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While a number of studies have looked at the relationship between outdoor and indoor particulate levels based on daily (24 h) average concentrations, little is known about the within-day variability of indoor and outdoor PM levels. It has been hypothesized that brief airborne particle excursions on a time scale of a few minutes to several hours might be of health significance. This article reports variability in measurements of daily (24 h) average PM$_{2.5}$ concentrations and short-term (15 min average) PM$_{2.5}$ concentrations in outdoor and indoor microenvironments. Daily average PM$_{2.5}$ concentrations were measured using gravimetry, while measurements of 15 min average PM$_{2.5}$ mass concentrations were made using a light scattering photometer whose readings were normalized using the gravimetric measurements. The measurements were made in 3 urban residential neighborhoods in the Minneapolis–St. Paul metropolitan area over 3 seasons: spring, summer, and fall of 1999. Outdoor measurements were made at a central monitoring site in each of the 3 communities, and indoor measurements were made in 9–10 residences (with nonsmoking occupants) in each community. Residential participants completed a baseline questionnaire to determine smoking status, sociodemographics, and housing characteristics. Outdoor PM$_{2.5}$ concentrations across the Minneapolis–St. Paul metropolitan area appear to be spatially homogeneous on a 24 h time scale as well as on a 15 min time scale. Short-term average outdoor PM$_{2.5}$ concentrations can vary by as much as an order of magnitude within a day. The frequency distribution of outdoor 15 min averages can be described by a trimodal lognormal distribution, with the 3 modes having geometric means of 1.1 μg/m^3 (GSD = 2.1), 6.7 μg/m^3 (GSD = 1.6), and 20.8 μg/m^3 (GSD = 1.3). There is much greater variability in the within-day 15 min indoor concentrations than outdoor concentrations (as much as ∼40-fold). This is most likely due to the influence of indoor sources and activities that cause high short-term peaks in concentrations. The indoor 15 min averages have a bimodal lognormal frequency distribution, with the 2 modes having geometric means of 8.3 μg/m^3 (GSD = 1.66) and 35.9 μg/m^3 (GSD = 1.8), respectively. The correlation between the matched outdoor and indoor 15 min average PM$_{2.5}$ concentrations showed a strong seasonal effect, with higher values observed in the spring and summer (R^2_{adj} = 0.49 ± 0.33) and lower values in the fall (R^2_{adj} = 0.13 ± 0.13).

INTRODUCTION

Recent epidemiological studies have shown an association between airborne particulate levels and morbidity and premature mortality (U.S. EPA 1996; Vedal 1997). There is also growing evidence that fine particles (PM$_{2.5}$, i.e., particulate matter <2.5 μm in aerodynamic diameter) may be responsible for these associations (Schwartz et al. 1996; Schwartz and Neas 2000; Samet et al. 2000). Most of these studies have focussed on evaluating changes in health endpoints associated with changes in 24 h average PM levels over 1–5 days (Pope 2000).

It is generally agreed that the indoor microenvironment is an important determinant of human exposure to PM. People spend a large fraction of their time indoors engaged in various activities that generate PM, such as wood combustion, smoking, and cooking (Wallace 1996). A number of studies have looked at the relationship between outdoor and indoor particulate levels based on 24 h average concentrations (Evans et al. 2000; Janssen et al. 1998, 2000). However, little is known about the within-day variability of indoor and outdoor PM levels. Several recent articles have hypothesized about the health significance of brief airborne particle excursions on a time scale of a few minutes to several hours, suggesting that these excursions might explain some of the excess mortality (Michaels 1996, 1997; Michaels

and Kleinman 2000). This hypothesis is based on a number of acute human exposure studies (∼25) cited in Michaels and Kleinman (2000) and recent animal studies (Godleski et al. 1999; Clarke et al. 2000) that show evidence of adverse health effects due to such brief excursions.

This article presents results from a study of 15 min average and 24 h average (referred to from now on as daily average) $PM_{2.5}$ concentrations measured concurrently at outdoor (central site) and indoor (residential) locations in the Minneapolis–St. Paul metropolitan area. We describe within-day variability in 15 min average $PM_{2.5}$ concentrations, correlations between simultaneous measurements of 15 min averages at different locations in the metro area, and factors affecting the short-term indoor to outdoor (I/O) ratios.

METHODS

Measurements of outdoor and indoor daily average and 15 min average $PM_{2.5}$ mass concentrations were made in 3 urban residential communities in the Minneapolis–St. Paul metropolitan area (population 2.6 million with 1.04 million housing units and a population density between ∼365/km^2 and ∼13,700/km^2 in 2002). The communities—Phillips, East St. Paul, and Battle Creek—are separated from each other by a distance of 8 to 11 km. Figure 1 shows a map of the metro area and the location of these 3 communities. The communities were chosen on the basis of a modeling study of Volatile Organic Chemicals (VOC) dispersion (Pratt et al. 1998). Healthy nonsmoking adults (23 females, 9 males; mean age 42 ± 10, range 24–64 years) were recruited in each neighborhood using a sampling scheme that ensured a geographically diverse distribution of residences around the outdoor central monitoring site. The participants completed a baseline questionnaire to determine smoking status, sociodemographics, housing characteristics, and a brief health history.

The measurements were made over 3 seasons—spring (April 26–June 2), summer (June 20–August 10), and fall (September 23–November 20) of 1999. The daily average and 15 min average $PM_{2.5}$ measurements were matched so that over
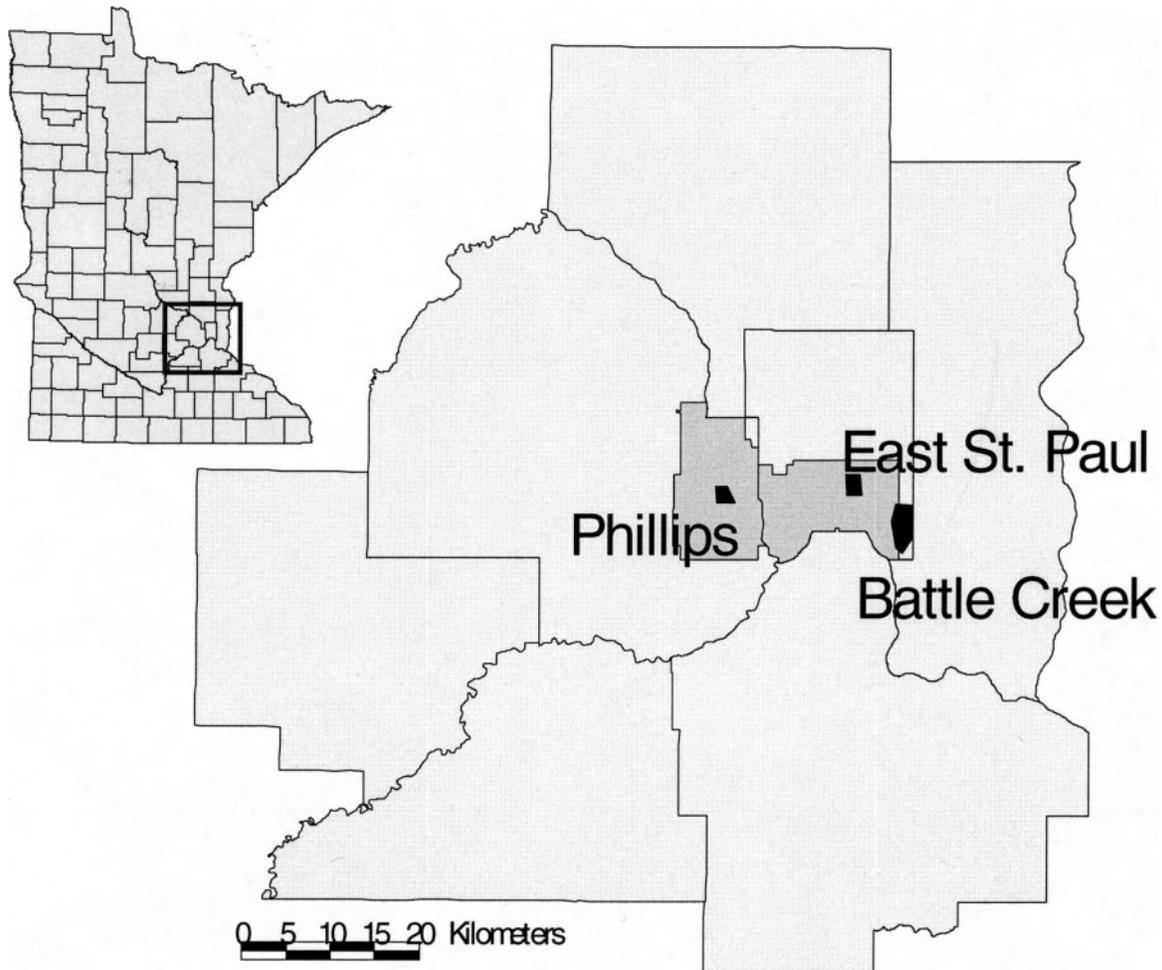

Figure 1. Map of the Minneapolis–St. Paul metropolitan area showing the location of the 3 communities where the study was conducted. The upper left side shows a map of Minnesota, and the metro (7-county) area boundaries are shown in the center. The darker gray areas represent the cities of Minneapolis and St. Paul.

each 24 h period, we obtained 1 daily gravimetric average mass concentration using a PM$_{2.5}$ impactor inlet and 96 15 min average mass concentrations using a collocated light scattering photometer. Outdoor measurements were made at a central monitoring site in each of the 3 communities. Matched indoor 15 min and daily average PM$_{2.5}$ mass concentrations were also measured concurrently in 9–10 residences (with nonsmoking occupants) in each community. Measurements were made on weekdays as well as weekends in all 3 communities. Thus for a given calendar day, we obtained 1 set of outdoor measurements from each community outdoor site and several sets of indoor residential measurements from each community. For each of the monitoring days, we obtained hourly meteorological measurements (temperature, relative humidity, dewpoint, pressure, wind direction and speed, and ceiling height) made at the National Weather Service station at the Minneapolis–St. Paul International Airport. These data were obtained from the National Climatic Data Center (Asheville, NC). A summary of the different types of measurements made during the course of this study is provided in Table 1. The details of the measurement methods are described in the following subsections.

Outdoor and Indoor PM$_{2.5}$ 24 h Average Gravimetric Measurements

Outdoor measurements were made at a central monitoring site in each of the 3 communities on a midnight-to-midnight schedule. These measurements were made on the roof of a school building in Battle Creek (~10 m from the ground), and on the top of a fire station in East St. Paul (~10 m from the ground), and on the roof of a community center in Phillips (~15 m from the ground). While the Phillips and East St. Paul sites are part of the State and Local Air Monitoring Stations (SLAMS) and National Air Monitoring Stations (NAMS) networks, respectively, the Battle Creek location is a special purpose site. The siting criteria are based on federal guidelines (U.S. EPA 1998). The sites were near the approximate geographic center of each neighborhood and were sufficiently far away from major streets and industrial sources. The outdoor daily average gravimetric concentrations were obtained with an Andersen RAAS2.5-300 at a sampling flow rate of 16.67 lpm (±5%). The samples were collected on 46.2 mm PTFE 2 μm pore-size filters (Whatman Inc., Clifton, NJ). This sampler meets the specifications for PM$_{2.5}$ federal reference method (FRM) samplers described in

Table 1

Summary data for valid PM$_{2.5}$ 24 h average and 15 min average concentrations and I/O ratios

	Number of valid 24 h measurement periods				Mean	Median	Standard deviation	(10th percentile, 90th percentile)
	Total	Spring	Summer	Fall				
Outdoor concentration								
Gravimetric 24 h average (μg/m^3)	271[1]	102	77	92	10.3	8.6	6.6	(3.7, 26.4)
DustTrak 15 min average (μg/m^3)	102[2]	26	20	56	10.5	8.4	8.4	(2.1, 22.1)
Indoor concentration								
Gravimetric 24 h average (μg/m^3)	294[3]	70	98	126	13.9	10.9	11.6	(5.2, 23.5)
DustTrak 15 min average (μg/m^3)	201[4]	36	66	99	14.2	9.7	22.1	(4.6, 23.2)
Matched I/O measurements								
I/O ratio based on 24 h average concentrations	248[5]	63	84	101	1.7	1.3	1.6	(0.6, 2.9)
I/O ratio based on 15 min average concentrations	33[6]	8	8	17	2.7	1.2	8.7	(0.6, 3.6)

[1] 336 total outdoor gravimetric samples attempted, with 65 (19%) invalidated due to equipment failure. Valid samples were obtained over 112 calendar days.

[2] 387 total outdoor 24 h DustTrak measurements attempted. 153 (39%) were invalidated because no corresponding gravimetric measurements were made for calibration purposes. 132 (34%) were invalidated due to unacceptable zero drift. Valid samples were obtained over 69 calendar days.

[3] 367 total indoor gravimetric samples attempted, with 62 (16.9%) of the measurements invalidated due to pump problems (e.g, flows outside of target range), and 19 (5.2%) of the measurements invalidated due to filter problems (e.g, punctures, mishandling). Valid samples were obtained over 96 calendar days.

[4] 246 total indoor 24 h DustTrak measurements attempted. These are a subset of the 367 gravimetric measurements attempted. 45 (18%) of the measurements were invalidated due to problems with the gravimetric measurements that are needed for calibrating the DustTrak. No sample losses were due to zero drift. Valid samples were obtained over 88 calendar days.

[5] These are measurement periods for which valid outdoor and indoor gravimetric measurements were available. Valid measurements were obtained over 86 calendar days. Outdoor measurements started at midnight, while indoor measurements started sometime in the evening (4–9 PM).

[6] Concurrent valid outdoor and indoor DustTrak measurements were available for 33 complete 24 h measurement periods. These are a subset of the 102 outdoor monitoring periods and 201 indoor monitoring periods. Both outdoor and indoor measurements started at midnight.

40 CFR Part 50, Appendix L (U.S. EPA 1997). The sampling period for each gravimetric sample taken at the community central sites was from midnight to the next midnight. Final concentrations were adjusted for passive loading. A detailed description of the sampling protocol and the procedure for correcting passive loading artifacts is given in Ramachandran et al. (2000). A valid outdoor measurement had to meet all aspects of the FRM and be corrected for passive loading artifacts.

Indoor residential measurements were obtained from 9 residences in the Phillips and East St. Paul communities and 10 residences in Battle Creek. These measurements were made on a 24 h basis, starting and ending in the evening (e.g, 4–9 PM) rather than at midnight for participant convenience and logistical reasons. The average overlap between indoor and outdoor measurements was 17.5 ± 1.9 h. The measurements were made in a room of the residence where the study participant spent most of his/her waking time, and the sampler was placed at approximately the subject's seated breathing height ($\sim$1.0–1.2 m high) in the room where he/she reported spending the majority of his/her waking hours and away from local particle sources. Daily average gravimetric samples were obtained using $PM_{2.5}$ inertial impactor PEM inlets (MSP, Inc., Minneapolis, MN), whose annular impaction surfaces were lightly coated with silicone grease. The samples were collected on 37 mm Teflon filters (Gelman Sciences, Ann Arbor, MI) with a 2 μm pore size on a polyolefin ring. A sampling flow rate of 10 lpm ($\pm$5%) was obtained using Buck-Genie Extra pumps (AP Buck, Inc., Orlando, FL). The sampler flow rate was calibrated at the beginning and end of each 24 h sampling period with a Mini-Buck Calibrator (Model M-30, AP Buck Inc., Orlando, FL). A valid indoor measurement had to meet all aspects of this protocol.

Quality Assurance for Gravimetric Measurements

Both outdoor and indoor filters were weighed in an automated microbalance system (Cahn Instruments, Cerritos, CA) accurate to 1 μg and located in a climate-controlled room, which met temperature and humidity guidelines prescribed by the FRM (relative humidity between 30 and 40%, with a variability of no more than 5% over 24 h and temperature between 20 and 23°C, with a variability of no more than 2°C over 24 h). Further details of the measurement protocol are provided in Ramachandran et al. (2000).

For outdoor samples, the average change in field blank weight was 10.9 μg ($n = 16$; SD $= 6.4$), and the detection limit, defined as 3 times the standard deviation of the field blanks divided by the average sampled air volume, was 0.8 μg/m^3. For indoor samples, the average change in field blank weight was 6.2 μg ($n = 37$, SD $= 17$ μg) and the detection limit was 3.6 μg/m^3.

We also compared the Andersen RAAS2.5-300 (blank subtracted and corrected for passive loading) and the PEM sampler (blank subtracted) by collocating them and obtaining daily average concentrations for 18 separate days. The differences between the concentrations obtained using the 2 instruments were analyzed with a t-test using the MEANS procedure in SAS

(Version 8.01, SAS Institute, Inc., Cary, NC). The differences were not statistically significant ($p > 0.34$).

15 Min Average $PM_{2.5}$ Mass Concentrations

DustTrak instruments (TSI Inc., Model 8520) with $PM_{2.5}$ inlets were used to measure 15 min average $PM_{2.5}$ concentrations indoors and outdoors. For the outdoor measurements, the DustTraks were placed in a wooden box to protect against rain and snow and collocated with the FRM samplers. Air was drawn in through a sampling tube extending through an opening in the box. For the indoor measurements, the DustTraks were collocated with the indoor PEM samplers and away from indoor sources and inside a closed box to prevent light intrusion and disturbance by participants. The DustTrak is a light scattering laser photometer that measures the light scattered at 90° using a solid-state silicon photodetector. The intensity of the scattered light is a function of the particle mass concentration, the size distribution of the aerosol, and its composition. The relationship between scattered light and the aerosol size distribution and refractive index is given by Mie theory (Kerker 1969). The laser diode used by the DustTrak has a wavelength of 780 nm, which limits the smallest detectable particle to about 0.1 μm. This instrument has an internal pump that continuously draws the aerosol through the sensing chamber at a flow rate of 1.7 LPM ($\pm$3%).

For a given particle size, scattered light is directly proportional to the number concentration of particles. Thus for a given constant aerosol mass concentration, intensity of scattered light decreases with increasing particle size. Since the DustTrak was calibrated using the respirable fraction of standard ISO 12103-1, A1 test dust (Arizona Test Dust; $MMAD = 4.4$ μm), the instrument response to a finer $PM_{2.5}$ aerosol will be biased 3–5 times higher than the true value due to the larger number of particles in the finer aerosol for the same mass concentration. This response can be theoretically calculated if one knows the particle size distribution and refractive index of the calibration aerosol as well as the test aerosol (Görner et al. 1995). Since this was not possible in a field study where a large number of environments were sampled and the aerosol characteristics changed with time, a more practical method of accounting for this bias was adopted. For each 24 h period that DustTrak measurements were collected for, a collocated 24 h average gravimetric sample was collected concurrently. The DustTrak measurements were scaled using a specific calibration factor for each 24 h period:

$$\text{Calibration Factor} = \frac{\text{24 h average gravimetric concentration}}{\text{24 h time integrated DustTrak concentration}}. \quad [1]$$

Thus the DustTrak measurements were normalized to the average aerosol concentration over each 24 h sampling period. Each 15 min average DustTrak measurement was then multiplied by this calibration factor to estimate the "true" 15 min average $PM_{2.5}$ concentration. The indoor DustTrak measurements

were corrected using the gravimetric concentrations determined with the 10 lpm 2.5 μm inlet, while the outdoor DustTrak measurements were corrected using the concentrations determined using the Andersen RAAS2.5-300 sampler. Other studies have reported that the DustTrak provides precise measurements of PM$_{2.5}$, although the measurements need to be corrected as described above (Ramachandran et al. 2000; Yanosky et al. 2002).

Quality Assurance for DustTrak Measurements

The PM$_{2.5}$ inlet was calibrated at TSI, Inc. using monodisperse emery oil droplets generated using a vibrating orifice aerosol generator. The impactor efficiency as a function of particle size was determined by measuring particle concentration upstream and downstream of the inlet using an aerodynamic particle sizer (APS Model 3321, TSI, Inc.).

The DustTrak samples the aerosol at ambient humidity. The concentration measurements of any light scattering instrument increase with relative humidity due to the increase in the average particle size associated with condensational growth of hygroscopic components of the aerosol (McMurry et al. 1996). This effect has been modeled by Lowenthal et al. (1995), and a relative humidity correction curve has been developed (presented in Laulainen (1993). The correction factor (*CF*) can be expressed by the empirical relationship

$$CF = 1 + 0.25 \frac{RH^2}{(1 - RH)},$$ [2]

where *RH* is the relative humidity. This model fits more recent data presented by Day et al. (2000) quite well. We used this correction curve to study the relationship between the DustTrak light scattering responses and gravimetric measurements. For the outdoor community site measurements, the DustTrak samples the aerosol at ambient humidity. Hourly relative humidity measurements were obtained from the local meteorological station (Minneapolis–St. Paul Airport), and these were used to correct the outdoor DustTrak measurements. The indoor measurements were not corrected, since we did not have relative humidity measurements for the residences. However, indoor relative humidities are typically <60%, and thus the effects of humidity on DustTrak measurements are likely to be small.

While the indoor DustTrak instruments were zeroed at the end of each 24 h sampling period, the outdoor instruments (at the community central sites) were zeroed once every 7 days, since the central sites, which were part of federal and state monitoring networks, were accessible only on selected days. These outdoor instruments underwent significant ambient temperature fluctuations, which caused drifts in the zero level of the instruments over the 7 day monitoring period. For days when the drift was >5 μg/m^3, the DustTrak data were discarded. This accounted for more than 50% of the outdoor measurements, and these data were not used in our analysis. When the drift was <5 μg/m^3, we corrected for it by assuming that the zero drifted in a linear fashion over the 7 day time period. Thus if the zero drifted

from 0 to some negative value *D* over a time interval *T*, then the corrected concentration at some intermediate time *t* is given by

$$C_{corr}(t) = C_{meas}(t) - \frac{D}{T} t.$$ [3]

No systematic differences were observed between days for which zero drift was <5 μg/m^3 and days for which the drift was greater, in terms of 24 h average PM$_{2.5}$ levels, and so the data set obtained by discarding days for which the drift was >5 μg/m^3 is not biased toward higher or lower PM$_{2.5}$ days. Zero drift was negligible for all the indoor measurements (<1 μg/m^3). Thus a valid DustTrak measurement was obtained when there was a corresponding 24 h gravimetric measurement to provide the calibration factor and the zero drift was <5 μg/m^3.

RESULTS AND DISCUSSION

Outdoor Measurements (Daily and 15 Min Averages)

A total of 271 valid outdoor, daily average PM$_{2.5}$ gravimetric concentrations were obtained over 112 calendar days across all 3 communities. The means and standard errors of the daily average gravimetric PM$_{2.5}$ concentrations were 9.6 ± 0.2 μg/m^3 for Phillips, 10.6 ± 0.2 μg/m^3 for East St. Paul, and 9.5 ± 0.2 μg/m^3 for Battle Creek. East St. Paul was statistically significantly higher than Phillips ($p < 0.05$) and Battle Creek ($p < 0.05$), both of which were not different significantly from each other. There was no significant effect of season on the concentration levels. Day-to-day variability was the dominant contributor to total variability. The gravimetric PM$_{2.5}$ concentrations had a standard deviation of 6.6 μg/m^3, with a range between 1.0 and 41.6 μg/m^3.

The gravimetric PM$_{2.5}$ concentrations measured at the 3 outdoor community central sites were strongly correlated with each other. Ordinary least squares linear regressions for these 3 sets of gravimetric PM$_{2.5}$ concentrations against each other were performed. The slopes of the regressions were 1.057 ± 0.052, 0.862 ± 0.035, and 1.019 ± 0.036 for East St. Paul (y) versus Phillips (x), Battle Creek (y) versus East St. Paul (x), and Battle Creek (y) versus Phillips (x), respectively. The intercepts were close to zero, and the adjusted coefficients of determination (R^2_{adj}) values were 0.85, 0.89, and 0.93, respectively. This provides some evidence that outdoor gravimetric PM$_{2.5}$ concentrations across the Minneapolis–St. Paul metropolitan area tended to be spatially homogeneous during the monitoring period.

Valid DustTrak measurements were obtained on a subset of 102 of the 271 daily measurement periods when gravimetric measurements were made (see Table 1). The daily gravimetric averages were highly correlated with the medians of the 15 min averages within a day ($R^2_{\text{adj}} = 0.89$), and a linear regression yielded a slope of 1.076 ± 0.038 and an intercept of -1.38 ± 0.45. This implies that the means and medians of the 15 min averages within a day were highly correlated. There was, however, significant variability in the 15 min concentrations

within a day. The 90th percentile of the 15 min averages within a day can be 5–6 times the median value and as much as 1–2 orders of magnitude greater than the 10th percentile. The average standard deviation of 15 min averages within a day was 4.8 μg/m^3 and the maximum within-day standard deviation was 15.2 μg/m^3.

On 27 calendar days we obtained concurrent 15 min averages for an entire 24 h period from at least 2 community central sites (see Table 1). These data were analyzed, along with hourly meteorological data for those days, to study the correlations between the PM$_{2.5}$ levels at the different community sites on a 15 min basis, as well as statistical associations between PM$_{2.5}$ levels and meteorological conditions. In general, the 15 min average PM$_{2.5}$ concentrations at different community sites over a 24 h period track each other closely, and the trends can generally be related to prevailing meteorological conditions over the metropolitan area. Figures 2a, b, and c illustrate this trend for selected days.

Figure 2a shows the 15 min average PM$_{2.5}$ concentrations at the Battle Creek and East St. Paul sites on 2 July 1999. Early on this day, southwesterly winds were accompanied by rising PM$_{2.5}$ concentrations at both sites, and although the peak values and the variability were higher at East St. Paul, the R^2_{adj} was quite high (0.78). Surface winds shifted to the north and northwest in the late morning, and trace rainfall amounts were also recorded over the region. PM$_{2.5}$ concentrations dropped sharply in the late morning, consistent with the wind direction shift, an incoming cleaner air mass, and removal of particles by precipitation.

Figure 2b shows 15 min average PM$_{2.5}$ concentrations in all 3 communities for 18 November 1999, a day when there was a high correlation among the 3 communities (Battle Creek and Phillips = 0.89; East St. Paul and Phillips = 0.66; Battle Creek and East St. Paul = 0.74). Winds were southeasterly on this day on the back side of a receding high pressure system. This type of meteorological condition is typically associated with high pollutant levels from transport into the region. The end of a declining temperature trend and the start of an increasing temperature trend from 1,200 h to 2,000 h coincides with increasing PM$_{2.5}$ levels over the same period. PM$_{2.5}$ concentrations reached 22–27 μg/m^3 at all 3 sites.

Figure 2c shows 15 min average PM$_{2.5}$ concentrations at the Phillips and East St. Paul sites on 20 November 1999. If the spikes at 1,000 and 1,300 h at East St. Paul and the spike at 2,000 h at Phillips are removed, then the concentrations at the 2 sites are well-correlated with each other ($R^2_{\text{adj}} = 0.56$). However, the spikes which may be indicative of very localized source activity, resulting in a low overall R^2_{adj} (0.04).

Considering all 33 daily periods (over 27 calendar days) where comparisons were possible, in 14 cases the value of R^2_{adj} was >0.5. In the remaining cases the R^2_{adj} was lower; however, the R^2_{adj} values do not always reflect the close tracking of PM$_{2.5}$ levels across communities. This maybe due to temporal lags in the PM$_{2.5}$ trends in different communities and the influence of localized PM$_{2.5}$ emissions causing short-lived spikes at one site, both of which may induce scatter in PM$_{2.5}$ concentrations.

Of the calendar days for which we have concurrent Dust-Trak measurements for at least 2 community central sites, we selected 18 days of measurements each for the Phillips, Battle Creek, and East St. Paul communities for further statistical analysis. The data from these days were first analyzed using a general linear model. The logarithm of the 15 min average PM$_{2.5}$ concentration was the dependent variable, and the measurement site (community) was the independent effect variable. The errors were assumed to be normally distributed but not independent, instead having an $AR(1)$ correlation structure that accounted for the autocorrelation between the 96 15 min averages measured within one day. However, correlations between measurements from day to day were ignored. Using the PROC MIXED feature in SAS (Version 8.01, SAS Institute, Inc., Cary, NC), the measurement sites did not have a statistically significant effect on PM$_{2.5}$ measurements (approximate F-test $F = 0.71$, $p = 0.50$). A general linear mixed model with random effects for the 18 days and for the 96 averages within days was then fit. This imposed a compound symmetry correlation (equicorrelation) structure between the 96 average concentrations and in addition allowed for day-to-day correlations. Again, it was found that measurement sites did not have a statistically significant effect on PM$_{2.5}$ measurements (approximate F-test $F = 0.65$, $p = 0.53$). The above results seem to indicate that, in general, PM$_{2.5}$ concentrations over the Minneapolis–St. Paul metropolitan area are homogeneously distributed over a 24 h time scale as well as on a 15 min time scale.

A frequency distribution of all the outdoor 15 min average concentrations (all days and all community central sites) is shown in Figure 3. A trimodal lognormal distribution was fit to this distribution. The smallest mode contained 12.5% of all measurements and had a geometric mean (GM) of 1.1 μg/m^{3}and a geometric standard deviation (GSD) of 2.2. This may be interpreted as a low concentration background aerosol that is observed relatively rarely. The second mode contained 60.2% of all measurements, had a GM of 6.7 μg/ m^3 and a GSD of 1.6, may be interpreted as the most commonly observed ambient aerosol, and is at least a metropolitan area scale phenomenon. The third mode contained 27.2% of all the measurements, with a GM of 20.8 μg/ m^3 and a GSD of 1.3, and may be representative of high concentrations possibly due to a combination of localized sources of PM$_{2.5}$ and meteorological effectsconsisting of predominantly southerly winds that may be indicative of transported regional PM$_{2.5}$.

Indoor Measurements (Daily and 15 Min Averages)

The average indoor gravimetric PM$_{2.5}$ concentration across the 3 communities and over 3 seasons ($n = 294$ daily measurements made over 96 calendar days) was 13.9 μg/m^3 with a standard deviation of 11.6 μg/m^3. The concentrations ranged between 5.2 and 23.5 μg/m^3. Valid DustTrak measurements were obtained on a subset of 201 of the 294 daily measurement periods for which gravimetric measurements were made. The daily gravimetric averages were correlated with the medians of
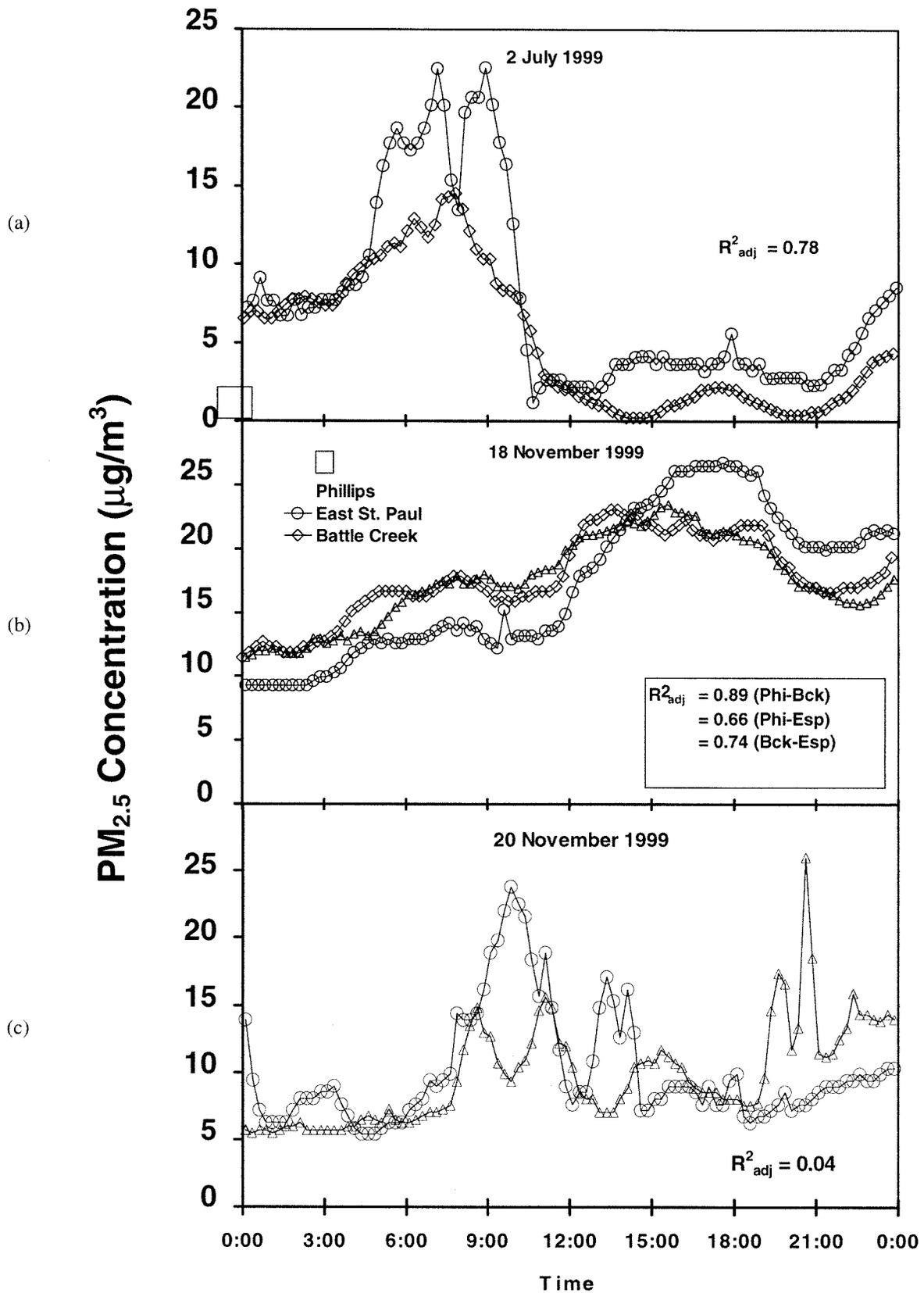
Figure 2. Simultaneous outdoor 15 min average PM$_{2.5}$ concentrations over a 24 h period measured at more than 1 community central site. (a) Battle Creek and East St. Paul on July 2, 1999; (b) Phillips, East St. Paul, and Battle Creek on November 18, 1999; (c) Phillips and East St. Paul on November 20, 1999.
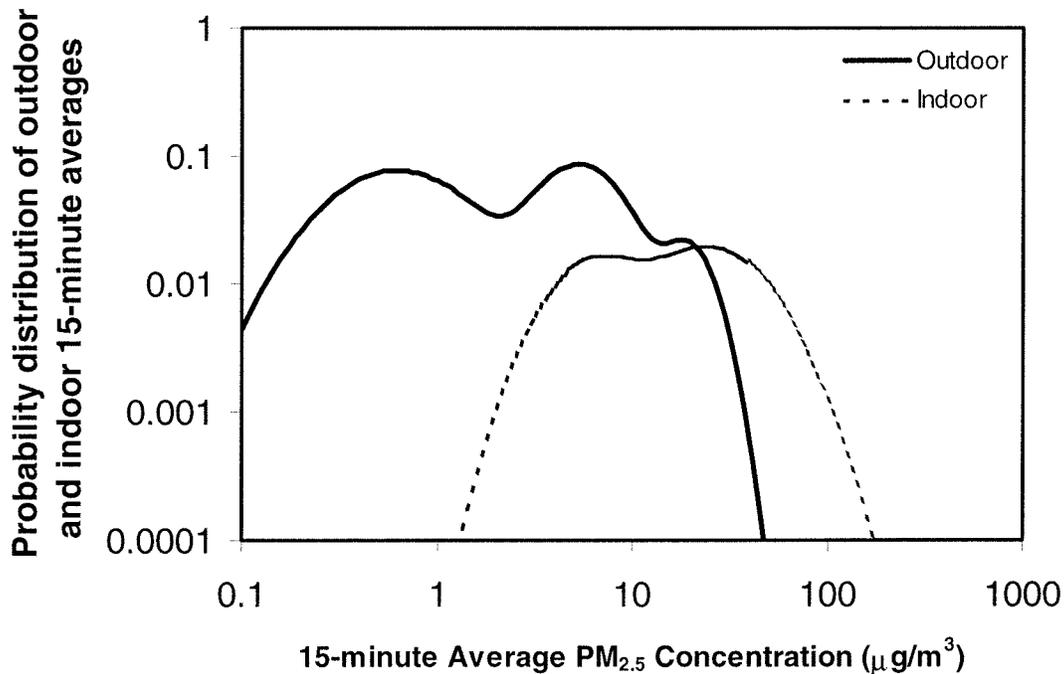
Figure 3. Probability distributions of outdoor and indoor 15 min average PM$_{2.5}$ concentrations.

the 15 min averages with a $R^2_{\text{adj}} = 0.63$, and a linear regression yielded a slope of 0.59 ± 0.03 and an intercept of 3.05 ± 0.59. The lower value of R^2_{adj} (compared to outdoor measurements) is most likely due to the effect of indoor activities that generate a number of short-term peaks in PM$_{2.5}$ concentrations that increase the means more than the medians. The fact that the mean concentration was greater than the median concentration indicates a skewed distribution of 15 min average concentrations within a day.

There was much greater variability in the 15 min indoor concentrations than outdoor concentrations within a day. The 90th percentile of the 15 min averages within a day can be 20-fold greater than the median and ∼40-fold greater than the 10th percentile. The average standard deviation of 15 min averages within a day was 9.3 μg/m^3, which is comparable in magnitude to the day-to-day (calendar day) variability in daily concentrations (standard deviation 11.6 μg/m^3). The maximum within-day standard deviation was 121 μg/m^3. The highest observed concentrations were almost 300 μg/m^3.

A frequency distribution of all the indoor 15 min average concentrations (all days and all residences) is shown in Figure 3. A bimodal lognormal distribution was fit to this distribution, with 14% of the measurements falling under the first mode (GM = 8.3 μg/m^3 and GSD = 1.66) and 86% of the measurements forming a second mode (GM = 35.9 μg/m^3 and GSD = 1.8). One possible interpretation of these 2 modes is that the first mode reflects the influence of the outdoor aerosol on the indoor aerosol and the second mode reflects the more dominating influence of emissions from human activities and indoor sources.

Comparison of Integrated DustTrak Readings with Gravimetric Measurements

A linear regression of 24 h integrated DustTrak concentrations against daily gravimetric concentrations for the outdoor data (corrected for relative humidity) has a $R^2_{\text{adj}} = 0.63$, with a slope of 2.7. A similar regression for the indoor data (uncorrected) has a $R^2_{\text{adj}} = 0.71$, with a slope of 2.3. For both regressions, the intercept was not significantly different from zero. The value of the slope depends on the DustTrak response to aerosol properties, such as its size distribution and composition. The similar slopes suggest that on average the DustTrak response to the outdoor aerosol was not much different from that for the indoor aerosol.

In addition, the variability in the ratio of daily integrated DustTrak concentration to the daily gravimetric concentration for indoor measurements was used as an indicator of day-to-day variability in aerosol characteristics. We did not attempt to separate out the contributions due to size distribution and aerosol refractive index. There was a statistically significant difference between the 3 communities ($p < 0.001$): the average value of this ratio was 1.9 ± 0.1 for Battle Creek, 2.6 ± 0.1 for East St. Paul, and 2.2 ± 0.2 for Phillips. A statistical analysis using a general linear model showed that this was associated with differences in the age of housing stock in the 3 communities ($p < 0.05$). Both East St. Paul and Phillips had significantly older residential stock (40–60 year old houses) than Battle Creek (20–30 year old houses). The statistically significant differences in the value of the ratio between homes ($p = 0.001$) is probably due to the different types and sizes of aerosols that might be

generated in each home. In season 1 (April 26–June 19), the average ratio of the 24 h time-integrated DustTrak concentration to the daily gravimetric concentration was about 1.9 ± 0.2; in season 2 (June 20–August 10) it was about 2.7 ± 0.1; in season 3 (September 23–November 20) it was 2.0 ± 0.1. Thus season 1 (spring) was not different from season 3 (fall), but both spring and fall were statistically different from summer. One possible explanation could be the greater influence of outdoor air on indoor environments in summer.

The most significant source of variability was the within-home day-to-day variability, which could cause the ratio to vary dramatically from one day to the next (e.g, from 0.5 to 3.5) within the same residence. This variability is most likely due to indoor human activities, such as cooking and cleaning, and within-home combustion sources.

Indoor to Outdoor Concentration Relationships (Daily and 15 Min Averages)

Daily average I/O ratios were measured over 248 periods (86 calendar days) and ranged between a minimum of 0.2 and a maximum of 14.9. These ratios were distributed lognormally with a $GM = 1.3$ and a $GSD = 2.0$. When interpreting these ratios, it should be kept in mind that the outdoor and indoor gravimetric measurements typically began and ended at different times, so that the time overlap between them was not complete (17.5 ± 1.9 h). A more detailed picture emerged when we

considered only the 32 daily (24 h) periods for which we had matched 15 min I/O ratios. These 32 periods were obtained in 21 homes over 25 calendar days: seven 24 h periods were during the first season, eight were during the second season, and 17 were during the third season. Matched I/O DustTrak concentrations over a complete 24 h period were available for such a relatively small number of days because of 2 problems: indoor measurements were not always made on the same consecutive days as the outdoor measurements, and indoor measurements began at different times than outdoor measurements. Each day is composed of 96 matched pairs of indoor and outdoor 15 min average PM$_{2.5}$ concentrations, which allows for calculation of 96 I/O concentration ratios. The 15 min I/O ratios for these days ($n = 33 \times 96 = 3168$) had a minimum of 0.14 and a maximum of 140 and showed a bimodal lognormal distribution. The dominant mode (87.5% of the measurements) had a $GM = 1.1$ and a $GSD = 1.7$ and was very similar to the distribution of daily I/O ratios. A smaller mode (12.5% of the measurements) had a $GM = 7.3$ and a $GSD = 2.5$ and could possibly represent short-term spikes in the I/O ratio, which may be due to elevated indoor concentrations or low outdoor concentrations or some combination of the two.

For each of these 32 daily periods, we examined the correlation between the 96 matched outdoor and indoor 15 min average PM$_{2.5}$ concentrations and calculated the adjusted squared correlation coefficient (R^2_{adj}). Figure 4 provides an example of 15 min
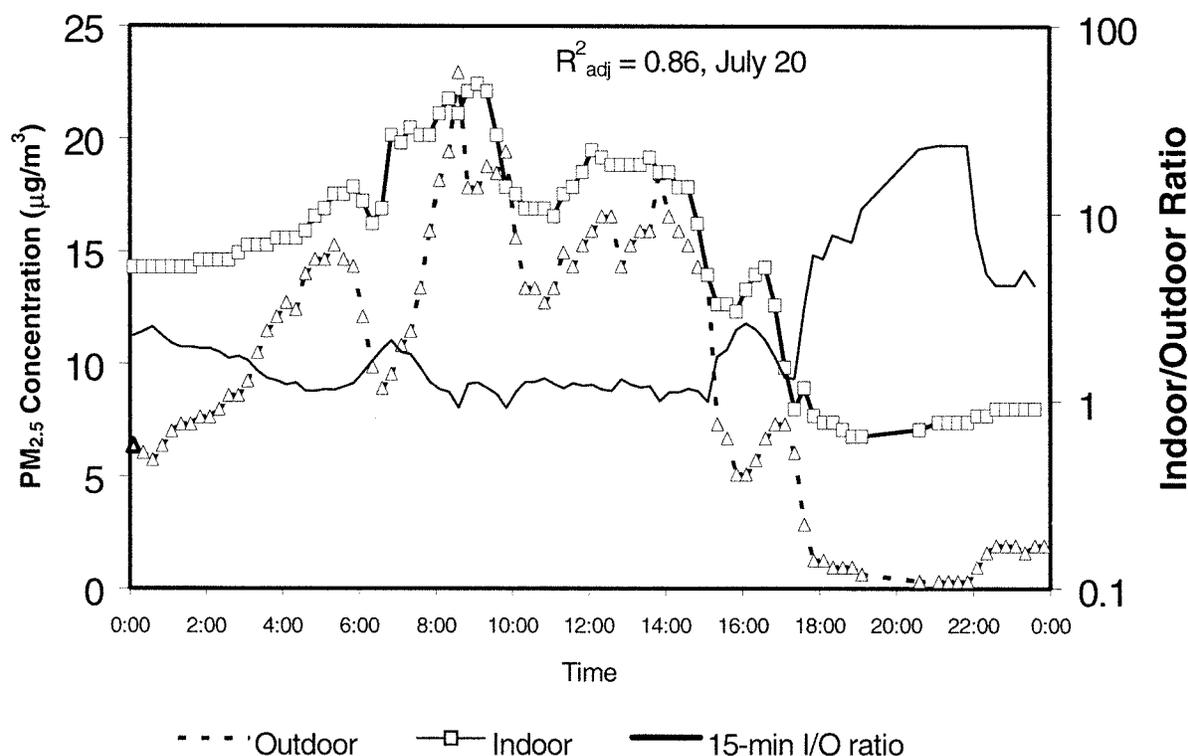

Figure 4. Real-time outdoor and indoor PM$_{2.5}$ concentrations and I/O ratios over 1 day in a residence (July 20, 1999). There is a high correlation between indoor and outdoor levels.

Table 2
Characteristics of 21 homes for which matched indoor and outdoor 15 min averages are available for a 24 h period

Sampling date	Community/ residence	R^2_{adj}[1]	Residence type/age[2]	Hours windows open	Hours spent at home	Air conditioning	Annual income
May 6	BCK-10	0.37 (0.66)	A (30–40 yr)	6	21.50	Central	5
May 15	BCK-5	0.46 (0.89)	A (40–50 yr)	15	14.00	Central	3
May 18	ESP-7	0.33	A (10–15 yr)	4	13.50	None	2
May 21	ESP-8	0.10 (0.56)	A (>60 yr)	24	16.50	Central	2
May 24	PHI-17	0.08	A (10–15 yr)	10	11.00	Central	6
May 27	BCK-7	0.03	A (40–50 yr)	2	12.50	Central	4
June 2	BCK-15	0.68	B (0–10 yr)	2	13.33	Central	3
June 20	BCK-4	0.26	A (40–50 yr)	24	18.00	Central	5
June 23	BCK-15	0.91	B (0–10 yr)	24	13.08	Central	3
June 23	ESP-5	0.8	A (40–50 yr)	24	20.50	None	5
June 26	BCK-8	0.67	A (50–60 yr)	24	14.00	None	5
June 26	PHI-18	0.53	B (>60 yr)	24	23.00	None	2
June 29	PHI-31	0.0001	A (>60 yr)	8	13.00	Window	2
July 8	BCK-7	0.28 (0.45)	A (40–50 yr)	Not reported	Not reported	Central	4
July 20	PHI-31	0.86	A (>60 yr)	24	14.00	Window	2
Oct 6	ESP-8	0.36	A (>60 yr)	8	19.00	Central	2
Oct 9	ESP-8	0.05	A (>60 yr)	18	20.00	Central	2
Oct 9	ESP-9	0.61	A (>60 yr)	16	21.33	Window	4
Oct 15	ESP-7	0.009	A (10–15 yr)	7	24.00	None	2
Oct 15	ESP-9	0.05	A (>60 yr)	9	20.16	Window	4
Oct 18	ESP-4	0.16	A (>60 yr)	2	14.00	None	5
Oct 21	PHI-16	0.15	B (>60 yr)	0	21.00	Central	5
Oct 27	BCK-5	0.015	A (40–50 yr)	18	17.41	Central	3
Oct 27	BCK-17	0	C (20–30 yr)	Not reported	15.00	Window	3
Nov 2	BCK-2	0.22	A (20–30 yr)	0	21.00	Central	6
Nov 2	BCK-10	0.0001	A (30–40 yr)	0	23.00	Central	5
Nov 5	PHI-17	0.135	A (10–15 yr)	15	16.00	Central	6
Nov 8	PHI-18	0.002	B (>60 yr)	14	14.00	None	2
Nov 11	PHI-8	0.21	A (>60 yr)	24	19.50	Window	4
Nov 17	BCK-1	0.008	A (30–40 yr)	0	18.33	Central	Not reported
Nov 17	BCK-11	0.17	A (0–10 yr)	0	24.00	Central	3
Nov 17	ESP-17	0.1	A (>60 yr)	0	22.25	None	1

[1]For some days, deleting data for a portion of the day when sudden spikes in indoor concentrations occurred due to indoor sources significantly increased the values of R^2_{adj}. These values are provided in parentheses.
[2]A: single family detached home; B: townhouse/duplex; C: apartment. Annual income is divided into 6 categories in increasing order 1: $10K–$20K; 2: $20K–$30K; 3: $30K–$40K; 4: $40K–$50K; 5: $50K–$75K; 6: $75K–$100K.

average indoor and outdoor concentrations for a 24 h period and the resultant I/O ratios and overall R^2_{adj} value. Figure 4 shows a day when the correlation between indoor and outdoor levels was relatively high ($R^2_{adj} = 0.86$). The house was more than 60 years old, and this was a day in summer (July 20) when the windows in the residence were open all 24 h. The daily I/O ratio was 0.78, although the 15 min average I/O ratios vary over an order of magnitude.

The indoor concentration tends to be driven by human activities rather than the outdoor concentration (Wallace 1996). The R^2_{adj} values for all 32 monitored days, along with relevant

characteristics of the monitored residences obtained from the questionnaire administered to the residents, are summarized in Table 2 and plotted in Figure 5. All 21 residences were heated with gas, although one house had supplemental electric heating. All occupants were nonsmokers.

In Figure 5, a strong seasonal effect on the I/O R^2_{adj} can be seen, whereby higher values are observed in the spring and summer ($R^2_{adj} = 0.49 \pm 0.33$) and lower values are seen in the fall ($R^2_{adj} = 0.13 \pm 0.13$). A stepwise forward regression using a general linear model that included season, community, age of the building, availability of central air conditioning, availability
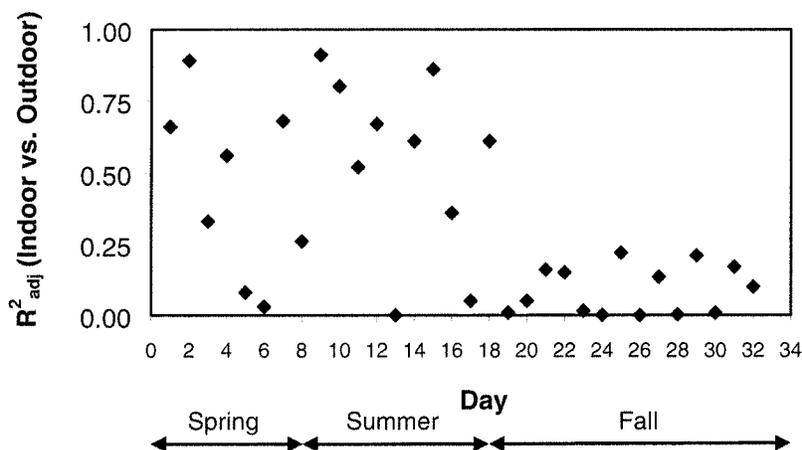
Figure 5. The R^2_{adj} between the 15 min indoor and outdoor concentrations over a 24 h period for all days in the study for which such data are available.

of central heating, and income level showed that only season is a statistically significant factor affecting R^2_{adj}. This statistical model had a $R^2_{\text{adj}} = 0.80$. As we suspected, the seasonal effect was explained using a linear model by the number of hours that windows are open over the course of a 24 h period ($p = 0.09$). During spring and summer, on 9 days out of 15, the value of R^2_{adj} was >0.5. Excluding periods when there were sharp peaks in indoor concentrations (possibly due to human activities and indoor sources) increased this to 13 out of 15 days. In contrast, during winter R^2_{adj} was >0.5 on only 1 out of 17 days.

This explanation does not hold for some days when the value of R^2_{adj} is relatively high despite windows being closed for most of the day (e.g, BCK-15, June 2, and BCK-10, May 6). This could potentially be due to the presence of other pathways into the building that provided easy entry of outdoor air. On some days the value of R^2_{adj} is relatively low despite windows being open most of the day (e.g, ESP-8, October 9). This might be due to the effect of indoor human activities.

It is also instructive to see the variability in R^2_{adj} value within the same residence. For example, a R^2_{adj} value of 0.53 was observed on June 26 in PHI-18. Windows were open for 24 h for this day. The same residence showed a R^2_{adj} value of 0.002 on November 8, when windows were open for only 14 h. An additional factor that might have caused the low correlation on November 8 was the increased level of human activity on that day compared to June 26. Another example is PHI-31 on June 29, when $R^2_{\text{adj}} = 0.0001$, when windows were open for <8 h. The same residence showed a R^2_{adj} value of 0.86 on July 20, when windows were open for 24 h.

Using a general linear model, only availability of central heating was associated with the I/O ratio ($p < 0.05$). Season, community, and age of building were not found to be statistically associated. The I/O ratio was lower (1.3 ± 0.2) if central heat was used and higher (1.9 ± 0.3) if central heat was not used. These observations show that, as expected, indoor activities, type of house, and the extent to which doors and

windows are open or closed on a particular day can affect I/O correlations.

SUMMARY AND CONCLUSIONS

1. Outdoor PM$_{2.5}$ concentrations across the Minneapolis–St. Paul metropolitan area appear to be spatially homogeneous. The daily gravimetric averages of PM$_{2.5}$ measured at the 3 community central sites track each other closely. Simultaneous 15 min average DustTrak measurements at these community central sites also track each other closely. This suggests that the spatial homogeneity of PM$_{2.5}$ holds even on a 15 min time scale. The time trend of 15 min average PM$_{2.5}$ measurements also appears to be related to prevailing meteorological conditions over the metropolitan area. However, it appears that localized PM$_{2.5}$ emissions, temporal lags in PM$_{2.5}$ concentrations in different communities, and significant scatter in PM$_{2.5}$ concentrations tend to reduce the strength of this association.

2. Fifteen-minute average outdoor PM$_{2.5}$ concentrations can vary by as much as an order of magnitude within a day. The means and medians of the within-day 15 min averages are very similar.

3. The frequency distribution of outdoor 15 min averages can be described by a trimodal lognormal distribution. The 3 modes ($GM = 1.1$ μg/m^3 ($GSD = 2.1$), 6.7 μg/m^3 ($GSD = 1.6$), and 20.8 μg/m^3 ($GSD = 1.3$), respectively) can be interpreted as (a) background aerosol that can be seen on clean days, (b) the most commonly observed ambient aerosol in the metro area, and (c) high concentrations possibly due to a combination of localized sources of PM$_{2.5}$ and meteorological effects.

4. Fifteen-minute average indoor PM$_{2.5}$ concentrations were observed to vary by a factor of as much as ~40 within a day. Thus there is much greater variability in the within-day 15 min indoor concentrations than in outdoor concentrations.

5. Within-day 15 min indoor concentrations have a skewed distribution with means being much larger than medians. This is most likely due to the influence of indoor activities, other than smoking, that cause high short-term peaks in concentrations, which affect the mean values more than the median values.

6. The variability in the ratio of 24 h integrated DustTrak concentration to the daily gravimetric concentration for indoor measurements can be used as an indicator of day-to-day variability in indoor aerosol characteristics. This ratio was higher for East St. Paul and Phillips (closer to the value of the ratio for outdoor measurements), which also had much older housing stock. The ratio was also much closer to outdoor values in the summer as compared to the spring and winter. These two findings illustrate the effect of outdoor air on indoor environments, which depends on the season as well as the housing stock.

7. The distribution of 15 min I/O ratios was bimodal. The dominant mode (87.5% of the measurements) is very similar to the distributions of daily I/O ratios. A smaller mode (12.5% of the measurements) likely represents short-term spikes in the I/O ratios due to human activities and indoor sources.

8. The correlation between the matched outdoor and indoor 15 min average $PM_{2.5}$ concentrations showed a strong seasonal effect, with higher values observed in the spring and summer and lower values in the fall. This is mainly due to doors and windows being open for more time during the spring and summer, thus letting in the outside air. However, factors such as indoor activities and the presence of other air pathways into the building may also be important.

When interpreting these results it is important to keep in mind that (a) measured outdoor concentrations were relatively low compared to many other urban areas in the U.S. (e.g, the 1999 annual average $PM_{2.5}$ concentration in Minneapolis–St. Paul, 11.3 μg/m^3, ranked 25th out of the 30 largest Statistical Metropolitan Areas), and (b) all study participants were non-smokers living in households with other nonsmoking occupants. It is likely therefore that measured indoor and outdoor $PM_{2.5}$ concentrations are on the low end of exposures typically encountered in urban environments and are probably illustrative of quasi-baseline conditions. Nevertheless, findings clearly demonstrate that the more refined characterization of both indoor and outdoor concentrations made possible by continuous $PM_{2.5}$ measurements allows for a more detailed and hence more complete exposure assessment. Furthermore, these data show that there can be substantial short-term variability over a given 24 h period for both outdoor and indoor $PM_{2.5}$ concentrations.

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