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# Personal, Indoor, and Outdoor Concentrations of Fine and Ultrafine Particles Using Continuous Monitors in Multiple Residences

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**Concentrations of airborne continuous fine particulate matter or ($PM_{2.5}$), black carbon (BC), and ultrafine particles (UFP) were continuously measured over 5 days in winter and summer both indoors and outdoors at residences for forty-eight adults in 2005 and forty-seven asthmatic children in 2006. During 2006, personal concentrations of $PM_{2.5}$ were also measured continuously. All 4 continuous instruments employed performed well both in laboratory and field conditions. Mean outdoor concentrations of $PM_{2.5}$, BC, and UFP were significantly higher than either indoor or personal concentrations. Air exchange rates were low (median value only 0.2/h), there was widespread use of central forced air and high-quality furnace filters. Outdoor concentrations of all particle-related pollutants showed overnight decreases followed by increases during the morning rush hours. Afternoon concentrations increased for UFP and decreased for BC, with $PM_{2.5}$ staying about the same. Between 5:00 pm and 7:00 pm, indoor UFP and $PM_{2.5}$ concentrations exceeded their mean daily values by 160% and 60%, respectively, suggesting that cooking is an extremely important source for these two pollutants. However, BC values did not increase at these hours. The highest indoor–outdoor ratios were observed for UFP suggesting that indoor sources were relatively more important for UFP than for other particle components. BC measurements in Windsor agreed moderately well ($R^2 = 41\%$) with an independent measure of elemental carbon (EC) in Detroit. This large residential air pollution study has provided data making it possible to identify short-term variations and possible sources that can influence the relationships between pollutants and environments.**

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## INTRODUCTION

Exposure to outdoor airborne particulate matter (PM) is known to be associated with morbidity and mortality (Dockery et al. 1993). More recent studies have implicated other components of PM with reductions in both respiratory and cardiovascular health (Delfino et al. 2004; 2005; Timonen et al. 2006; Beelen et al. 2008), as well as increased mortality (Ostro et al. 2010).

However, ultrafine, fine, and coarse particles are a complex mixture of sulfates, nitrates, trace elements, and organic and elemental carbon, and the presence of a causal agent, or agents, is unknown. Although some pathways are better understood for some of these PM components, such as ultrafine particles, which have been shown to be toxic (Öberdorster et al. 2005) and connected with oxidative stress-induced DNA damage and mortality (Bräuner et al. 2007; Stölzel et al. 2007), the components responsible for these health effects still remain unknown (National Academy of Science, 1998). Recent exposure studies that have examined residential continuous PM measurements are limited in terms of number of homes and days of measurements available. A number of studies have measured continuous fine particulate matter ($PM_{2.5}$) (Abt et al. 2000; Howard-Reed et al. 2000; Long et al. 2001; Allen et al. 2003, 2004; 2007; Liu et al. 2003; Wallace et al. 2003a, 2003b, 2006a, 2006b; and Williams et al. 2003; 2008; Delfino et al. 2004; Rodes et al. 2010). Continuous black carbon (BC) has been included in only three studies (LaRosa et al. 2002; Wallace 2005; and Järvi et al. 2008). Studies of ultrafine particles (UFP) have also been restricted to a relatively small number of homes (Abt et al. 2000; Long et al. 2001; Wallace and Howard-Reed 2002; Morawska et al. 2003; Wallace 2005; Zhu et al. 2005; Weichenthal et al. 2007; and Wallace and Ott 2010).

The Windsor study provides one of the first multi-home studies where continuous instruments have measured personal, indoor, and outdoor $PM_{2.5}$, UFP, and BC concentrations. One of the objectives of the study was to examine the relationships between indoor and outdoor concentrations and personal exposures to a variety of air pollutants. Temporal variability on a daily, seasonal, and year-to-year basis was assessed, and important sources of exposure were identified. The availability of time-activity diary (TAD) data in conjunction with indoor and outdoor measurements was applied to model personal exposures using time spent indoors and outdoors.

## MATERIALS AND METHODS

### Study Design

The study design has been fully described in Wheeler et al. (2011). In 2005 and 2006, Health Canada and the University of Windsor conducted a personal exposure study in Windsor, in which study participants were recruited from the larger Windsor Children's Respiratory Health Study (Dales et al. 2009). Forty-eight adults (parents of the study participants in the Dales et al. 2009 study) participated in 2005, and 48 asthmatic children (aged 10–13 years) participated in 2006. Residential indoor and outdoor exposures were assessed over a period of 10 days, with a total of 5 sampling days each in the winter (January–March) and summer (July–August) of each year. In addition, personal exposures were measured for the children in 2006.

### Household and Participant Characteristics

The Windsor homes were nonsmoking, mostly detached, and had forced air gas furnaces used for heat (Table 1). More than half reported a furnace-installed air cleaner, most of which were high efficiency filters, HEPA filters, or electrostatic precipitators. Over 60% had air conditioners. About 75% had electric stoves. Forty-eight adults were originally recruited for both the winter and summer 2005 sampling sessions. However, as five participants withdrew from the study after the winter session due to moving, renovating homes, or summer travel plans, two new additional participants were recruited for the summer. Therefore, the total sample size was 48 and 45, in winter and summer, respectively, with 43 homes participating in both seasons. There were 5 male and forty-five female adult participants in total. During the winter and summer of 2006 the total sample size for the asthmatic children was forty-eight for each season, with forty-five individuals participating in both seasons. The children were between 10 and 13 years of age with the majority of them ($N = 31$) being males.

TABLE 1
Household characteristics

| Characteristic | N | Fraction of positive answers |
|---|---|---|
| Detached house | 88 | 0.93 |
| Gas furnace* | 29 | 0.78 |
| Forced air | 90 | 0.95 |
| Storm windows | 11 | 0.13 |
| Gas hot water heater | 84 | 0.94 |
| Air cleaner | 47 | 0.53 |
| Air conditioner | 60 | 0.63 |
| Electric stove | 70 | 0.74 |
| Kitchen exhaust fan* | 31 | 0.69 |
| Major renovations* | 21 | 0.48 |

*From 2005 questionnaire only.

Pollutants included in the study were nitrogen dioxide ($NO_2$), volatile organic compounds (VOC), ozone ($O_3$), UFP, BC, $PM_{2.5}$, and $PM_{10}$. Ancillary measurements included air exchange rates (AER), temperature, relative humidity, and settled dust, as well as respiratory health measures collected for the asthmatic children in 2006. Sampling began on Monday evenings at approximately 4:00 pm, and ended on Saturday evenings at approximately the same time. At the end of each 24 $\pm$ 3-h interval, teams of two technicians visited each home to refurbish sampling equipment, check for mechanical malfunctions, and administer questionnaires. All data were collected over 8 consecutive weeks per season, with a total of six homes being sampled concurrently. Personal sampling was conducted in 2006 by having participants carry a small backpack containing air pollution monitoring equipment. Integrated and continuous monitors were employed to measure indoor and outdoor particles and BC. This manuscript addresses only personal, indoor, and outdoor continuous monitoring results for UFP, BC, and $PM_{2.5}$. The monitoring instruments employed in each season are shown in Table 2.

## MEASUREMENTS

### DustTrak

Continuous indoor and outdoor $PM_{2.5}$ was measured using the DustTrak (TSI, St. Paul, MN). In both seasons in 2005 and in the winter 2006, outdoor DustTrak units were housed indoors with a 4-ft Tygon sampling tube going to the outdoors through a sealed window opening. In summer 2006, outdoor units were placed in outdoor sampling boxes with a Nafion® dryer to control humidity levels in the sampled air. All DustTraks used a 2.5-$\mu$m size-selective inlet and operated at a flow rate of 1.7 L/min (LPM); data was recorded at a time interval of 3 min. A single calibration factor for all DustTraks was determined by comparison with co-located gravimetric monitors: DustTrak = 2.87 (grav) $-$ 5.2 $\mu g/m^3$ (Wallace et al. 2010). All DustTrak readings have been adjusted using this equation, which was based on 799 24-h averages ($R^2 = 90\%$).

TABLE 2
Particulate components, monitors, and time periods employed

| PM component | Instrument | Media | Winter 2005 | Summer 2005 | Winter 2006 | Summer 2006 |
|---|---|---|---|---|---|---|
| $PM_{2.5}$ | DustTrak | Indoor/outdoor | x | x | x | x |
| $PM_{2.5}$ | pDR | Personal | | | x | x |
| BC | Aethalometer | Indoor/outdoor | x | x | x | |
| UFP | P-Trak | Indoor/outdoor | | x | x | x |

*Note*: All seasonal campaigns included six homes measured concurrently for 5 days each week (Tuesday–Saturday). Each seasonal period lasted for 8 weeks. All continuous monitors measured on a 3-min schedule except for the P-Trak, which measured every 30 s for 10 min each hour.

### Personal DataRAM

In both seasons of 2006 asthmatic children carried a personal DataRAM (pDR-1000; ThermoScientific, Waltham, MA) fitted with a $PM_{2.5}$ inlet and a battery-operated pump (1.8 LPM) in a backpack. Data were recorded at 3-min intervals. The pDR uses a laser at 880 nm to determine particle volume. A single calibration factor for all pDRs was determined by comparison with co-located gravimetric monitors: pDR = 1.92 (grav) − 4.6 $\mu g/m^3$ (Wallace et al. 2010). All pDR readings have been adjusted using this equation, based on 306 24-h averages ($R^2$ = 70%).

### Aethalometer

In both seasons in 2005 and in the winter of 2006 a 2-wavelength (880 and 370 nm) Aethalometer (Model AE42, Magee Scientific Ltd., Berkley, CA) was used to collect semi-continuous measures of BC indoors and outdoors at each home. One unit was placed indoors and a switching mechanism was used to collect air from indoors and outdoors by having one tube sampling indoor air and another tube (through a sealed window) sampling outdoor air. The units were programmed to switch alternately between indoors and outdoors every 30 min from 7:00 am to 22:00 pm, and every 60 min from 23:00 pm to 6:00 am. They operated at a flow rate of 2.0 LPM with 3-min logging intervals in the winter of 2005 and 4.0 LPM with 3-min logging intervals in summer 2005 and winter 2006.

### P-Trak

The P-Trak (Model 8525, TSI, St. Paul, MN) was used to collect semi-continuous measures of UFP both indoors and outdoors during summer 2005 and in both seasons in 2006. The P-Trak measures particles from 20 nm to about 1 $\mu$m; however, as normally at least 80% of particles are smaller than 100 nm (Wu et al. 2008), the P-Trak is considered to be mainly an ultrafine monitor. In summer 2005 and winter 2006 two P-Traks were placed inside each house; one sampled indoor air and another outdoor air using a tube placed through a sealed window. The wick was replaced with a new alcohol saturated wick each day. In the last season (summer 2006), one P-Trak was housed in a weather-proof enclosure in the participant's backyard to measure outdoor air. In order to keep the alcohol in the P-Traks from running dry in the 24-h sampling period these were programmed to measure for 10 min in each hour at a time interval of 30 s.

All four types of continuous instruments (DustTraks, pDRs, Aethalometers, and P-Traks) were assessed by placing all units in the same location in a laboratory at the University of Windsor to see how they compared to each other before and after each of the field visits to establish their limits of detection, bias, and precision (Wallace et al. 2010). The median precision for the continuous instruments ranged from 6–10% (Wallace et al. 2010). The accuracies of the Aethalometer and P-Trak were not determined due to the lack of a recognized standard for black carbon and ultrafine particles.

### Air Exchange

Daily air exchange was measured in each of the homes using perfluorocarbon tracer gas (Dietz and Cote 1982). Four sources of the tracer gas and one activated carbon receptor unit were installed on the main floor of the home. These were placed away from any potential ventilation or heating sources. New receptors were installed daily, and the total amount of tracer gas adsorbed on the receptors was used to calculate a daily average air exchange flow for each home. House volume was determined from municipal property tax records and combined with the measured air flow rate to obtain a daily air exchange rate (per hour).

### Time-Activity Diaries

In both years participants completed a TAD throughout the day, noting their activities such as watching television, reading, riding a bike, as well as their presence in various locations at 15- and 30-min intervals for adults and children, respectively. The adult diary included details on whether the individual cooked or cleaned; these activities were deemed less likely for the children and were removed from their diaries. Both diaries included information on whether the participants were in close proximity to any smokers, and for how long. Locations noted on the diaries included at home, outside home, in transit, at work/school, outside away from home, and inside away from home. Multiple responses could be included if activities or locations changed within the time interval. All surveys and diaries were independently entered twice and compared electronically to each other to identify any discrepancies in the data entry.

## DATA ANALYSIS

### Data Screening and Calculation of Averages

Several different datasets were prepared for the analyses of the continuous monitoring instruments: continuous, hourly, and daily. The raw data for the continuous monitors were examined for isolated peaks corresponding to electronic noise; error rates were low at less than 0.01% of the total data. This process revealed that the Aethalometer was affected during the first one or two 3-min measurements following a change in the sampling location: switching from outdoor to indoor air or vice versa resulted in large spikes in the data. The instrument may have required this time to adjust to the changed environmental conditions (temperature, RH). These values, amounting to about 15% of the total data, were removed from all of the BC-generated datasets. The short monitoring times of 10 min in each hour for the UFP resulted in concentrations that were biased low by about 5% due to the warm-up period for the instrument (Wallace et al. 2010). Reviews of plots of concurrent outdoor $PM_{2.5}$ concentrations were effective in identifying a small number of cases with questionable data over some portion of the 5-day monitoring period. These data were replaced with the median values of the remaining concurrent DustTrak 3-min average measurements, provided that at least three other DustTraks had valid data for that time.

Hourly datasets were prepared using the corrected continuous data. Hourly averages were calculated for all three pollutants where at least 75% of the possible individual hourly measurements were recorded and these were used to create the diurnal graphs.

The daily datasets included the 24-h averages and again 75% of the data was required for the dataset to be valid. In all the datasets, all raw data were included as well as the corrected data and flags describing the changes to the data. Summary statistics were calculated using Statistica 9.1 (StatSoft Inc.). Graphs were created using Excel 2007 (Microsoft Inc.) and Statistica v.8 (StatSoft Inc.). All measurement distributions were tested for log-normality in each season using the Chi-squared test (Statistica 9.0). Regressions were calculated using ordinary least squares (OLS) or reduced major axis (RMA) approaches (Ayers, 2001). OLS regressions provide the best estimate of the dependent variable given a value (with measurement error) of the independent variable, whereas RMA regressions provide the best estimate of the "true" relation between two variables with known measurement error.

### Estimation of 2005 Personal $PM_{2.5}$ Concentrations

Continuous data were available for indoors and outdoors at the residences for both years. However, only in 2006 were personal continuous $PM_{2.5}$ exposures measured using the pDR. As such, the 2005 adults' personal $PM_{2.5}$ concentrations were modeled using their TAD data and the indoor and outdoor $PM_{2.5}$ (i.e., DustTrak) concentrations. The times spent in the three indoor environments (indoors at home, indoors away, and at school) and the three outdoor environments (outdoors at home, outdoors away, and in vehicles) were summed separately and multiplied by the concentrations measured indoors at home and outdoors at home to estimate personal exposure. This method was also applied to the 2006 children's activity pattern data and then compared with the measured personal exposures to validate the approach.

## RESULTS

### Quality Assurance and Basic Statistics

Positive and negative zero offsets and electronic drift of the DustTrak and pDR were observed leading to a substantial loss of data (18%) for the latter instrument. Limits of detection (LOD) for the DustTraks and pDRs were found to be 5 $\mu g/m^3$ and 5.5 $\mu g/m^3$, respectively (Wallace et al. 2010). LODs could not be determined for the Aethalometer and the P-Trak. No instruments were found to have consistent positive or negative biases compared to the median values of multiple co-located units. Most (42 of 54 seasonal distributions) were consistent with being drawn from a log-normal distribution. Of all potential data, 96–97% of the indoor and outdoor $PM_{2.5}$ and UFP data and 83–85% of the personal $PM_{2.5}$ and indoor and outdoor BC data were validated and available for analysis.

### Air Exchange Rate and House Volumes

Ninety-seven homes had estimated house volumes either through measurement, homeowners' estimates, or tax records. The volumes of the homes ranged from 160 $m^3$ to 1200 $m^3$, with a median value of nearly 500 $m^3$. A total of 673 24-h average measurements of air flow were collected from 93 homes, but delays in delivery of the PFT emission tubes across customs resulted in no air exchange measurements in the first 2 weeks of both winter seasons. The median air exchange rate, calculated from the measured air flow and the estimated house volume, was 0.35/h in winter and 0.18/h in summer (Table 3).

### Comparison of Outdoor Windsor BC with Detroit EC (Elemental Carbon)

In the Windsor study, the Aethalometer was used to measure continuous BC, but no other method was available in Windsor for comparison to assess bias. However, the Detroit Exposure Aerosol Research Study (DEARS) measured 24-h average elemental carbon (EC) at the Allen Park site in Detroit (Williams et al. 2008). There were eighty-four days in which both measurement methods overlapped. The measurements of EC and BC are defined operationally and therefore the relationship of the two is not necessarily 1:1; however, they should be similar. The situation is complicated by the fact that EC and BC are strongly affected by roadway diesel emissions and therefore have more spatial variability than $PM_{2.5}$. Nonetheless, when the two methods were compared between cities, there was agreement for the mean values for each method to within 4% and within the standard error of the measurements (Table 4). The BC

TABLE 3
Air exchange rates (per hour) by season

| Season | Year | N | Mean | Std. Dev. | Standard error | Geometric mean | Percentile | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | | | Min | 10 | 25 | Median | 75 | 90 | Max |
| Winter | 2005 | 136 | 0.37 | 0.23 | 0.02 | 0.31 | 0.06 | 0.14 | 0.19 | 0.27 | 0.49 | 0.61 | 0.99 |
| | 2006 | 131 | 0.44 | 0.38 | 0.03 | 0.34 | 0.07 | 0.16 | 0.22 | 0.30 | 0.47 | 1.09 | 2.02 |
| Summer | 2005 | 209 | 0.29 | 0.58 | 0.04 | 0.16 | 0.04 | 0.07 | 0.10 | 0.14 | 0.21 | 0.52 | 4.27 |
| | 2006 | 197 | 0.24 | 0.24 | 0.02 | 0.17 | 0.03 | 0.07 | 0.09 | 0.14 | 0.28 | 0.48 | 1.39 |

measurements made using the Aethalometer showed somewhat greater variability, which might be expected due to the fact that they were taken at approximately ninety-six locations throughout Windsor and therefore more likely to be influenced by the local emissions, whereas the EC measurements were made at a single location. The association between the two methods was moderate, with a Spearman rank correlation of 0.58 and an $R^2 = 41\%$.

## Descriptive Statistics

Mean 24-h outdoor concentrations of all three particulate-related variables were significantly greater than the mean indoor concentrations for all years and seasons (Tables 5 and 6) with the single exception of the summer 2006 UFP concentrations. Mean personal exposures for both adults and children were slightly higher than concurrent indoor concentrations. The TAD data indicated that both populations spent approximately 75% of their time indoors at home, 5% in transit, and 5% outdoors.

## Exposure Model

Estimated children's mean personal exposures in 2006 using the TAD and $PM_{2.5}$ (i.e. DustTrak) indoor and outdoor data were compared with the actual measurements, which agreed fairly well (within 8% in winter and 12% in summer). The RMA regression of the estimated personal exposures using the time-weighted indoor and outdoor DustTrak values showed very good agreement, with a slope of 1.02 (95% CI range = 0.96–1.08) and a nonsignificant intercept of −0.58 (−1.26 to +0.10) with an $R^2 = 71\%$ (Figure 1). This suggests that estimating personal exposures for the adults in 2005 using this time-weighted method is acceptable. As on average participants spent nearly 90% of their time indoors, an even simpler model of estimating personal exposure from the indoor concentrations alone was nearly as good as the time-weighted model.

## Diurnal Variation

Hourly data for $PM_{2.5}$, BC, and UFP were investigated in an effort to explore the impact of different activities that occurred throughout the day both indoors and outdoors. The relationships between each of the pollutants at the time of particulate matter generating activities were investigated. Daily variations of the indoor and personal particle components for both years and seasons around their 24-h mean are shown in Figure 2a. The peak indoor values occurred around 6:00 pm and were likely due to dinnertime cooking; these data were captured on a daily basis using participant questionnaires. Ultrafine particles also showed the strongest peaks at this time, consistent with studies showing that both gas and electric stoves generate UFP (Dennekamp et al. 2001; Long et al. 2001; Weichenthal et al. 2007; Wallace et al. 2008). Black carbon appeared to show a relative decline at this time for unknown reasons. As the 2006 personal pDR data showed similar trends to the other indoor methods, it is likely that the personal $PM_{2.5}$ was highly influenced by similar sources. Personal $PM_{2.5}$ returned to baseline overnight when

TABLE 4
Comparison of ambient EC and BC measurements ($ng/m^3$)

| | N (days) | Mean | Std. Dev. | Geometric mean | Percentile | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | Min | 10 | 25 | Median | 75 | 90 | Max |
| Elemental carbon (Detroit) | 84 | 696 | 335 | 600 | 45 | 382 | 476 | 636 | 890 | 1077 | 1724 |
| Black carbon (Windsor) | 84 | 667 | 443 | 558 | 184 | 276 | 356 | 569 | 879 | 1192 | 2394 |

TABLE 5
$PM_{2.5}$ measurements from DustTrak monitors ($\mu g/m^3$)

| Year | Season | Monitor type | N | Mean | Std. Dev. | Geometric mean | GSD | Percentile | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | | | | Min | 10 | 25 | Median | 75 | 90 | 95 | 99 | Max |
| 2005 | Winter | Outdoor | 234 | 17.1 | 13.8 | 13.5 | 1.96 | 3.3 | 5.5 | 8.9 | 13 | 20 | 33 | 48 | 73 | 76 |
| | | Indoor | 230 | 7.9 | 9.7 | 6.4 | 1.72 | 2.4 | 3.6 | 4.5 | 5.8 | 8.5 | 14 | 17 | 26 | 136 |
| | | Personal* | 225 | 8.6 | 9.8 | 7.0 | 1.74 | 2.5 | 3.9 | 4.7 | 6.3 | 9.6 | 15 | 20 | 29 | 134 |
| | Summer | Outdoor | 151 | 19.7 | 12.4 | 16.3 | 1.87 | 4.2 | 7.7 | 10 | 16 | 26 | 37 | 45 | 61 | 62 |
| | | Indoor | 202 | 10.2 | 6.4 | 8.9 | 1.64 | 3.2 | 4.8 | 6.1 | 8.6 | 12 | 17 | 22 | 38 | 49 |
| | | Personal* | 140 | 10.5 | 5.6 | 9.4 | 1.58 | 3.2 | 5.4 | 6.7 | 9.2 | 13 | 17 | 20 | 35 | 40 |
| 2006 | Winter | Outdoor | 232 | 12.5 | 6.4 | 11.0 | 1.65 | 4.2 | 5.7 | 7.1 | 11 | 16 | 23 | 25 | 30 | 30 |
| | | Indoor | 228 | 8.0 | 5.6 | 6.9 | 1.64 | 2.8 | 4.0 | 4.8 | 6.3 | 9.1 | 14 | 18 | 32 | 53 |
| | | Personal* | 225 | 8.3 | 5.2 | 7.3 | 1.59 | 2.9 | 4.3 | 5.1 | 6.9 | 9.5 | 14 | 17 | 29 | 49 |
| | Summer | Outdoor | 228 | 14.3 | 7.5 | 12.6 | 1.64 | 4.8 | 6.7 | 8.3 | 12 | 20 | 24 | 28 | 40 | 46 |
| | | Indoor | 218 | 8.4 | 6.9 | 7.3 | 1.61 | 2.9 | 4.4 | 5.1 | 6.9 | 9.3 | 13 | 16 | 26 | 70 |
| | | Personal* | 218 | 9.2 | 6.6 | 8.1 | 1.58 | 3.0 | 4.9 | 5.8 | 7.6 | 10 | 14 | 18 | 25 | 66 |

*Estimated from indoor and outdoor measurements and time spent indoors and outdoors.

there were no noted sources; it then increased in the mornings at around 9:00 am and stayed consistently elevated until around 4:00 pm, then spiked along with the indoor $PM_{2.5}$ and UFP when cooking occurred around dinnertime.

In contrast, outdoor concentrations for both years and seasons showed a sharp rise between 6:00 am and 9:00 am corresponding to typical morning traffic emissions (Figure 2b). Both BC and $PM_{2.5}$, as measured by the DustTrak, fell below their daily mean in the afternoon. However, UFP remained elevated until about 6:00 pm. This is consistent with observed mid-day nucleation bursts (Zhang et al. 2004; Kearney et al. in press).

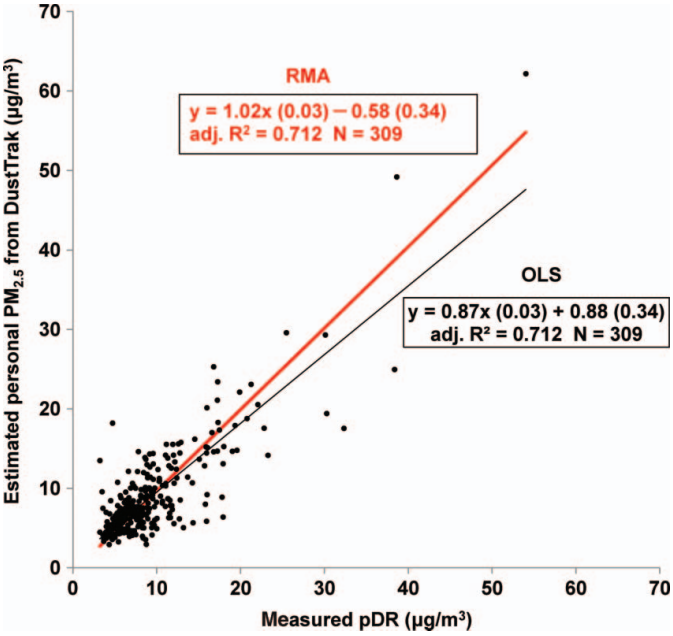


FIG. 1. Estimated personal concentrations of $PM_{2.5}$ using time-weighted indoor and outdoor DustTrak measurements compared with personal concentration measurements by the pDR. Numbers in parentheses are standard errors. The RMA regression has a slope not significantly different from 1 and an intercept not significantly different from zero. (Figure provided in color online.)

The six outdoor DustTrak values typically agreed quite well throughout all the weeks, with median hourly relative standard deviations ranging from 8–18% across the seasons (Figures S1a and S1b in online Supplemental Information). These showed rapid temporal variation, as well as slow variations over 1–2-day period. However, spatial variation was minimal for these homes, which were typically separated by 5–15 km. Using these plots, it was possible to identify a small number of the outdoor DustTrak monitors that showed clear evidence of malfunction, and their data were either deleted or replaced by the median of the remaining monitors. Less than 5% of the data was affected in this way.

## Seasonal Variation

During 2005 there were higher concentrations of outdoor $PM_{2.5}$ than were found in 2006 (Figure 3a). In all four seasons, the outdoor mean levels were the highest, with personal and indoor concentrations lower in 2005 (approximately 18 $\mu g/m^3$ compared with 9 $\mu g/m^3$), but in 2006 outdoor concentrations were approximately 13 $\mu g/m^3$ compared with 8 $\mu g/m^3$. Summer mean outdoor values were approximately 15% higher than winter in both years. Personal concentrations were always slightly higher than the indoor concentrations.

Black carbon concentrations were measured in the first three seasons, with summer 2005 having the highest mean outdoor levels, followed by winter 2005 and winter 2006 (Figure 3b). Outdoor BC concentrations exceeded indoor concentrations in

TABLE 6
Ultrafine particles, black carbon, and personal $PM_{2.5}$ from continuous monitors (24-h averages)

| | | | | | | | | | Percentile | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Pollutant | Year | Season | Type | N | Mean | Std. Dev. | Geometric mean | GSD | Min | 10 | 25 | Median | 75 | 90 | 95 | 99 | Max |
| UFP (# $cm^{-3}$) | 2005 | Summer | Outdoor | 206 | 12514 | 4017 | 11887 | 1.4 | 4551 | 7783 | 9796 | 11768 | 14624 | 19054 | 20498 | 22237 | 23751 |
| | | | Indoor | 213 | 7704 | 6919 | 5337 | 2.5 | 461 | 1673 | 2753 | 5774 | 10898 | 16479 | 20660 | 26518 | 59897 |
| | 2006 | Winter | Outdoor | 220 | 14350 | 6288 | 13252 | 1.5 | 4139 | 7958 | 10423 | 13370 | 16595 | 21924 | 25543 | 33156 | 57928 |
| | | | Indoor | 222 | 9645 | 8629 | 7123 | 2.2 | 785 | 2679 | 4306 | 6996 | 11394 | 19893 | 27189 | 42930 | 61684 |
| | | Summer | Outdoor | 196 | 7764 | 2771 | 7245 | 1.5 | 2588 | 4283 | 5645 | 7627 | 9772 | 11422 | 12482 | 14850 | 15031 |
| | | | Indoor | 206 | 7877 | 6233 | 5651 | 2.4 | 364 | 1876 | 3017 | 6016 | 11016 | 17186 | 20928 | 25861 | 27991 |
| BC ($ng/m^3$) | 2005 | Winter | Outdoor | 219 | 575 | 511 | | 2.7 | −16 | 123 | 241 | 418 | 711 | 1309 | 1665 | 2347 | 2983 |
| | | | Indoor | 221 | 299 | 292 | | 3.1 | −151 | 36 | 134 | 250 | 353 | 628 | 766 | 1126 | 2804 |
| | | Summer | Outdoor | 204 | 969 | 683 | 793 | 1.9 | 96 | 371 | 542 | 815 | 1160 | 1689 | 2120 | 3851 | 4761 |
| | | | Indoor | 204 | 420 | 791 | | 2.5 | −91 | 93 | 150 | 241 | 446 | 855 | 1088 | 2490 | 8800 |
| | 2006 | Winter | Outdoor | 216 | 389 | 407 | 278 | 2.3 | 24 | 89 | 170 | 310 | 512 | 793 | 917 | 1420 | 4804 |
| | | | Indoor | 219 | 282 | 449 | 187 | 2.3 | 9 | 66 | 106 | 200 | 311 | 467 | 671 | 2451 | 5117 |
| $PM_{2.5}$ ($\mu g/m^3$) | 2006 | Winter | Personal | 177 | 7.8 | 4.8 | 6.9 | 1.6 | 2.8 | 4.0 | 5.1 | 6.4 | 8.5 | 14 | 16 | 35 | 36 |
| | | Summer | Personal | 147 | 10.4 | 6.8 | 9.0 | 1.7 | 3.4 | 4.8 | 5.9 | 8.9 | 13 | 18 | 22 | 34 | 58 |

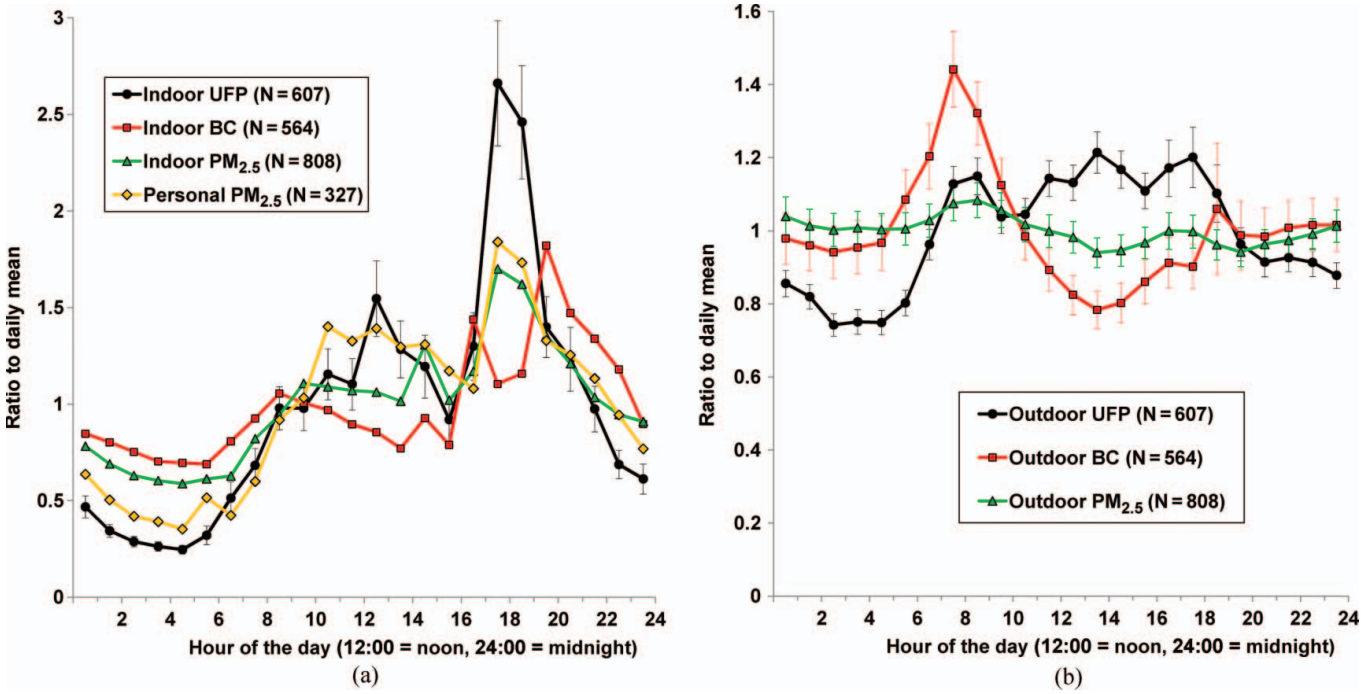


FIG. 2. (a) Diurnal variation around the daily mean for indoor and personal particle components. Averaged over all seasons. Standard errors shown for one variable; (b) diurnal variation around the daily mean for outdoor particle components. Averaged over all available years and seasons. Standard errors shown for all variables.

all three seasons suggesting that outdoor sources are important for this pollutant.

Ultrafine particle concentrations were measured during the last three seasons of the study. Mean outdoor concentrations were the highest in winter 2006, about 14,000/cm$^3$ (Figure 3c). Mean outdoor values in summer 2005 (more than 12,000/cm$^3$) were higher than in summer 2006 (less than 8000/cm$^3$). Summer 2005 and winter 2006 had significantly higher outdoor than indoor concentrations, whereas the summer 2006 had similar values for both indoor and outdoor concentrations.

## Indoor/Outdoor Ratios

Hourly indoor/outdoor ratios show a 60% increase for UFP during the period that cooking occurred, typically around 5:00–7:00 pm (Figure 4). The $PM_{2.5}$ mean ratio at the same time was also elevated (by about 25%) yet not by the same amount of time or magnitude. Relative standard errors for these ratios ranged between 11% and 32%.

The daily indoor–outdoor ratios also show UFP with the highest mean ratio (0.55), with more than 25% of 24-h average ratios exceeding unity (Figure 5). The $PM_{2.5}$ and BC mean ratios were lower (0.5 and 0.45, respectively) and exceeded unity for 15% of the measured values.

## DISCUSSION

All continuous instruments used for personal, indoor, and outdoor residential monitoring performed well. These data have made it possible to identify short-term variations and possible sources that can influence the relationships between pollutants and environments for one of the largest residential air pollution studies. The large dataset has made it possible to show consistent trends in the pollutants over both temporal and spatial periods.

The morning traffic-related outdoor peak was recorded by all three outdoor instruments (i.e., BC, $PM_{2.5}$, and UFP). The cooking peak between 5:00 pm and 7:00 pm was observed for indoor and personal $PM_{2.5}$ and indoor UFP. The apparent depression of the indoor BC concentrations at this time of day is an interesting observation that may require additional study (Figure 2a).

Mean personal $PM_{2.5}$ exposures, both estimated and measured, were similar to those observed in other studies in relatively unpolluted locations such as Seattle and Helsinki (Koistinen et al. 2001; Liu et al. 2003). Mean estimated personal exposures in 2006 ranged from 2.9–49 $\mu$g/m$^3$ and 3.0–66 $\mu$g/m$^3$ in winter and summer, respectively, and were within 8–12% of the mean measured exposures, showing that this estimation method performs well during different seasons when activity patterns and aerosols can vary.

Mean outdoor concentrations were considerably higher, typically a factor of two higher than mean personal concentrations, which were in turn also slightly higher than mean indoor concentrations. This finding is unusual; of eleven recent studies, most showed mean personal exposures that were higher than mean outdoor concentrations (personal/outdoor ratios ranging

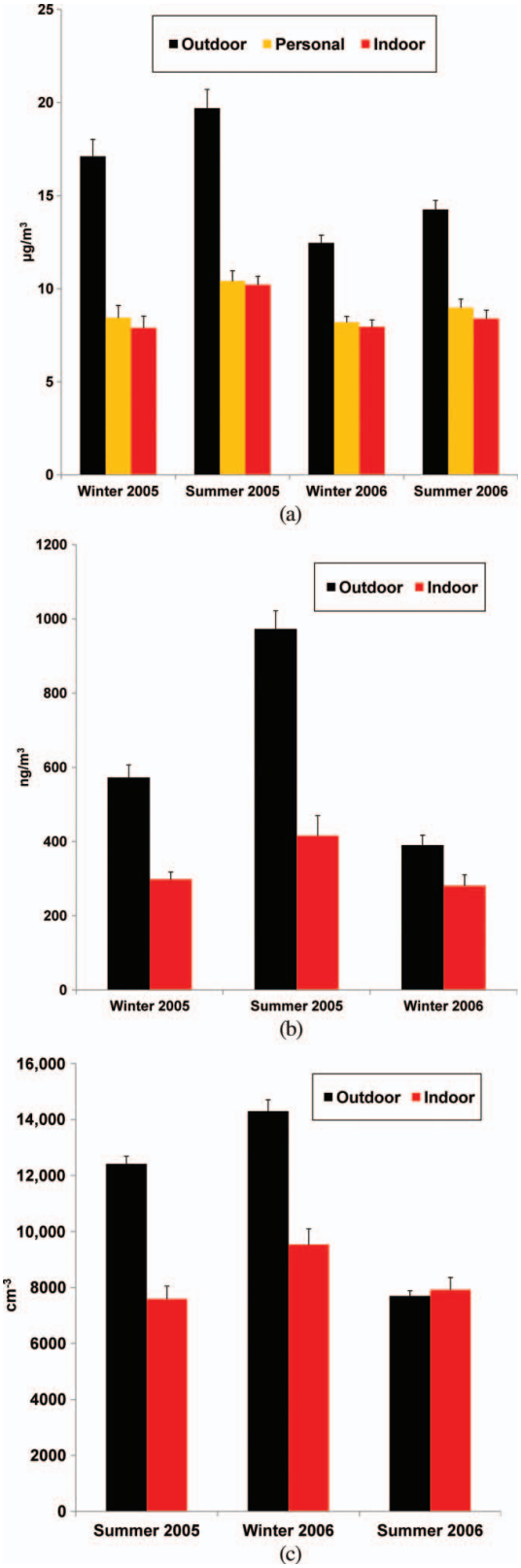


FIG. 3. (a) $PM_{2.5}$ concentrations in four seasons; (b) BC concentrations during three seasons; (c) UFP concentrations during three seasons. Error bars are standard errors. (Figure provided in color online.)

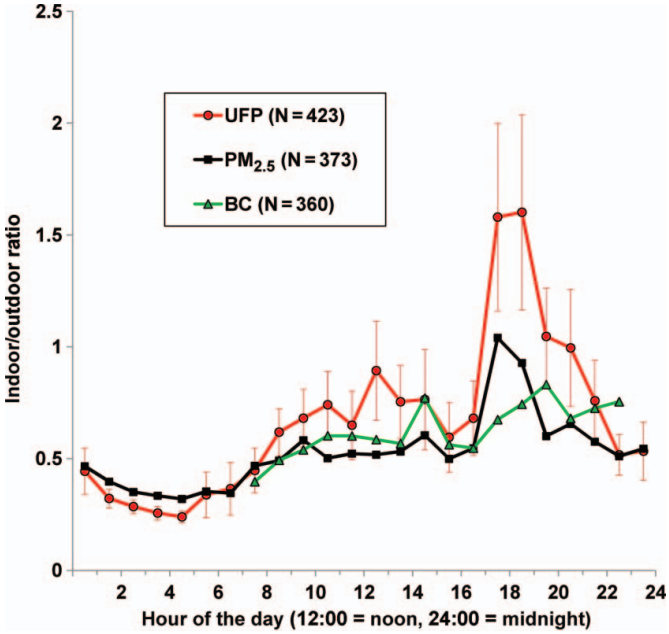


FIG. 4. Hourly ratios of indoor and outdoor UFP, $PM_{2.5}$, and BC for summer 2005 and winter 2006. (Figure provided in color online.)

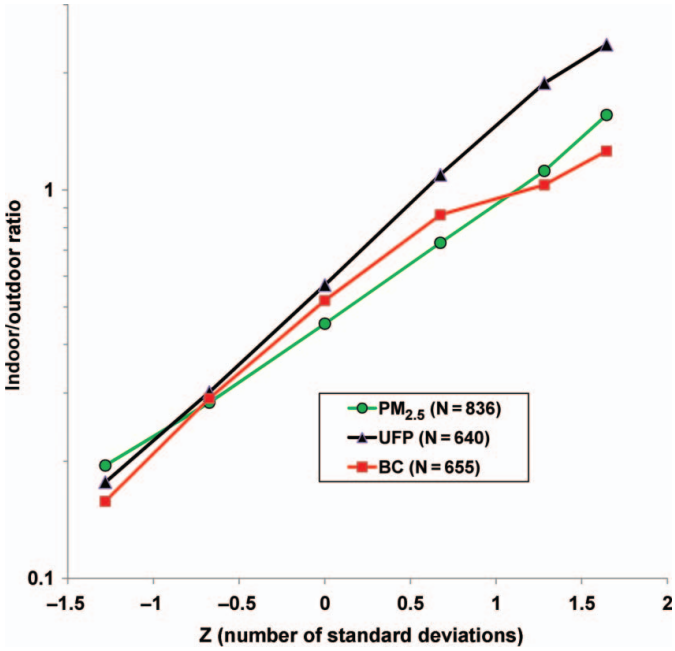


FIG. 5. 24-h Indoor/outdoor ratios for three particle components. Shown are the 10th, 25th, median, 75th, 90th, and 95th percentiles. (Figure provided in color online.)

from 0.8 to 2.5, compared to our ratio of 0.6, the lowest of all the studies) (Jannsen et al. 1999; 2000; Pellizzari et al. 1999; 2001; Rojas-Bracho et al. 2000; Williams et al. 2000; 2003; Weisel et al. 2005; Turpin et al. 2007; Brown et al. 2008). The DEARS study in the neighboring city of Detroit took place in the same years and resulted in similar mean outdoor $PM_{2.5}$ concentrations of about 16 $\mu g/m^3$ (Williams et al. 2008), which is nearly identical to the mean of 15.9 $\mu g/m^3$ in Windsor; yet the mean indoor concentrations in Detroit were 19 $\mu g/m^3$ compared with 8 $\mu g/m^3$ in Windsor. Possible reasons for this difference include the very low air exchange rates in Windsor (median = 0.20/h), reflective of well-constructed and insulated homes, and the wide use of high-grade filters in many of the Windsor homes. Some homes had in-duct air cleaners, including electrostatic precipitators, the most effective air cleaners (Riley et al. 2002; Wallace et al. 2004). If the finding of an important role for air cleaners is verified, it could be significant in suggesting ways to reduce PM exposure.

Annual and seasonal variations were found with the outdoor mean $PM_{2.5}$ concentrations in 2005 being about 40% higher than in 2006. In both years, summer mean outdoor $PM_{2.5}$ concentrations were about 15% higher than winter mean values. For the adult participants in 2005, the outdoor mean concentrations were very close to double the indoor levels. For the asthmatic children in 2006, the difference was less marked, but was still greater than 50%. In contrast, relatively small seasonal and annual variability was found for the indoor and personal measures. This study demonstrates the value of including multiple years and seasons to investigate the relationship between pollutants and how the personal and indoor concentrations are impacted by changes in outdoor PM concentrations. These are important considerations when assessing exposure to PM and components of PM in health studies.

Summer 2005 mean levels of outdoor BC were more than 70% higher than either winter season. In contrast, other studies have shown an increase in the fall and winter, which have been attributed to lower mixing heights (Järvi et al. 2008) to wood burning (LaRosa et al. 2002) to cold-start emissions and home heating (Demerjian and Mohnen, 2008). Windsor had little to no wood burning in the homes where measurements were made. Lower mixing heights were seen in winter (947 m) than in summer (1268 m). Indoor means were about half the outdoor levels in 2005, but were 70% of the outdoor levels in winter 2006. This is consistent with a 2-year study showing an indoor/outdoor BC ratio of 0.53 (LaRosa et al. 2002). Ultrafine particle concentrations outdoors were the highest in winter 2006, somewhat lower in summer 2005, and barely half as high in summer 2006. Unlike both $PM_{2.5}$ and BC, for which outdoor concentrations were nearly always higher than indoor levels, indoor concentrations of UFP actually slightly exceeded outdoor concentrations in the summer of 2006. On average, indoor/outdoor ratios of UFP were higher, at about 0.7 than indoor/outdoor ratios for either $PM_{2.5}$ or BC (about 0.5). This suggests the relatively greater importance of indoor sources, such as cooking, for UFP than for $PM_{2.5}$ or BC as was seen in Figure 4.

Very few studies have measured UFP in a large number of homes over extended periods, although they were included in early studies of 4 homes by Abt et al. (2000), 10 homes by Long et al. (2001), fourteen homes by Morawska et al. (2003), and thirty-six homes by Weichenthal et al. (2007). All of these studies noted the importance of cooking as a source of UFP during evening hours. The present results show that indoor ultrafine concentrations exceeded outdoor levels during the dinnertime hours, whereas the indoor $PM_{2.5}$ concentrations seldom exceeded outdoor levels at any time for these participants. These results are useful in determining the relative importance for human exposure from indoor and outdoor sources of UFP. For example, the relatively greater importance of cooking for UFP compared with $PM_{2.5}$ increases the impact of this major indoor source. Likewise, the low air exchange rates in Windsor lessen the impact of outdoor air infiltration (due to allowing more time for infiltrated particles to deposit on surfaces, filters, or ductwork) and increase the impact of indoor air sources (due to not allowing them to exfiltrate as rapidly).

The UFP indoor/outdoor ratios were generally higher than those for either $PM_{2.5}$ or BC. This suggests that indoor sources were relatively more important for UFP than for other particle components. This finding is particularly significant, as UFP have higher deposition rates than $PM_{2.5}$ and also deposit in cracks more readily while traversing the building envelope, leading to lower penetration coefficients (Nazaroff, 2004). Therefore in the absence of indoor sources, one would expect indoor/outdoor ratios for UFP to be lower than for $PM_{2.5}$ or BC, and this has in fact been observed for overnight hours (Kearney et al. in press). However, higher indoor–outdoor ratios were observed for UFP during the dinnertime hours, suggesting a relatively stronger response to cooking for UFP than for BC or PM2.5.

Limitations of the study include the non-probabilistic selection of participants, making it impossible to extrapolate findings to a larger population; the unknown accuracy of the BC and UFP instruments (i.e., Aethalometer and P-Trak) due to lack of standard reference methods; loss of a season of data for the UFP due to late arrival of instruments; and loss of about 4 weeks of air exchange data due to delayed arrival of equipment past customs.

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