Factors affecting personal exposure to thoracic and fine particles and their components

Shao-I Hsu[1], **Kazuhiko Ito**[2], **Michaela Kendall**[3], and **Morton Lippmann**[2]

[1]Columbia University, Public Health, New York, New York, USA

[2]Department of Environmental Medicine, NYU School of Medicine, New York, New York, USA

[3]Exeter University, Department of Chemistry, Truro, Cornwall, UK

Abstract

Central monitoring site (CMS) concentrations have been used to represent population-based personal exposures to particulate matter (PM) of ambient origin. We investigated the associations of the concentrations of $PM_{2.5}$ and PM_{10} and their elemental components for elderly clinic patients with chronic obstructive pulmonary disease in two cities with different PM compositions, that is, New York City (NYC) and Seattle. Daily measurements of CMS, outdoor residential, and indoor PM_{10} and $PM_{2.5}$ concentrations, as well as personal PM_{10}, were made concurrently for 12-consecutive winter days at 9 NYC and 15 Seattle residences, as well for 9 NYC residences in summer. Filters were analyzed for elemental components using X-ray fluorescence (XRF), and for black carbon (BC) by light reflectance, and outdoor–indoor–personal relationships of PM components were examined using mixed-effect models. Using sulfur (S) as a tracer of PM of ambient origin, the mean contributions of outdoor $PM_{2.5}$ was 55.2% of the indoor concentrations in NYC, and 80.0% in Seattle, and outdoor $PM_{2.5}$ in NYC and Seattle were 19.7 and 18.5% of personal $PM_{2.5}$ concentration. S was distributed homogeneously in both cities ($R^2 = 0.65$), whereas nickel ($R^2 = 0.23$) was much more spatially heterogeneous. Thus, CMS measurements can adequately reflect personal exposures for spatially uniform components, such as sulfate, but they are not adequate for components from more local sources.

Keywords

personal exposure; population-based studies; environmental monitoring; exposure modeling; metals; particulate matter

Correspondence to: Dr. Morton Lippmann, 57 Old Forge Road, Tuxedo, NY 10987, USA. Tel.: +1-845-731-3558. Fax: +1-845-351-5472. morton.lippmann@nyumc.org.

CONFLICT OF INTEREST
The authors declare no conflict of interest.

INTRODUCTION

Owing to typical resource constraints for exposure assessment, information on characteristics of personal exposure to particulate matter (PM) remains sparse. Clayton et al.[1] and Thomas et al.[2] conducted field studies of human exposure that examined the relationships between ambient air PM mass concentrations and total personal PM exposures, but little is known about exposure to PM components. Central monitoring site (CMS) data are now available for the concentrations of $PM_{2.5}$ components on a 1-in-3 or 1-in-6 day basis, but not for components of PM_{10} or $PM_{10-2.5}$. Even for $PM_{2.5}$, with relatively high spatial uniformity, their adequacy for representing PM component exposures and their temporal or spatial variation is questionable. One of the research priorities on PM health effects of the US National Research Council[3] was assessing the relationship between ambient PM concentrations and personal exposures.

Previous Indoor–Outdoor–Personal PM Exposures Studies

Most of the prior personal PM mass exposure studies used cross-sectional designs, and obtained measurements from individuals over a limited number of days. Weak cross-sectional correlations were generally found between outdoor PM mass concentrations and personal PM mass exposures, and most reports concluded that the ambient PM concentrations were inadequate representations of personal PM exposures.

The personal–ambient relationship is a function of: penetration factor, air exchange rate, and particle deposition rate. Advances in source apportionment methods have provided quantitative tools for evaluating how much the different sources affect the personal exposures to PM.

Inter-personal and between-season variations make the quantification of the personal–ambient relationships a complex task. Furthermore, these variations also pose a challenge when addressing measurement error issues in time-series studies. Personal exposure studies have only been conducted in a handful of the cities, and none in New York City (NYC), which has a very large population with some atypical housing characteristics.

Objectives

The primary objectives of this research were to: characterize personal PM exposures for panels of respiratory disease patients; improve our understanding of chemical sources of PM; and to assess the appropriateness of using CMS ambient PM measurements in epidemiological air pollution health effect studies, given that personal PM exposures and their health-related effects differ by city, season, and cohort. The research was focused on the following aims:

1. To examine the personal exposures to two particle size fractions of PM_{10} ($PM_{2.5}$ and $PM_{10-2.5}$) of panel members residing in two urban areas (NYC and Seattle).

2. To quantify the relationships among personal PM_{10} and CMS, outdoor residential, and indoor $PM_{2.5}$ and $PM_{10-2.5}$ and their component concentrations measured in these two different cities.

3. To investigate the compositions of outdoor, indoor, and personal particles and assess the contributions of outdoor and indoor PM to personal exposures in these two communities with different climatic conditions and air pollution mixtures.

METHODS

Populations

Ambulatory elderly patients with physician-diagnosed moderate chronic obstructive pulmonary disease (COPD) or asthma were recruited primarily through Pulmonary Clinics at Bellevue Hospital in NYC and Asthma Clinics in Seattle. Some were referred by Clinic participants via patient support groups. The NYC cohort consisted of 9 COPD elderly patients, and the Seattle cohort consisted of 15 elderly patients with COPD, asthma, or both. All were current non-smokers. A preliminary cardiopulmonary test was conducted and an overview of the study was presented to each potential subject before they agreed to participate. A written informed consent was obtained from all before their participation in the study.

Air Sampling Methods

Home sampling strategy—Personal PM_{10} exposures for all participants were measured for 12 consecutive days in summer (July 2000–October 2000) and winter (November 2000–January 2001) in NYC, but only in winter (November 2002–March 2003) in Seattle. Concurrent indoor, outdoor residential, and CMS PM_{10} and $PM_{2.5}$ concentrations were measured daily throughout each sampling session, using Harvard Impactors (HIs) on 37-mm Teflon filters operating at 10 l/min for 24 h. Oiled impactor plates were used to minimize particle bounce and penetration to and collection of particles larger than 10 μm and 2.5 μm. Indoor samplers were located in the room where each participant spent most of their time. The sampling inlet was at a height of about 2 feet above the floor and away from walls. Outdoor residential samples were drawn in through probes that extended outdoors without affecting air exchange between outdoor and indoor air, with the sampling inlet kept away from the exterior wall to prevent interference from air eddies. Personal PM_{10} exposures were monitored using personal exposure monitors (PEMs) on 37-mm Teflon filters operating at 4 l/min for 24 h. The PM_{10} PEMs were equipped with battery-powered BGI pumps. We did not collect personal $PM_{2.5}$ samples because that would have required the wearing of two personal samplers and would have been too much of a burden for the elderly volunteer subjects.

Participants wore a vest that minimized the collection of PM_{10} resuspended from their clothes. They removed the personal PM_{10} sampler during prolonged periods of inactivity (e.g., sleeping, reading, and watching television) or during activities were wearing it would be too inconvenient (e.g., showering). When the vest was not worn, they kept the sampling inlet as close as possible to their breathing zones. They recorded the type and duration of their activities in daily diaries, which provided space to indicate specific microenvironments that they occupied. Confounding exposures included proximity to sources such as nearby tobacco smoking and cooking activities.

During each sampling season, field staff members visited the participants' residences every day to change sampling filters, replace personal pump batteries, and calibrate airflows. They also recorded the readouts of the cardiopulmonary function instruments used by the participants,[4] administered a brief questionnaire documenting ventilation status and potential indoor sources within the residences, and reviewed participants' time-activity diaries from the previous 24 h.

Central site monitoring—Ambient concentration data were obtained from a CMS in each city during each study period. At each CMS, which was located within 3 miles of the patients' residences, we collected ambient PM_{10} and $PM_{2.5}$ using HI samplers. The sites were located in midtown Manhattan in NYC (the 8th floor of the NYC Public Health Laboratory Building at East 26th Street and 1st Avenue), and at the Beacon Hill site in Seattle, and the sample collections coincided with those of the indoor and personal samples.

Air Sample Analyses

Gravimetric analysis—Before and after field sampling, all NYC filters were acclimated for at least 24 h in an environmental controlled weighing facility, which was maintained under negative pressure and at environmental equilibration at 25 °C (±5 °C) and 40% relative humidity (±5%). All PM masses were determined using gravimetric analysis with a micro-balance (Mettler MT5). Static charges on the filters were eliminated before weighing by using a ^{210}Po neutralizer.

For Seattle, filter mass measurements were conducted in the University of Washington weigh-room facility in a method consistent with that used by NYU. All filters were equilibrated for at least 24 h before weighing in a controlled environmental chamber with constant relative humidity (34.7%±2.5%) and temperature (22.4±1.9 °C). The weighing procedures were calibrated by applying certified standard weights, and laboratory blanks were used for quality assurance.

Trace element analysis—After the masses of the filters were determined, all filters were analyzed for the elemental composition using X-Ray fluorescence (XRF) spectroscopy at NYU. The NYU facility is equipped with an energy dispersive XRF spectrometer (Model6600AF, Jordan Valley, Austin, TX, USA) using a rhodium anode X-ray tube as the energy source to determine areal concentration of trace elements deposited on the filters. XRF spectroscopy is non-destructive technique capable of quantitative analysis of elements from Na to lead. XRF samples were blank-corrected and areal concentration was converted to ambient concentration.

Black carbon (BC) analysis—The filters were then analyzed for carbonaceous particles using a Reflectometer (Model M43M, Diffusion Systems, Harwell, London). The method for measuring particle reflectance is based on the Black Smoke protocol, and the darkness of the PM sample can be an indication of the amount of BC on the filter. Each filter was measured three times, and the average was used. The amount of reflected light was transformed to absorption coefficients. The conversion of the amount of reflected light and absorption coefficient was based on an international standard (ISO9835, 1993), and the

apparent BC concentration using this technique was well-correlated with elemental carbon (EC) measurements.

Data analyses—Data were organized and analyzed using the statistical package SAS (version 9; SAS Institute, Cary, NC, USA) and R (version 2.5; R-project.org). The components of the data analyses were: (1) characterization of outdoor, indoor concentrations, and personal exposures for the COPD cohorts from the two cities; and (2) examination of the contribution of ambient PM concentrations to corresponding indoor and personal exposures.

Characterization of PM

PM concentrations from CMS, indoor, outdoor, and personal samples were characterized using descriptive statistics, graphical depictions, and analysis of variance. Means, medians, and standard deviations were reported for all pollutant concentrations and exposures. Unusually high and low concentrations were examined, and were removed if they were the results of sampling error. Data values below the limit of detection, and negative pollutant concentration values were included in data analyses, as measured, in order to reduce possible bias in estimating relations among measurements. PM mass concentrations are reported in $\mu g/m^3$, and the elemental concentrations are reported in ng/m^3.

Mass concentration data were tested for normality using Shapiro–Wilks tests, and are presented as histogram plots. As pollutant concentration distributions were moderately skewed, we used non-parametric measures of association. Spearman correlation coefficients were used to examine the inter-relationship between outdoor, indoor, and personal PM concentrations, and for characterizing elemental concentration data. Time-series plots and boxplots were used to characterize the seasonal patterns, and to compare the findings across groups.

Contribution of Ambient Air Particles to Indoor and Personal Exposures

Quantitative assessment of the outdoor origin and indoor origin of PM to total indoor concentrations and personal exposures can be used to evaluate the implications of exposure errors, and for developing effective strategies for controlling outdoor exposures and mitigating indoor exposures. A regression approach was used to calculated home-specific values for PM infiltration.

Infiltration Factor using Sulfur as an Index Pollutant

Sulfur (S) was considered a valid tracer of outdoor-generated particles, because there were no known indoor S sources in our study. Using indoor and outdoor S to determine an infiltration factor has been shown to be successful.[5,6]

Estimation of Overall Ambient Contributions to Indoor Microenvironments and Personal Exposures

To estimate the overall contribution of ambient or outdoor PM to corresponding indoor PM in our study residences, mixed-model regression analysis was used in order to take into account of the repeated measurements on the same participant. In a similar manner, the

contributions to personal exposure from outdoor-generated PM were estimated using the same approaches.

Estimation of Source Contributions to PM Mass Concentrations

To examine the contributions of major PM species to PM mass in outdoor, indoor, and personal samples, elemental components were categorized into different source-related factors. "Soil" is the sum of the refractory oxides (Si, Ca, Ti, and Fe). BC was converted from the light reflectance readings. "Trace" represents the trace element grouping that includes sodium (Na), chlorine (Cl), Nickel (Ni), vanadium (V), zinc (Zn) and others present at low mass concentrations. The category classified as "Other" is defined as the difference between the measured mass and the sum of the measured species, and mainly constitutes organic materials, water, and nitrate.

RESULTS

Household Activities and Meteorological Patterns

The participants were non-smokers, with few exposed to environmental tobacco smoke (ETS) in their homes (i.e., <3% of the time); with the cohorts in NYC more likely to be exposed to ETS compared with the Seattle counterparts. For NYC, for 53% of the sampling days' meals were cooked at home, 64% of the sampling days had opened windows, and 27% of the sampling days had pets indoors. Windows were open more frequently in summertime than wintertime in NYC (36% *vs* 28%), and air-cleaning devices were used more often (15% *vs* 6%) summer and winter, respectively. There were no other seasonal differences in frequency for of behaviors such as cleaning and cooking. In general, Seattle subjects cooked less, and kept the windows closed most of the time, compared with those in NYC (19% for cooking and 22% had windows opened, respectively).

In the NYC summertime, the predominant winds came from the west (28% of the sampling time) and from the east (35% of the sampling time). High-speed winds (>10 mph) tended to came from the east. In wintertime, winds tended to came from the west (60% of the days), and average wind speed was between 5 and 10 mph. In the Seattle wintertime, winds blew from the south on >50% of the sampling days, and high winds (>10 mph) tended to come from that direction. The wind-rose plots for three sampling sessions are presented in Supplementary Figure 1.

Measurements of PM Mass Concentrations

New York City—Overall, the rate of successful sample collection ranged from 86% for the CMS PM_{10} to 95% for indoor $PM_{2.5}$ samples. Depending on the type of samples, the successful collection rates for each participant ranged from a minimum of 7 to all 12 days, with >50% of the participants having complete sampling data for all 12 days.

The summary statistics of all successful samples of personal, residential indoor, residential outdoor, and CMS PM_{10} and $PM_{2.5}$ data are presented in Table 1, whereas summary data for $PM_{10-2.5}$, which are based on subtraction of $PM_{2.5}$ from PM_{10}, are presented in Table 2. The distributions are reported by sampling locations for each city and separated by seasons.

Overall, summer PM concentrations were higher for all type of samples for both PM_{10} and $PM_{2.5}$. Maximum concentrations were typically found among the personal samples, with minima usually observed among CMS samples. The CMS and residential outdoor concentrations were more variable in summer than in winter, with coefficients of variation (CVs) of 0.56 *vs* 0.47 for residential outdoor $PM_{2.5}$ and 0.55 *vs* 0.54 for CMS $PM_{2.5}$ measurements.

Personal PM_{10} exposures were significantly higher than co-located indoor and outdoor PM_{10} concentrations, and personal > indoor > outdoor for both seasons. The overall distributions of concentrations by locations and by seasons are presented in Supplementary Figure 4. Day-to-day personal PM mass measurements varied more in both summer, and in winter were comparable to the residential outdoor and CMS measurements. The daily personal PM_{10} concentration was substantially higher than the concurrent daily indoor and outdoor PM_{10} concentrations for all participants, suggesting that personal activity, resulting in re-suspension of settled dust, contributed to the total personal mass concentrations. Furthermore, greater person-to-person variation in personal PM_{10} concentrations was observed compared with those for outdoor PM_{10} samples.

The coarse thoracic PM ($PM_{10-2.5}$), computed as the difference between the co-located PM and PM concentrations, had larger mean concentrations in indoor than the ambient samples, suggesting contributions from indoor sources, as shown in Table 2.

$PM_{2.5}$ was, on average, 90% of the PM_{10} mass in NYC, in both summer and winter. Distributions of the ratios ($PM_{2.5}/PM_{10}$) by sampling types are shown in Supplementary Table 1. Mean indoor ratios were lower than those outdoors in both seasons (0.77 *vs* 0.91 for summer, and 0.82 *vs* 0.90 for winter), indicating a greater influence of $PM_{10-2.5}$ on indoor PM_{10}. In addition, the mean mass contribution of outdoor air $PM_{10-2.5}$ to indoors was ~ 2 $\mu g/m^3$ both in summer and in winter. Furthermore, the interquartile range of the $PM_{2.5}/PM_{10}$ ratio was wider for indoor than for outdoor PM.

As shown in Supplementary Table 1, Spearman correlations showed that the personal PM correlated with indoor PM (r =0.72, 0.68, and 0.24 for $PM_{2.5}$, PM_{10}, and $PM_{10-2.5}$, respectively) for summer, but not for winter. Wintertime personal PM_{10} correlated better with outdoor PM_{10} measurements than with indoor PM_{10}. Good correlations were found between CMS and outdoor sites, suggesting that our pre-selected CMS could adequately represent the ambient PM mass concentrations at residential outdoors sites (r =0.7, and 0.9, winter and summer, respectively). Detailed correlation matrix summaries of sampling location by season are shown in Supplementary Tables 2 and 3, showing cross-sectional correlations between different PM metrics. There were variations in individual correlations between subjects, with longitudinal Spearman correlations between personal PM_{10} exposures and CMS PM_{10} measurements ranging between –0.29 and 0.56 in summer, and between –0.29 and 0.90 in winter. This between-subject variation may be attributed to different type, location, and magnitude of personal activities.

Seattle—Personal exposures varied widely, with the means being significantly greater than indoor and outdoor concentrations. The distributions and the patterns of personal, indoor,

and outdoor relationships are depicted in Supplementary Figure 3. The average indoor $PM_{10–2.5}$ concentration was 1.9 times higher than the coincident outdoor concentration, indicating potential indoor sources. Conversely, much higher $PM_{10–2.5}$ concentrations were found at the CMS, compared with residential outdoors, indicated a local $PM_{10–2.5}$ source near the CMS.

A majority of the outdoor and CMS PM in Seattle was $PM_{2.5}$, 78% and 73%, outdoor and CMS, respectively. By comparison, only about 66% of indoor PM was $PM_{2.5}$. Moreover, $PM_{10–2.5}$ contributed 2.8–3.3 $\mu g/m^3$ in outdoor PM mass, whereas contributing 4.6–3.2 $\mu g/m^3$ to the indoor PM mass.

The highest correlation for PM_{10} and $PM_{2.5}$ measurements was for residential outdoors and CMS ($r =0.76$). Personal PM_{10} correlated fairly well with indoor measurements ($r =0.55$, 0.42, and 0.56 for PM_{10}, $PM_{2.5}$, and $PM_{10–2.5}$, respectively), but correlated poorly with ambient measurements of all three of the size fractions ($r<0.2$). Correlations between personal PM_{10} and CMS PM_{10} among panel members varied widely, with r ranging from −0.18 to 0.79. For some participants, the personal exposures were primarily influenced by ambient PM levels, whereas for others, indoor and personal activity sources were the main contributors.

Comparisons between NYC and Seattle in PM Mass, S, and BC Concentrations

Similarities in NYC and Seattle—Correlations among PM measurements obtained at residential outdoors and PM concentrations measured at the CMS's in the two cities in the wintertime were similar. High correlations were observed for residential outdoor PM_{10} and CMS PM_{10}, that is, 0.72 for NYC and 0.71 for Seattle. For $PM_{2.5}$, it was 0.69 for both NYC and Seattle winter. Uniform spatial distributions were more pronounced for S, with distributions ranging from 0.68 in NYC winter to 0.91 in NYC summer.

The mean $PM_{2.5}/PM_{10}$ mass concentration ratios obtained at outdoor sites were higher than at co-located indoor sites, and for the three sampling seasons, indicating that the indoor concentrations are higher in PM10 and that indoor-generated sources are responsible for most of the $PM_{10–2.5}$.

In both cities, ambient $PM_{2.5}$ was correlated highly with ambient S. However, the S contribution in $PM_{2.5}$ varied with location. In both cities, the average indoor S concentration was lower than the corresponding mean outdoor and personal S concentrations, and the average personal S concentration was less or equal to the mean indoor S concentration. Moreover, the average personal S concentration was about 60% of the mean outdoor S concentration, for the three sampling sessions. S and BC concentrations constituted significant fractions of total $PM_{2.5}$ and PM_{10} mass in outdoor samples. On average, they constituted 18.8% of outdoor PM_{10} mass (range 16.9–21.5%), and 20.6% of outdoor $PM_{2.5}$ mass (range 17.1–23.0%).

There were higher correlations between pairs of indoor and outdoor co-located samples for S and BC than for $PM_{2.5}$ mass in both cities. The high correlation for S, which ranged from 0.73 to 0.87, is consistent with the results of studies conducted in the United States, Europe,

and Canada. Higher correlations were also observed between pairs of outdoor and personal samples for S and BC than for $PM_{2.5}$. The high correlation found for S and EC is consistent with them having few or no indoor sources, and small particle sizes, and their high degree of penetration from outdoors into homes. Outdoor $PM_{2.5}$ sulfate (calculated as 4.125 times the S concentrations based on the ratio of molecular weights for ammonium sulfate and elemental S) constituted 34% of outdoor $PM_{2.5}$ mass concentrations in summer, but decreased to 20% in winter. Moreover, mean S and BC concentrations in PM_{10} were similar to those in the $PM_{2.5}$, indicating that most of the S and BC were in $PM_{2.5}$. BC concentrations in the $PM_{10-2.5}$ were more than 10-fold lower than BC concentrations in the $PM_{2.5}$.

Differences between NYC and Seattle—Despite the similarities, discussed above, there were some key differences. Although the ambient $PM_{2.5}$ levels measured at the CMS stations in NYC and Seattle were very similar, the average outdoor $PM_{2.5}$ concentrations collected from resident's homes were more variable. The variability in residential outdoor concentrations could be due to differences in wind directions and potential local sources. A major difference between NYC and Seattle was the relationship among personal, indoor, and outdoor measurements. In NYC, average personal PM_{10} was significantly higher and the range of concentration was larger than indoor and outdoor PM_{10} levels, and indoor PM_{10} was greater than outdoor PM_{10}, and this personal > indoor > outdoor relationship was more pronounced in summer than in winter. In Seattle, although personal PM_{10} was higher than indoor and outdoor PM_{10}, the indoor and outdoor PM_{10} concentrations were similar. The much higher and more variable personal PM_{10} concentration found in NYC, compared with Seattle, could be attributed to the characteristics of the participants. In NYC, two study subjects required nebulizer therapy on a daily basis, and one subject carried an oxygen tank. These lifestyles and activities may have skewed the average personal PM exposures in the cohorts. Another possible explanation would be that the NYC cohort was slightly younger, may rely more on walking or public transportation when doing daily errands, and have more direct local traffic-related exposures than those in Seattle.

Elemental Components of PM

New York City—There were seasonal differences for some elements, with summer > winter for S, whereas winter BC was higher for both size fractions. Ni had much higher concentrations in winter than in summer. In addition, the contributions of S, BC, and Ni to PM_{10} and to $PM_{2.5}$ were essentially equal, indicating that these components were mainly within the $PM_{2.5}$. In contrast, soil elements, such as silica (Si), calcium (Ca), and iron (Fe) were mainly in the $PM_{10-2.5}$, as demonstrated in their larger percentage contributions to $PM_{10-2.5}$. Soil-derived elements, such as Ca and Si, were higher indoors than outdoors in both cities, and throughout the three sampling periods. A significantly higher personal concentration of Si was found compared with outdoor or indoor Si concentration, indicating a personal source of unknown origin. The personal to outdoor Si ratio was about 2.6 across the three sampling periods, whereas the ratio of personal to indoor Si was more variable, ranging from 1.2 in NYC winter to 2.3 in Seattle winter. No clear seasonal pattern was found in the concentrations for these crustal elements. Graphic representations of the seasonal pattern in temporal domain and boxplots are shown in Supplementary Figure 4.

For indoor and personal PM_{10}, the contributions from S and BC to the total mass were lower than their contributions to outdoor mass. In general, indoor and personal PM_{10} were heavily influenced by elements generated from personal activities; this was shown by comparing elemental mean concentrations found indoors and outdoors. For Si, the indoor and personal concentrations were 2–3 times higher than their concurrent outdoor concentrations. For elements that lack indoor sources, such as S and Ni, the indoor and personal concentrations were significantly lower than the corresponding outdoor concentrations. In addition, the personal and indoor concentrations of elements, lacking indoor sources tended to correlate well with their outdoor concentrations. Results of outdoor concentrations for selected elements in NYC are illustrated in Figure 1.

Spatial distributions of the CMS and residential outdoors elements indicated uniform distributions for PM_{10} and $PM_{2.5}$ S. Higher correlation for $PM_{2.5}$ S was found in summer than in winter, $r = 0.87$ and 0.71, respectively, and may be related to increased summertime transport of $SO_4^=$ to the region. The seasonal differences may be related to weather patterns and mixing heights. However, for BC and Ni, the magnitude of spatial correlation was lower ($r < 0.66$), suggesting that local sources were more influential. Moreover, for crustal elements (Ca, Si, and Al), spatial correlations between CMS and residential outdoor sites were generally quite low. The correlation for all summer data was <0.11, and it increased to <0.31 after removing three potential outliers. The spatial correlation for winter Al was higher than for its summer counterpart ($r = 0.23$ and 0.45, respectively). Low spatial correlations for crustal elements indicated not only local sources for these elements, but also that most crustal PM was in the $PM_{10-2.5}$, which did not travel long distances.

Overall, sulfate and BC contributed higher percentages to PM mass in outdoor than to indoor samples. Outdoor sulfate concentrations in both size fractions were more than twice those of indoor and personal samples. Outdoor and indoor BC concentrations exhibited similar contributions in both size fractions (10% and 4.5%, outdoor, and indoor respectively), and contributed 2.8% for personal PM_{10} samples. However, soil elements contributed more to the PM_{10} size fraction compared with the $PM_{2.5}$ size fraction, and this was more pronounced for indoor samples than for outdoor samples, with 10.6% *vs* 7.4% for outdoor samples, and 9.6% *vs* 4.6% for indoor samples. Trace elements concentrations in indoor and personal samples were >6 times higher than in outdoor samples, suggesting substantial indoor sources. When combining trace elements and other categories, these two categories explained >70% of the total mass concentrations in indoor and personal samples.

Seattle—BC was an abundant component in the outdoor PM in Seattle for both size fractions. Na, Cl, and S, had the next largest contributions in both outdoor PM_{10} and $PM_{2.5}$. Soil-related elements, by comparison, only comprised small fractions of the outdoor total PM mass.

Average personal PM_{10} Ca and Si concentrations were significantly higher than corresponding indoor and outdoor concentrations, with personal-to-indoor and personal-to-outdoor ratios ranging from 2.2 to 6.2, suggesting personal sources. In comparison, concentrations of indoor and personal S and BC, which lack known indoor sources, were lower, and similar to their outdoor concentrations.

The comparison of elemental concentrations in PM_{10} and $PM_{2.5}$ showed that, for combustion source elements such as S, the ratio of PM_{10} to $PM_{2.5}$ was close to unity for the outdoor PM and the indoor PM samples. In contrast, for soil-related elements, such as Ca, and Si, concentrations in PM_{10} were much higher than those in $PM_{2.5}$ (ratios of 4.0–5.1 for outdoor PM and 3.4–4.8 for indoor).

Outdoor $PM_{2.5}$ mass was highly correlated with outdoor BC, K, S, and Zn in the $PM_{2.5}$, with $r = 0.73, 0.82, 0.68,$ and 0.71, respectively. The strength of correlation was similar for outdoor $PM_{2.5}$ mass concentrations and those elements in the PM_{10} fraction (range 0.68 to 0.78). However, outdoor $PM_{2.5}$ mass correlated poorly with elements in the coarse size fraction (–0.07 to 0.21), indicating that BC, K, S, and Zn in ambient were primarily in the $PM_{2.5}$.

Outdoor PM_{10} and $PM_{2.5}$ mass concentrations were negatively correlated with outdoor Na and Cl for both for residential outdoor and CMS samples. One possible explanation is that during clean days, winds tended to blow from Puget Sound inland, resulting in particles enriched in Na and Cl, but containing less mass. Wind-rose plots of Na and Cl concentrations by direction and speed provided confirmation. Furthermore, during those days when winds tended to blow from the south to southwest directions (from Puget Sound); the wind speeds were generally >5 mph.

Soil elements in PM_{10} and $PM_{10–2.5}$ (Si, Ca, Ti, and Fe) were highly correlated, but they were not for $PM_{2.5}$, indicating that the soil-related elements were dominant in the coarse fraction. Ambient S $PM_{2.5}$ correlated well with K and V ($r = 0.53$ and 0.60, respectively), and moderately with BC ($r = 0.49$). Ambient K levels correlated highly with $PM_{2.5}$ BC concentrations ($r > 0.79$), but not with $PM_{10–2.5}$ BC concentration ($r = –0.08$) suggesting that the two elements may be emitted from the same source, with this source being predominantly in the fine mode. Furthermore, there was a high correlation between Na and Cl ($r > 0.80$), indicating these two elements were travelling together, and were generated from a common source.

Indoor PM_{10} mass concentrations correlated highly with $PM_{10–2.5}$ Ca and Fe ($r = 0.69$ and 0.78, respectively), and for indoor $PM_{2.5}$ mass, there was a high correlation with K and BC ($r = 0.63,$ and 0.76, respectively). Indoor PM_{10} K was strongly correlated with indoor $PM_{2.5}$ K ($r = 0.97$), but did not correlate with K in the $PM_{10–2.5}$ ($r = 0.02$), indicating that indoor K was primarily in the $PM_{2.5}$. Similar results were observed for S and BC. In contrast, soil-related elements (Al, Si, and Ca) correlated highly with $PM_{10–2.5}$ ($r = 0.83–0.98$), and correlated less with $PM_{2.5}$ ($r = 0.29–0.73$), indicating soil-related elements were primarily in the $PM_{10–2.5}$. Indoor Cl correlated highly with Na, and also with Br, suggesting the influence of an indoor source of Br and Cl, such as aerosol medications.

Personal PM_{10} mass correlated moderately with indoor soil-related elements (Al, Si, Ca, Sb, and Fe) both in the PM_{10} and in the $PM_{10–2.5}$ with $r = 0.33–0.50$ for PM_{10}, and $r = 0.25–0.46$ for $PM_{10–2.5}$. Poor correlations were found between personal PM_{10} mass and soil-related element concentrations in the ambient PM samples, indicating that the soil-related elements found indoors and those in ambient originated from different emission sources.

Elements from long-range transported PM tended to be distributed homogeneously in the atmosphere, at least in comparison with locally generated PM. Regressing S concentrations measured at the CMS on S concentrations at residential outdoor sites yielded a high degree of agreement ($R^2 = 0.86$), whereas components from combustion sources, such as BC and K, which are known tracers of biomass burning, were predominately in the $PM_{2.5}$, and had less homogeneous concentrations across the city ($R^2 = 0.5$ for BC and $R^2 = 0.54$ for K).

The difference in $PM_{2.5}$ composition in the two cities reflects their different pollutant sources. Coal-fired power plants in the eastern United States are the major source of S emissions that impact NYC, whereas local wood burning contributes significantly to BC concentrations in Seattle winters. The higher S contribution found in NYC summer could be attributed to predominant winds from the west or southwest. In winter, the predominant winds blow from north or northwest. The high BC contributions in NYC are attributed to local motor vehicle traffic, and to both motor vehicles and wood burning in Seattle. The difference in BC sources in NYC and Seattle is supported by the high correlation between BC and K for Seattle winter ($r > 0.80$) as compared with NYC winter ($r < 0.35$). A major difference between the two cities was the ambient Ni concentration, which was 10 to 29 times higher in NYC compared with Seattle, with the mean ambient Ni $PM_{2.5}$ concentration in NYC winter being 2.7 times higher than that in NYC summer.

The seasonal difference in Ni in NYC is due to different emission source strengths. Ni is a well-known tracer of residual oil combustion emissions from electric power generation and ocean-going ships, and it usually travels together with V. However, in NYC, there was little difference in mean V concentration in the two seasons, 11.3 ng/m^3 (summer) and 13.8 ng/m^3 (winter). Ni exhibited a very weak CMS–outdoor association in NYC ($R^2 = 0.23$) compared with that for S ($R^2 = 0.65$). The higher Ni concentration in NYC winter can only be explained by greater emissions from large commercial and residential buildings that burned residual oil for space and water heating, as reported by Peltier et al.[7] and Peltier and Lippmann.[8] They noted that residual oil combustion effluent during wintertime has a much higher Ni to V ratio compared with the residual oil burned in power plants.

DISCUSSION AND CONCLUSIONS

This research characterized the elemental concentrations of $PM_{2.5}$ and $PM_{10-2.5}$ measured at a CMS in each city, at series of residential sites outdoors and indoors, as well as personal PM_{10} exposures, for two cities with different PM components, in elderly pulmonary disease clinic patients. The primary analyses were conducted to examine and compare the similarity and the differences of $PM_{2.5}$ and $PM_{10-2.5}$ and their elemental components' concentration–response relationships for the NYC and Seattle patients. Temporal variation among personal, indoor, and outdoor PM concentrations were observed in both cities.

Representativeness of CMS Measurements

One uncertainty in exposure assessment is how representative a CMS needs to be when used in an epidemiological health effects study. Debate centers on the nature and extent of the time that people spend indoors and the often poor correlation between personal exposures and the CMS concentrations. Some studies have shown that higher personal–CMS

correlations improved when examining longitudinal data.[1,9,10] Consistent with these findings, we also found higher longitudinal personal–indoor– outdoor correlations for PM_{10} when compared with cross-sectional correlations, and this pattern remained across our two cities and seasons. The correlation was even stronger for S. The results have provided evidence supporting the usage of CMS concentrations as surrogates of population exposure to ambient $PM_{2.5}$, especially in the cities where regionally transported pollutants are the dominant sources. For cities where local sources might be more pronounced, the CMS could still serve population exposures, but using CMS data is likely to introduce greater measurement error.

In order to assess the bias introduced by the usage of CMS data as surrogates for personal exposure to ambient-generated PM, and to quantify this potential measurement error, we assessed the relationship between outdoor PM and indoor PM of outdoor origin.

Linear regression of CMS measurements on residential outdoor concentrations yielded a high degree of comparability, indicating the homogeneity of $PM_{2.5}$ concentrations within the NYC metro area, and to a lesser extent in Seattle. The slopes of the CMS–outdoor $PM_{2.5}$ regressions were 0.85 and 0.49, for NYC and Seattle, respectively. The higher NYC slope indicated that its $PM_{2.5}$ was mainly due to long-range transport, whereas the $PM_{2.5}$ in Seattle was due to both long-range transported and locally generated components. As a result, the CMS measurements collected in NYC may better characterize the PM mass concentrations across the whole city in comparison with measurements collected from the Seattle CMS. Furthermore, the CMS–outdoor relationship varied with the $PM_{2.5}$ composition. For example, the spatial gradients using S measurements showed $PM_{2.5}$ to be distributed relatively uniformly across NYC (R^2 =0.43 and 0.65, $PM_{2.5}$ and S, respectively). A similar pattern was observed for Seattle, where homogeneous spatial distributions were found both for $PM_{2.5}$ and S, but with a higher degree for S than $PM_{2.5}$. On the other hand, when we examined data by each sampled home, wide variations of the relationship were observed across homes. For example, slopes ranged from 0.52 to 1.21 for $PM_{2.5}$ measurements, and from 0.40 to 1.34 for S concentrations in NYC. The NYC variations may be attributed to different elevations of the sampled apartments, which were mostly in high-rise buildings, and possibly to the effects of the street canyons on wind velocities.

In contrast, Ni exhibited a very weak CMS–outdoor association in NYC (R^2 =0.23) compared with that for S (R^2 =0.65). Ni sources in NYC are much more locally variable than $PM_{2.5}$ mass and S, which are attributable to long-range transport. The indoor–outdoor, personal–outdoor, and personal–indoor relationships for Ni and S were similar in NYC, but the CMS–outdoor relationships were much higher for S than for Ni. Moreover, another component generated primarily from local sources, that is, BC from vehicular exhaust, did not have uniform spatial distribution. The slope of CMS–outdoor relationship for BC in NYC was 0.67, but this was higher than the slope for Ni of 0.46. The lower slope for Ni indicated that the Ni emissions in NYC came from fewer point sources. The results suggested that the S measurements obtained from the CMS can be used to reasonably predict the S measurements across NYC, whereas those for Ni and BC cannot.

One critical question in epidemiological health effect research is whether the concentrations from CMS are appropriate surrogates for personal exposures to PM. Using S as an outdoor tracer and computing the attribution of ambient PM to indoors, the indoor PM can be further partitioned into outdoor-origin PM and indoor-generated PM. To test how CMS concentrations represent personal exposures, the relationships between CMS and outdoor-origin PM, and CMS and indoor-generated PM were examined. First, regression was conducted for $PM_{2.5}$ mass concentrations measured at CMS on indoor $PM_{2.5}$ measurements. The regression models had low R^2 values, that is, 0.03 and 0.17, for NYC and Seattle, respectively. The relationships improved for CMS concentrations with indoor $PM_{2.5}$ of outdoor-origin, with R^2 values of 0.27 and 0.25 for NYC and Seattle, respectively. Furthermore, no relationships were found when regressing CMS measurement on indoor-generated $PM_{2.5}$.

Correlations between Indoor, Outdoor, and Personal Measurements

This study showed that personal PM_{10} concentrations were moderately correlated with indoor PM_{10}, but less so with outdoor or CMS PM_{10} concentrations. Individual personal to indoor correlation ranged from $r = -0.7$ to $r = 0.9$ and personal to outdoor correlation ranged from $r = -0.7$ to $r = 0.7$ for our two cohorts.

Our correlation values are lower than those from the Baltimore study conducted by Williams et al.[11] and a European study conducted by Brunekreef et al.,[12] but are compatible with those from other personal exposure studies.[13,14] The Williams et al. study was conducted in a retirement facility in which participants' communal lifestyle and the facility's uniform ventilation may have contributed to the higher correlation. The European study was done in Amsterdam and Helsinki, where the airsheds differed from our study sites, as did the PM compositions. We found indoor and outdoor PM concentrations in NYC differ more during the summer than in the winter, possibly due to the air conditioning usage in some apartments during the summer. Although mean summer concentrations of $PM_{2.5}$ and PM_{10} were higher than the winter, the differences were not significant, and this finding contrasts with the findings from previous studies carried out in the northeastern United States. The higher concentrations in the NYC winter, compared with those of other northeastern cities, may be due to the unique residual oil combustion effluents from residential and commercial buildings in NYC in the wintertime.[7,8]

We found similar, but slightly higher, average central site $PM_{2.5}$ and PM_{10} concentrations in Seattle sampling compared with those reported by Liu et al.[13] (13.7 *vs* 10.1 μg/m^3 and 18.1 *vs* 17.3 μg/m^3, $PM_{2.5}$ and PM_{10}, respectively). Our higher concentrations may be due to the more prevalent firewood burning during the colder winter months in Seattle. Liu et al. reported Spearman correlations of personal $PM_{2.5}$ to indoor $PM_{2.5}$ and personal $PM_{2.5}$ to outdoor PM_{10} of 0.65 and 0.56, respectively, whereas we reported 0.51. Our slightly lower value may be attributed to our collection of samples during the winter season only, whereas the Liu et al. study was conducted both in summer and winter seasons.

The mean ambient $PM_{2.5}$ and PM_{10} concentrations from Seattle were similar to those of Ebelt et al.[9] in Vancouver. However, the mean personal PM_{10} concentration from our Seattle cohort was 1.6 times greater than those in Vancouver. The differences in personal

exposure can be attributed to variations in the participants' activity patterns and the sampling season, or to lower concentrations in Vancouver. In addition, it should be noted that our cohort included both asthmatic and COPD patients, whereas the Vancouver cohort was restricted to COPD patients. Moreover, our study was conducted during the winter season, while the Ebelt et al. study was conducted in spring and summer.

Overall, ambient concentrations of PM_{10} were significantly associated with personal PM_{10} exposures for participants from both cities. The degree of individual correlation varied greatly, ranging from large negative to large positive values. The improvement using longitudinal association compared with cross-sectional association is consistent with results from other recent panel PM exposure studies.[9,15,16] It is believed that cross-sectional analyses result in lower correlations between personal and ambient concentrations potentially because the interpersonal variability may mask the intrapersonal correlations.

Characterization of Elemental Concentrations

Most of the prior panel-based exposure studies have focused on characterizing mass concentrations. Some studies have reported the $SO_4^=$ concentrations and their indoor, outdoor relationships, but few studies had detailed elemental characterization of the PM. One unique aspect of this research is its investigation of the compositions of both PM_{10} and $PM_{2.5}$ from indoor, outdoor, and CMS sites, and PM_{10} for personal samplers. Hence, this research could provide insights in understanding the PM components from different sources and origins (indoor, outdoor, and personal).

Ambient $PM_{2.5}$ is relatively rich in S and BC, but less so in PM_{10}. The S contribution to ambient $PM_{2.5}$ was more pronounced in NYC, whereas the BC contribution was more pronounced in Seattle. In addition, outdoor Na and Cl of the PM_{10} size fraction were enriched in Seattle, but not in NYC. Thus, there are different source impacts in the two cities. NYC, which is situated far downwind of most coal burning power plants, has elevated S concentrations compared with western US cities that are not impacted by upwind coal-burning power plants. On the other hand, Seattle, a west coast city, is heavily impacted by marine aerosol. Although the outdoor PM samples had higher BC concentrations than the indoor samples in both cities, the sources of the BC differ. In NYC, BC is a marker for traffic emissions, whereas in the Seattle winter, BC may also be generated from wood combustion. The higher BC emission and the wood smoke source in Seattle winter was noted by Larson et al.[17] when they apportioned the Seattle PM. Prior PM exposure studies have suggested that some potential indoor sources include re-suspended soil, cooking fumes, and ETS. In our study, we found a substantially higher contribution to PM_{10}, but not to $PM_{2.5}$, of Si, in indoor PM compared with outdoor PM. This enriched indoor PM_{10} Si is consistent with the presence of re-suspended dust in indoor environments. Elevated Na and Cl fractions in PM mass were significantly higher in some indoor and personal samples than in outdoor samples. The higher Na and Cl concentrations may have been due to the saline solutions used in nebulizer therapy by our study subjects in both NYC and Seattle, but was more pronounced in NYC. In Seattle, more of the outdoor Na and Cl may be contributed by marine aerosols.

Elemental correlation patterns were significantly different in indoor, outdoor, and personal PM, and varied by city and season. Outdoor PM_{10} was highly correlated with S and crustal elements (Si, Ti, and Ca) in NYC summer, but not in NYC winter. Outdoor PM_{10} mass, which includes $PM_{2.5}$, was highly correlated with S and V, but only moderately with soil components. This was expected, because pollution sources can differ by season. In the northeast United States, transported sulfate from coal-fired power plants dominated in summer, whereas residual oil burning in NYC was a dominant source in NYC in winter. The elemental profile varies by region as well. This geographical source difference is evident in that the outdoor PM_{10} correlated best with S, K, and BC in Seattle winter samples, but not in NYC samples. Another difference between NYC and Seattle is that high correlations were found between outdoor PM_{10} mass and soil components of the PM_{10} and $PM_{2.5}$ size fractions, however, the PM_{10} mass correlated poorly with soil components in the $PM_{10-2.5}$ fraction. In contrast, high correlations of PM_{10} mass and soil elements of the $PM_{10-2.5}$ were observed in Seattle samples. Furthermore, the PM_{10} Si was correlated moderately to highly with $PM_{2.5}$ Si in NYC, but not in Seattle, suggesting that the particle size distribution of soil elements was broader in Seattle than in NYC.

For the indoor and personal samples, mass of PM_{10} and $PM_{2.5}$ correlated highly with crustal elements (Si, Ca, Ti, Fe, Ba, and Al), indicating the contribution of soil sources. One unexpected observation was the good correlation between PM mass and Na, Cl, and Br, especially for NYC summer samples, suggesting a contribution from aerosol medication usage.

Spatial relationships of PM components at the participants' outdoor locations and the CMS indicated that S had little spatial variability in either NYC or Seattle compared with soil elements (Si, Ca, and Fe). These findings are consistent with results from other studies. The spatial distribution of K, Na, and Cl showed significantly different patterns between NYC and Seattle. Little-to-moderate spatial variability of these elements was found in Seattle, whereas it was high in NYC.

As in other studies, outdoor S correlated highly with indoor and personal S concentrations. The relationships were more pronounced in summer than the winter for NYC, probably because of the higher air exchange rate during the summer. Beside S, high indoor–outdoor and personal–outdoor correlations were found for BC, V, Se, and Ni, indicating that they likely originated from outdoor sources. For the sources located indoors, such as Na, Cl, and Br from aerosolized medications, there were weak indoor–outdoor correlations, but high indoor–personal correlations.

As part of major European Exposure and Risk Assessment for Fine and Ultrafine Particles in Ambient Air (ULTRA) study, Brunekreef et al.[12] reported indoor, outdoor, and personal $PM_{2.5}$ components for cardiovascular patients from Amsterdam and Helsinki. They found higher personal–outdoor correlations for BC and S concentrations compared with using $PM_{2.5}$ mass alone, which is consistent with our findings. As in our study, their distributions of indoor–outdoor ratio of soil elements (Si and Ca) were above 1 and were more variable compared with S and BC distributions. This suggests there are substantial indoor soil-related sources, and that their influence varies from house-to-house. Furthermore, coincident with

our findings, personal elemental concentrations correlated well with the indoor elemental concentrations, and the strong personal–indoor relationships apply to both outdoor-origin and indoor-origin elements. The personal–indoor relationship was similar but slightly lower compared with outdoor–indoor relationship for those major outdoor-origin elements (S and BC). This was expected because the participants usually spend >80% of their time indoors each day, and thus personal exposure is highly influence by indoor concentrations.

Implications of the Study Findings

Although people spend most of their time in indoor environments, this research has demonstrated that day-to-day fluctuations in ambient PM concentrations affect day-to-day PM exposures at the personal level. The total personal PM exposure is attributable to both ambient air and non-ambient air contributions. The subject-to-subject variation of ambient contributions is due, in part on residential ventilation rates. The air exchange rate is generally lower when using air-conditioning compared with when windows are open, and thus outdoor air infiltration is lower. Season is a modifying factor for infiltration.

Our results provide some support for using ambient $PM_{2.5}$ concentrations measured at CMS as surrogates of population exposures, but show that the appropriateness of such usage varies with geographical location. In a Northeast urban city, secondary sulfate is a dominant source of the $PM_{2.5}$ mass, and S can serve as a reasonably reliable surrogate for estimating the ambient $PM_{2.5}$ contribution to personal exposures. In other regions, where predominant sources and the size distribution of the $PM_{2.5}$ mixture differ from Northeast PM mixtures, using S may not be an appropriate surrogate for ambient $PM_{2.5}$ exposures at personal levels. For example, in a separate paper that examined associations of daily variations of $PM_{2.5}$ and $PM_{10-2.5}$ and their elemental component concentrations with short-term changes in cardiac and pulmonary functions in our COPD cohorts, we reported that the only significant associations that we observed were those between heart rate and the concentrations of Ni in outdoor residential, indoor, and personal PM in NYC, and there was no such association for the NYC CMS samples.[4] There were no significant associations in the Seattle cohort, which had much lower Ni concentrations.

When using CMS speciation data in epidemiological studies that investigate the association between ambient PM component concentrations and increased morbidity and mortality, the impacts of spatial and temporal concentration variations need to be considered, and our findings illustrate the complexity of this issue, especially for components emitted by local sources. The personal–outdoor relationships characterized in this research represent an initial attempt to assess the strengths and limitations of CMS data in two urban communities with quite different PM mixtures. In any case, our results offer evidence that, depending on the constituents of the PM, different elements contribute different degrees of measurement errors inherent in using CMS data. Further research in characterizing the extent of the measurement error, and systematically improving our models for exposure, would minimize the uncertainty in observational air pollution studies.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

Acknowledgments

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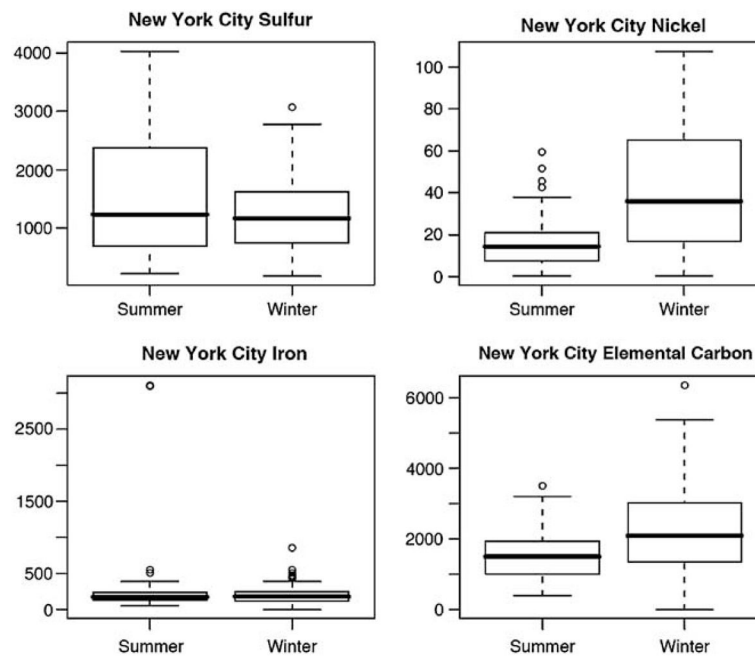
Figure 1.
Boxplots of outdoor $PM_{2.5}$ concentrations for selected elements in NYC. The median value is represented by the dark bar, 25th to 75th percentile is represented by the box, and 10th to 90th percentiles are represented by the whiskers. The concentrations on the *y* axes are in ng/m³.

Table 1

PM mass concentrations for different types of samples.

	N	Min	1 Qt	Mean	3 Qt	Max
NY summer PM$_{10}$ (µg/m³)						
CMS	91	4.08	7.59	14.67	17.71	51.99
Outdoor	94	6.89	12.44	20.59	26.47	75.53
Indoor	95	8.45	22.92	37.18	46.18	109.1
Personal	91	8.45	38.81	71.09	92.42	260.1
NY summer PM$_{2.5}$						
CMS	89	4.02	7.49	12.94	17.08	45.13
Outdoor	91	5.54	10.14	18.58	24.27	62.00
Indoor	96	5.63	15.93	28.83	38.13	95.19
NY winter PM$_{10}$						
CMS	72	5.61	11.33	15.50	18.92	31.29
Outdoor	86	4.28	12.92	18.83	23.79	42.75
Indoor	11.22	19.68	28.72	32.66	32.66	106.2
Personal	84	8.99	32.97	52.62	61.66	280.0
NY winter PM$_{2.5}$						
CMS	76	2.58	8.09	13.17	17.79	29.92
Outdoor	85	4.11	11.28	16.71	20.67	37.69
Indoor	87	7.05	14.22	21.63	25.23	81.76
Seattle winter PM$_{10}$						
CMS	166	2.83	8.42	18.13	26.07	51.00
Outdoor	168	2.91	7.52	13.64	17.08	42.67
Indoor	170	1.00	7.80	12.32	15.19	50.61
Personal	141	1.75	16.34	30.18	37.31	108.1
Seattle winter PM$_{2.5}$						
CMS	168	1.56	5.86	13.68	18.83	44.35
Outdoor	166	2.50	4.99	10.70	13.97	35.59
Indoor	172	1.57	4.33	7.55	8.81	37.30

Table 2

Summary of $PM_{10-2.5}$ mass concentrations.

	N	Mean	SD
NYC summer			
CMS	87	1.75	6.11
Outdoor	89	2.12	5.98
Indoor	95	8.34	9.70
NYC winter			
CMS	74	1.94	5.29
Outdoor	80	2.38	5.80
Indoor	82	5.61	7.86
Seattle winter			
CMS	160	4.14	3.59
Outdoor	162	2.84	3.30
Indoor	166	4.64	3.24