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Residential indoor and outdoor ultrafine particles in Windsor, Ontario

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ABSTRACT

UFPs in the 20–100 nm size range were measured for 10 mins every hour for 5 consecutive days in 45 homes of nonsmoking adults in summer 2005 and in 49 homes of asthmatic children in summer and winter 2006. Median hourly outdoor levels across all measurements were 10,800, 12,000 and 6300 cm^{-3} and median indoor levels were 2700, 3730 and 2580 cm^{-3} in summer 2005, winter 2006 and summer 2006, respectively. Outdoor levels generally exceeded indoor levels but indoor concentrations were higher around 5–7pm, suggesting a strong influence of cooking. Daily and weekly infiltration factors were estimated for each home using three methods. Weekly infiltration factors (F_{inf}'s) based on a censored indoor/outdoor ratio method varied widely across homes; median F_{inf}'s across homes were 0.16 (summer 2005), 0.21 (winter 2006) and 0.26 (summer 2006). Large indoor peaks and low infiltration of ambient PM resulted in the indoor sources generally contributing more than infiltrated ambient UFPs to indoor concentrations. Median estimates of the percentage of indoor-generated contribution to total indoor levels were 58% (summer 2005), 65% (winter 2006) and 69% (summer 2006). The proportion of homes having more than half of their indoor concentrations provided by indoor sources was 66% (summer 2005), 67% (winter 2006) and 79% (summer 2006). Median deposition rates ranged from 0.61–0.79 h^{-1} across the 3 sampling sessions. Spatial variability was higher for outdoor UFPs than concurrently measured $PM_{2.5}$. The median correlations of hourly averaged outdoor UFPs between pairs of homes were moderate in the three sampling sessions (0.56–0.65), but were considerably lower than corresponding $PM_{2.5}$ correlations. The wide range of infiltration factors across homes as well as the spatial variability and moderate between-home correlations of outdoor UFPs could cause measurement error in epidemiology studies that use central site UFP measurements as a surrogate for personal exposure to ambient UFPs.

1. Introduction

Ultrafine particles (UFP) have diameters < 100 nm. Their small size allows them entry into all parts of the lung including the alveoli as well as other human cells of various tissue types. The main outdoor sources are auto exhaust and atmospheric reactions (Kittelson, 1998; Gaydos et al., 2005). Indoor sources include cooking (Dennekamp et al., 2001; Wallace et al., 2008), indoor chemical reactions of ozone and terpenes (as found in various cleaning products) (Singer et al., 2006), candles and incense burning (Klepeis et al., 2003) and electric motors (Szymczak et al., 2007). Animal and human studies have shown effects on various health endpoints, including oxidative damage to DNA repair mechanisms (Bräuner et al., 2007) and total and cardio-respiratory mortality (Stölzel et al., 2007). In assessing human exposure to outdoor-generated UFPs while indoors, it is important to know how well a central site monitor captures the temporal and spatial variability of ambient UFP concentrations and to understand the magnitude and variability of infiltration of ambient UFPs into homes. Some studies of indoor and outdoor UFP concentrations have been published (e.g. Long et al., 2001); however, only a few have involved indoor and outdoor measurements at multiple homes (He et al., 2005; Puustinen et al., 2007; Weichenthal et al., 2007; de Hartog et al., 2010). Deposition rates are another important parameter in determining human exposure to particle, however, few residential measurements have been made for UFPs.

In 2005–06 a personal exposure study was carried out in Windsor, Ontario to assess levels of exposure to various air pollutants and to investigate the relationships between personal, indoor and outdoor levels. Windsor, a city of 216,500 residents (2006 census, City of Windsor, 2010) is across the Detroit River

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from Detroit, Michigan. It is the home of the major Canadian automotive manufacturers and related industries and there is significant diesel truck traffic to and from the U.S. over the Ambassador Bridge. Previous publications have reported the study methods (Wheeler et al., in press-a), quality assurance results for the continuous measurements (including UFP) (Wallace et al., 2010) and summary findings on the particle-related continuous measurements including $PM_{2.5}$, UFP, and black carbon (Wheeler et al., in press-b). This manuscript provides a more in-depth analysis of the indoor and outdoor UFP continuous measurements, including analysis of temporal and spatial variability, estimation of the UFP infiltration factors, deposition rates and the ambient and non-ambient components of indoor UFPs.

2. Methods

In summer 2005 (Jul–Aug), indoor and outdoor UFP measurements were obtained from 45 homes of non-smoking adults in Windsor, ON. In winter 2006 measurements were taken in 47 homes of asthmatic children (10–13 y). The following summer, measurements were taken in 46 homes; 44 of which had participated in the winter 2006 season. In each of the 3 sampling sessions, which occurred over an 8 week period, 6 homes were sampled concurrently each week, generally from Monday evening to late afternoon on Saturday. Further details of the study methods are presented in Wheeler et al. (in press-a).

This study was approved by the Human Research Ethics boards at Health Canada and the University of Windsor. All participants provided informed consent.

2.1. Measurements

Indoor and outdoor UFP levels were measured over 5 consecutive days in summer 2005, winter 2006 and summer 2006 using a portable condensation particle counter (CPC) (P-Trak, Model 8525, TSI, Inc., St. Paul, MN) capable of measuring ultrafine particles as small as 20 nm in diameter. The alcohol reservoir in the P-Trak has a capacity allowing 6–8 h of continuous operation. In order to collect data over the 24-h monitoring period, P-Traks were programmed to operate for 10 min of each hour on a 30-s sampling interval; alcohol was replenished by a technician every day. In summer 2005 and winter 2006, both P-Traks were located indoors. In summer 2006, the outdoor units were placed outdoors in a shelter.

Indoor and outdoor $PM_{2.5}$ was also measured continuously (3-min) using DustTrak instruments (TSI, St. Paul, MN) fitted with 2.5 μm size-selective inlets. Side-by-side sampling indicated that the DustTrak was highly correlated with a gravimetric instrument but overestimated according to a relationship reported in Wheeler et al. (in press-a). The statistical correlations and other results reported here do not require correction by the calibration factor.

Air exchange was measured daily using perfluorocarbon tracer gas (Dietz and Cote, 1982). On the main floor of each home, four sources of the tracer gas and one receptor unit were installed away from any potential ventilation or heating sources. New receptors were installed daily, and the total amount of the tracer gas absorbed on the receptors combined with the square footage of each home (obtained from tax records) was used to calculate a daily average air exchange rate for each home.

2.2. Quality assurance

The 14 P-Traks as well as the DustTrak units used in the study were tested side by side on four occasions, indoors at a University of Windsor laboratory, to assess relative bias and precision (Wallace et al., 2010).

2.3. Data cleaning

Invalid outdoor data were identified by inspection of plots of concurrent measurements; a small number of cases when a monitor appeared to be performing badly were noted. Hourly average datasets were prepared based on at least 15 valid 30-s measurements (75% completeness). Data cleaning and management was carried out using Excel 2007 (Microsoft Inc.) and SAS (v 9.1, SAS Inc.). Statistical analyses were carried out using SAS; graphs were prepared in Statistica v9.0 (Statsoft Inc.) and Excel 2007.

2.4. Data analysis

2.4.1. Descriptive statistics, temporal and spatial variability, Spearman correlations

The indoor and outdoor UFP distributions were typically skewed, with some very high observations, particularly for the indoor measurements. Hence, nonparametric statistics (medians, percentile distributions, Spearman correlations and rank tests) have been used for many analyses.

Temporal variability of indoor and outdoor levels was examined by plotting the indoor and outdoor levels over the weekly period for each home, and calculating the coefficient of variation (CV) for both indoor and outdoor UFPs for each home over each week and over each year-season.

Spearman correlation coefficients were calculated for all possible pairs of homes using the hourly data from each home; the median and range are presented for each week and for each year-season. While UFP levels at different locations may be correlated, spatial variability (differences in the absolute levels across the sites) may still exist. Spatial variability was examined by summarizing the hourly differences between all possible pairs of homes for each week and year-season. As well, the coefficient of variation across the concentrations from each of the 6 homes sampled concurrently was calculated for each hour, and summarized over each week and year-season. The coefficient of divergence (COD) was also used as a relative summary measure of the differences of measurements over time between two locations (Wilson et al., 2005; Cyrys et al., 2008). It is defined as

$$\mathrm{COD}_{jk} = \sqrt{\frac{1}{p}\sum_{i=1}^{p}\left[\left(x_{ij}-x_{ik}\right)/\left(x_{ij}+x_{ik}\right)\right]^2},$$

where x_{ij} and x_{ik} represent the hourly average UFP concentration for hour i at homes j and k, and p is the number of hours. By definition, CODs range between 0 and 1; sites with similar measures have a COD closer to 0. For this analysis, the COD between all possible pairs of homes was calculated for each day (or week) and summarized across each week and year/season.

In order to compare these results with $PM_{2.5}$, within-home CVs, between-home CVs, CODs and Spearman correlations were also calculated for the hourly $PM_{2.5}$ measurements.

2.4.2. Indoor–outdoor relationships

The relationship between indoor and outdoor UFPs was examined using time series indoor–outdoor plots and calculating longitudinal Spearman correlations between the indoor and outdoor hourly UFP averages for each home. The correlations and indoor/outdoor ratios and differences were summarized over each week, during each year/season.

2.4.3. F_{inf} and ambient-infiltrated and indoor-generated components of indoor UFP concentrations

Three approaches were developed to estimate daily and weekly infiltration factors for each home. The infiltration factor, F_{inf}, is the fraction of outdoor particles that penetrate a building and stay suspended (Wilson et al., 2000). This can be derived from the mass balance equation in the absence of indoor sources (Shair and Heitner, 1974):

$$\frac{dC_{in}}{dt} = PaC_{out} - (a+k)C_{in} \quad (1)$$

where C_{in} = Concentration indoors [# cm^{-3}]; C_{out} = Concentration outdoors [# cm^{-3}]; P = penetration coefficient [dimensionless]; a = air exchange rate [T^{-1}]; k = deposition rate [T^{-1}].

Using the definition of the derivative (before taking the limit as Δt goes to zero),

$$\frac{C_{in}(t+\Delta t) - C_{in}(t)}{\Delta t} \approx PaC_{out}(t) - (a+k)C_{in}(t) \quad (2)$$

Combining terms and multiplying through by Δt,

$$C_{in}(t+\Delta t) \approx PaC_{out}(t)\Delta t + C_{in}(t)(1-(a+k)\Delta t) \quad (3)$$

For time steps of size Δt, this becomes the following recursive difference equation:

$$C_{in_{i+1}} = Pa\Delta tC_{out_{i+1}} + (1-(a+k)\Delta t)C_{in_i} \quad (4)$$

At equilibrium, $C_{in_{i+1}} = C_{in_i}$ and $C_{out_{i+1}} = C_{out_i}$, so

$$C_{in_i} = Pa\Delta tC_{out_i} + (1-(a+k)\Delta t)C_{in_i} \quad (5)$$

Combining terms, the indoor–outdoor relation at equilibrium is

$$C_{in} = \frac{Pa}{a+k}C_{out} = F_{inf}C_{out} \quad (6)$$

The recursive model in equation (4) was used to estimate a daily F_{inf} for each home using the measured daily air exchange rates. Since the model assumes no indoor sources, censored indoor concentrations were used in equation (4); these were obtained through a censoring algorithm described below. Since we had data for only 10 min for each hour, the model was based on one-hour intervals over the 5-day monitoring period. Cases with at least 4 consecutive days with a complete set of indoor and outdoor measured UFP concentrations were analyzed. Outdoor concentrations were imputed (linear interpolation) if no more than 6 consecutive hours were missing. Missing indoor concentrations were not imputed. Missing air exchange data were imputed for one day if the data showed low variability over the other 4 days. The analysis was done using the Excel Solver function (Excel 2007) with no constraints on the two unknown parameters P and k. The best estimates of P and k were obtained for the full 4- or 5-day period by minimizing the sum of the absolute differences between observed and predicted indoor concentrations. Since the starting value can affect the final estimate (e.g., by causing the search routine to settle at a local minimum), random starting values for P and k between 0 and 1 were provided and 50 runs for each dataset were carried out. The largest and smallest of the 50 minimum sums of errors were compared as a measure of the uncertainty of the estimates. In more than 80% of cases, all 50 runs converged on the same P and k values. The P and k estimates were used to calculate a weekly F_{inf}. Daily estimates of F_{inf} were obtained using the P and k estimates and the corresponding daily air exchange rates, a (equation (6)). Two of the 86 weekly F_{inf} and six of the 417 F_{inf} estimates were negative; these values were included in subsequent analyses.

The censoring algorithm was written in SAS (Sas Inc.) to remove peaks due to indoor sources from the hourly indoor UFP time series. The start of an indoor peak was identified when the indoor concentration increased at least 4000 cm^{-3} from the previous hour and either the indoor concentration increase exceeded a concurrent outdoor increase in concentration or the indoor concentration was greater than the outdoor concentration. The latter criteria were used to avoid identifying any peaks associated with a concurrent increase in outdoor levels. The 'background' concentration was considered to be the concentration in the hour prior to the start of the peak. The end of a peak was identified when the indoor concentration had declined back to within 500 cm^{-3} of the background concentration, or when the next 3 hourly concentrations were within 500 cm^{-3} of each other. All hours within the peak were assigned a 'censored' value equal to the 'background' concentration. The censored data were re-examined and a few values were manually censored, for instance, in situations where the algorithm did not capture changes in the indoor levels that were likely due to severe outdoor fluctuations (i.e. the algorithm had 'over-censored' the data).

The censored indoor data was also used directly to estimate the daily and weekly average F_{inf} for each home as the ratio of the mean value of the censored indoor concentrations to the mean value of the outdoor concentrations (herein called the censored I/O ratio method). This assumes that a daily or weekly mean ratio will be close to the equilibrium ratio. Although the hourly ratio is seldom equal to the equilibrium ratio due to the lag of the indoor concentrations behind the outdoor values, over time the mean values will approach the equilibrium ratio. Censored indoor/outdoor ratios should be less than 1; however, all estimates <1.2 were considered in the analyses, allowing for measurement error. Three of the 622 daily estimates were excluded from the analyses because they were greater than 1.2. No weekly estimates were greater than 1.

A third estimate of F_{inf}, the indoor/outdoor ratio during overnight hours, assumes that no indoor sources are active, and that the indoor/outdoor relationship is near equilibrium (Long et al., 2001). The night-time period of 2–6am was generally the period of lowest UFP levels in the homes so the median of the four hourly I/O ratios beginning at 2am was used as an estimate of F_{inf}. Again, hourly I/O ratios >1.2 were excluded from the analyses ($<1\%$). The median of the 5 daily estimates was used to estimate a weekly F_{inf}.

In order to gain a better understanding of the relative importance of indoor-generated and infiltrated ambient-generated UFPs that make up the total indoor concentration, the F_{inf} estimates were used to estimate separately, for each home a) the average concentration of ambient UFPs that have infiltrated indoors and b) the average concentration of indoor UFPs that have originated from indoor sources (Wilson et al., 2005). An estimate of the weekly-averaged ambient-infiltrated UFP concentration was calculated for each home by multiplying the weekly F_{inf} estimate (based on the censored I/O ratios) by the weekly mean outdoor UFP concentration. The indoor-generated UFP concentration was then estimated by subtracting the infiltrated ambient estimate from the weekly mean indoor concentration. The percentages of the indoor concentration were calculated for each component. In two cases, the indoor-generated component was negative; in these cases the percentage contribution for the indoor-generated component was assumed to be zero.

2.4.4. Estimating UFP deposition rates

UFP deposition rates were estimated by measuring the rate of decline of particle concentrations following individual peaks resulting from indoor sources (equation (1). This rate equals $a+k$, and was determined by regressing the logarithms of the

background-corrected concentrations against the decay time. The deposition rate k was then determined by subtracting the measured air exchange rate a.

To estimate the decay rate, it is desirable to have at least several hours of uninterrupted decay of the observed concentrations. This requires a high initial concentration; if the concentration is too low, high decay rates will not extend over a sufficient time, and the results will be biased low. Therefore peaks were required to be greater than 20,000 cm^{-3} and the decay was required to extend over at least four hours. A second restriction was imposed requiring the regression to explain at least 90% of the observed variance ($R^2 > 0.9$).

2.4.5. Relationships with co-pollutants

Spearman correlations between the hourly UFP and residential $PM_{2.5}$ and central site $PM_{2.5}$, O_3, SO_2, NO_2, NO, and NO_x measurements were examined for each week in each year-season. The latter measurements were taken by the Canadian National Air Pollution Surveillance (NAPS) Network at two Windsor locations, one in the west end and one downtown.

3. Results and discussion

3.1. Home characteristics

Participant's homes were located throughout Windsor; the median distance between pairs of homes was 6.4 km in 2005 and 5.3 km in 2006 (ranging from 0.2 to 18.8 km). Homes were primarily single-family, detached dwellings with non-smoking occupants. The majority of the 94 homes with measurements used in this analysis relied on forced air furnaces (96%) for heating, fuelled predominantly by natural gas. More than half of the homes reported a furnace-installed air cleaner (premium filter, HEPA filter or electrostatic precipitator) and 68% had air conditioners. The majority of homes had an electric stove for cooking (74%). The median measured air exchange rate was 0.20 h^{-1} with an inter-quartile range of 0.12–0.37 h^{-1}. Further details regarding home characteristics are presented in Wheeler et al. (in press-b).

3.2. Quality assurance of P-Trak measurements

Quality assurance results for the P-Trak measurements from the co-located studies are described fully in Wheeler et al. (in press-a). Bias relative to median values ranged from −17% to +8%, with only one of 14 instruments displaying bias in the same direction in all the co-location studies. No seasonal or year effects in the bias estimates were observed. Bias-corrected precision ranged from 4% to 15% (mean 10%).

Although the P-Trak counts all particles from 20 nm to 1 μm, most common aerosol mixtures have >80% of particles in the ultrafine size range (<100 nm) (e.g., Wu et al., 2008); therefore the P-Trak is generally considered to be an ultrafine particle monitor. Fresh emissions from vehicle exhaust or from electric and gas stoves typically contain particles less than 10 nm (Zhu et al., 2006; Kittelson, 1998; Wallace et al., 2008); therefore the P-Trak will underestimate levels of freshly produced UFP.

Approximately 99% of both the indoor and outdoor 30-s measurements were used in the analyses. For the outdoor data, 14,989 out of 15,602 hours (96%) attained at least 75% completeness for calculating hourly (i.e. 10-min) averages. The comparable completeness percentage for indoor measurements was 94% (14,625/15,602). Despite limitations in the particle size range, slight losses during a short warm-up period and the limited sampling time, the P-Trak instrument provided useful measurements in this field study.

3.3. Residential outdoor and indoor UFP levels

Descriptive statistics on outdoor and indoor UFP levels over each year and season are provided in Table 1; weekly data are summarized in Supplemental Tables S1 (outdoor) and S2 (indoor). Boxplots of indoor and outdoor levels for each home, year and season are provided in Supplemental Fig. 1. Outdoor levels were similar in summer 2005 and winter 2006, but considerably lower in summer 2006. Median indoor levels were similar in all 3 sampling sessions. Indoor levels were generally less than outdoor levels but were more highly skewed; indoor 99th percentiles greatly exceed the outdoor 99th percentiles.

Median outdoor levels in this study were similar to levels reported at background urban sites in Rochester, NY (Jeong et al., 2006) and Helsinki, Augsberg, and Stockholm (Aalto et al., 2005). Median UFP levels were lower than those reported in Los Angeles (Westerdahl et al., 2005) and Europe (Cyrys et al., 2008; Aalto et al., 2005; Belleudi et al., 2010).

Median indoor levels in this study were similar to levels of 7 nm–1 μm particles in the homes of asthmatics and COPD patients in Helsinki (3700 cm^{-3}) (de Hartog et al., 2010). They were lower than in homes of asthmatic and COPD patients in Athens, Amsterdam and Birmingham (UK) (de Hartog et al., 2010) and in evening hours (11,039–17,064 cm^{-3}) in 36 homes in Pembroke and Montreal, Canada (but some of these were homes with smokers) (Weichenthal et al., 2007).

3.4. Outdoor and indoor temporal variability

3.4.1. Time series plots and within home CV

Time series plots of hourly indoor and outdoor UFP measurements were created for each home; examples from two homes are provided in Fig. 1. The first example is of a home where outdoor levels remained higher than indoor levels throughout the week with little evidence of strong indoor sources. In the second example, there were frequent large indoor peaks reflecting significant indoor sources. The indoor peaks typically decayed to original levels over 3–10 h. The median within-home CV for hourly indoor UFPs was 193% compared with 56% for outdoor UFPs, also

Table 1
Descriptive statistics of hourly (10-min) averages of UFP number concentration measurements (# cm^{-3}).

	Outdoor			Indoor		
	2005 Summer	2006 Winter	2006 Summer	2005 Summer	2006 Winter	2006 Summer
N homes	44	46	45	45	47	45
N hours	4750	5130	4730	5000	5200	4760
Min	974	860	16	105	160	69
10%	5000	5450	2680	613	1250	631
25%	7230	8200	4130	1230	2100	1220
Median	10,800	12,000	6320	2700	3730	2580
75%	15,900	17,900	9660	6750	6970	6430
90%	22,200	24,600	14,300	16,600	17,800	16,200
95%	27,000	31,400	17,600	28,000	38,600	30,000
99%	39,200	55,700	26,200	101,000	146,000	111,000
Max	354,000	365,000	138,000	370,000	448,000	297,000
Mean	12,600	14,500	7650	7990	10,300	7990
Stdev	9340	12,300	5600	20,200	27,700	20,400
C.V. (%)	74	85	73	253	270	255
GeoMean	10,600	11,800	6170	3020	4220	2900
GSD	1.8	1.9	2.0	3.6	3.1	3.7

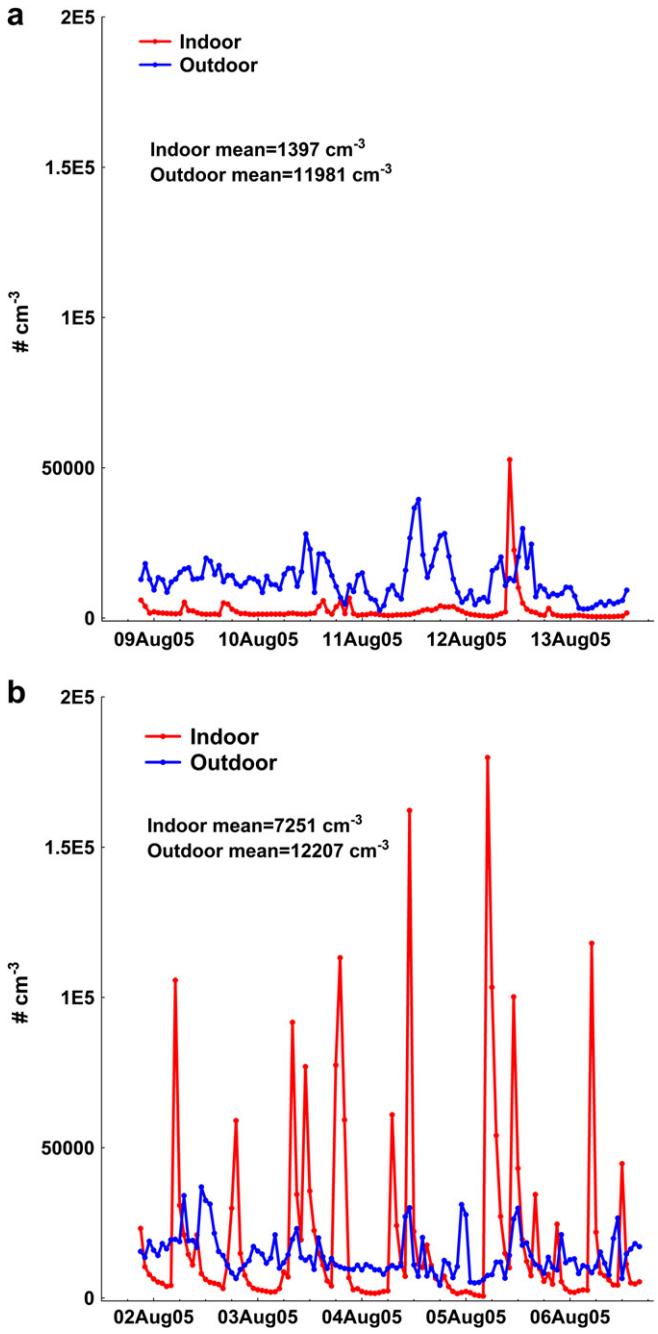

Fig. 1. Weekly time series plots of hourly indoor and outdoor UFPs at two homes showing differences in frequency and strength of indoor sources (# cm^{-3}).

indicating more variation in the indoor levels over time compared with the outdoor levels. The median CVs of the daily-averaged data ranged from 22–29% for outdoor UFPs and 44–46% for indoor UFPs; however it is important to note that there were only 5 daily average values per home.

3.4.1.1. Diurnal plots. Diurnal plots of hourly indoor and outdoor UFPs show slightly different patterns in the three sampling sessions (Fig. 2). In all three sampling sessions outdoor levels were lowest during the early morning hours followed by an increase in outdoor levels to a peak around 7–8am, possibly due to rush hour traffic. In winter and summer 2006, another smaller peak was observed around 6pm most likely associated with evening traffic. Summer 2005 had a different diurnal pattern with the highest peak around noon hour. This may be due to more frequent occurrences of mid-day events in 2005 where there was a fairly steep rise in UFP levels at all homes followed by a gradual decline (Fig. 3), likely due to a mid-day nucleation burst. These phenomena have been noticed at many locations with ambient monitors measuring UFP (McMurry and Woo, 2002; Stanier et al., 2004).

Indoor UFP levels were lowest overnight, indicating the absence or reduction of indoor sources. Peaks around noon and 6pm, particularly in 2006, probably reflect cooking sources. The variability across homes and individual days is also much lower for the overnight periods and highest for the peak periods. In a study in homes of COPD and asthmatic patients in Helsinki, Athens, Amsterdam and Birmingham, minimum indoor UFP levels were also seen from 2 to 5am and highest levels were seen around 9am, 1pm and 6pm (Hoek et al., 2008).

3.5. Outdoor UFP spatial variability and correlation between homes

3.5.1. Concurrent outdoor time series plots

Time series plots of the hourly data were created for each week showing the concurrent outdoor UFP measurements for each of the 6 homes sampled in each week. In most weeks there was considerable temporal variability across the week and in many weeks levels at different homes appeared to track each other. Examples of this are provided in Fig. 4, showing the temporal variability of UFPs in two different sampling weeks. The temporal variability and correlation evident in these plots is similar to that reported in Cyrys et al. (2008) at 4 background urban sites in Augsberg, Germany and at 4 sites in Helsinki (Buzorious et al., 1999).

3.5.2. Spatial variability

The median and 90th percentile of absolute differences in hourly outdoor UFP levels across all possible pairs of homes for each week are presented in Supplemental Table S3. The median and 90%ile's of the differences were highest in winter 2006 and lowest in summer 2006. In all 3 sampling sessions, the median absolute differences between homes were positively correlated with the distance between homes (Spearman $r = 0.53$, 0.25 and 0.19 in summer 2005, winter 2006 and summer 2006 respectively). The median and range of the hourly CVs (across the 6 homes) and the median of CODs between all possible pairs of homes for each week are presented in Supplemental Table 3. The medians for each season-year are provided in Table 2, as well as the between-home CVs and CODs for $PM_{2.5}$ for comparison. Wilson et al. (2005) has used a COD less than 0.2 criterion to distinguish between 'homogenous' and 'heterogeneous' spatial variability, but did indicate that 'there is no formal regulative or scientific definition of what constitutes uniformity at the intra-urban spatial scale'. Again, the median CV and COD are slightly larger in winter 2006, indicating slightly more variability between homes in that season. In all 3 sampling sessions, the COD was positively correlated with the distance between homes (Spearman $r = 0.64$, 0.35 and 0.40 in summer 2005, winter 2006 and summer 2006 respectively).

Both the UFP CVs and CODs are roughly twice those for $PM_{2.5}$, indicating that the spatial variability of the ultrafine particles was higher than for the concurrently measured $PM_{2.5}$.

Median CODs found in this study were similar to those from hourly measurements at 4 background urban sites in Augsberg, Germany (0.16–0.25 (winter), 0.18–0.32 (summer) (Cyrys et al., 2008). Moore et al. (2009) reported slightly higher median CODs (0.30–0.35) based on hourly means across various sites in the San Pedro Harbour area in Los Angeles, which is heavily impacted by nearby ship activity and traffic.

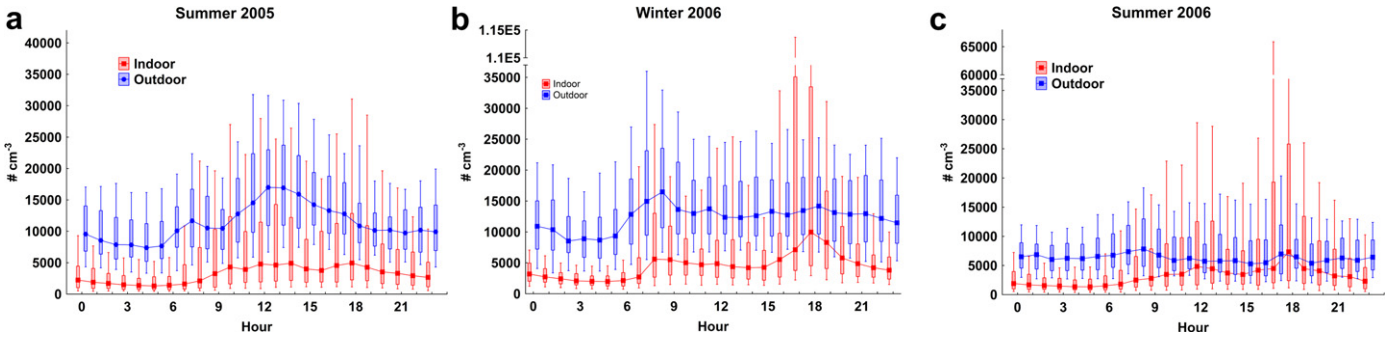

Fig. 2. Diurnal patterns of hourly indoor and outdoor UFPs by year and season. Boxplots show medians, 25–75%iles (box) and 10–90%iles (whiskers) (# cm^{-3}).

3.5.3. Correlations between homes

The median and range of the Spearman correlation coefficients across all possible pairs of homes for each week are also presented in Supplemental Table 3. The median pair-wise correlations across the 3 sampling sessions were similar, ranging from 0.56 to 0.65 (Table 2), indicating moderate correlations between homes. However, there was considerable variation in the correlations between weeks, ranging from 0.35–0.83, suggesting that in some weeks regional meteorology, atmospheric chemistry or possibly regional sources, were affecting the UFPs similarly at the various homes. In the week with the highest median correlation of 0.83 (summer 2005, week 4), the median difference between homes was 3075 cm^{-3}, similar to the median of the entire summer, indicating that while the correlation was high, the absolute levels were still varied across the homes. In all 3 sampling sessions, the Spearman correlations between homes were negatively correlated with the distance between homes (Spearman $r = -0.40$, -0.49 and -0.19 in summer 2005, winter 2006 and summer 2006 respectively).

The median correlations of UFPs levels between homes were lower than those for $PM_{2.5}$, which were greater than 0.90 for each sampling session (Table 2).

Median correlations for UFPs found in this study are similar to those reported for urban background sites in Helsinki (0.60–0.88; Buzorious et al., 1999) Higher correlations were reported by Cyrys et al. (2008) between hourly measurements at four background urban sites in Augsberg, Germany (0.77–0.92 (winter) but lower correlations were reported by Tuch et al. (2006) in Leipzing, Germany. Median correlations based on the daily average UFPs for each year-season in Windsor ranged from 0.6 to 0.8 (Supplemental Table S4), which were slightly higher than that reported between individual homes and a central site location in four European cities (0.56–0.66; Puustinen et al., 2007). Puustinen et al. (2007) also reported lower correlations between sites for UFP compared with $PM_{2.5}$.

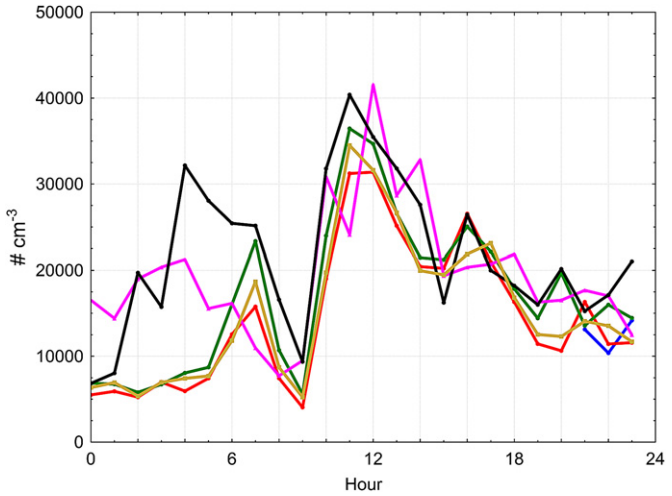

Fig. 3. Time series plot of hourly outdoor UFPs at 5 homes on Thurs 28 Jul 2005 showing possible late morning nucleation event (# cm^{-3}).

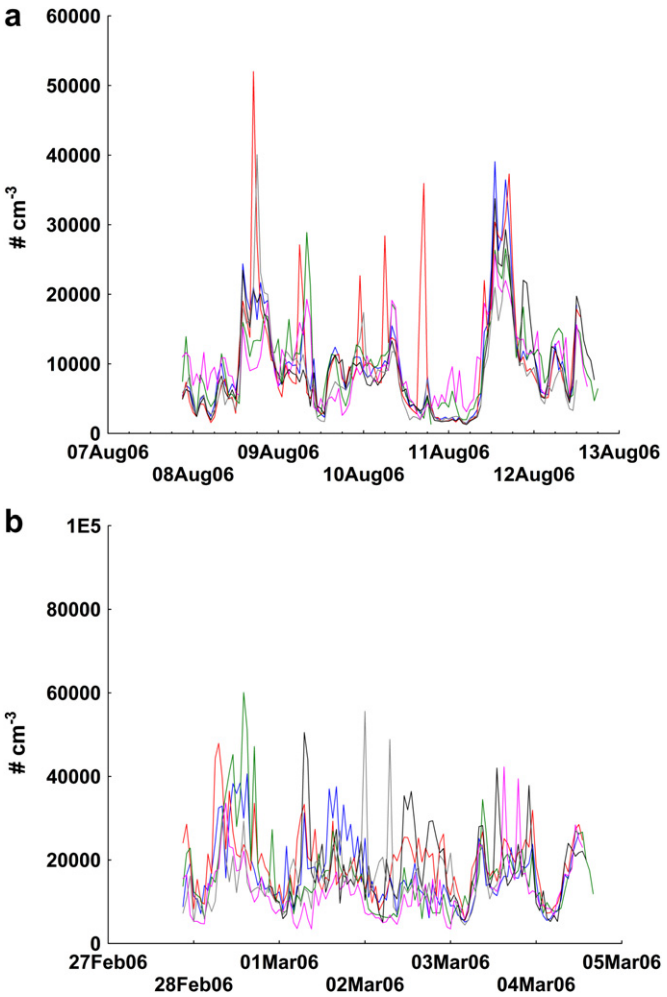

Fig. 4. Hourly outdoor UFPs measured concurrently at six homes in two different seasons (# cm^{-3}).

Table 2
Median UFP and $PM_{2.5}$ CVs, CODs and Spearman r's by year and season.

	Hourly coefficients of variance (CV) across all homes (%)			Coefficients of divergence (COD) across pairs of homes			Spearman correlations across pairs of homes (r)		
	N	Median	Range	*N*	Median	Range	*N*	Median	Range
UFPs									
Summer 2005	929	28	0–196	103	0.23	0.14–0.39	103	0.65	0.15–0.93
Winter 2006	920	34	0–198	110	0.26	0.14–0.48	110	0.56	0.07–0.89
Summer 2006	909	30	2–205	105	0.24	0.10–0.45	105	0.62	0.00–0.96
PM2.5									
Summer 2005	667	14	0–103	78	0.12	0.05–0.39	78	0.95	0.70–0.99
Winter 2006	943	18	0–202	115	0.12	0.02–0.32	115	0.96	0.62–1.00
Summer 2006	956	8	0–225	110	0.08	0.02–0.19	110	0.96	0.79–1.00

Table 3
Median hourly UFP I/O ratios, O–I differences and I–O correlations.

	I/O ratio[a]			Median O–I difference[b]		Median longitudinal I–O Spearman correlation[c]	
	Hourly averaged data	Daily averaged data		Hourly averaged data	Daily averaged data	Hourly averaged data	
	Median	Median	Range	Median	Median	Median	Range
2005 Summer	0.27	0.52	0.04–3.85	6331	5877	0.40	−0.04–0.66
2006 Winter	0.31	0.48	0.08–4.45	7199	6144	0.42	−0.14–0.72
2006 Summer	0.39	0.81	0.07–12.07	3047	1600	0.22	0.16–0.68

[a] Median of all I/O ratios across all hourly or daily averaged observations for each year season.
[b] Median of all O–I differences across all hourly or daily averaged observations for each year season.
[c] Median and range of Spearman I–O correlations calculated for each home over the year, season.

Considerable temporal and spatial variability in the hourly outdoor ultrafine particle concentrations was experienced at all homes. There was a median absolute difference of approximately 2000–3600 cm^{-3} between homes based on the hourly data, but this varied considerably for individual pairs of homes indicating that exposure characterization of absolute levels for individual homes may not be captured with one or two monitoring locations. However, the median correlations between homes were moderate (0.56–0.65), providing evidence that city-wide common sources and/or meteorology and atmospheric chemistry are driving 'background' UFP levels in the Windsor region. Other more localized sources or factors are presumably responsible for some spatial variability in absolute concentrations between homes. Calculations based on daily average data showed reduced median differences, CVs and CODs and generally increased correlations (Supplemental Table 4) compared to the hourly data, indicating that it is important to consider the averaging period when quantifying spatial variability and correlation. These daily average measures must be interpreted with caution however, since there were only 5 daily average measurements per home.

3.6. Indoor–outdoor relationships

3.6.1. Indoor/outdoor ratios, differences and correlations

Outdoor levels exceeded indoor levels 75–86% of the time; this was lowest in summer 2006 when outdoor levels were lowest for all seasons measured. Indoor/outdoor ratios and indoor–outdoor differences were calculated for the hourly and daily averaged data for each home. The median values for each year and season and longitudinal indoor–outdoor Spearman correlations are provided in Table 3. The lower indoor–outdoor difference in summer 2006 (3047 cm^{-3}) vs. 6331 cm^{-3} in summer 2005 was due to the lower outdoor levels in 2006 (the indoor levels were similar). The median I–O correlations (0.22–0.42) are fairly low, similar to those

Table 4
F_{inf} estimates (weekly-averaged censored I/O ratio).

Method	Summer 2005			Winter 2006			Summer 2006		
	Recursive model	Censored I/O	Nighttime I/O	Recursive model	Censored I/O	Nighttime I/O	Recursive model	Censored I/O	Nighttime I/O
N (homes)	33	44	44	23	46	46	30	43	44
Min	−0.08	0.03	0.03	0.04	0.07	0.04	−0.66	0.08	0.04
10%ile	0.05	0.09	0.06	0.07	0.11	0.12	0.11	0.17	0.12
25%ile	0.09	0.11	0.12	0.15	0.16	0.17	0.14	0.22	0.17
Median	0.11	0.16	0.19	0.18	0.21	0.25	0.21	0.26	0.27
75%ile	0.22	0.34	0.36	0.25	0.28	0.33	0.28	0.41	0.38
90%ile	0.38	0.43	0.54	0.32	0.36	0.44	0.40	0.60	0.57
95%ile	0.50	0.58	0.64	0.34	0.38	0.45	0.60	0.66	0.61
99%ile	0.77	0.79	0.66	0.35	0.45	0.51	0.62	0.87	0.90
Max	0.77	0.79	0.66	0.35	0.45	0.51	0.62	0.87	0.90
Mean	0.18	0.23	0.26	0.19	0.22	0.26	0.21	0.33	0.30
StdDev	0.17	0.18	0.19	0.09	0.09	0.11	0.21	0.18	0.18
StdErr	0.03	0.03	0.03	0.02	0.01	0.02	0.04	0.10	0.03
GeoMn	0.14	0.18	0.20	0.17	0.20	0.23	0.20	0.28	0.25
GeoSd	2.19	2.01	2.22	1.78	1.57	1.65	1.73	1.74	1.98

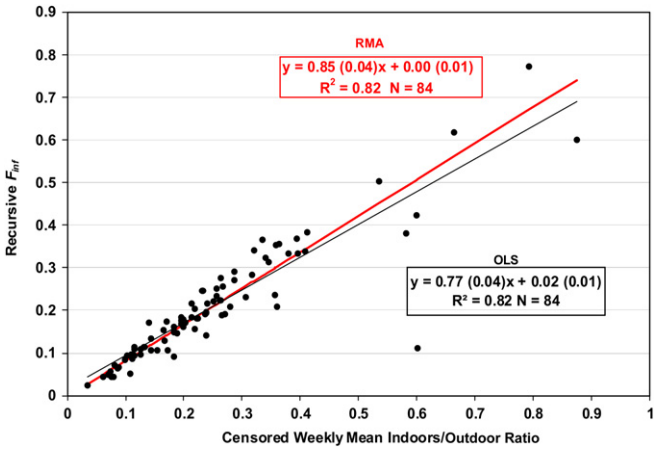

Fig. 5. Comparison of weekly F_{inf}'s: recursive model vs. censored I/O ratio using reduced major axis and ordinary least squares regression.

reported for UFPs 7 nm-3 μm at homes in 4 European cities (ranging from 0.25–0.62) (Hoek et al., 2008). This is not surprising because indoor sources obscure the indoor–outdoor relationship.

3.6.2. Infiltration factor estimates

Table 4 provides summary statistics on the daily and weekly F_{inf} estimates from the three methods: recursive model, censored I/O ratios and overnight I/O ratios. The recursive model estimates were available from a fewer number of homes due to missing air exchange data. The highest Spearman correlation and regression R^2 were between the estimates from the recursive model and censored I/O ratios (e.g. Spearman $r = 0.93$, $R^2 = 0.82$ (weekly data). The lowest correlations were between the estimates from the overnight I/O ratio and the other two methods (Spearman $r \leq 0.8$, $R^2 < 0.6$). Median estimates were generally highest for the overnight I/O ratio and lowest for the recursive model (Table 4). The reduced major axis regression parameters indicate that the recursive estimates were, on average, 85% of the censored I/O ratio estimates (Fig. 5).

In general, in the absence of sources, indoor levels lag behind outdoor levels, causing the I/O ratio to be either an underestimate or overestimate of the true equilibrium ratio depending on whether the outdoor concentration is increasing or decreasing. Over a 24-h or 120-h period, it is likely that the mean censored indoor concentration divided by the mean outdoor concentration would be close to the true equilibrium value. The recursive model also depends on the censored values, but the outdoor relationship is modelled more accurately. It is not clear why the recursive model generally produced estimates that are slightly (about 15%) less than the censored I/O method.

Overnight I/O ratios may overestimate F_{inf} due to ambient-infiltrated UFPs lagging behind the drop in outdoor UFPs typically seen during the early hours. Also any late-night indoor source may affect levels for 5–10 h. Indeed, in summer and winter 2006, the I/O ratio showed a decline from the 2–3am hour until the 5–6am hour, suggesting that the early hours of the morning may have been influenced by lingering elevated concentrations from earlier indoor source events, causing the overnight I/O ratio to be an overestimate of F_{inf}.

Because of the fewer missing values, and the strong correlation between the censored I/O ratio and recursive model F_{inf} estimates, plots showing the median daily and weekly F_{inf} estimates based on the censored I/O ratios for each home are presented in Fig. 6. The medians across all homes were 0.16 (summer 2005), 0.21 (winter 2006) and 0.26 (summer 2006), however the maximum weekly F_{inf} was considerably lower in winter 2006 (0.45) compared to the two

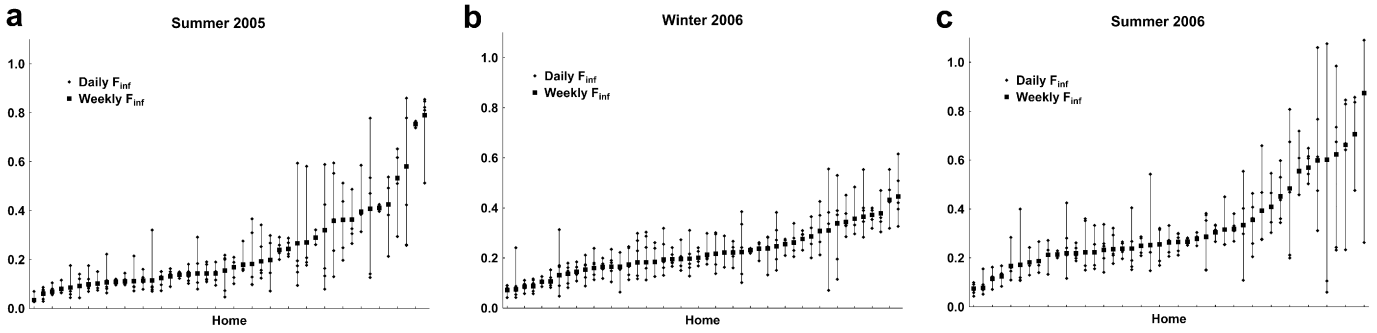

Fig. 6. F_{inf} estimates for each home (daily estimates and 5-day median for each home).

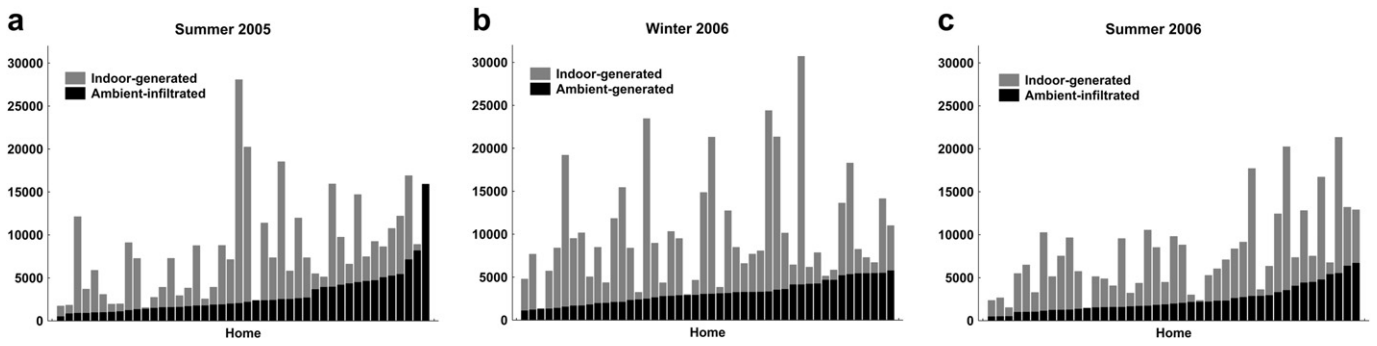

Fig. 7. Estimates of indoor-generated and ambient-infiltrated components of indoor UFP concentrations (time-averaged over the 5-day sampling period) (# cm^{-3}).

Table 5
UFP deposition rates (h^{-1}) by season.

	N of peaks	Min	10%ile	25%ile	Median	75%ile	90%ile	Max	Mean	SD	Geo Mean	GSD
Deposition rate (k)												
2005 Summer	97	0.06	0.39	0.61	0.76	1.00	1.32	1.96	0.82	0.37	0.73	1.76
2006 Winter	79	0.08	0.26	0.41	0.61	0.85	1.15	1.87	0.68	0.37	0.58	1.91
2006 Summer	92	0.21	0.42	0.59	0.79	1.15	1.41	2.30	0.87	0.40	0.78	1.63

summer periods (0.79, 0.87). The lower maximum F_{inf} in winter probably reflects homeowner attempts to reduce heat loss (e.g. closed windows).

UFP infiltration estimates from other studies vary significantly. The values found in this study are similar to the I/O ratios reported by Hahn et al. (2009) in a near-highway urban 3-story unoccupied test house in Brooklyn, NY (0.10–0.33) and by Zhu et al. (2005) for particles 10–20 nm (0.1–0.4). However, higher ratios have been reported in a number of other studies (0.75 for 80–300 nm particles, 0.5 for 20–30 nm particles in Sarnat et al., 2006; 0.34 in Hussein et al., 2005). The median UFP F_{inf}'s in this study are lower than those reported for $PM_{2.5}$ which typically range from 0.4 to 0.7 (Wallace et al., 2006; Liu et al., 2003; Landis et al., 2001). Barn et al. (2007) reported a F_{inf} of 0.27 for $PM_{2.5}$ in winter in Prince George, BC.

3.6.3. *Relative contribution of ambient and indoor sources to total indoor concentrations*

The censored I/O ratio F_{inf} estimates were used to calculate the weekly-averaged ambient-infiltrated and indoor-generated components of indoor PM for each home (Fig. 7). The median contribution of infiltrated ambient UFP to indoor concentrations was 42%, 35% and 31% in summer 2005, winter 2006 and summer 2006 respectively, however individual homes varied significantly.

3.7. *Indoor UFP deposition rates*

The median deposition rates (0.76, 0.79 h^{-1}) agreed very closely for two different sets of 38 homes in the two summer seasons (Table 5). The winter season had a somewhat lower value of 0.61 h^{-1}. The range of deposition rates (k) is provided for one season in Fig. 8. Individual median estimates of k from summer and winter 2006 were poorly correlated (Spearman $r = 0.20$). These values are larger than typical estimates of deposition rates for $PM_{2.5}$, such as the value of 0.39 h^{-1} from the PTEAM study (Özkaynak et al., 1996). Larger values are expected because the smaller particles have higher diffusion rates and therefore higher probability of striking surfaces indoors.

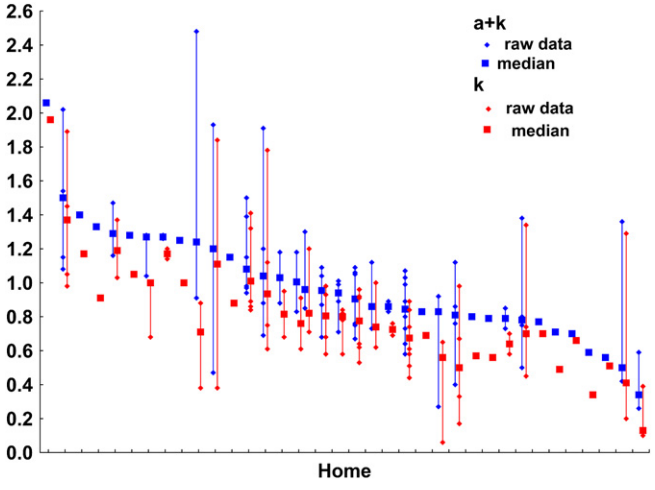

Fig. 8. Individual estimates of $(a + k)$ and k – Summer 2006 (raw data and median for each home).

As can be seen from Fig. 8, the uncertainty of these estimates is high. The reasons include a) the small number of points (only 4 in many cases) available to determine the total decay rate, b) the uncertainty in the estimate of the air exchange rate, which depends not only on measurement of the flow rate but also on an accurate estimate of the effective volume of the well-mixed portion of the house, c) the fact that only a 24-h average of the air exchange rate is available, thus an assumption must be made that the air exchange rate applies to the decay period, and d) unknown factors that might affect deposition during the measurement, such as having a forced air furnace fan turned on at certain times, which would add an additional loss factor due to filtration and deposition in the ductwork.

3.8. *Relationships of outdoor UFPs with co-pollutants*

The weekly median Spearman correlations (across 6 homes) of UFPs with the $PM_{2.5}$ measured outside the home and various co-pollutants measured at two central site NAPS locations, are provided in Table 6. Correlations are generally low, even negative for O_3 in some sampling sessions. The gases (SO_2, NO, NO_2, and

Table 6
Weekly Spearman correlation coefficients between UFP and co-pollutants.

		Summer 2005 (44 homes)		Summer 2006 (45 homes)		Winter 2006 (46 homes)	
		Median	Range	Median	Range	Median	Range
At homes outdoors $PM_{2.5}$ (Dusttrak)		0.24	−0.27 0.73	0.10	−0.39 0.46	0.18	−0.33 0.69
	Naps site						
$PM_{2.5}$ (TEOM with DRYER)	60,211	0.31	−0.15 0.75	0.23	−0.12 0.42	0.23	−0.01 0.56
	60,204	0.25	−0.17 0.75	0.22	−0.13 0.48	0.25	0.00 0.58
O_3	60,211	0.19	−0.37 0.45	−0.17	−0.62 0.34	−0.26	−0.65 0.35
	60,204	0.19	−0.30 0.42	−0.17	−0.61 0.31	−0.34	−0.70 0.24
SO_2	60,211	0.40	−0.04 0.75	0.29	−0.23 0.54	0.15	−0.37 0.53
	60,204	0.38	−0.02 0.69	0.28	−0.05 0.51	0.24	−0.01 0.53
NO_2	60,211	0.14	−0.32 0.50	0.34	−0.12 0.70	0.33	−0.20 0.69
	60,204	0.23	−0.17 0.62	0.39	−0.12 0.77	0.49	0.03 0.76
NO	60,211	0.31	−0.10 0.50	0.25	−0.23 0.49	0.45	−0.09 0.65
	60,204	0.28	0.05 0.60	0.18	−0.09 0.50	0.51	−0.07 0.73
NO_x	60,211	0.17	−0.31 0.54	0.35	−0.14 0.72	0.41	−0.19 0.72
	60,204	0.27	−0.09 0.67	0.43	−0.14 0.76	0.52	−0.01 0.75

NO_x) tend to have slightly higher median correlations with UFPs than with $PM_{2.5}$. SO_2 was more highly correlated with UFP in summer 2005 than in the 2006 sessions, while the median correlations between UFPs and NO_2, NO and NO_x were generally higher in the winter seasons.

Jeong et al. (2006) also found negative correlations between O_3 and UFPs at a downtown Toronto site. In that study, UFP correlations were also higher with NO and NO2 than $PM_{2.5}$ but correlations with SO_2 were lower than with the other pollutants. For all four pollutants, correlations were higher in the winter than summer. Our findings indicate that UFPs may share some common sources and factors affecting levels with other co-pollutants; however none of them appear to be strong surrogates for UFPs.

4. Conclusions

This is one of the first studies that has estimated UFP infiltration and deposition rates for a large number of homes. Based on censored indoor/outdoor ratios, the median weekly UFP infiltration factors were 0.16, 0.26 and 0.21 in the three sampling sessions, however values ranged from 0.04 to 0.86 across individual homes. For more than 65% of homes, indoor-generated UFPs provided more of the total indoor UFP concentration than infiltrated ambient UFPs. Mean deposition rates across the three sampling seasons ($0.61–0.79\ h^{-1}$) were considerably higher than previously published estimates for $PM_{2.5}$. Spatial variability in outdoor UFPs appeared to be greater than for $PM_{2.5}$, as reflected in larger between-home coefficients of divergence and lower between-home correlations. The wide range of infiltration factors across homes as well as spatial variability and moderate between-home correlations of outdoor UFPs could cause measurement error in epidemiology studies using outdoor UFP measurements at a central location as a surrogate for personal exposure to ambient UFPs. Median hourly correlations between residential outdoor UFPs and co-pollutants measured at the home or at two central monitoring sites were generally low indicating that these other pollutants would not be appropriate surrogates for UFPs in epidemiological studies.

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Appendix. Supplementary data

Supplementary data associated with this article can be found in the online version, at doi:10.1016/j.atmosenv.2010.11.002.

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