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Comparison of a Passive Aerosol Sampler to Size-Selective Pump Samplers in Indoor Environments

The objective of this work was to investigate the ability of the Wagner-Leith passive aerosol sampler to measure indoor exposures over periods of 24 hours to 2 weeks. An automated analysis technique was developed so that lower aerosol concentrations could be sampled over shorter time periods. A test of the new analytical method against a manual method showed good agreement. The passive sampler was tested alongside three pump-operated, size-selective samplers in indoor environments. Generally, good correlation with the active samplers was observed. Correlation with a personal impactor with uncoated substrates was not statistically significant, but the cyclone, MS&T impactor, and overall correlations had R^2 values of 0.73–0.88. Combining these data with a previous study produced an R^2 of 0.96 between passive and active results. Large discrepancies (up to 147%) between passive and personal impactor results were observed and were attributed to particle bounce in the impactor, passive sampler imprecision due to few collected fine particles, and problems with detection of organic particles in the passive sampler. The Wagner-Leith sampler has now been tested over five orders of magnitude in mass concentration, in which it has proved useful for obtaining aerosol size distributions, mass fractions, qualitative elemental analysis, and morphology of individual particles. The sampler has several limitations, including increased sensitivity to contamination when fewer particles are collected, uncertainties in sampling semi-volatile particles, and the need for some expertise and expense to analyze the passive samples.

Keywords: aerosol sampling, indoor air, passive sampling, sampler comparison, SEM

The passive aerosol sampler described by Wagner and Leith⁽¹⁾ is a potentially useful tool for exposure assessments. The sampler (Figure 1) does not require a pump and is capable of sampling for periods of minutes to weeks, depending on the ambient aerosol concentration. The passive sampler is silent and unobtrusive, weighs 1.7 g, and is 1.5 cm wide. Thus, it interferes minimally with human activities.

The sampler consists of a standard scanning electron microscope (SEM) stub, a collection substrate, and a protective mesh cap. During sampling, particles are transported by gravity, diffusion, and inertia through the 157 μm -diameter holes of the mesh cap and deposit on a substrate mounted on the stub. The stub is oriented such that the substrate is horizontal. After sampling, the mesh cap is removed, the stub is placed in

an SEM, and the particles are counted and sized to determine the particle flux. (Other microscope types may be used, but SEM is most convenient.) A semiempirical, particle size-dependent deposition velocity model is used in conjunction with the measured flux to obtain the ambient mass size distribution:

$$C = \frac{F}{v_{\text{dep}}} = \frac{F}{v_{\text{amb}}\gamma_m} \quad (1)$$

where C is the average mass concentration over the sampling period; F is the mass flux (particle mass/time/substrate area); v_{dep} is the deposition velocity; v_{amb} is the ambient deposition velocity; and γ_m is the mesh factor. All are functions of particle aerodynamic diameter, d_a . The ambient deposition velocity represents the theoretical deposition rate to a flat plate.⁽¹⁾ The mesh factor represents the effect of the mesh on the particle

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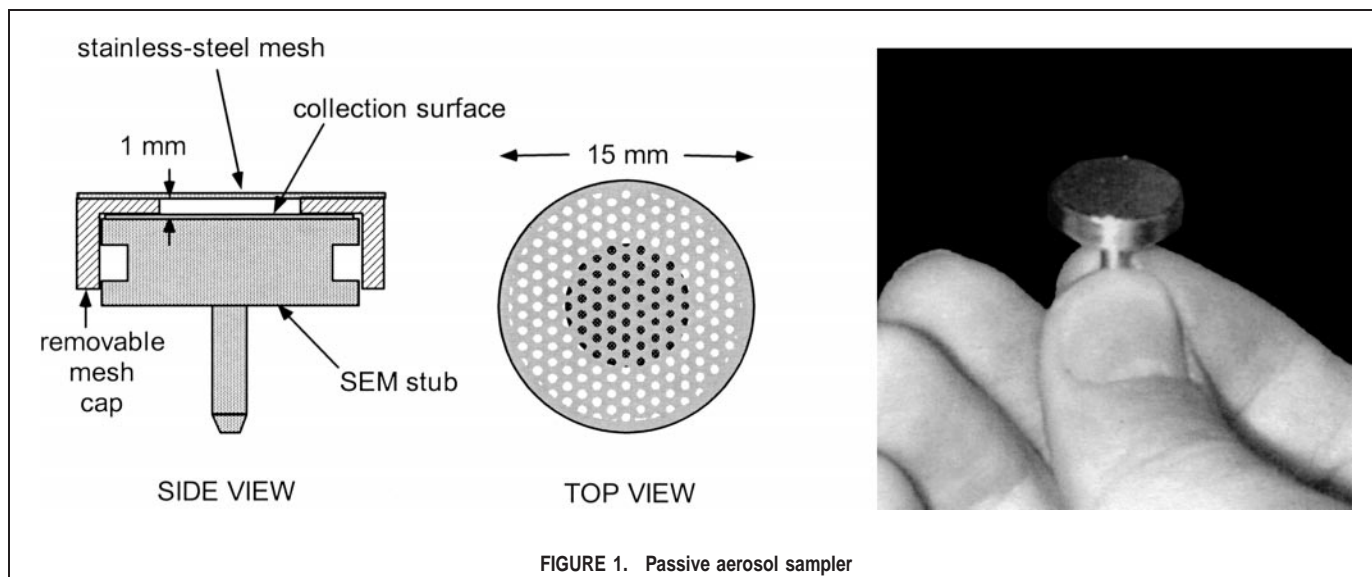


FIGURE 1. Passive aerosol sampler

deposition rate and was determined with wind-tunnel testing for $0.1 < d_a < 10 \mu\text{m}$.⁽²⁾ These tests also verified that the deposition rate was independent of wind speed at low turbulence levels, such as would be found commonly indoors.

Comparison of the passive sampler to conventional samplers in various environments is needed to demonstrate its strengths and limitations. Previously, the passive sampler had been field-tested only in an industrial environment with high-concentration, coarse-mode dusts.⁽³⁾ Results correlated well with those of an eight-stage cascade impactor. Discrepancies were attributed to the small number of fine-particle counts in the passive sampler and particle bounce in the impactor.

Aerosols in indoor environments such as homes and offices offer a significantly different challenge to the passive sampler. Typically, these aerosols are present at much lower levels than problem areas in industrial workplaces. Indoor aerosols often contain a variety of organic and inorganic species and a significant fraction of fine and submicron particles.⁽⁴⁾

The objective of this work was to investigate the passive aerosol sampler's ability to measure indoor particle exposures over periods of 24 hours to 2 weeks. The work consisted of two tasks. First, an automated analysis technique was developed so that large portions of the collection surface could be examined more efficiently. This capability allowed lower aerosol concentrations to be sampled over shorter time periods. Second, the passive sampler was tested alongside three conventional, pump-operated, size-selective samplers in indoor environments. By integrating passive sampler size distributions over the appropriate size ranges, comparisons could be made to the size fractions measured by each active sampler. This article describes the findings and their implications for the utility of the passive sampler.

METHODS

Automated SEM Analysis

The number of particles collected by the passive sampler depends linearly on the particle concentration and sampling time. When few particles are collected, observation of more microscope fields is necessary to measure the flux. In previous tests of the passive sampler,^(2,3) aerosol concentrations were high, leading to large

numbers of collected particles. The small number of SEM images needed to count and size the particles were obtained and processed manually. In this work lower aerosol concentrations, lower particle counts, and the need for more microscope images were anticipated.

To obtain microscope fields more efficiently, an automated analysis method was developed using a Philips XL30 ESEM (FEI Co., Eindhoven, The Netherlands) and VANTAGE Digital Microanalysis System (Thermo NORAN, Middleton, Wisc.). A program written with the VANTAGE software controlled movement of the microscope stage across a matrix of positions. At each position the program also set the microscope magnification, acquired an image, converted the gray-scale image to binary (black and white), and counted and sized any detected particles.

The program rejected particles that touched the top or left edges of any field. Similarly, particles were not counted that were fully contained within a "guard region" constructed along the right and bottom edges. Particles that partially overlapped the guard region were considered valid. This method yields unbiased counting and sizing of edge particles.⁽⁵⁾ The guard region width for a given magnification was set to equal $10 \mu\text{m}$ or one-fourth of the field width, whichever was smaller.

For each valid particle detected, the program measured a projected-area equivalent diameter, d_{pa} , where

$$d_{pa} = (4 \times \text{projected area} / \pi)^{1/2} \quad (2)$$

Particles with d_{pa} larger than the width of the guard region were automatically excluded from the analysis. To prevent undercounting or inaccurate sizing of small particles at high magnifications, particles with $d_{pa} < 7$ pixels were excluded as well. These criteria effectively set the minimum and maximum particle diameters measurable at a given magnification. As such, magnifications must be chosen so that their measurable size ranges overlap slightly and cover the particle sizes of interest. To measure $d_a = 0.1 - 10 \mu\text{m}$, magnifications of $225\times$, $385\times$, $1270\times$, and $12,001\times$ were selected.

Because the computer differentiates particles from background using image intensity, particle and background intensities must be clearly distinct. This condition was achieved using the SEM's back-scattered electron (BSE) detector, which responds primarily to differences in atomic number. Although the secondary electron (SE)

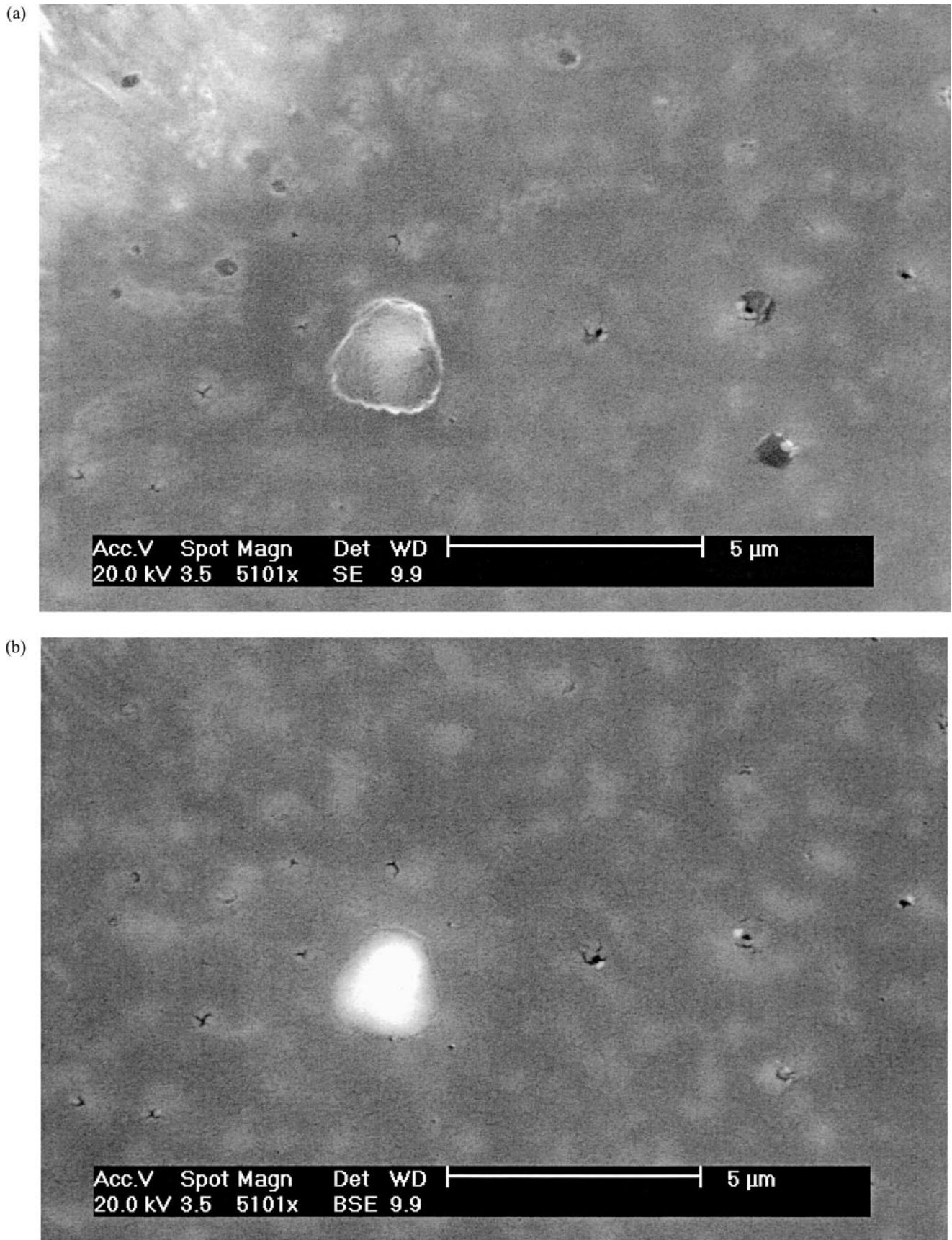


FIGURE 2. The secondary electron image (a) shows more topography than the back-scattered electron image (b). Topography is an undesirable quality for automated particle detection.

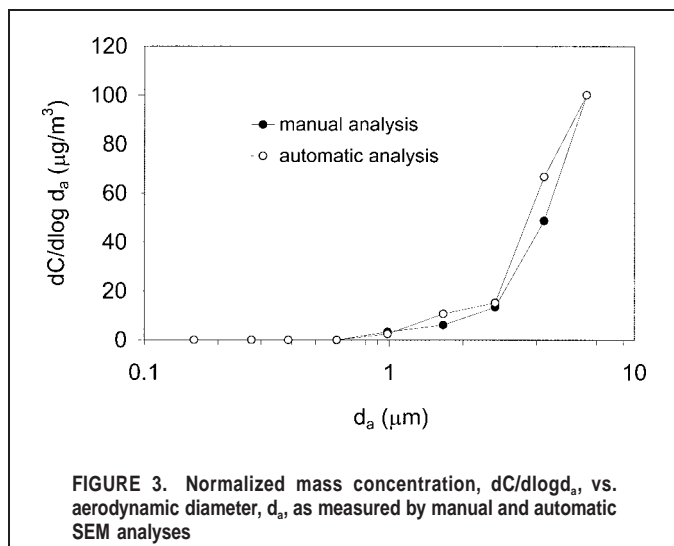


FIGURE 3. Normalized mass concentration, $dC/d\log d_a$, vs. aerodynamic diameter, d_a , as measured by manual and automatic SEM analyses

detector yields higher-resolution images, its sensitivity to surface topography yields images that are more ambiguous for particle detection. Figure 2 shows images for the same particle taken with SE and BSE detectors. The SE image has wide overlapping ranges of particle and substrate intensities, whereas the BSE image more closely resembles a binary image. The substrate used for these tests was a conductive carbon tab (model 16084-1, Ted Pella Inc., Redding, Calif.), so particles with atomic number >12 were imaged with good contrast.

To verify the accuracy of the automated analysis method, a preliminary passive sample was taken in the Oakland sampling location. This sample was analyzed both automatically and with the manual method of Wagner and Leith.^(2,3) In the latter method SEM images were obtained manually, particles were identified subjectively, and binary images were created. An image analysis program (Sigma Scan Pro, Jandel Scientific, San Rafael, Calif.) was used to count and size the identified particles. Size distributions obtained with the two methods are plotted in Figure 3. The automated and manual results showed good agreement.

In practice, each analysis began by manually obtaining a good high-magnification image of a particle in the passive sample. From this image the threshold intensity for particle detection was determined manually and entered into the computer. The proper threshold value was crucial for obtaining accurate results. After selecting the beam parameters for each magnification, the analysis was started for unattended operation. When the analysis was completed, the program shut off the electron beam.

Sampling Overview

In each of five sampling events three passive samplers were tested alongside active, size-selective samplers (Table I, Figure 4). Three

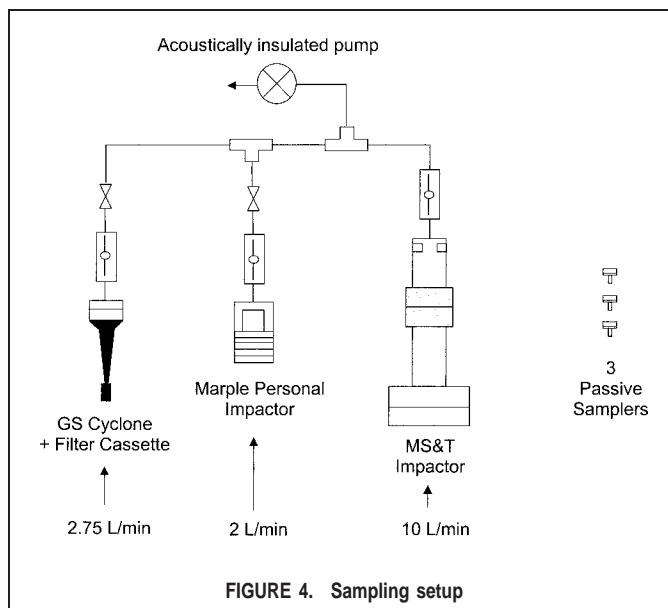


FIGURE 4. Sampling setup

active samplers were used: a Marple 290 Personal Cascade Impactor (Andersen Instruments, Smyrna, Ga.), a GS Cyclone/filter cassette (SKC, Eighty Four, Pa.), and a Harvard-type impactor (MS&T Area Sampler, Air Diagnostics and Engineering, Harrison, Maine). One pump (model SP-280, Air Diagnostics and Engineering) was used for all three active samplers using a sampling manifold. The passive samplers were mounted face-up on a horizontal aluminum strip that was attached to a ring stand. The ring stand supported the cyclone, personal impactor, rotameters, and valves. The acoustic insulation of the pump and compactness of the setup minimized disruption to the study participants. The ring stand was placed on tables or dressers at heights of 1 to 1.5 m above the ground. The residents of each indoor environment were asked to check active-sampler flow rates and adjust them if necessary. Indoor wind speeds and relative humidities were measured periodically using a handheld anemometer (Velocichек 8830, TSI, St. Paul, Minn.) and hygrometer (Fisher Scientific, Pittsburgh, Pa.), respectively.

Sampling was conducted at three residences in northern California during August–October 2000 (Table II). Sampling periods ranged from 24 hours to 2 weeks. The Oakland and Richmond locations were urban sites, whereas the Soledad site was rural. All sampling was conducted in carpeted rooms, and no smoking occurred at any of the locations.

The first three sampling events occurred in Oakland. The sampling site was in a small bedroom in a third-floor apartment occupied by two persons and two cats. The building was within 150 m of a freeway off-ramp and a smaller highway that served as a local truck route. The bedroom window was left open frequently

TABLE I. Samplers Used for Comparison

Sampler	Manufacturer	Flow Rate (L/min)	No. of Size Fractions	d_p Range (μm)	Filter Type
GS cyclone	SKC, Inc.	2.75	1	0–4	Teflon
MS&T impactor	Air Diagnostics and Engineering	10	1	0–10	Teflon
Marple personal cascade impactor	Andersen Instruments, Inc.	2	5	0–(>10)	Mylar for stages 3–6; PVC for final filter
Passive aerosol sampler	(Wagner and Leith ⁽³⁾)	—	9	0.1–10	—

TABLE II. Sampling Conditions

Sampling Event	Sampling Time	Avg. Relative Humidity (%)	Avg. Temperature (°C)	Room Dimensions (m)	Activity in Room	Residence Location	Outdoor Surroundings
1	24 hours	56	24	3 × 3 × 2.6	yes	Oakland, Calif.	urban, near highways
2	2 weeks	57	23	3 × 3 × 2.6	yes	Oakland, Calif.	urban, near highways
3	24 hours	56	24	3 × 3 × 2.6	yes	Oakland, Calif.	urban, near highways
4	1 week	45	27	9 × 12 × 2.7	yes	Soledad, Calif.	rural
5	2 weeks	57	22	3 × 3 × 2.7	no	Richmond, Calif.	urban, near construction

during sampling. During Event 1, a candle was lit in the room for approximately 20 min.

The Soledad sampling was conducted in a single-story house occupied by two persons, a cat, and a dog. The house was at the edge of a small subdivision of single-family dwellings that were otherwise surrounded by open spaces and agricultural lands. The large, open sampling area included a kitchen, living room, and two doors to the outside. Both doors were periodically left open for ventilation. Candles occasionally were lit on the side of the space opposite the samplers.

The Richmond site was in a small spare room on the second floor of a semidetached apartment occupied by one person and two cats. The complex was approximately 500 m from the San Francisco Bay. The neighboring property was the site of a large construction project. The window in the test room was left open continuously.

Passive Samplers

Passive sampler stubs, mesh caps, and carrying cases were cleaned with soap and water, alcohol, and compressed air. After double-sided-adhesive carbon tabs were mounted on the stubs, the mesh caps were pressed onto the carbon tabs, which provided firm but temporary bonds to the undersides of the caps (Figure 1). The carbon tab was also used as the particle collection substrate. The passive samplers were kept in plastic carrying cases whenever they were not sampling.

Before SEM analysis, the mesh caps were removed from the passive samplers and the collection surfaces were sputter coated with gold (model 9000, Pelco International, Redding, Calif.). SEM analyses were performed in high-vacuum mode at a working distance of 10 mm and accelerating voltage of 20 kV. Images were acquired by the Vantage system with a dwell time of 5000 ns and a resolution of 512 × 512 pixels. Each final image was the result of a moving average of 10 frames (Kalman averaging).

Passive sampler concentrations were determined using the automated analysis described previously; 100–200 images were acquired per sample.

Each analysis resulted in a report of the particles detected in each field, each particle's d_{pa} , and other supplemental information. These reports were then combined into histograms in a Microsoft® Excel® (Seattle, Wash.) spreadsheet. The d_{pa} bins were converted to d_a bins using assumed dynamic- and volume-shape factors of 1.4 and 1.6, respectively. Counts were converted to particle mass using an assumed density of 2.0 g/cm³. These values are representative of those compiled by Wagner and Leith⁽¹⁾ for heterogeneous aerosols. Because the sampled environments were observed to have low turbulence, the deposition velocity was assumed to be independent of wind speed.⁽²⁾ Equation 1 was used to calculate size distributions between 0.1 and 10 μm for each passive sampler.

One of the three passive samples in sampling Event 2 was mishandled during analysis, and its results were disregarded. In addition, a hygroscopic contaminant was introduced inadvertently to the first several samples during analysis. The contamination resulted in unrealistically large numbers of particles at high magnifications. To correct for this effect, fields with >3 times the particle count expected for each magnification (assuming concentrations <100 μg/m³) were disregarded automatically.

One "blank" passive sampler was analyzed with every three passive samples. The blank sampler was transported in the same carrying case as the other three and remained in the case while the others were sampling. Each passive sample result was corrected by the size distribution measured for the corresponding blank.

Active Samplers

Table I lists the flow rates, aerodynamic particle size ranges, and filter types used for the three active samplers. The personal impactor was used with Mylar™ substrates and stages 3, 4, 5, and 6 in place (respective 50% cut points at 2 L/min of 9.8, 6.0, 3.5, and 1.55 μm).⁽⁶⁾ The Mylar substrates were not coated, as coating would have interfered with an endotoxin analysis performed as part of an associated study. For the final filter, 37-mm PVC filters (Polyvic Type BS, Millipore, Bedford, Mass.) were used, which collected particles <1.55 μm.

The cyclone sampler was used with 37-mm Teflon™ filters (no. SA240PR TEF-DISC with polyolefin support ring, Andersen Instruments, Smyrna, Ga.). According to the manufacturer the sampler approximates the respirable efficiency curve at 2.75 L/min, with a 50% cut point of 4 μm.

The MS&T impactor was used with 37-mm Teflon filters (described previously). This sampler yields a relatively sharp size cut at 10 μm when operated at 10 L/min.

Prior to sampling, rotameters were calibrated downstream of each active sampler using a Mini-Buck calibrator (A.P. Buck, Orlando, Fla.) for flows <5 L/min and a 1-L glass bubble meter for flows >5 L/min. An attachment was constructed to connect the flow meter directly upstream of the inlet to the personal impactor. The cyclone could not be connected directly to a flow meter. Similarly, the in-line attachment provided by the manufacturer of the MS&T impactor was found to contribute a significant pressure drop and was not used. For these two samplers rotameter calibration was achieved by measuring the pressure drop across each sampler with a pressure gage as a function of flow. Then a valve was substituted for each sampler, a flow meter was connected upstream, and the valve was adjusted to match the measured sampler pressure drops.

Active sampler concentrations were determined gravimetrically. Filters were conditioned in a weighing chamber for 24–48 hours before pre- and postsample weighing. Temperature and relative humidity in the chamber were maintained at 21.0 ± 0.8°C and

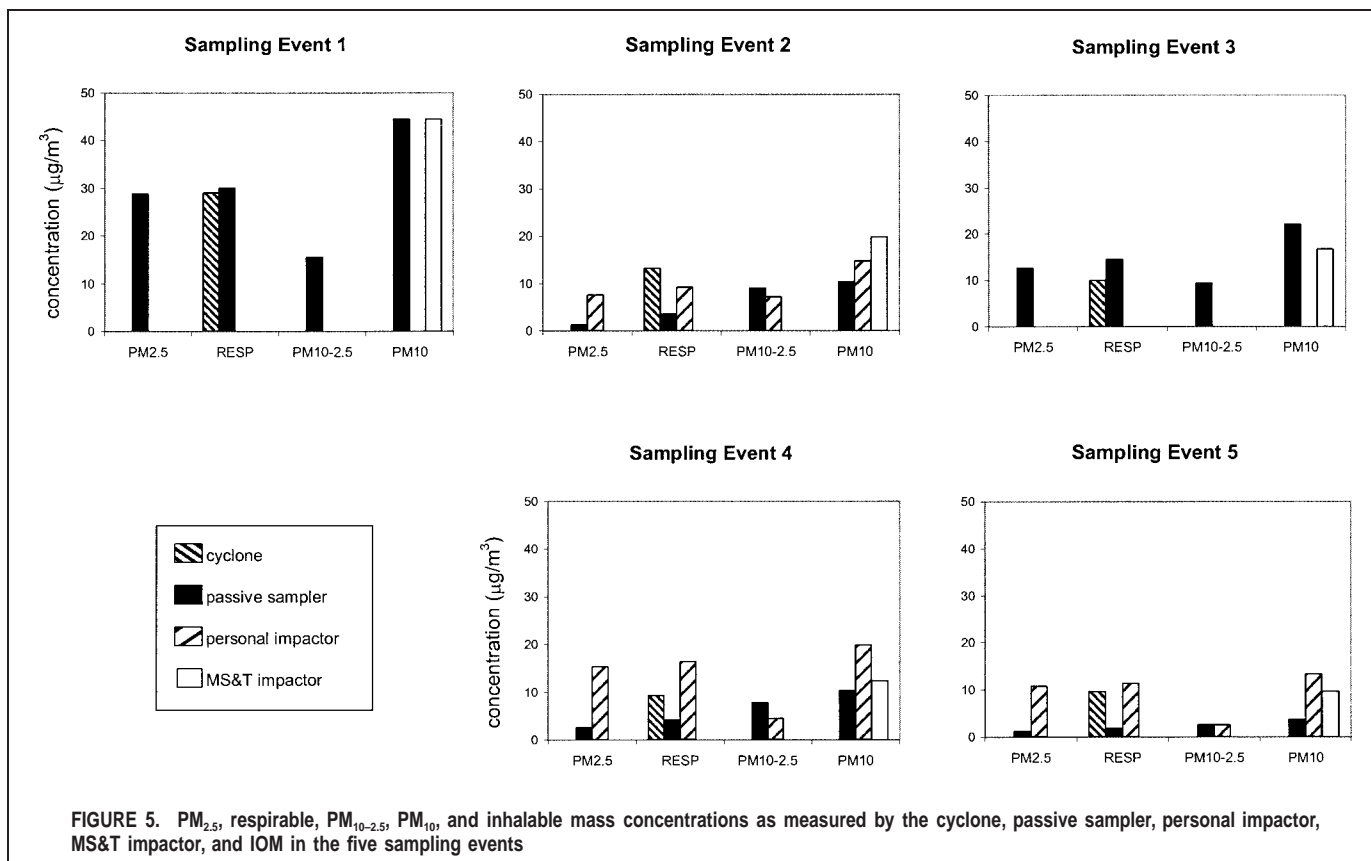


FIGURE 5. $PM_{2.5}$, respirable, $PM_{10-2.5}$, PM_{10} , and inhalable mass concentrations as measured by the cyclone, passive sampler, personal impactor, MS&T impactor, and IOM in the five sampling events

$47.9 \pm 2.2\%$, respectively. Filters were weighed with a Cahn 26 Microbalance (Cerritos, Calif.) after passing each filter over a charge-neutralizing Po^{210} strip (NRD Inc., Grand Island, N.Y.). “Substitution weights” were used to tare out amounts slightly less than the mass of each filter type. Then, more precise balance ranges could be chosen that corresponded to the sampled particulate mass alone. Substitution weights of 20 and 100 mg were used

when weighing the PVC and Teflon filters, respectively. No substitution weights were required for the Mylar substrates. All filter masses were measured to within 1 μg . Two filter blanks for each filter type were used, and the average weight change was subtracted from the corresponding sample weights.

Personal impactor substrates were not weighed after the 24-hour tests, because this was judged to be an inappropriate sampling interval for this device. Extrapolations from the 2-week results confirmed that the 24-hour masses would have been indistinguishable from the filter blank values.

Rotameters were monitored only periodically during sampling. Although the pump adjusted the total flow for changes in total

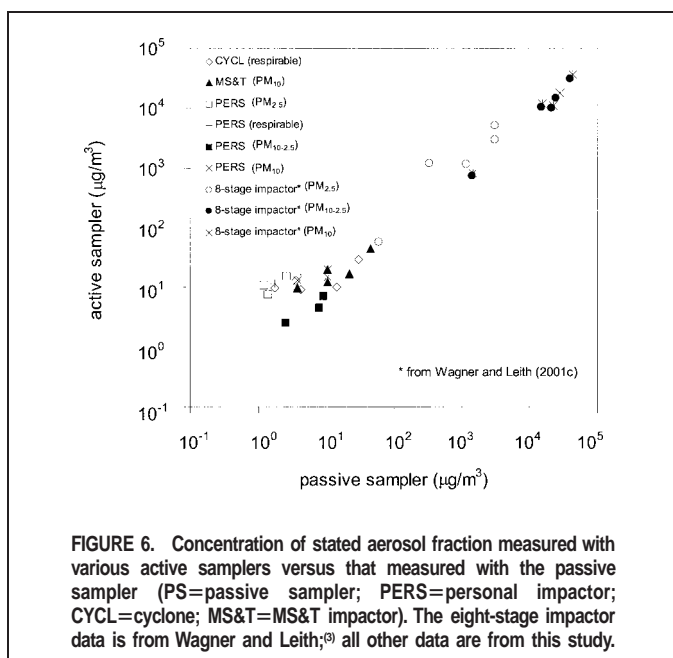


FIGURE 6. Concentration of stated aerosol fraction measured with various active samplers versus that measured with the passive sampler (PS=passive sampler; PERS=personal impactor; CYCL=cyclone; MS&T=MS&T impactor). The eight-stage impactor data is from Wagner and Leith;⁽⁹⁾ all other data are from this study.

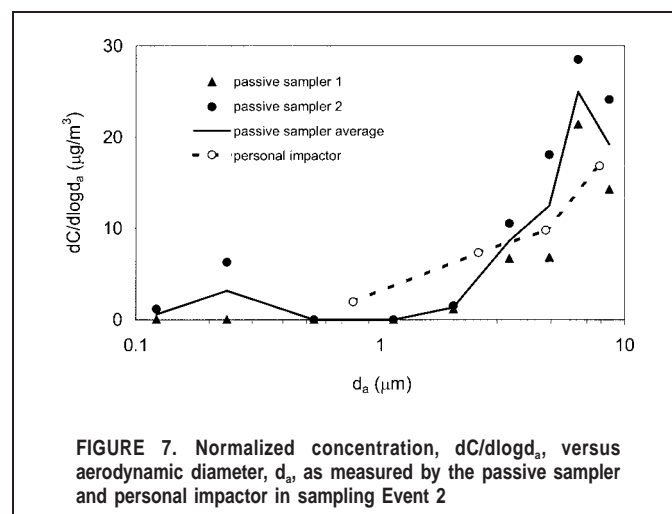


FIGURE 7. Normalized concentration, $dC/d\log d_a$, versus aerodynamic diameter, d_a , as measured by the passive sampler and personal impactor in sampling event 2

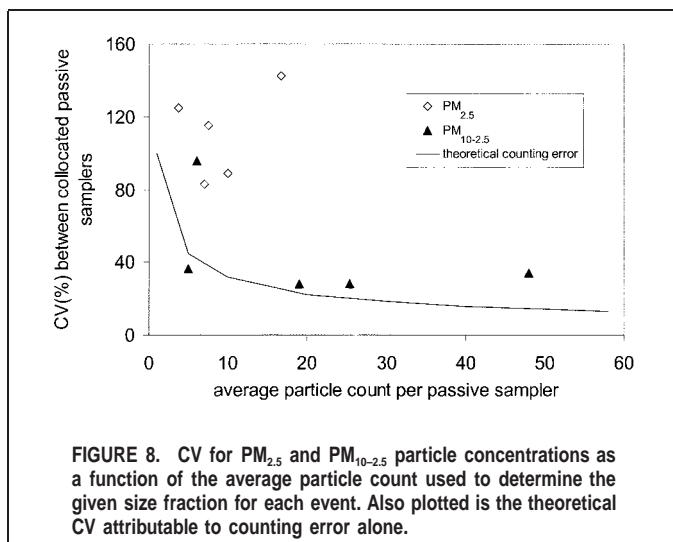


FIGURE 8. CV for $PM_{2.5}$ and $PM_{10-2.5}$ particle concentrations as a function of the average particle count used to determine the given size fraction for each event. Also plotted is the theoretical CV attributable to counting error alone.

pressure drop, it could not correct each sampler's individual flow if its pressure drop changed in relation to the others. As a result, flow deficits of 5% were sometimes observed before the valves were manually adjusted.

Calculations

Passive sampler and personal impactor size distributions were integrated over the appropriate particle sizes to calculate fine ($PM_{2.5}$, $d_a < 2.5 \mu m$), coarse ($PM_{10-2.5}$, $2.5 \mu m < d_a < 10 \mu m$) and fine-plus-coarse (PM_{10} , $d_a < 10 \mu m$) mass concentrations.

To calculate respirable mass concentrations for the passive sampler and personal impactor, the respective size distributions were multiplied by the ACGIH/CEN/ISO respirable penetration curve. A simplified approximation of this curve was used:⁽⁷⁾

$$RF = IF \times (1 - e^{-Y}) \quad (3)$$

where RF is the respirable fraction, $Y = e^{(2.54 - 0.681 d_{ai})}$; d_{ai} is the mean aerodynamic diameter of size bin i ; and the inhalable fraction, IF, is

$$IF = 0.5 (1 + e^{-0.06 d_{ai}}) \quad (4)$$

Theoretical counting error was calculated for comparison with the observed precision of the collocated samplers. Assuming a Poisson distribution for the number of particle counts per microscope field, the expected coefficient of variation (CV) is $N^{-1/2}$, where N is the total number of particle counts used to calculate the given size fraction.⁽⁸⁾

RESULTS

Average relative humidities and temperatures for the five sampling events are given in Table II. All wind speed measurements were < 0.01 m/sec, the detection limit for the anemometer. Qualitative XRF analyses of particles collected by the passive sampler yielded common dust constituents such as silicon, aluminum, calcium, sodium, potassium, chloride, sulfur, titanium, carbon, and oxygen, as well as magnesium, zinc, bromium, and niobium.

Figure 5 shows particle concentrations determined with the various sampler types. Figure 6 shows the agreement between passive and active samplers for all events. Also plotted on Figure 6 are the results of Wagner and Leith.⁽³⁾ Figure 7 shows passive sampler and personal impactor size distributions for sampling Event 2.

Linear regressions were performed on the data in Figures 5 and 6 (Table III). The correlation between the passive and cyclone samplers for respirable PM was $R^2 = 0.77$ ($p = .05$). Regression of passive sampler and MS&T impactor PM_{10} yielded an R^2 of 0.88 ($p = .02$). Correlations between the passive sampler and the personal impactor for $PM_{2.5}$, respirable PM, $PM_{10-2.5}$, and PM_{10} ranged from $R^2 = 0.21$ – 0.83 ; none were significant at the .05 level (Table III). Combining the data sets from the three sampler comparisons yielded an overall regression of $R^2 = 0.73$ ($p < .0001$). When the Wagner and Leith⁽³⁾ data from Figure 6 were added, the overall R^2 was 0.96 ($p < .0001$). Also listed in Table III are the slopes and intercepts from these regressions. As each regression was performed with the active sampler as the independent variable and the passive sampler as the dependent variable, non-zero intercepts can be interpreted as bias in the passive sampler.

Paired t-tests were performed to determine the agreement between the various sampler measurements (Table IV). A positive percentage difference in Table IV means that the first sampler of the pair had a higher mean than the second. All differences between passive and active samplers were statistically insignificant except those between the passive sampler and the personal impactor. The differences between the means for these two samplers were 147% for $PM_{2.5}$ ($p = .04$), 119% for respirable PM ($p = .04$), and 65% for PM_{10} ($p = .04$). The difference between passive sampler and personal impactor $PM_{10-2.5}$ was statistically insignificant.

Figure 8 shows the precision of the collocated passive samplers in each event. The CV is plotted as a function of the average particle count used to determine the given size fraction in each event. The theoretical counting error also is plotted as a function of count.

DISCUSSION

All wind speeds were very low in these tests, consistent with the low-turbulence assumption made for the deposition velocity model. This result agrees with previous measurements in a well-ventilated occupational environment that showed negligible turbulence levels.⁽⁹⁾

All environments possessed comparable temperatures, relative humidities, and aerosol concentrations, with the exception of sampling Event 1 (Figure 5). In this small room the lighting of a candle produced a particle concentration that was approximately twice that of the other events.

Sampling Event 5 exhibited the lowest aerosol concentrations. No activity occurred in this room during sampling, and a large fraction of the sampled aerosol probably came in through the open window from the construction site next door. The filters from this event were noticeably darker in color, consistent with the carbonaceous particles likely produced by soil excavation and emissions from heavy machinery and diesel trucks. This event also showed the worst agreement between passive and active samplers, which is also consistent with a large fraction of carbonaceous material. A carbonaceous particle on a carbon substrate shows little atomic number contrast in a BSE image, and thus such particles are virtually invisible. For this reason it is likely that carbonaceous particles were detected poorly by the automated SEM analysis. Figure 9a is an SE image of two large particles collected by the passive sampler. Figure 9b is the corresponding BSE image, in which only the fine, higher-atomic-number constituents of the particles are visible. To avoid this limitation in the future, passive samplers with both carbon and aluminum substrates could be used simultaneously. The aluminum-substrate passive samplers should give

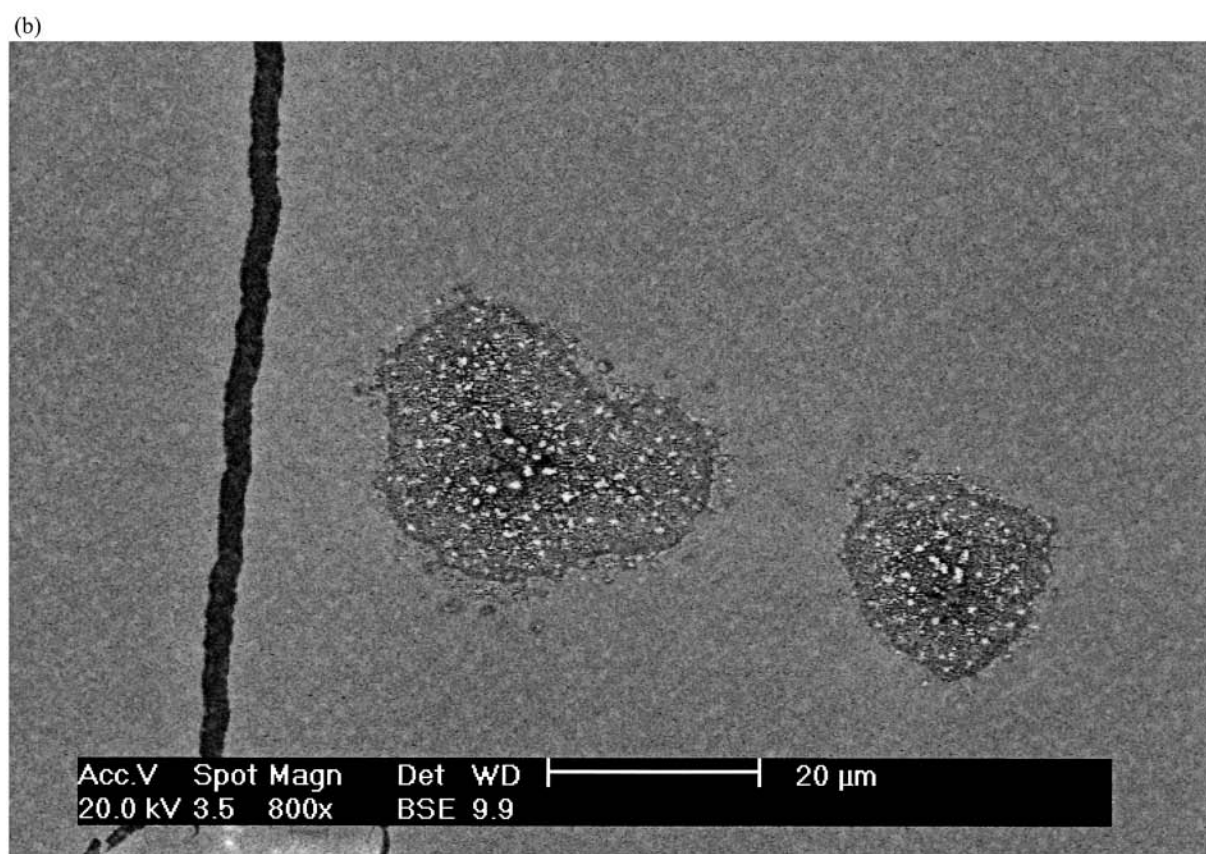
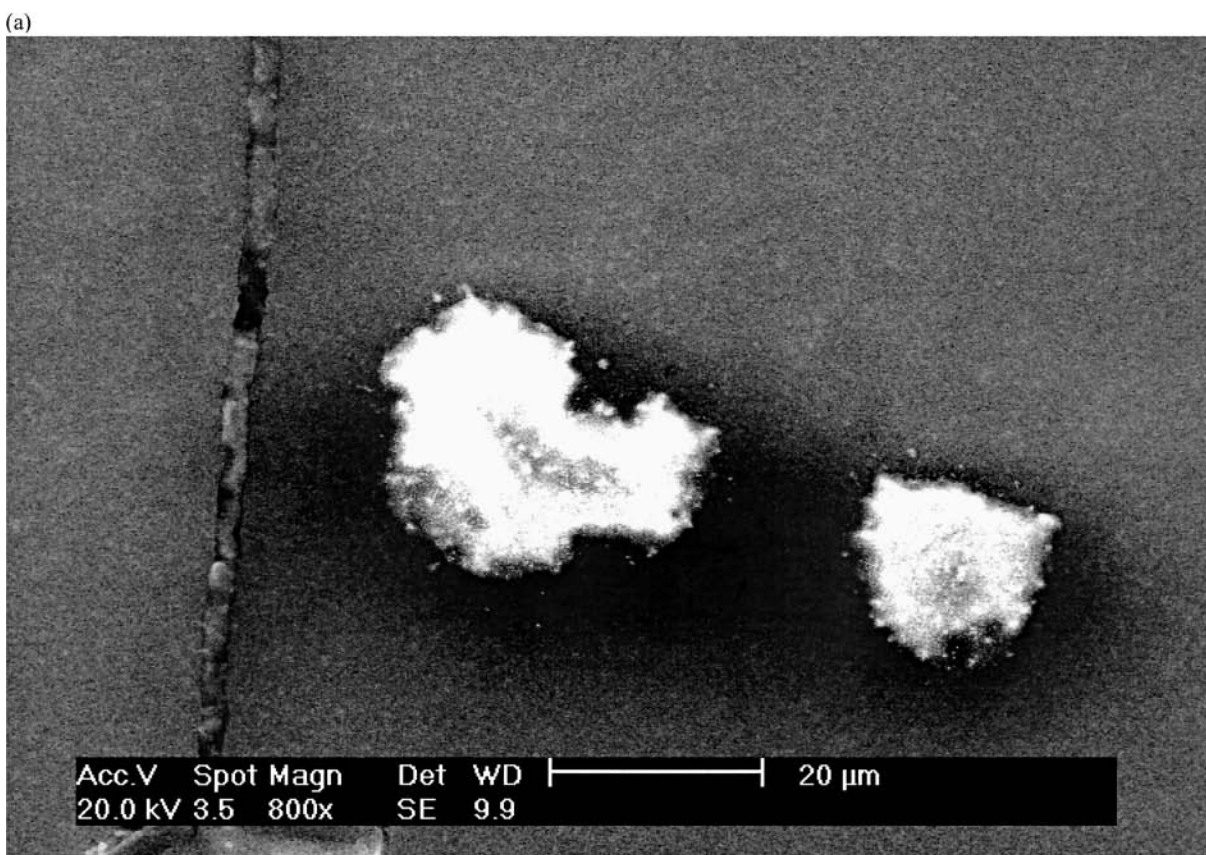


FIGURE 9. The secondary electron image (a) reveals two heterogeneous particles with a significant organic component, whereas the back-scattered electron image (b) shows only the fine, high atomic number constituents.

TABLE III. Correlations Between Results for the Passive Sampler and Various Active Samplers

	Cyclone Respirable	MS&T Impactor PM ₁₀	Personal Impactor				Overall	
			PM _{2.5}	Respirable	PM _{10-2.5}	PM ₁₀	This Study	This Study Plus Wagner and Leith ⁽³⁾
No. of data points	5	5	3	3	3	3	22	32
R ²	0.77 ^A	0.88 ^A	0.73	0.21	0.83	0.45	0.73 ^A	0.96 ^A
p-value	0.05	0.02	0.35	0.69	0.27	0.53	≪0.001	≪0.0001
Slope ^B	1.2	1.1	0.17	0.16	1.3	0.75	1.0	1.3
Intercept (μg/m ³) ^B	-6.8	-4.2	-0.17	1.2	0.10	-3.9	-4.9	320

^ACorrelation statistically significant at .05 level.

^BCalculated with the active sampler as the independent variable.

good BSE detection of particles containing lower-atomic-number elements such as elemental carbon and organic compounds.

In addition, sampling Event 5 was dominated by small particles, because the PM_{2.5} and respirable PM concentrations were nearly equal to the PM₁₀ values. As a result, many particles could have been at or below the SEM's limit of resolution, and thus missed by the passive sampler. SEM analysis has diminishing accuracy for smaller particles, with a practical lower limit of approximately $d_{pa}=0.1$ μm. This limitation could be eliminated with an alternative analysis technique with higher resolution, such as transmission electron microscopy or atomic force microscopy.

An additional source of the discrepancy between passive sampler and personal impactor PM_{2.5} was likely particle bounce in the personal impactor, as the Mylar substrates could not be coated with an adhesive. If high-inertia large particles bounced from their nominal collection stages into lower ones, underestimation of the coarse fraction and overestimation of the fine fraction would result. The observed discrepancies between passive sampler and personal impactor PM_{2.5} and PM_{10-2.5} (Figures 5 and 6 and Table IV) are consistent with this theory, as are the "flattened" personal impactor size distributions that were observed (see, for example, Figure 7).

Despite some discrepancies between the passive and active samplers, Figure 6 and Table III show generally good correlations between them. The cyclone, MS&T impactor, and overall correlations with the passive sampler were relatively high ($R^2=0.73-0.88$). The corresponding slopes for these correlations were all within 20% of unity. The intercepts of these regressions were all approximately -5 μg/m³, suggesting that the passive sampler was slightly negatively biased.

Correlations between passive and personal impactor size fractions were not significant, probably due to the smaller number of available comparisons and particle bounce. The personal impactor data also yielded nonsignificant correlations and nonunity slopes when regressed against the cyclone data ($R^2=0.62$, $p=.42$, slope= -0.5) and MS&T impactor data ($R^2<0.01$, $p=.97$, slope= -0.1).

The combination of this study's data with that of Wagner and Leith⁽³⁾ spanned five orders of magnitude, a fact that is partially

responsible for the high overall correlation observed between passive and active samplers ($R^2=0.96$). The high-concentration data dominated the overall regression, leading to a much higher slope and intercept (1.3 and 320 μg/m³, respectively) than the regression performed on this study's data alone.

Figure 8 shows that the precision of the passive sampler is generally worse for results determined with lower particle counts. This trend is consistent with the theoretical counting error, but the uncertainty in counting does not account for all of the imprecision observed. Possibly, differing amounts of contamination arose from the preparation and handling of the passive samplers, producing slightly different results in the three samplers for each event. This effect would be more prominent in cases with few "legitimate" counts.

The issue of low particle counts in indoor environments was the motivation for developing the automated passive sampler analysis. The number of particles detected by the automated analysis was enough to provide a good correlation with the other samplers, but not enough to provide good precision. The sampler's precision and accuracy should improve if more microscope fields are acquired. However, because a portion of the imprecision does not appear to be due to counting error, averaging the results of multiple, collocated passive samplers is recommended. Sampling with multiple passive samplers creates no added burden while sampling, as the samplers are inexpensive, consume no power, and require little space. The burden of additional analysis is not negligible, but it can be minimized using the automated analysis.

CONCLUSIONS

Indoor aerosol samples measured at 24-hour, 1-week, and 2-week intervals were taken with the Wagner-Leith passive sampler. Generally, good correlation with active samplers was observed. The largest discrepancies between passive and active samplers were observed for fine and carbonaceous particles. The ability of the passive sampler to measure these particles would be improved by the use of a higher-resolution analysis technique, such as transmission

TABLE IV. Differences Between Sampler Measurements and p-values From Paired t-tests

	PM _{2.5} PS/PERS	Respirable			PM _{10-2.5} PS/PERS	PM ₁₀		
		PS/PERS	PS/CYCL	PERS/CYCL		PS/PERS	PS/MST	PERS/MST
No. of pairs	3	3	5	3	3	3	5	3
Difference (%)	-147 ^A	-119 ^A	-27	14	31	-65 ^A	-13	13
p-value	0.04	0.04	0.39	0.66	0.21	0.04	0.39	0.64

Note: PS = passive sampler, PERS = personal impactor, CYCL = cyclone, MST = MS&T impactor.

^ADifference statistically significant at .05 level.

electron microscopy, and the simultaneous use of carbon and aluminum substrates. Large, statistically significant discrepancies (up to 147%) between the passive sampler and personal impactor were attributed partially to particle bounce from the uncoated impactor substrates.

An automated analysis method was developed for the passive sampler. The new method agreed well with the results of the manual method. Using the automated method, 100–200 SEM images per sampler were obtained across four magnifications.

The highest concentrations were measured over 24 hours in a small bedroom in which a candle had been lit, whereas the lowest were measured over 2 weeks in a room with no activity. The generally low aerosol concentrations in these indoor environments translated into low particle counts in the passive sampler analysis. Because sampler precision was found to worsen with decreasing counts, the results of multiple, collocated passive samplers should be averaged if low counts are expected.

In combination with the results of Wagner and Leith,⁽³⁾ the passive aerosol sampler now has been tested over five orders of magnitude in mass concentration. Generally, the passive aerosol sampler has proved useful for obtaining aerosol size distributions, mass fractions, qualitative elemental analysis, and individual particle morphology.

The Wagner-Leith passive sampler has several limitations. First, the presence of background contamination complicates the analysis when particle counts are low. This limitation is analogous to that of filter-based samplers, in which the blank filter weight gain may be similar to the sampled mass in low-concentration environments. Second, uncertainties in sampling submicron and semivolatile particles remain. Although filter-based samplers often perform at least as poorly for semivolatile particles, they are currently more reliable for submicron particles. Third, although the automated analysis reduces the amount of labor required per passive sample, the analysis still requires some expertise and expense. This cost is in contrast to filter-based sampling, in which the capital and personnel costs (e.g., equipment, maintenance of flows and pressure drops) are incurred before and during sampling. Ultimately, the utility of the passive sampler depends on the anticipated sampling period, aerosol composition and concentration, and requirements for sampler unobtrusiveness.

Preliminary work suggests the Wagner-Leith sampler may be useful for identifying bioaerosols such as fungal spores and pollen grains and will be pursued further. Other future work includes the use of alternate collection substrates and microscopy techniques to expand the sampler's capabilities for fine particles and the use of the sampler as a personal sampler.

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