# Particle Concentrations in Urban Microenvironments

*Jonathan I. Levy,[1] E. Andres Houseman,[2] Louise Ryan,[2] Dejuran Richardson,[2] Students from the 1998 Summer Program in Biostatistics,[*] and John D. Spengler[1]*

[1]Department of Environmental Health; [2]Department of Biostatistics, Harvard School of Public Health, Boston, Massachusetts, USA

**Although ambient particulate matter has been associated with a range of health outcomes, the health risks for individuals depend in part on their daily activities. Information about particle mass concentrations and size distributions in indoor and outdoor microenvironments can help identify high-risk individuals and the significant contributors to personal exposure. To address these issues in an urban setting, we measured particle count concentrations in four size ranges and particulate matter ≤ 10 μm ($PM_{10}$) concentrations outdoors and in seven indoor microenvironments in Boston, Massachusetts. Particle counts and $PM_{10}$ concentrations were continuously measured with two light-scattering devices. Because of the autocorrelation between sequential measurements, we used linear mixed effects models with an AR-1 autoregressive correlation structure to evaluate whether differences between microenvironments were statistically significant. In general, larger particles were elevated in the vicinity of significant human activity, and smaller particles were elevated in the vicinity of combustion sources, with indoor $PM_{10}$ concentrations significantly higher than the outdoors on buses and trolleys. Statistical models demonstrated significant variability among some indoor microenvironments, with greater variability for smaller particles. These findings imply that personal exposures can depend on activity patterns and that microenvironmental concentration information can improve the accuracy of personal exposure estimation.** *Key words*: **air pollution, exposure assessment, indoor air, microenvironments, particulate matter.** *Environ Health Perspect* **108:1051–1057 (2000). [Online 16 October 2000]**
*http://ehpnet1.niehs.nih.gov/docs/2000/108p1051-1057levy/abstract.html*

Recent epidemiologic studies have established relationships between particulate matter and both morbidity and premature mortality and presented evidence that fine particulate matter (the fraction of particulate matter < 2.5 μm in diameter) may be responsible for these adverse outcomes (*1–6*). Although some physiologic and toxicologic evidence exists (*7–9*), most of the evidence for particulate matter health effects is taken from epidemiologic studies that use fixed-site ambient measurements as estimates of exposure.

Critics of the positive epidemiologic findings consider the disconnect between ambient monitors and actual exposure to be a potential source of error (*10,11*). However, although some studies have found poor correlations between personal exposure and ambient concentrations (*12–14*), the correlations have been stronger when evaluated within individuals across time (*15,16*). Furthermore, any errors induced by using fixed-site monitors to represent personal exposure would likely be "Berkson errors," which would not induce bias if the dose–response relationship were linear (*17*).

Despite these facts, knowledge about personal exposure to particulate matter is crucial in a risk assessment and public policy context. Estimates of the distribution of exposures can help identify high-risk individuals and risks to susceptible subpopulations, and understanding the primary contributors to personal exposure can lead to well-designed control policies. Because individuals spend a significant fraction of the day indoors, with variable ventilation rates and differing indoor sources, the differences in personal exposures between individuals represented by the same fixed monitor could be substantial.

Because it would be implausible to measure the personal exposures of a significant number of people, a theoretically sound alternative is to measure concentrations in a number of microenvironments and determine the time spent by individuals in these microenvironments. A microenvironment can be defined as a physical compartment or defined space with relatively homogeneous air pollution concentrations (*18*). Simple microenvironmental models could involve estimates of indoor and outdoor concentrations and the amount of time spent in each of these two settings. For more complex models, such as the Probabilistic National Ambient Air Quality Standards Exposure Model (pNEM) (*19*) or the Simulation of Human Air Pollutant Exposure (SHAPE) for carbon monoxide (*20*), there is a need to understand concentration patterns across a number of different microenvironments that have not been well characterized to date.

Along with particulate matter mass concentrations, there are compelling reasons to estimate the particle counts and the size distributions of those particles. From a health effects standpoint, it has been argued that particle surface area or number could be more important than particle mass, due to the potential impairment of macrophage functions associated with clearance (*21*). In addition, because different source types provide different particle sizes and chemical compositions, understanding the sizes of particles present in different microenvironments can establish a framework for source attribution. Recent evidence that particulate matter from combustion sources may have greater effects than crustal particulate matter (*22*) gives source attribution added significance.

Continuous real-time monitoring can provide the information necessary to detect the influence of a local source or changes in local circumstances on particulate matter counts or mass concentrations. In addition, continuous monitoring allows us to evaluate short-term particle exposures, a topic for which very little exposure or health information has been collected (*23*).

To address these issues, we continuously monitored particulate matter mass concentrations and particle counts in a number of indoor microenvironments in an urban area over a period of 4 days, with outdoor measurements taken outside each microenvironment. We considered building or source factors as well as temporal trends in pollution concentrations to determine the significance of differences among indoor microenvironments. By determining microenvironments that might contribute significantly to personal exposure and by capturing the degree of microenvironmental variability in an urban area, we provide a template to better estimate personal exposures to particulate matter.

Address correspondence to J.I. Levy, Harvard School of Public Health, 665 Huntington Avenue, Boston, MA 02115 USA. Telephone: (617) 432-4845. Fax: (617) 432-4122. E-mail: jilevy@hsph.harvard.edu

*B. Acosta-Garcia (University of Puerto Rico), M. Cote (Harvard/Main Program for Children), T. Jones (LeMoyne-Owen College), R. Moore (Bennett College), M. Price (Albany State University), R. Sebro (Morehouse College), K. Taylor (Hampton University), and I. Williams (Howard University).

We thank H. Özkaynak for providing valuable feedback, and we are indebted to all of the Summer Program students for their interest and assistance.

Funding for this research was provided in part by the National Cancer Institute (CA48061), NIH (R25 GM55353), U.S. EPA Cooperative Agreement CR825267-01, and by a U.S. EPA EMPACT grant. In addition, students in the Summer Program in Biostatistics were supported by NIEHS (5 T35 ES07293-05).

This paper has not been subjected to review by the above agencies and does not necessarily reflect their views.

Received 17 April 2000; accepted 5 July 2000.

## Methods

We measured particle counts and mass concentrations in seven indoor microenvironments as well as outdoors over a 4-day period in June 1998. To measure particle counts, we used an APC-1000 Airborne Particle Counter (Biotest Diagnostics, Denville, NJ). The APC-1000 is a light-scattering device that simultaneously measures particle counts above four device-specified size thresholds: 0.3 µm, 0.5 µm, 1.0 µm, and 5.0 µm in diameter. According to the manufacturer, the theoretical upper bound for the largest particle size category is on the order of 1,000 µm, although particle counts are generally minimal above 5 µm because particles much larger would not be suspended in ambient air. The APC-1000 also provided measurements of temperature and relative humidity and was factory-calibrated using isotropic polystyrene spheres within the year before use. Past exposure assessment studies have found the APC-1,000 to be useful for evaluating short-term concentration peaks and making preliminary source attributions (*23*).

We measured particulate matter mass concentrations using a DustTrak 8520 (TSI, Minneapolis, MN), a laser photometer designed to measure particles between 0.1 and 10 µm. The DustTrak was factory-calibrated using A1 test dust (Arizona Test Dust, ISO 12103-1) within the year before use and was calibrated to a zero filter during the sampling period.

Nine students from the Harvard School of Public Health Summer Program in Biostatistics were trained in the operation of all equipment and divided into three sampling groups. The Summer Program in Biostatistics is a short-term program funded by the National Institute of Environmental Health Sciences, which is intended to introduce undergraduate mathematics majors from underrepresented minority groups to biostatistics, environmental health, and public health research.

On each of the 4 sampling days, each group sampled for up to three sessions in designated microenvironments in the Boston, Massachusetts, area. To normalize for temporal trends, measurements were taken between 1100 and 1700 hr on all 4 days. The students followed a detailed script that directed them to spend 20–40 min in each of several sampling locations. Along with the monitoring data, the groups recorded other information about site characteristics that might affect particulate matter concentrations, including the presence of smokers or air conditioning, whether windows were kept open or closed, and the distance from the street.

The seven indoor microenvironments selected were categorized as bus, gymnasium, hospital, museum, restaurant, store, and subway (Table 1). In Boston, the buses are largely diesel fueled, and the subway line is electric and consists of both a street-level and an underground section. These microenvironments were chosen strategically to represent typical activities of urban residents that had not been previously incorporated into many microenvironmental models. The students mimicked the typical behaviors within microenvironments (i.e., walking in stores, sitting in restaurants), so that their personal influence on particle concentrations would not alter true exposure patterns. In addition, outdoor measurements were taken in the vicinity of all microenvironments, with additional measurements taken in parks and on sidewalks.

Because the APC-1000 requires a 15-sec standby period between measurements, we used a 2-min averaging time for the APC-1000 and a 135-sec averaging time for the DustTrak. This ensured that the two instruments were synchronized throughout the measurement period. The sampling interval was selected to provide a reasonable sample size within each microenvironment while dampening the effects of short-term spikes in concentrations.

## Statistics

Aside from estimating particle mass and count concentrations and size profiles within different microenvironments, the primary goal of this study was to understand the degree of variability between microenvironments, controlling for common factors. However, standard statistical comparisons between microenvironments are impeded by two aspects of our study design. First, environmental measurements were taken with short averaging times, making it unlikely that the measurements are independent of one another within microenvironments (a required assumption for many standard statistical methods). Second, when comparing a number of microenvironments, random variability could be the cause of significant deviations, and adjustments for multiple comparisons are needed.

The anticipated autocorrelation between sequential measurements was confirmed by exploratory analysis using the variogram technique (*24*). Given this, we sought a statistical model that could account for the autocorrelation. We considered using generalized estimating equations (GEEs) within our regression analysis, but preliminary simulations suggested that a linear mixed effects (LME) model (*25*) had better properties, particularly in terms of Type I error. Consequently, we applied LMEs within our regression analysis, assuming an AR-1 autoregressive correlation structure within each session. Thus, for each session $i$ and replicate $j$, we statistically modeled each measurement $y_{ij}$ as

$$y_{ij} = x_i\beta + b_i + u_{ij},$$

where $x_i$ is a row-vector consisting of the covariates of interest, $\beta$ is a corresponding set of regression coefficients to be estimated, $b_i$ is a normally distributed random effect intercept, and $u_{ij}$ is a normally distributed error with AR-1 structure

$$u_{ij} = \rho u_{ij-1} + e_{ij},$$

(each $e_{ij}$ being independent and normally distributed). We constructed separate models for indoors and outdoors, given differences in relevant factors as well as a desire to determine the degree of heterogeneity among both types of microenvironments. All particle count and mass concentration parameters were log-transformed to more closely approximate normal distributions.

For indoor microenvironments, the predictors considered for the model include

**Table 1.** Descriptions of indoor and outdoor microenvironments.

| Microenvironment | Description | No. of sessions (indoor, outdoor) |
|---|---|---|
| Bus | Diesel-fueled city buses and medical area shuttle (all urban travel) | 15 (7, 8) |
| Subway | Electric-powered subway traveling both above ground (on-street trolley) and underground | 21 (12, 9) |
| Gymnasium | Athletic facility located near medical area | 2 (1, 1) |
| Hospital | Two hospitals located across the street from one another, along bus route | 6 (3, 3) |
| Museum | Art museum in urban area | 2 (1, 1) |
| Restaurant | Small pizza place, fast-food restaurant, coffee shop | 10 (5, 5) |
| Store | Three large shopping malls downtown, shops near medical area and other urban areas | 15 (8, 7) |
| Sidewalk | High-traffic roads near medical area, other urban areas | 8 (0, 8) |
| Park | Park areas downtown, close to traffic | 6 (0, 6) |

indicator variables for microenvironment, open windows, and presence of central air conditioning, number of people nearby, and temperature and relative humidity. For outdoor microenvironments, we considered indicator variables for microenvironment, presence of smokers nearby, as well as number of people nearby and temperature and relative humidity. It should be noted that no smokers were present in any indoor microenvironments, explaining its exclusion from the indoor model. In addition, we incorporated a term for date of measurement into both models, to account for meteorological or other differences that could influence ambient concentrations.

Once the LME models have established whether there are significant differences among microenvironments in indoor or outdoor settings, we need to determine which microenvironments differ significantly from one another. To make this comparison, we used Wald tests on the estimated group means for different microenvironments. However, this technique can result in specious statistical findings if the effect of multiple comparisons is not properly accounted for. In ordinary least-squares regression, multiple comparison methods such as the Tukey and Scheffe tests have been developed to control the Type I error probability without significantly increasing the likelihood of a Type II error (*26*). However, these techniques are not applicable to LME.

To address this issue, we conducted multiple comparisons using two procedures at opposite extremes. First, we effectively ignored the multiple comparisons issue, rejecting the null hypotheses if $p < 0.05$. This adequately controls for Type II errors but could yield extremely high Type I errors. On the other extreme, we used the Bonferroni probability of $0.05/n$ (where $n$ is the number of comparisons) to yield an overall Type I error rate of 0.05 while increasing Type II errors. These two extreme methods should bracket the correct statistical inferences.

For all statistical assessments, we evaluated the particle counts within specified size ranges rather than the measured threshold values. In other words, the APC-1000 measures the number of particles per unit volume that have diameters of at least 0.3 µm, 0.5 µm, 1.0 µm, and 5.0 µm. We analyzed the differences between these categories, reflecting the particle size ranges of 0.3–0.5 µm, 0.5–1.0 µm, 1.0–5.0 µm, and > 5.0 µm. Past studies have found that these ranges correspond to aerodynamic diameters of 0.9–1.2 µm, 1.2–1.7 µm, 1.7–3.7 µm, and > 3.7 µm (*23*).

## Results

In total, the microenvironmental sampling yielded 578 measurements taken within 85 measurement sessions. Due to equipment issues, there were only 381 measurements for which all four particle size counts from the APC-1000 and the $PM_{10}$ concentration using the DustTrak were valid (66%). Because of our interest in evaluating variability in and predictors of particle counts within specified size ranges, we focused our analysis on these 381 measurements, even though this reduced the statistical power of our analysis. Descriptive statistics as well as statistical models did not differ significantly when applied to the full set of data when appropriate.

For all descriptive statistics, we estimated the geometric mean and geometric standard deviation to account for the logarithmic distribution of pollution concentrations and to increase comparability with the study by Brauer and colleagues (*23*). Taken across all microenvironments, outdoor $PM_{10}$ concentrations ranged between 10 and 90 µg/m$^3$, with a geometric mean of 19 µg/m$^3$ and a geometric standard deviation (GSD) of 1.9 (Table 2). Indoor $PM_{10}$ concentrations were generally higher and more variable, with a geometric mean concentration of 35 µg/m$^3$ (range 0–380 µg/m$^3$) and a GSD of 3.0. The patterns are similar for the four particle size ranges, with higher geometric mean count concentrations as well as greater variability indoors.

To compare the two instruments, we followed the methodology of Brauer and colleagues (*23*) and used an assumed particle density of 2.8 g/cm$^3$ (*7*) to convert particle counts into particle mass. Assuming spherical shape and using the midpoint of the particle diameter ranges (7.5 µm assumed for the largest size range), the calculated particle mass from the APC-1000 is well correlated with the $PM_{10}$ mass concentration measures ($r = 0.87$). In addition, we can compare our measurements to concentrations from fixed monitors. Although $PM_{10}$ data are only available every 6 days from the nearest U.S. Environmental Protection Agency (U.S. EPA) monitoring station (Kenmore Square, Boston), the ambient concentration was approximately 20 µg/m$^3$, similar to our geometric mean outdoor value.

When we consider some simple comparisons by site characteristic, we see some systematic differences (Table 2). Particle counts and mass concentrations tend to be greater with higher temperatures and higher relative humidity, both in indoor and outdoor microenvironments. Particle counts and mass concentrations are slightly higher in outdoor microenvironments with smokers and in indoor microenvironments with central air conditioning, although few measurements were taken indoors without air conditioning ($n = 10$).

Stratifying by microenvironment, we can see some systematic differences between indoor and outdoor particle mass concentrations (Figure 1). Indoor $PM_{10}$ concentrations

**Table 2.** Particle count and mass concentrations aggregated across urban microenvironments.

| | Sample size | $PM_{0.3–0.5}$ (particles/cm$^3$) | | $PM_{0.5–1.0}$ (particles/cm$^3$) | | $PM_{1.0–5.0}$ (particles/cm$^3$) | | $PM_{5.0+}$ (particles/cm$^3$) | | $PM_{10}$ (µg/m$^3$) | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | GM | GSD | GM | GSD | GM | GSD | GM | GSD | GM | GSD |
| Outdoors | | | | | | | | | | | |
| All | 147 | 1.1 | 2.0 | 0.4 | 2.4 | 0.08 | 1.8 | 0.004 | 2.0 | 19 | 1.9 |
| RH ≤ 30% | 66 | 0.9 | 1.9 | 0.4 | 2.3 | 0.07 | 1.9 | 0.004 | 1.8 | 15 | 1.8 |
| RH > 30% | 81 | 1.4 | 1.9 | 0.3 | 2.5 | 0.09 | 1.8 | 0.003 | 2.2 | 24 | 1.9 |
| Temperature ≤ 24°C | 79 | 1.0 | 1.9 | 0.3 | 2.4 | 0.08 | 1.9 | 0.004 | 2.0 | 18 | 1.7 |
| Temperature > 24°C | 68 | 1.3 | 2.1 | 0.5 | 2.3 | 0.08 | 1.8 | 0.004 | 2.1 | 21 | 2.1 |
| Nonsmoking | 47 | 1.1 | 1.8 | 0.3 | 2.4 | 0.07 | 1.8 | 0.004 | 1.7 | 17 | 1.7 |
| Smoking | 100 | 1.2 | 2.1 | 0.4 | 2.4 | 0.08 | 1.9 | 0.004 | 2.2 | 20 | 2.0 |
| Indoors | | | | | | | | | | | |
| All | 234 | 1.5 | 2.4 | 0.8 | 3.8 | 0.15 | 2.5 | 0.007 | 2.9 | 35 | 3.0 |
| RH ≤ 30% | 62 | 1.5 | 2.2 | 1.2 | 3.3 | 0.17 | 2.6 | 0.008 | 2.5 | 33 | 2.8 |
| RH > 30% | 172 | 1.5 | 2.5 | 0.7 | 3.8 | 0.15 | 2.5 | 0.007 | 3.0 | 36 | 3.0 |
| Temperature ≤ 24°C | 143 | 1.3 | 2.6 | 0.6 | 3.6 | 0.12 | 2.4 | 0.005 | 2.5 | 30 | 3.2 |
| Temperature > 24°C | 91 | 1.9 | 2.1 | 1.4 | 3.3 | 0.23 | 2.4 | 0.012 | 2.8 | 46 | 2.4 |
| No AC | 10 | 1.2 | 1.5 | 0.4 | 1.5 | 0.10 | 1.5 | 0.003 | 1.4 | 21 | 1.4 |
| AC | 224 | 1.5 | 2.5 | 0.8 | 3.8 | 0.16 | 2.6 | 0.007 | 2.9 | 36 | 3.0 |

Abbreviations: AC, air conditioning; GM, geometric mean; RH, relative humidity.

appear to be elevated over outdoor concentrations in the subway, bus, and museum microenvironments, with occasional peaks within restaurants. Indoor $PM_{10}$ concentrations also appear to vary more substantially across microenvironments than outdoor concentrations, an expected finding given the similarity among the urban outdoor settings. To determine whether the outdoor mass concentrations were homogeneous and could therefore be considered a single microenvironment, we applied a Wald test to the LME model for outdoor $PM_{10}$ measurements, controlling for temperature, relative humidity, and date of measurement. With this model, there was no evidence of outdoor microenvironment heterogeneity ($p$ = 0.75), a finding that was not altered by the inclusion of additional covariates.

Similarly, outdoor particle count concentrations and size distributions appear relatively similar across microenvironments (Figure 2). Wald tests on the outdoor particle count data (controlling for temperature, relative humidity, and date of measurement) found relatively little evidence of significant heterogeneity ($p$ = 0.03 for 0.3–0.5 µm; $p$ = 0.005 for 0.5–1.0 µm; $p$ = 0.11 for 1.0–5.0 µm; $p$ = 0.97 for 5.0 µm). Although we cannot reject heterogeneity for the smaller particles, multiple comparisons reveal that no differences are significant at the Bonferroni 5%. For pairwise 5% comparisons, only the park microenvironment differs significantly from other microenvironments for 0.3–0.5 µm, with a small number of significant comparisons for 0.5–1.0 µm. Thus, the overall evidence suggests that our outdoor count measurements are relatively homogeneous, and we therefore consider the outdoors as a single microenvironment in the multiple comparisons below.

Within indoor microenvironments (Figure 3), both the total particle count concentrations and the size distributions appear to differ significantly. For example, the subway microenvironment has its highest median particle count concentration within the 0.5–1.0 µm range, whereas the store microenvironment has relatively more coarse particles. In general, particle counts for larger particle sizes are greater in microenvironments with significant pedestrian traffic (i.e., museum and store), whereas particle counts for smaller particle sizes are highest near combustion sources (i.e., subway, bus, and restaurant).

Considering indoor/outdoor ratios stratified by microenvironment (defined as the ratio between the geometric mean concentrations indoors and outdoors, measured sequentially) helps to emphasize the differences among microenvironments (Table 3). For the subway, bus, and restaurant microenvironments, indoor count concentrations were most significantly elevated over outdoor concentrations for $PM_{0.5–1.0}$, a particle-size range associated with fuel combustion (both from proximity to traffic and indoor combustion sources). In the remaining four microenvironments, the greatest elevation occurred for $PM_{5.0+}$, indicative of dust or other coarse particles generated by human activities. The information from particle size counts provides more extensive evidence of source contributions than $PM_{10}$ mass measures. For example, the $PM_{10}$ indoor/outdoor ratios are almost identical in the museum and subway microenvironments, despite the differences in the types of particles penetrating from the

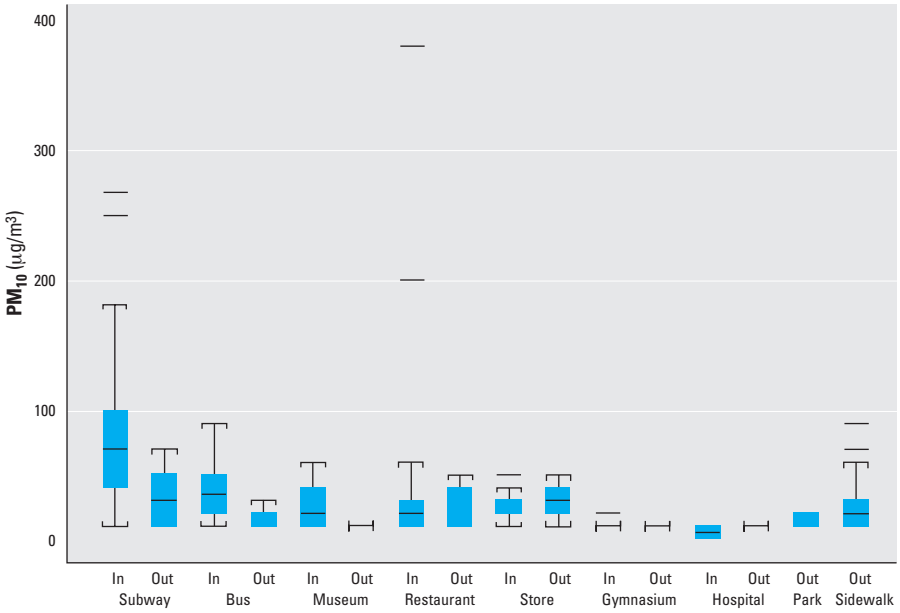


**Figure 1.** Distribution of indoor and outdoor $PM_{10}$ mass concentrations (µg/m$^3$), stratified by microenvironment. Upper and lower quartiles (box); median, black line in box; 1.5 interquartile ranges (bar); and outliers (—).

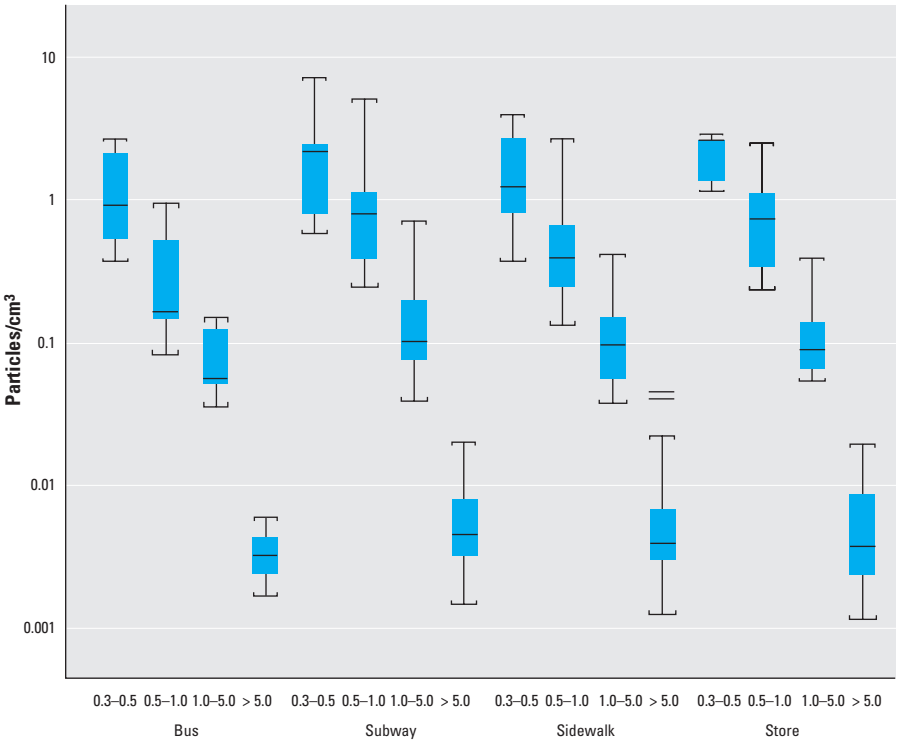


**Figure 2.** Log-scale box plot of particle counts (particles/cm$^3$) in four size ranges (0.3–0.5 µm, 0.5–1.0 µm, 1.0–5.0 µm, > 5.0 µm) for selected outdoor microenvironments. Upper and lower quartiles (box); median, black line in box; 1.5 interquartile ranges (bar); and outliers (—).

outdoors and generated indoors. In general, the fact that the indoor/outdoor ratio is < 1 only for the store microenvironment indicates that significant particle exposures occur in many indoor environments.

To move from these qualitative descriptions to quantitative comparisons between microenvironments, we constructed LME models for the eight microenvironments (seven indoor microenvironments and the pooled outdoor microenvironment). For our primary model, we controlled for date of measurement, temperature, and relative humidity. The indoor microenvironments with hypothesized proximity to combustion sources (subway, bus, and restaurant) tended to have significantly greater particle counts and mass concentrations than other microenvironments, particularly for smaller particle sizes (Table 4). For particles > 5.0 µm, store and museum had the highest count concentrations, although none of the differences between indoor microenvironments were statistically significant. In general, greater microenvironmental variability was seen for $PM_{0.3–0.5}$ and $PM_{0.5–1.0}$ than for larger particles.

To determine whether the differences among indoor microenvironments could be related to site characteristics, we constructed LME models including presence of open windows and central air conditioning. Using Wald tests, there was no evidence of significant heterogeneity associated with either of these parameters for $PM_{10}$ concentrations or particle counts ($p > 0.05$ for all).

## Discussion

In general, we found that mass concentrations of particulate matter as well as particle counts differed significantly among some indoor microenvironments, with higher levels found in close proximity to motor vehicles or other combustion sources. Our measurements of particle counts in different size ranges as well as particle mass concentrations allow for preliminary conclusions regarding the sources responsible for levels in different microenvironments. For example, the indoor subway microenvironment had significantly greater $PM_{10}$ concentrations than the indoor hospital microenvironment, with significantly greater particle count concentrations for particles < 5 µm and no significant differences for particles > 5 µm, despite similar outdoor levels. This result could be explained by the closer proximity to traffic for the street-level trolley and the better air filtration system in the hospital. In addition, $PM_{10}$ concentrations and fine particle counts were significantly higher within the subway than outdoors, implying that particulate matter could be concentrating within this microenvironment.

Along with the microenvironmental concentrations and patterns, our study applied a statistical model that accounted for autocorrelation between sequential measurements, an aspect that is often overlooked in continuous monitoring settings. Use of ordinary least squares for these data would have likely overstated the significance of our findings, given the significant autocorrelation. Simulations run under a variety of "true" correlation structures corroborate this statement, with ordinary least squares underestimating standard errors by approximately a factor of three. Furthermore, the use of GEEs to account for autocorrelation yielded significant Type I error for simulated data, implying that GEE models would conclude that greater heterogeneity existed among microenvironments than actually was found.

Along with the relevant information for personal exposure modeling, our study also provided useful benefits for the students participating in our Summer Program in Biostatistics. Students with good mathematics training but relatively little exposure to statistical analysis techniques were able to directly experience data collection and the issues inherent in field experiments. By participating in both the data collection and elementary statistical analyses, the students were able to gain a greater perspective on the interplay between these project elements.

Although our study demonstrated microenvironmental variability and estimated relative particle mass concentrations and counts for the microenvironments studied, the study design and methods imply some limitations in the generalizability of our findings. First, the sampling was conducted over a relatively limited time period, and the exposure patterns during these four summer days may not be representative of general exposure patterns. In addition, measurements were taken within the Boston area,

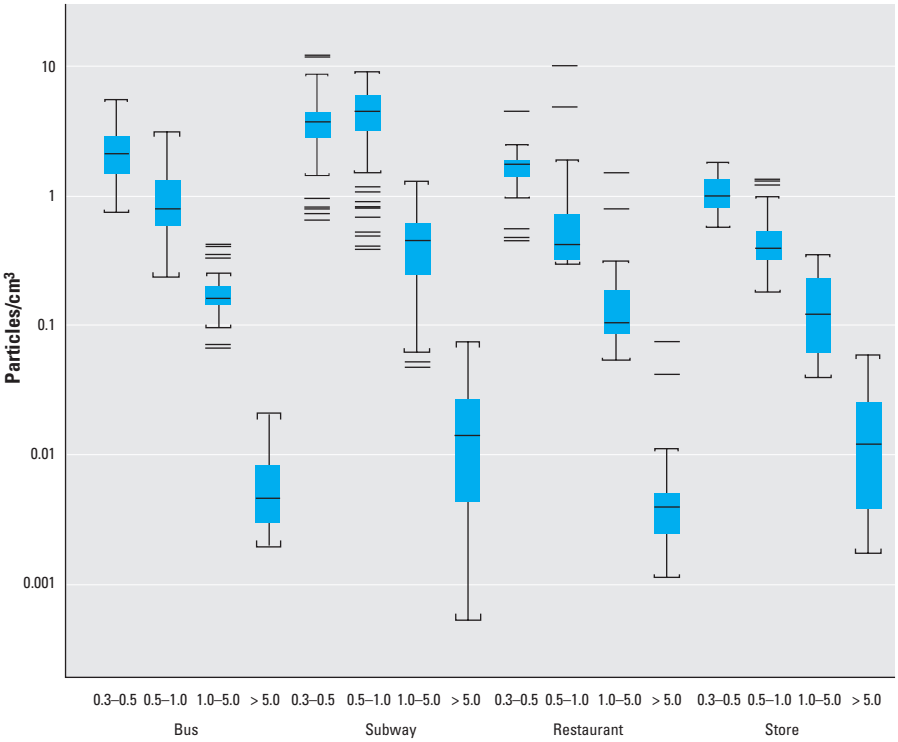


**Figure 3.** Log-scale box plot of particle counts (particles/cm$^3$) in four size ranges (0.3–0.5 µm, 0.5–1.0 µm, 1.0–5.0 µm, > 5.0 µm) for selected indoor microenvironments. Upper and lower quartiles (box); median, black line in box; 1.5 interquartile ranges (bar); and outliers (—).

**Table 3.** Ratios between geometric mean indoor and outdoor particle counts (particles/cm$^3$) and $PM_{10}$ concentrations (µg/m$^3$), stratified across urban microenvironments.

| | $PM_{0.3–0.5}$ | $PM_{0.5–1.0}$ | $PM_{1.0–5.0}$ | $PM_{5.0+}$ | $PM_{10}$ |
|---|---|---|---|---|---|
| Subway | 2.0 | 4.5[a] | 3.0 | 2.2 | 2.3 |
| Bus | 1.8 | 3.3[a] | 2.2 | 1.6 | 2.1 |
| Restaurant | 1.0 | 1.4[a] | 1.2 | 0.9 | 1.2 |
| Hospital | 0.8 | 1.4 | 1.7 | 1.8[a] | 1.0 |
| Gymnasium | 0.8 | 0.7 | 0.8 | 0.9[a] | 1.1 |
| Museum | 0.6 | 1.9 | 4.0 | 4.1[a] | 2.2 |
| Store | 0.5 | 0.6 | 1.1 | 2.5[a] | 0.8 |

[a]The largest particle count concentration ratio within each microenvironment.

and the microenvironments evaluated in our study may not correspond directly to similar microenvironments elsewhere. In other words, subway systems may be configured differently in different cities, restaurants may have different smoking laws, and stores may be located differently relative to busy streets. We attempted to include covariates that would control for some of these differences, but it is difficult to determine whether the lack of significance for many of the covariates implies that they have relatively small effects on particle mass concentrations or counts or whether this is simply a function of a relatively small sample size.

Interpretation of our particle count concentration data is limited by the fact that the APC-1000 has a lower limit of 0.3 µm, implying that ultrafine particles (including many combustion particles) were not measured in our analysis. It is also difficult to determine whether the "personal clouds" of the investigators contributed to the measured particulate concentrations, although their activities mimicked typical microenvironmental activities and the "persons nearby" term was statistically insignificant. Finally, although the categories of microenvironments were chosen to reflect common urban activities, the specific locations were not randomly selected according to systematic protocols. Thus, the concentrations that were allocated to a specific microenvironment may not be representative of the average concentrations in that microenvironment across the city of Boston. Nevertheless, the patterns in particle counts and mass concentrations are demonstrative of relationships that would be expected given source types and ventilation systems, which indicates that the general findings would likely remain consistent in a broader investigation.

**Table 4.** Multiple comparisons among microenvironments using LME model.[a]

| | Subway | Bus | Restaurant | Outdoor | Gymnasium | Store | Hospital | Museum |
|---|---|---|---|---|---|---|---|---|
| $PM_{0.3–0.5}$ (no./cm$^3$) | | | | | | | | |
| Subway | = | = | > | >> | >> | >> | >> | >> |
| Bus | = | = | = | > | > | >> | >> | >> |
| Restaurant | < | = | = | = | = | > | >> | > |
| Outdoor | << | < | = | = | = | >> | >> | > |
| Gymnasium | << | < | = | = | = | = | = | = |
| Store | << | << | < | << | = | = | = | = |
| Hospital | << | << | << | << | = | = | = | = |
| Museum | << | << | < | < | = | = | = | = |
| $PM_{0.5–1.0}$ (no./cm$^3$) | Subway | Bus | Restaurant | Outdoor | Store | Museum | Gymnasium | Hospital |
| Subway | = | > | >> | >> | >> | >> | >> | >> |
| Bus | < | = | = | > | > | = | > | > |
| Restaurant | << | = | = | = | = | = | = | > |
| Outdoor | << | < | = | = | = | = | = | > |
| Store | << | < | = | = | = | = | = | = |
| Museum | << | = | = | = | = | = | = | = |
| Gymnasium | << | < | = | = | = | = | = | = |
| Hospital | << | < | < | < | = | = | = | = |
| $PM_{1.0–5.0}$ (no./cm$^3$) | Subway | Bus | Restaurant | Museum | Store | Outdoor | Gymnasium | Hospital |
| Subway | = | = | > | = | >> | >> | >> | >> |
| Bus | = | = | = | = | = | > | > | > |
| Restaurant | < | = | = | = | = | = | = | > |
| Museum | = | = | = | = | = | = | = | = |
| Store | << | = | = | = | = | = | = | = |
| Outdoor | << | < | = | = | = | = | = | = |
| Gymnasium | << | < | = | = | = | = | = | = |
| Hospital | << | < | < | = | = | = | = | = |
| $PM_{5.0+}$ (no./cm$^3$) | Store | Museum | Subway | Restaurant | Bus | Hospital | Outdoor | Gymnasium |
| Store | = | = | = | = | = | = | > | = |
| Museum | = | = | = | = | = | = | = | = |
| Subway | = | = | = | = | = | = | > | = |
| Restaurant | = | = | = | = | = | = | = | = |
| Bus | = | = | = | = | = | = | = | = |
| Hospital | = | = | = | = | = | = | = | = |
| Outdoor | < | = | < | = | = | = | = | = |
| Gymnasium | = | = | = | = | = | = | = | = |
| $PM_{10}$ (µg/m$^3$) | Subway | Bus | Museum | Restaurant | Outdoor | Store | Gymnasium | Hospital |
| Subway | = | > | = | >> | >> | >> | >> | >> |
| Bus | < | = | = | = | > | > | = | >> |
| Museum | = | = | = | = | = | = | = | = |
| Restaurant | << | = | = | = | = | = | = | > |
| Outdoor | << | < | = | = | = | = | = | > |
| Store | << | < | = | = | = | = | = | > |
| Gymnasium | << | = | = | = | = | = | = | = |
| Hospital | << | << | = | < | < | < | = | = |

[a]<<, significant difference at Bonferroni 5% = 0.139%; <, significant difference at pairwise 5%, insignificant at Bonferroni 5%; =, no significant difference. The microenvironments with the highest concentrations are listed at the top; therefore, > indicates that the row element is greater than the column element; < means that the row element is less than the column element.

Future studies should focus on a more systematic collection of predictive covariates to allow for more substantive conclusions about relevant sources. Quantification of distance from combustion sources and information about air filtration systems would add substantially to the power of a microenvironmental exposure protocol. In addition, the lack of smokers in indoor microenvironments within our sample precluded us from drawing conclusions about the role of environmental tobacco smoke in indoor microenvironmental variability. Despite the omission of these parameters, the findings regarding microenvironmental variability as well as the microenvironments with highest and lowest concentrations were relatively robust with respect to the parameters included in the LME regression.

Particle size distributions in different microenvironments should also be investigated at greater length. In a recent study by Brauer and colleagues using the APC-1000 (*23*), the geometric mean indoor particle count concentrations in microenvironments in Vancouver were found to be 1.5 particles/cm$^3$ between 0.3 and 0.5 µm, 3.1 particles/cm$^3$ between 0.5 and 1.0 µm, 0.5 particles/cm$^3$ between 1.0 and 5.0 µm, and 0.04 particles/cm$^3$ > 5.0 µm. Thus, Brauer's study found a bulk of the particles to be in the 0.5–1.0 µm range, whereas our study found relatively more particles in the 0.3–0.5 µm range. This disparity could indicate geographic differences between Vancouver and Boston, differences in selected microenvironments, or could potentially be an instrumentation issue. In our analysis, $PM_{0.5–1.0}$ was greater than $PM_{0.3–0.5}$ only within the indoor subway or restaurant microenvironments. Because Brauer and colleagues focused on both transit and indoor microenvironments with cooking, these two microenvironments may be more representative of the microenvironments evaluated in Vancouver than other microenvironments, provided that the instrument implementation was identical. The peak indoor/outdoor ratios for subway, bus, and restaurant in our study were found in the $PM_{0.5–1.0}$ size range, which would be supportive of combustion peaks in this size range.

Finally, a logical extension of this research would involve collecting activity diaries and determining whether these microenvironmental concentration patterns improve the ability to predict personal exposures beyond simple models based on indoor and outdoor levels. In theory, many individuals

may spend a significant fraction of their time either at home, at work, or within microenvironments measured within this study or similar to those measured. This would allow for an accurate prediction of personal exposures as well as a preliminary determination of which sources contribute greatly to the exposure of a given individual.

## Conclusions

We have measured particle counts and mass concentrations within a range of indoor and outdoor microenvironments, finding significant variability among some indoor microenvironments and relative homogeneity among outdoor microenvironments. Higher $PM_{10}$ concentrations and fine particle counts were found in indoor microenvironments in close proximity to significant combustion sources (including subway, bus, and restaurant), indicating that these sources could contribute to variability in particulate matter exposure. Our findings indicate that information about time spent in indoor microenvironments would be useful in estimating personal exposure and in understanding which segments of the population are more likely to be highly exposed to both indoor and outdoor particle sources. Further investigations should focus on expanding the microenvironments investigated and the predictive covariates collected to determine the degree of microenvironmental variability and improve the ability to predict personal exposures.

## References and Notes


1. Dockery DW, Pope CA III. Acute respiratory effects of particulate air pollution. Annu Rev Public Health 15:107–132 (1994).
2. Schwartz J. Air pollution and daily mortality: a review and meta-analysis. Environ Res 64:36–52 (1994).
3. Levy JI, Hammitt JK, Spengler JD. Estimating the mortality impacts of particulate matter: what can be learned from between-study variability? Environ Health Perspect 108:109–117 (2000).
4. Schwartz J, Dockery DW, Neas LM. Is daily mortality associated specifically with fine particles? J Air Waste Manag Assoc 46:927–939 (1996).
5. Schwartz J, Neas LM. Fine particles are more strongly associated than coarse particles with acute respiratory health effects in schoolchildren. Epidemiology 11:6–10 (2000).
6. Schwartz J, Norris G, Larson T, Sheppard L, Claiborne C, Koenig J. Episodes of high coarse particle concentrations are not associated with increased mortality. Environ Health Perspect 107:339–342 (1999).
7. Churg A, Brauer M. Human lung parenchyma retains $PM_{2.5}$. Am J Respir Crit Care Med 155:2109–2111 (1997).
8. Dreher K, Jaskot R, Richards J, Lehmann J, Winsett D, Hoffman A, Costa D. Acute pulmonary toxicity of size fractionated ambient air particulate matter [Abstract]. Am J Respir Crit Care Med 153:A15 (1996).
9. Godleski JJ, Verrier RL, Koutrakis P, Catalano P. Mechanisms of Morbidity and Mortality from Exposure to Ambient Air Particles. Health Effects Institute Research Report No. 91. Cambridge, MA:HEI, 2000.
10. Gamble JF. $PM_{2.5}$ and long-term mortality in prospective cohort studies: cause–effect or statistical associations? Environ Health Perspect 106:535–549 (1998).
11. Moolgavkar SH, Luebeck EG. A critical review of the evidence on particulate air pollution and mortality. Epidemiology 7:420–428 (1997).
12. Dockery DW, Spengler JD. Personal exposure to respirable particulates and sulfates. J Air Pollut Control Assoc 31:153–159 (1981).
13. Sexton K, Spengler JD, Treitman RD. Personal exposure to respirable particles: a case study in Waterbury, Vermont. Atmos Environ 18:1385–1398 (1984).
14. Özkaynak H, Xue J, Weker R, Butler D, Spengler J. The Particle TEAM (PTEAM) Study: Analysis of the Data, Volume III [Draft Final Report]. EPA contract 68-02-4544. Research Triangle Park, NC:U.S. Environmental Protection Agency, 1993.
15. Janssen NAH, Hoek G, Harssema H, Brunekreef B. Personal exposure to fine particles in children correlates closely with ambient fine particles. Arch Environ Health 54:95–101 (1999).
16. Wallace L. Indoor particles: a review. J Air Waste Manag Assoc 46:98–126 (1996).
17. Carrothers TJ, Evans JS. Assessing the impact of differential measurement error on estimates of fine particle mortality. J Air Waste Manag Assoc 50:65–74 (2000).
18. Ott WR, Mage DT, Thomas J. Comparison of microenvironmental CO concentrations in two cities for human exposure modeling. J Expo Anal Environ Epidemiol 2:249–267 (1992).
19. Law PL, Lioy PJ, Zelenka MP, Huber AH, McCurdy TR. Evaluation of a probabilistic exposure model applied to carbon monoxide (pNEM/CO) using Denver personal exposure monitoring data. J Air Waste Manag Assoc 47:491–500 (1997).
20. Ott WR. Exposure estimates based on computer generated activity patterns. J Toxicol Clin Toxicol 21:97–128 (1984).
21. MacNee W, Donaldson K. Particulate air pollution: injurious and protective mechanisms in the lungs. In: Air Pollution and Health (Holgate ST, Samet JM, Koren HS, Maynard RL, eds). London:Academic Press, 1999;653–672.
22. Laden F, Neas LM, Schwartz J. The association of crustal particles in the fine particulate fraction with daily mortality in six cities [Abstract]. Am J Respir Crit Care Med 159:A332 (1999).
23. Brauer M, Hirtle RD, Hall AC, Yip TR. Monitoring personal fine particle exposure with a particle counter. J Expo Anal Environ Epidemiol 9:228–236 (1999).
24. Diggle PJ, Liang KY, Zeger SL. Analysis of longitudinal data. Oxford, UK:Clarendon Press, 1995.
25. Davidian M, Giltinan DM. Nonlinear Mixed Effects Models for Repeated Measurement Data. London:Chapman and Hall, 1995.
26. Kleinbaum DG, Kupper LL, Muller KE, Nizam A. Applied Regression Analysis and Other Multivariable Methods. 3rd ed. Pacific Grove, CA:Duxbury Press, 1998.