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PENETRATION OF AEROSOLS THROUGH MINICAM PRECONCENTRATOR TUBES

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The principle goal of this study was to investigate the use of MINICAM preconcentrator tubes (PCTs) as aerosol sampling devices. A series of penetration tests was conducted with a low-volatility liquid aerosol challenge on PCTs preloaded with several candidate sorbent bed materials. The results of this study show that commercial MINICAM PCTs were effective collectors for aerosol particles with aerodynamic diameters greater than 1 µm; however, significant breakthrough occurred for sub-micron aerosols with all four sorbent materials tested. Based on these findings, the use of PCTs for aerosol sampling is acceptable over the restricted range of laboratory-generated aerosol challenges indicated in this report.

15. SUBJECT TERMS

MINICAM preconcentrator tubes	Liquid aerosol challenge	Aerosol penetration
Direct aerosol sampling	Sorbent bed materials	Sampling recommendations

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PREFACE

The work described in this report was authorized under Project No. CA06DET504. This work was started in January 2010 and completed in September 2011.

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PENETRATION OF AEROSOLS THROUGH MINICAM PRECONCENTRATOR TUBES

1. INTRODUCTION

Sorbent beds are commonly used to sample vapors and gases from exposure test chambers to quantify challenge concentrations. The chief advantages of this sampling method include increased sensitivity for evaluation of low-level concentrations and relative ease of use in comparison to bubbler processing (*1*). In general, this sampling approach has not been adopted for the collection of aerosols because of the potential for particle breakthrough, particularly in the submicron size range (*2*). It is possible, however, that the sorbent sampling method could be extended to the sampling of aerosol and mixed aerosol-vapor atmospheres by appropriate selection of sorbent material, bed volume, and packing density and by control of challenge aerosol particle size distributions in a laboratory setting.

A series of detector tests was planned in which the sampling of aerosol test atmospheres was to be conducted using preconcentrator tubes (PCTs) in conjunction with the Miniature Continuous Air Monitoring System (MINICAM). The principal goal of this study was to assess the aerosol filtration characteristics of commercial PCTs preloaded with several candidate sorbent bed materials. The information obtained was used to establish efficacy and guidelines for the use of the PCT as an aerosol-sampling device.

2. EXPERIMENTAL MATERIALS AND METHODS

Using the test system shown in Figure 1, commercial PCTs (3 × 65 mm; Camsco, Inc., Houston, TX) were tested with four types of sorbent bed materials: Tenax TA (35/60 mesh), Tenax GR (60/80 mesh), Carboxen 569 (20/45 mesh), and Chromosorb 106 (60/80 mesh). Aerosols of diethyl sebacate (DES; Sigma-Aldrich, St. Louis, MO) were generated with a single jet Collison nebulizer operated at 20 psig. Output aerosol concentration from the generator was reduced by means of a bleed-off valve and through the introduction of filtered dilution (makeup) air before the aerosols entered a multiport test manifold. Total flow exhausted through the test manifold was approximately 20 L/min. PCTs to be tested were attached to one of the test manifold ports. An aerodynamic particle size spectrometer (APS, model 3321; TSI, Inc., Shoreview, MN) was used to measure aerosol concentration and particle size upstream and downstream to the PCTs, enabling an evaluation of particle penetration in principle over its measurement size range (0.5 to 20 μm in 52 size channels). All tests were conducted at a PCT flow rate of 300 cc/min by supplementing the APS sampling flow rate of 5 L/min with 4.7 L/min of filtered dilution air.

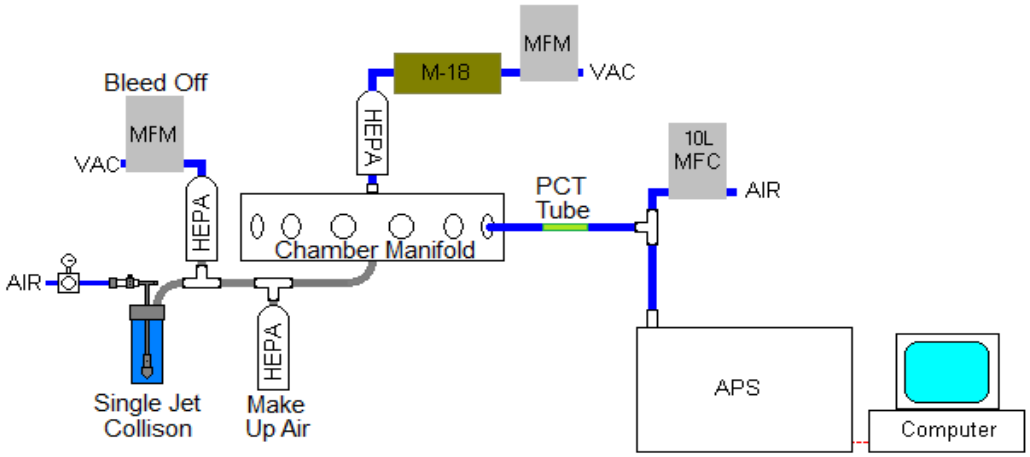

Figure 1. Test apparatus.

Initially, a measurement of upstream particle number concentration (U_{Ni}), as a function of particle size (i), was determined by sampling directly from the test manifold with an open bore tube substituted in place of the PCT. Immediately after this, a candidate PCT was put into position, and the downstream particle number concentration (D_{Ni}) was determined as a function of particle size. PCT flow rate was verified by means of a Gillibrator-2 airflow calibrator (Sensidyne LP; Clearwater, FL) each time a tube was changed. For each concentration measurement, a series of 10 consecutive 20 s APS samples were obtained and averaged. The fraction of aerosol penetrating the sorbent bed for a given particle size (i) was defined as the ratio of the averaged downstream and upstream concentrations for that channel:

$$P_i = \frac{D_{Ni}}{U_{Ni}} \quad (1)$$

Total aerosol penetration through a PCT in terms of particle counts was determined by calculating the ratio of the sum total averaged downstream and upstream channel concentrations for all channels:

$$P_{total\ count} = \frac{\sum_i D_{Ni}}{\sum_i U_{Ni}} \quad (2)$$

The Aerosol Instrument Manager software (version 8, 2006) also provided a calculated upstream and downstream mass concentration by assuming spherical particles and multiplying the particle counts by the particle volume (V_i) in a given particle size bin (i). In this manner, eq 3 allowed a determination of total aerosol penetration in terms of aerosol mass:

$$P_{total\ mass} = \frac{\sum_i D_{Ni} \times V_i}{\sum_i U_{Ni} \times V_i} \quad (3)$$

3. RESULTS AND DISCUSSION

DES challenge aerosols within the test manifold had, on average, a number-weighted median aerodynamic diameter of 0.77 µm with a geometric standard deviation (GSD) of 1.45 (Figure 2). This distribution of particle sizes, which constitutes the upstream challenge aerosol, was sufficiently broad and concentrated in the test manifold to allow accurate assessment of particle penetration through the PCT over a particle size range of 0.5 to 3.0 µm.

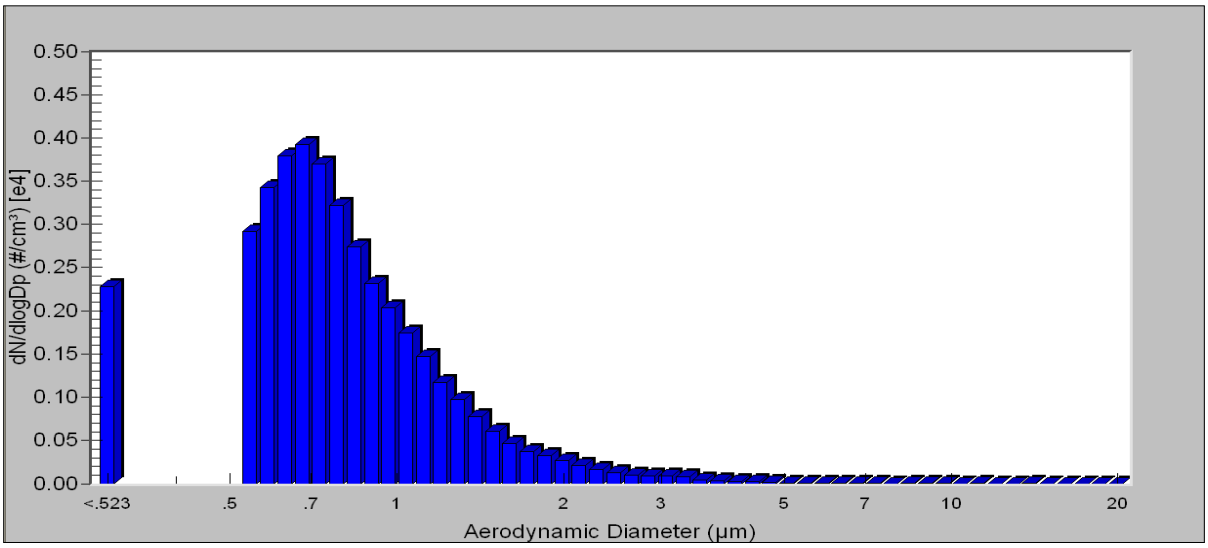

Figure 2. Typical number-weighted distribution of upstream challenge aerosol.

Aerosol penetration through the PCT as a function of particle size is shown in Figures 3 through 6 for each of the four different sorbent-packed varieties tested at a sampling flow rate of 300 cc/min. Each penetration value shown represents the average result from three representative PCTs with standard deviation bars shown. In every case, aerosol penetration was negligible for aerodynamic particle sizes greater than 1 µm. Penetration, however, became significantly more pronounced as particle size decreased below 1 µm. As much as 62% of the aerosol in the 0.542 µm–size channel was not captured in the PCTs packed with Tenax TA sorbent. The penetration fraction in the submicron particle range tended to be greatest with Tenax TA sorbent and least with Chromosorb 106 and Tenax GR; the latter both had a penetration fraction of 37% at 0.542 µm. The penetration fraction for the Carboxen 569–packed PCTs was midway between the high and low values, at 50% in the 0.542 µm–size channel.

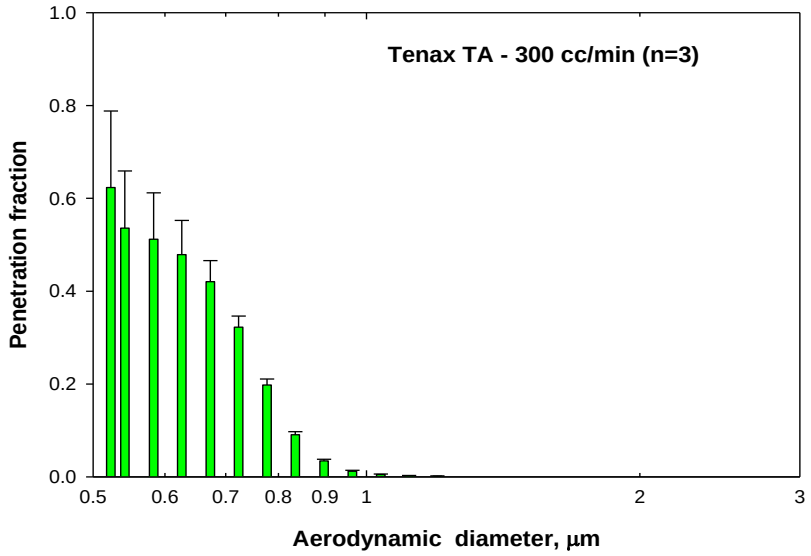

Figure 3. Aerosol filtration characteristics of MINICAM PCTs with Tenax TA sorbent at a sampling flow rate of 300 cc/min (n = 3); standard deviation bars are shown.

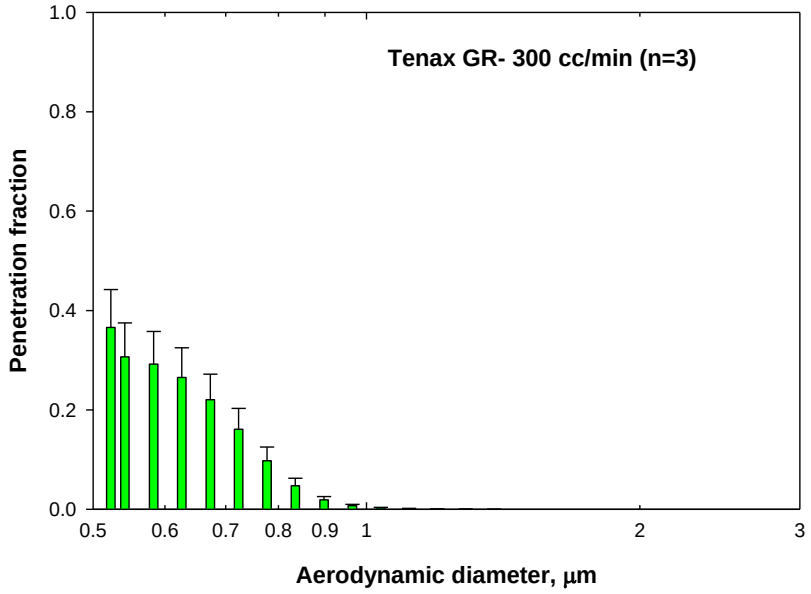

Figure 4. Aerosol filtration characteristics of MINICAM PCTs with Tenax GR sorbent at a sampling flow rate of 300 cc/min (n = 3); standard deviation bars are shown.

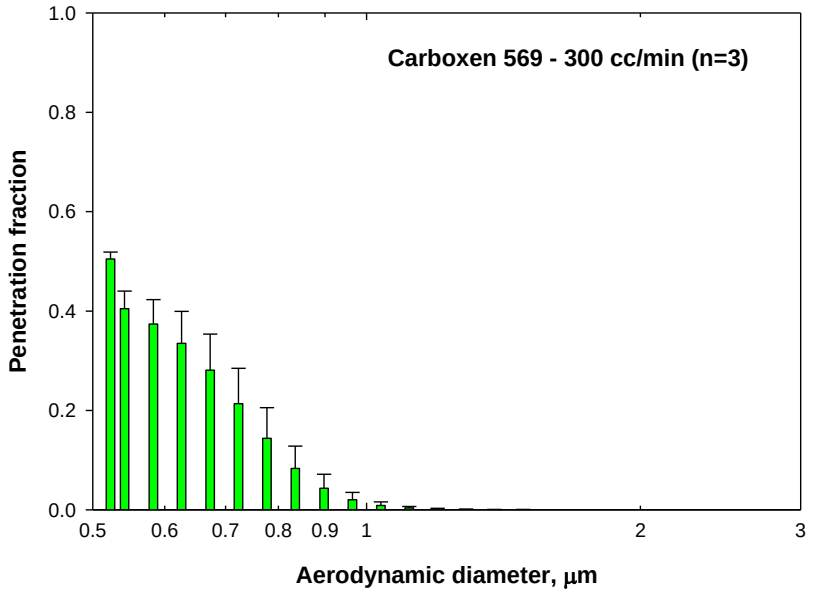

Figure 5. Aerosol filtration characteristics of MINICAM PCTs with Carboxen 569 sorbent at a sampling flow rate of 300 cc/min (n = 3); standard deviation bars are shown.

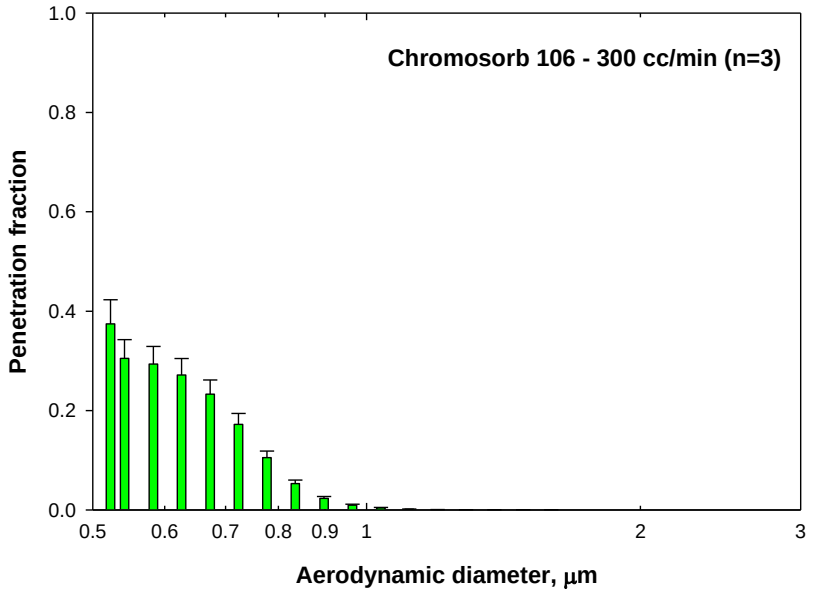

Figure 6. Aerosol filtration characteristics of MINICAM PCTs with Chromosorb 106 sorbent at a sampling flow rate of 300 cc/min (n = 3); standard deviation bars are shown.

Figure 7 depicts the overall mass breakthrough of the DES test aerosols through the various PCTs resulting from the mechanical penetration of particles under the given test conditions. In comparing the different PCTs, overall mass breakthrough was greatest (4%) with Tenax TA and Carboxen 569. The breakthrough for the PCTs containing Chromosorb 106 was 3%. The least mass breakthrough (2.2%) occurred in the Tenax GR–loaded PCTs, and the difference was statistically significant.

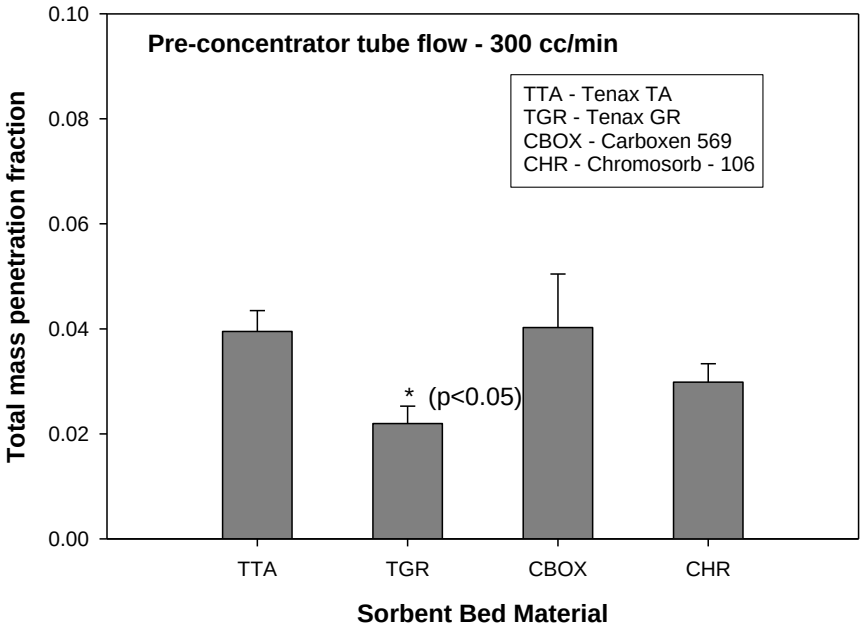

Figure 7. Effect of sorbent bed material on the mass breakthrough of DES test aerosols through MINICAM PCTs at a sampling flow rate of 300 cc/min (n = 3); standard deviation bars are shown; all aerosol challenges were generated with a Collison nebulizer.

In considering the aerosol penetration results, it is important to note that because of the measurement limitations of the APS, it was not possible to assess penetration of aerosol particles smaller than 0.5 µm. Within the given test conditions, it is highly likely that the aerosol penetration fraction increases for particles sizes smaller than 0.5 µm, and the present testing system does not account for the mechanical breakthrough of these smaller particles. In fact, on the basis of previous results for silica gel sorbent tubes (*2*), the most penetrating particle size is probably in the vicinity of 0.1 to 0.3 µm. Hence, the overall fractional penetration of particles in terms of number concentration, i.e., particles per unit volume, was greatly underestimated. Nevertheless, because 99% of the total mass of the DES challenge test aerosols resides in particles with sizes greater than 0.52 µm, in this case, the APS measurements do provide a reasonable account of the mechanical aerosol breakthrough in terms of total aerosol mass.

In accordance with the U.S. Standard Sieve Series, the 60/80 mesh Tenax GR and Chromosorb 106 sorbents consist of a range of granule sizes from 177 to 250 µm, whereas the Carboxen 569 and Tenax TA sorbent sizes range from 354 to 850 µm and 250 to 425 µm, respectively. The trends observed in the PCT aerosol mass breakthrough results show some correlation with the mesh sizes of the sorbent bed materials tested, with the finer grain bed materials being associated with reduced aerosol breakthrough. The fraction of aerosol collected by the glass wool plugs that hold the bed material in place relative to the amount collected by the sorbent bed material is not known at this time; however, it could be substantial. The relative tightness of these plugs could contribute to the variability identified in the mass breakthrough among materials with similar mesh sizes.

Aerosol particles may be captured in the PCTs by a host of filtration mechanisms including direct interception, inertial impaction, diffusion, gravitation settling, and electrostatic effects. At the relatively high PCT sampling flow rate of 300 cc/min used in this study, sorbent bed approach velocity was in excess of 70 cm/s and tube residence time was less than 0.4 s. Under these conditions, inertial impaction and interception are likely the dominant collection mechanisms for particles of 1 µm aerodynamic diameter and larger. This implies that PCT collection efficiency for particles in this range will be reduced at lower sampling flow rates, and significant increases in particle mass breakthrough may occur. Therefore, caution should be used in extending these results to significantly lower sample flow rates.

The aerosol penetration data presented in this report suggest that as long as the aerodynamic size of the particles under test is greater than 1 µm, the PCT is an effective collection device. In practice, real-world aerosols consist of a range of particle sizes. It would be useful to model PCT performance as a function of challenge aerosol characteristics, to establish some guidelines or limits of acceptable operation. To accomplish this, the Tenax TA penetration data (Figure 3) were expressed mathematically using a third-order linear regression ($r^2 = 0.97$). The resulting penetration function was used in combination with various lognormal aerosol distribution functions to calculate the fractional mass of aerosol that would penetrate the PCT. Figure 8 is a compilation of these calculations for a range of hypothetical challenge aerosols of differing mass median aerodynamic diameter and geometric standard deviation. Simply stated, the model shows that as the mass median aerodynamic size of the challenge aerosol increases, a broader size distribution (i.e., larger geometric standard deviation) can be tolerated while a 95% or better collection efficiency is maintained. Conversely, as the median size of the aerosol approaches 1 µm, the aerosol distribution must be tighter if acceptable mass penetration is to be maintained. In summary, if the challenge aerosol characteristics fall within the cross-hatched area in Figure 8, mass collection efficiency is predicted to be greater than 95% on the basis of penetration performance alone. However, potential sampling artifacts can further limit the effective usage of the PCT as an aerosol-sampling tool, as discussed.

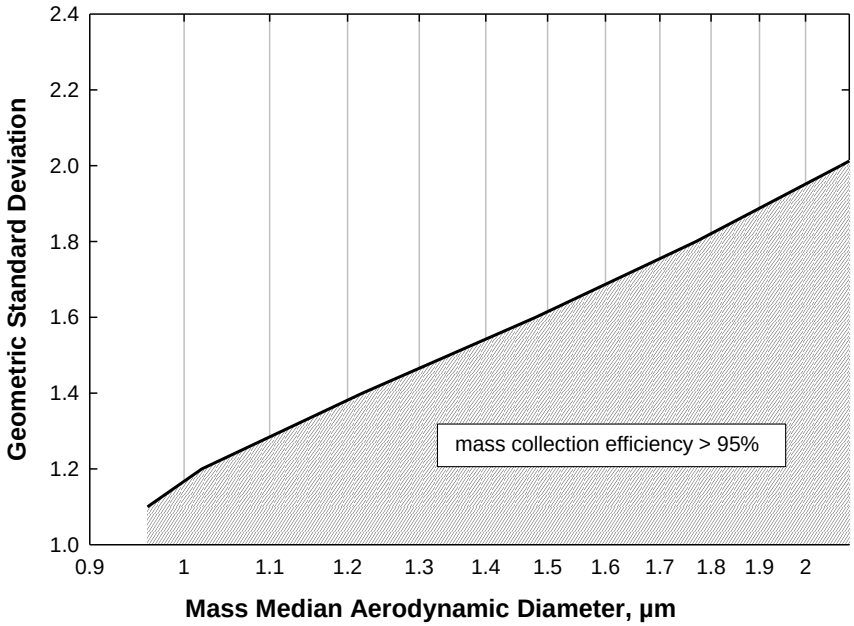

Figure 8. Effect of challenge aerosol size distribution on PCT particle collection efficiency at a sample flow of 300 cc/min. This prediction is based on the performance of PCTs loaded with Tenax TA sorbent and assumes a lognormal distribution for the challenge aerosol. Cross-hatched area represents a "safe operating zone" where the calculated aerosol mass collection efficiency is greater than 95%. Note: This chart is based solely on PCT filtration properties and does not take into account any potential sampling bias due to particle entry effects (see discussion).

Another point worth mentioning is that the small inlet diameter (1 mm) of the PCT creates a potential problem with respect to representative sampling of larger aerosol particles. Larger particles have greater inertia than smaller particles; therefore, larger particles can deviate from the flow streamlines and slip past the inlet to the PCT. This effect can lead to a sampling probe "entry bias" that results in underestimation of the actual concentration of larger particles. Predicting the particle size at which probe entry bias becomes problematic under a given set of sampling conditions is somewhat controversial (*3*). For example, the stringent "Davies sampling criteria" predicts that for the PCT at a 300 cc/min sampling flow, entry bias would begin to occur for particles larger than 3 µm. On the other hand, the Agarwal-Liu criterion predicts that entry bias is not an issue for particle sizes less than 10 µm (*4*) under the same operating conditions. Even in applying the latter, less restrictive criteria, it would not be advisable to use the PCT for sampling aerosols larger than the sizes indicated in Figure 8 without first verifying that sample entry bias is not an issue.

The results of this study are based on a very limited sample size (n = 3) and, in that sense, they may be viewed as preliminary findings. No attempt was made, for example, to examine variations in aerosol penetration from different production batches of PCTs. Increasing the sorbent packing density or simply stacking multiple PCTs in series would almost certainly improve the aerosol filtration characteristics, albeit at the expense of increased flow resistance. Still, such approaches cannot overcome the potential sample entry bias issues previously mentioned. Further work is planned on testing the aerosol penetration characteristics of larger sorbent tubes that could potentially overcome some of the sampling and particle collection issues associated with the PCTs.

4. CONCLUSIONS AND RECOMMENDATIONS

The results of this study show that commercial MINICAM PCTs were effective collectors for aerosol particles with aerodynamic diameters greater than 1 µm; however, significant breakthrough occurred for submicron aerosols with all four sorbent materials tested. The small inlet diameter of the PCT potentially places additional restrictions on its effectiveness as an aerosol-sampling device. In conclusion, the use of PCTs for aerosol sampling, while acceptable over a very limited range of laboratory-generated aerosol challenges, is not recommended for general purpose aerosol-sampling applications.

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