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Preferred sampler inlet configurations for collection of aerosolized nano-scale materials

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ABSTRACT

Background: Due to the lack of standard industrial hygiene sampling protocols for collection of nano-scale materials, sampling inlet device selection is left to individual researchers and professionals.

Objective: The objective of this study was to compare nano-scale aspiration efficiency for common inlet configurations with that of an open-ended sampler tube that is a commonly used inlet for direct reading instruments such as a condensation particle counter.

Methods: A polydisperse aerosol was generated using an electric motor as the aerosol source. Typical aerosols generated by this method produced particles with geometric mean mobility diameters of approximately 30 nm with geometric standard deviations of approximately 2. Comparison of raw particle counts in size ranges measured with a scanning mobility particle analyzer was made by determining the fractional difference between the selected inlet and that of the open-ended tube.

Results: Particle size distributions were nearly identical for all inlet types. The same held true for numbers of particles collected with the exception that the needle inlet was highly variable.

Conclusions: When completing air monitoring for nano-scale materials, inlets on most collection devices (filters, tubing) do not impact aspiration efficiency. This means that it is not necessary to match inlet configurations when using multiple methods to collect and analyze nano-scale materials.

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Background

As the study and application of submicron particles (nano-scale and ultrafine) in research and industrial processes rise, the endeavor to effectively collect and characterize nano-scale particles has grown in interest. Currently, there is no generally accepted industrial hygiene standard sampling method for nano-scale aerosols[1]. In addition, no occupational exposure limits explicitly exist for nano-scale materials due to the limited toxicological data and the plethora of materials used. Since the toxicological health impacts are widely unknown, the type of air sampling (e.g. surface area, particle count, mass, and/or charge) to conduct is also in question. In many instances, air monitoring may be conducted by multiple methods to potentially correlate prospectively with toxicological studies [2–4].

From a traditional industrial hygiene perspective, the inherent size of nano-scale materials are classified as respirable aerosols, defined by the American Conference of Governmental Industrial Hygienists sampling convention and captured via air sampling with cyclones and filters[5]. A more comprehensive system to characterize f nano-size aerosols typically requires the use of some form of particle counting using a condensation nucleation process such as a condensation particle counter (CPC) or sizing using a scanning mobility particle analyzing system (SMPS). To determine if the aerosol thusly monitored is composed of the nano-scale material of interest, the aerosol must be collected in quantity and analyzed separately using multiple analytical techniques. In cases such as these, issues of sampling comparability become important considerations. This study investigates typical inlet types on industrial hygiene air sampling equipment to determine differences that may impact data (particle size and/or count) that is collected. For example, assigning a particle count from a direct reading instrument as "nano" based on an examination of particle morphology or chemistry as determined from an open or closed faced filter sample. A thorough analysis of a nano-scale aerosol requires completing air monitoring from a variety of analytical instruments.

Studies in aerosol science have focused on effects of samplers with larger (micron sized) or fibrous materials to provide theoretical background for sampler aspiration efficiency, but have not fully addressed

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nano-scale materials [6–9]. Particle samplers are known to possess an inherent sampling efficiency dependent on the physical characteristics of the particles, inlet velocity and inlet configuration, among others [10,11]. It has been suggested that nano-scale particles in the free molecular flow regime (Figure 1) are less dependent on these characteristics[7], presumably because of their relatively low inertia compared to larger particles. It follows that nano-scale particles, many of which are smaller than typical smoke particles, are moved in the direction and speed of air currents, as observed by the movement of a smoke plume in an ambient air flow. Therefore, for nano-scale particles, their low Stokes numbers suggest that inlet configuration should have little effect on measured particle size distribution. Calculation of inlet "Aspiration Efficiency" [10] for the open-ended tube and test inlets was performed using the freeware aerosol calculator provided by Baron[12]. All inlet aspiration efficiencies calculated thereby were near unity for all nano-scale particle sizes.

Nano-particle movement within a parcel of moving air is also affected by Brownian diffusion as well as other forces (e.g., electrical, thermal). The air molecule/particle collisions necessary to change a particle's direction occur at a rate of about 10^{12}/s for a 10-nm particle, and while net displacement is zero, root mean square displacement is greater than zero[13]. These considerations, while important for passive sampling, play a much smaller role in active sampling.

Based on the lack of standard industrial hygiene air monitoring techniques for nano-scale materials, as well as the potential difference in aerosol behavior for nano-scale materials, an analysis of sampler collection efficiency was developed to better refine industrial hygiene sampling techniques. This work examined air sampling inlets for an open-faced cassette, closed faced cassette and a restricted inlet diameter, e.g. a syringe needle. Collection by these inlets was then compared with a typical inlet on a direct reading instrument (represented as an open-ended tube at a typical flow rate of 1 lpm). A needle configuration was thought to represent an extreme case of the open-ended inlet velocities one might encounter if using a virtual impactor to remove large, non-nano sized particles, on various direct reading instruments such as a CPC or SMPS. A commonly applied working definition of "nano-scale particle" refers to one with a physical diameter of 100 nm or less in any dimension whether single or agglomerated [14]. This study examined aspiration efficiency of various inlet configurations when sampling nano-scale materials with mobility diameters up to 100 nm, extending well into the transition regime (Figure 1). Ultimately, this study considered the applicability of traditional industrial hygiene collection methods for use with nano-scale materials and their respective comparability.

Material and methods

Aspiration characteristics of three inlet geometries including a needle as a point source (N), 25-mm closed-face sampling cassette (CFC), and 25-mm open-face sampling cassette (OFC) were experimentally measured and compared to that of a standard half centimeter thick wall conductive open-ended

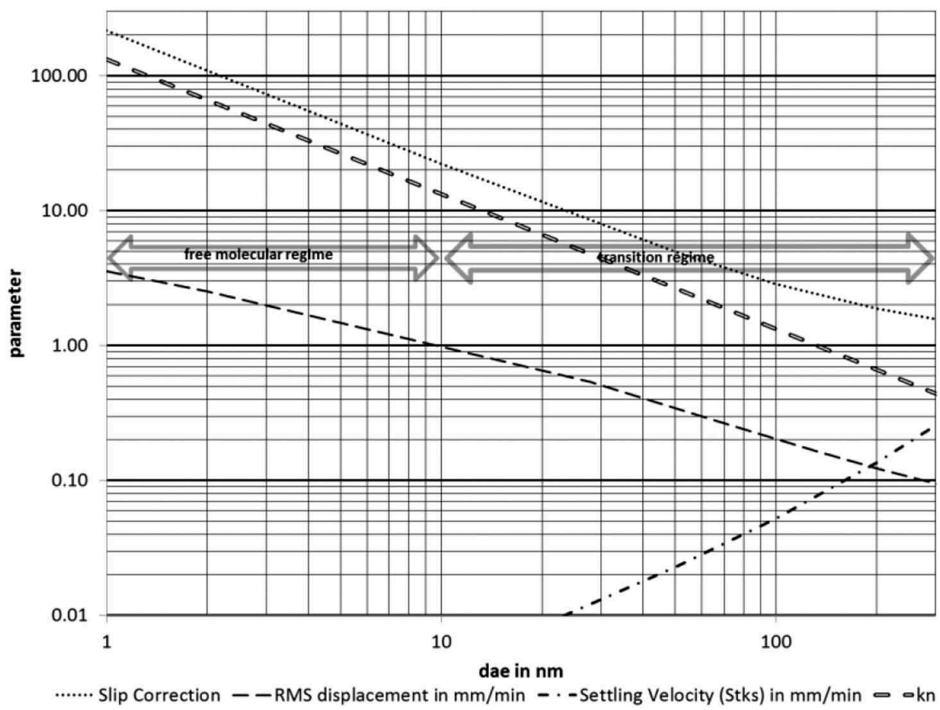


Figure 1. Aerodynamic equivalent diameter (Dae) Aerosol Parameters for nano-scale particles (Re $\ll 1$).

tube 0.5 cm in diameter (T) serving as the reference standard inlet for direct reading instruments.

A polydisperse carbon/copper aerosol (Figure 2a) was generated from the electrical contacts (brushes) of a small electric motor housed in a generation chamber stirred with filtered room air (Figure 2b). Particle emission was the result of a combination of spark emission and physical abrasion. The aerosol was then fed into a secondary mixing chamber where the inlets were housed for sampling (inlets facing downward) with a TSI model 3696 Scanning Mobility Particle Sizer (SMPS) and model 3080 classifier (Figure 2b). Tubing length, wall thickness, and diameter was the same for all trials. Although the tubing was composed of conductive material, it was not grounded during sampling. Sample collection and particle analysis were performed at a flow rate of 1 l per minute (lpm). This is the common flow rate of many commercially available particle counters. During the aerosol generation process, a Naneum Nano-ID™ NPS500 in CPC mode was used to monitor and ensure a consistent particle concentration inside the aerosol mixing chamber. Particles from the SMPS were then counted by a TSI™ model 3772 CPC and separated into particle bins, which were graphed using TSI 2009 Aerosol Instrument Manager (AIM) software. Data were exported to an Excel™ spreadsheet. Data analysis included particle size distribution and collection efficiency relative to the tubing inlet.

Simultaneous sampling by both the sample inlet and open-ended tube (reference) was not possible with only one SMPS available. Therefore, the sampling methodology employed was to alternate consecutive sampling in the manner of Tube – Inlet – Tube – Inlet and so on. Averaged values for each inlet type (n = 20) were then compared.

Centerline flows were too low to measure with available direct reading instruments, therefore, a two dimensional computational fluid dynamics (CFD) program (Tdyn 1.2™) based on the Finite Element Method was used to solve the Navier–Stokes equations describing the air flow into and around the various inlet configurations[15]. Air flow pattern entering the sampler inlet in combination with the average particle right angle displacement by diffusion was used to estimate an effective capture distance (ECD) which we define as the proximity of the source to sampler distance necessary for source sampling. The ECD is irrelevant for ambient sampling in well mixed environments.

The ECD was derived as follows:

- Inlet velocities at various centerline distances ($f(x)$) from the inlet were determined from the CFD model. Velocity (V) was plotted as a function of distance (X) and fitted to a power function and the equation of the line determined by a spreadsheet trend line function in Excel™.
- The velocity function was integrated between the distance at which inlet velocity was equal to 5 cm/s and the velocity at one inlet diameter (D) for each of the four inlets to obtain their average inlet velocities.

$$\bar{V} = \frac{dv\int_D^x f(x)dx}{\Delta x}, \quad \text{cm/s particles with mobility diameter up to 300 nanometers,} \tag{1}$$

- The time a 1-nm particle could be displaced a distance of $D/2$ cm, where D is the inlet diameter and 0.32227 is the rms displacement in the first second for a 1-nm particle at right angles to the inlet centerline.

$$t = \left(\frac{\frac{D}{2}}{0.3227}\right)^2, \text{s} \tag{2}$$

- The average velocity from Equation (1) is used to determine the first ECD (Equation (3)) for the initial x condition stated above. The ECD thus obtained is substituted for x back into Equation

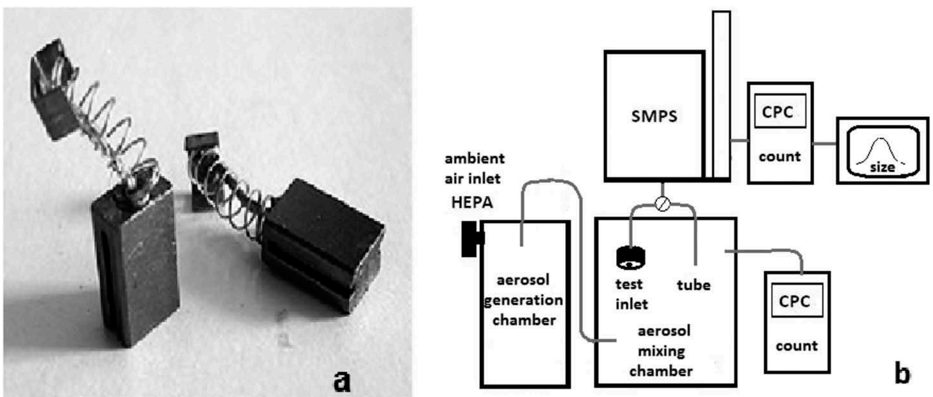


Figure 2. Aerosol generation and sampling system.

(1) to obtain an adjusted average velocity. This iteration is repeated until x and ECD converge.

$$ECD = \bar{V}\left(\frac{\frac{D}{2}}{0.3227}\right)^2, \text{cm} \quad (3)$$

Results

The experimental aerosol particle aspiration characteristics in terms of size distribution are presented in Figure 3, for each type of inlet. The nano-scale particle size distributions were essentially the same for all inlet types. In terms of particle number concentrations (Table 1), the needle inlet configuration was the only inlet tested that over-collected particles with respect to the open-ended tube; however, that difference did not achieve statistical significance. The closed face and open face cassettes slightly under sampled, and the differences (−3 and −6 percent, respectively) did achieve statistical significance, but may be of little practical significance. A compilation of sampler inlet velocity characteristics determined by CFD and particle size estimates measured by SMPS is found in Table 2.

Discussion

Evaluating sampling data from an industrial hygiene perspective requires thoughtful consideration of the material being sampled, controls used and the ambient conditions (background aerosol, air turbulence, and mixing). The lack of occupational health exposure limits and specific sampling techniques requires investigators to identify the presence/absence of nano-scale material by looking for chemical, morphological or numerical count differences from the background aerosol. Linking the presence of the non-background material with time and motion studies provides an indicator of exposure. Filter samples are particularly advantageous when electron microscopy with analytical capabilities is needed to confirm/deny a specific nano-scale material's presence/absence in the ambient aerosol.

The choice of inlet (Figure 3 and Table 2) has minimal impact on aerosol collection in terms of either size distribution or number concentration for particles in

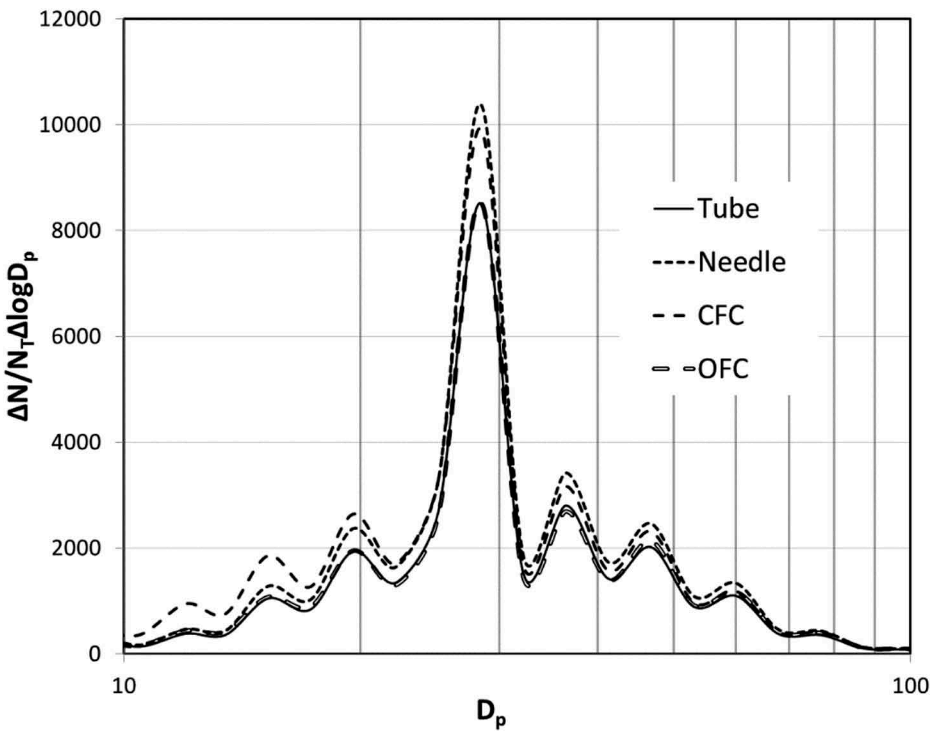


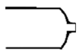
Figure 3. Single comparison of particle size distributions by inlet type.

Table 1. Particle counts: t-test paired two sample for means ($\alpha = 0.05$), $n = 20$.

Inlet	% Difference in mean particle count	P (two tail)	Statistically different
Tube (reference)	–	–	–
Open face cassette	−6	<0.01	Yes
Closed face cassette	−3	<0.01	Yes
Needle	+22[a]	0.14	No

[a] This value corresponds to the data from Figure 3. Needle collection varied from −51% to 128%

Table 2. Inlet velocity characteristics.

Inlet type velocity vectors	$f(x)$	Inlet diameter "D" (cm)	X @ V = 5 cm/s (cm)	$\bar{V} = \frac{dv\int_0^x f(x)dx}{\Delta x}$ (cm/s)	$ECD = \bar{V}\left(\frac{\frac{D}{2}}{0.3227}\right)^2$ (cm)	Expected aspiration efficiency	Geometric mean particle Size, nm and (geometric standard deviation)
Thick-walled tube 	$V = 41.8\ d_x^{-1.004}$	0.5	3	14	9	Reference	30.9 (1.89)
Open faced cassette 	$V = 10.6\ d_x^{-0.601}$	2.5	3	2	32	0.94	31.1 (1.90)
Closed face cassette 	$V = 32.3\ d_x^{-0.812}$	0.4	3	17	6	0.97	29.4 (1.90)
Needle 	$V = 21.3\ d_x^{-1.1}$	0.07	2	54	1	1[a]	28.3 (1.76)

[a]The trend was for increased collection but differences did not achieve statistical significance.

sizes extending well into the transition regime (Figure 1). When simple direct reading of aerosol concentration/particle size of nano-scale particles is required, the unaltered open-ended tube inlet of the instrument is a reasonable inlet configuration to use without concern that the particles are being size selected. Either the closed or open face cassette can be assumed to collect the aerosol represented in the concentration determination by the open-ended tube without regard to matching inlet velocities (Table 2) which means that the aerosol concentration determined with a hand-held CPC or personal sampling pump can be represented in terms of size, shape and composition determined from filter collection, if the proximity to the source is considered. Furthermore, the open-ended tube inlet of most direct reading instruments does not require modification to match up with other concurrent air sampling devices such as the open or closed face cassette, unless pre-separation of larger particles to exclude them from filter collection is desirable. A needle configuration for nano-scale particle collection, although not tested in this study, could act as an inertial separator for Stokes regime particles minimizing large particle masking of smaller particles on photomicrographs.

Further enhancements to this investigation should simultaneously measure penetration of the test inlet and the reference inlet with their own SMPS, varying concentrations, and replacing the polydisperse aerosol with a series of monodisperse aerosols. These changes would reduce variability allowing for a more definitive determination of real differences between inlets if any. The ability of a needle type inlet to reduce large particle collection should also be explored.

Disclosure statement

No potential conflict of interest was reported by the authors.

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