (b) when the GM of the aerosol is 100 nm greater than the MPPS of the filter.

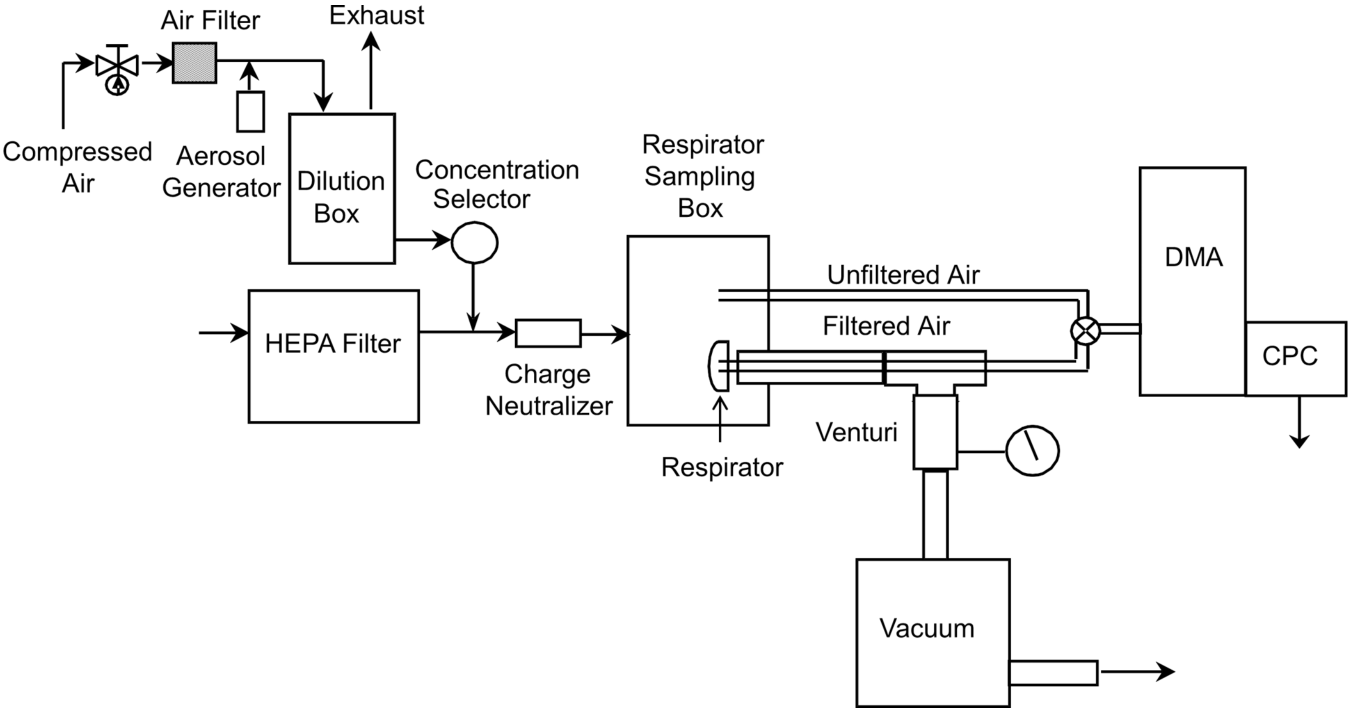


Figure 3.
Respirator testing system.

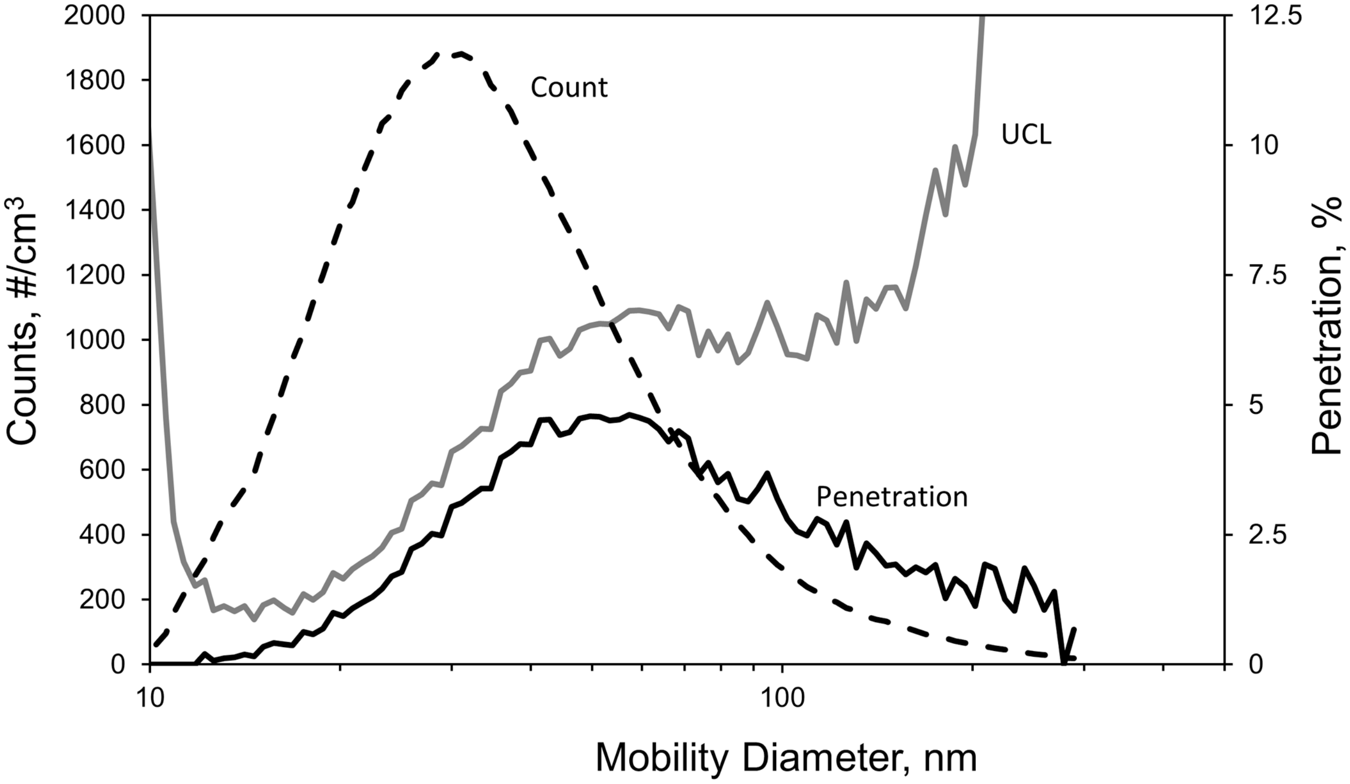
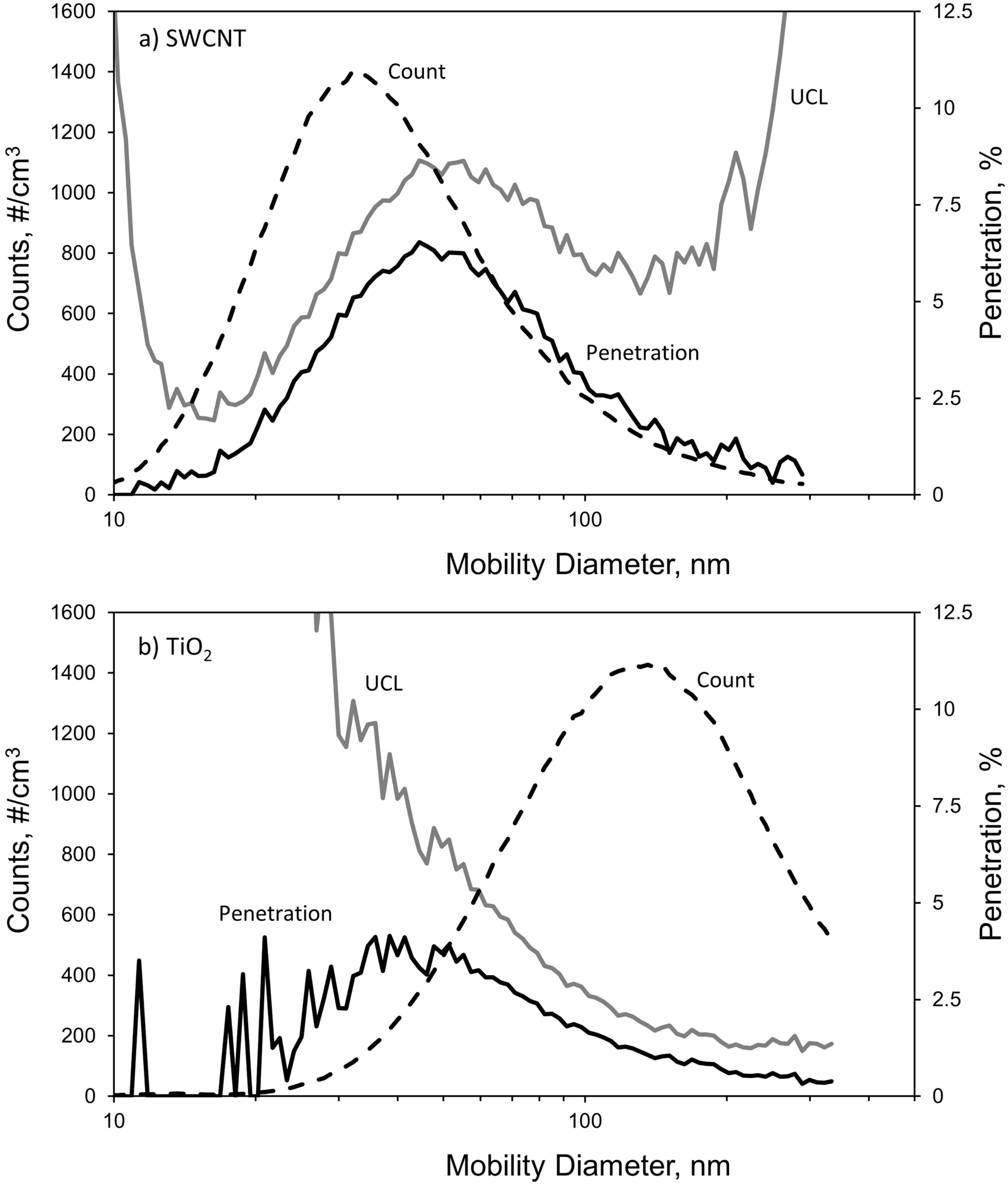


Figure 4.

Count distribution, penetration, and upper confidence limit (UCL) for an N95 respirator challenged with NaCl particles.


a) SWCNT
Count
UCL
Penetration
Counts, #/cm^3
Penetration, %
Mobility Diameter, nm
0
200
400
600
800
1000
1200
1400
1600
0
2.5
5
7.5
10
12.5
10
100
b) TiO$_2$
UCL
Count
Penetration
Counts, #/cm^3
Penetration, %
Mobility Diameter, nm
0
200
400
600
800
1000
1200
1400
1600
0
2.5
5
7.5
10
12.5
10
100

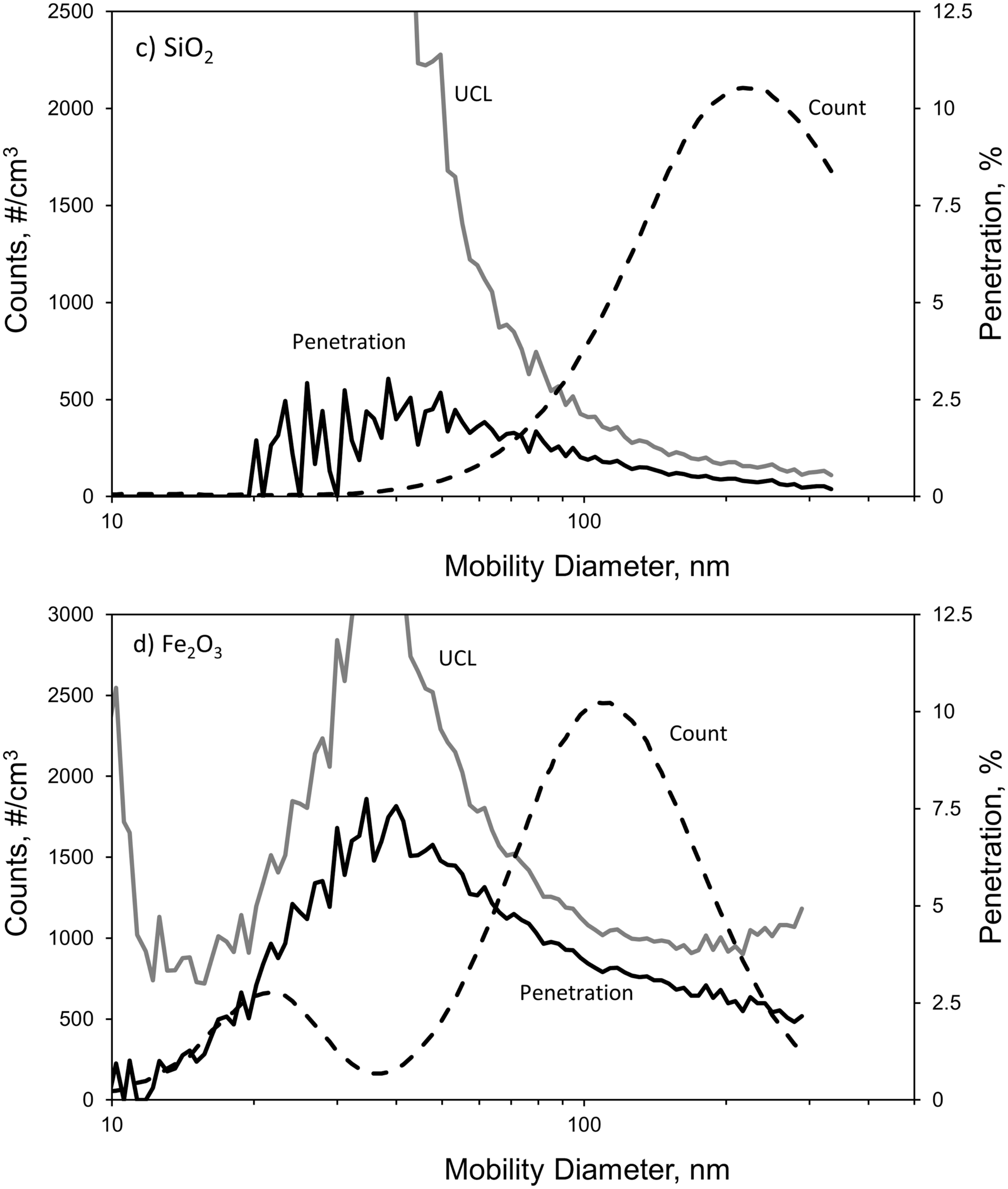

c) SiO_2
Counts, #/cm^3
Penetration, %
UCL
Count
Penetration
0
500
1000
1500
2000
2500
0
2.5
5
7.5
10
12.5
10
100
Mobility Diameter, nm
d) Fe_2O_3
UCL
Count
Penetration
0
500
1000
1500
2000
2500
3000
0
2.5
5
7.5
10
12.5
10
100
Mobility Diameter, nm

Figure 5.

Count distribution, penetration, and upper confidence limit (UCL) for an N95 respirator challenged with a) SWCNT, b) TiO_2, c) SiO_2, and d) Fe_2O_3 particles.

TABLE 1

Upstream counts needed to achieve a specified upper confidence limit relative to penetration.

	Penetration		
UCL	**5%**	**1%**	**0.03%**
2.00xP[1]	230	910	25400
1.50xP	700	2800	74600
1.25xP	2310	9300	265000
1.10xP	12500	51000	1400000

[1] Upper confidence interval expressed as multiples of the penetration (P)

TABLE 2

Filter penetration summary for four engineered nanoparticles and NaCl.

Particle	Constraint Index[1]	n[2]	$\bar{P}_{max}$[3] %	UCL_P[4] %	MPPS nm
NaCl	1.4	10	4.9	6.3	53
SWCNT	1.3	10	6.7	8.5	50
Fe_2O_3	2.0	6	7.0	7.9	35
TiO_2	2.2	4	4.0	4.8	46
SiO_2	7.8	0	---[5]	---[5]	---[5]

[1] Mean ratio of UCLp divided by the maximum penetration for ten trials

[2] Sample size of index values meeting the acceptance criterion: UCLp < 2P

[2] Mean of maximum penetrations oftrials meeting the criterion

[3] 95% upper confidence limit of the mean of trials meeting the criterion

[4] Mean of most penetrating particle size of trials meeting the criterion

[5] No trial met the acceptance criterion: UCLp < 2P