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Residential infiltration of fine and ultrafine particles in Edmonton

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HIGHLIGHTS

- Residential particle infiltration factors were estimated in Edmonton, Alberta.
- Considerable variability in infiltration factors was seen within and between homes.
- Median infiltration factors were higher in summer and higher for fine particles.
- On average, the majority of indoor fine particles originated outdoors.
- On average, the majority of indoor ultrafine particles were of indoor origin.

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ABSTRACT

Airborne indoor particles arise from both indoor sources and ambient particles that have infiltrated indoors. The intra-urban variability of infiltration factors (F_{inf}) is a source of measurement error in epidemiological studies estimating exposure from a central site measurement, hence information on the within and between-home variability of F_{inf} is useful to better characterize ambient PM exposure. The objective of this paper was to estimate magnitudes and predictors of daily residential infiltration factors (F_{inf}) and ambient/non-ambient components of indoor ultrafine particle (UFP) and fine particle (FP) concentrations. FPs and UFPs were measured continuously for 7 consecutive days in 74 Edmonton homes in winter and summer 2010 (50 homes in each season). Simultaneous measurements of outdoor (near-home) FP and ambient (at a central site) UFP concentrations were also measured. Daily infiltration factors were estimated for each home; considerable variability was seen within and between homes. For FPs, seasonal-averaged F_{inf} (the average of the 7 daily F_{inf} estimates) ranged from 0.10 to 0.92 in winter (median = 0.30, n = 49) and 0.31 to 0.99 in summer (median = 0.68, n = 48). For UFPs, the seasonal-averaged F_{inf} ranged from 0.08 to 0.47 across homes in winter (median = 0.21, n = 33 houses) and from 0.16 to 0.94 in summer (median = 0.57, n = 48). The higher median F_{inf} in summer was attributed to a high frequency of open windows. Daily infiltration factors were also estimated based on the indoor/outdoor PM_1 sulfur ratio. These estimates were poorly correlated with DustTrak-based FP infiltration factor estimates; the difference may be due to losses of volatile components on the PM_1 filter samples. Generalized linear mixed models were used to identify variables significantly associated with F_{inf} and the non-ambient component of indoor FP and UFP concentrations. Wind speed was consistently associated with F_{inf} across all seasons for both FPs and UFPs. The use of an air cleaner was associated with reduced UFP infiltration factors in summer, suggesting a potential method of reducing infiltrated UFPs. Various cooking activities and smoking were associated with the non-ambient component of indoor FP and UFP concentrations. On average, the majority of indoor FPs were of ambient origin while the majority of UFPs were of indoor

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origin. In summer, more of the indoor FP and UFP concentrations were from ambient origin, compared to winter, due to the higher infiltration factors. The variability in FP and UFP F_{inf} within and between homes may cause substantial exposure misclassification in epidemiological studies using only ambient measurements.

1. Introduction

Exposure to fine particles (FPs) (particles of aerodynamic diameter less than <2.5 μm) and ultrafine particles (UFPs) (particles of aerodynamic diameter less than <100 nm) has been associated with human health effects (for reviews see Health Effects Institute, 2013; US EPA, 2009). Human exposure to FPs and UFPs comes from both outdoor (e.g. vehicle, industrial, heating emissions and atmospheric chemical reactions) and indoor (e.g. cooking, candles, indoor chemical reactions, cleaning and dusting) sources. People spend most of their time in residential indoor environments (Matz et al., 2014; Schweizer et al., 2007) where they are exposed to particles of indoor (non-ambient) origin as well as outdoor particles that have infiltrated indoors; these particles may differ in composition and toxicity.

The infiltration factor (F_{inf}) is defined as the fraction of ambient particles that penetrates indoors and remains suspended under steady state conditions (Wilson et al., 2000). It is a function of the particle penetration efficiency (P), the air exchange rate (α) and the deposition rate (k):

$$F_{inf} = \frac{(P\alpha)}{(\alpha + k)}$$

Estimation of P, k and α is difficult (Switzer and Ott, 1992; Stephens and Siegel, 2012; Chao et al., 2003; Thatcher et al., 2012; Wallace et al., 2013; Long et al., 2001; Sherman, 1989; Lunden et al., 2012) but F_{inf} can be estimated as the indoor/outdoor ratio when there are no indoor sources or the indoor source signal has been censored. Consequently there is less uncertainty in measuring F_{inf} directly than in estimating F_{inf} from estimates of P, k and α.

F_{inf} is affected by numerous factors including a) building characteristics and occupant behaviors, including air exchange rates (determined by the tightness of the building envelope and other factors such as window/door openings (Rim et al., 2010, 2013a; Hystad et al., 2009; MacNeill et al., 2012)), forced air heating, exhaust fan use, air conditioning use (Clark et al., 2010; Allen et al., 2012; Rim et al., 2013b, Stephens and Siegel, 2013) and use of filtration devices (Barn et al., 2008), (b) meteorological parameters such as wind speed and indoor–outdoor temperature difference (Hahn et al., 2009; MacNeill et al., 2012), (c) particle size (Rim et al., 2010; Zhu et al., 2005; Long et al., 2001; McAuley et al., 2010) and (d) particulate matter (PM) composition, specifically where it contains semi-volatile components that can be lost indoors due to volatilization (Allen et al., 2012; Hodas et al., 2012; Meng et al., 2007; Sarnat et al., 2006; Sangiorgi et al., 2013; Lunden et al., 2003).

Estimates of daily infiltration factors from multiple homes in multiple cities are useful for risk assessment as inputs to models of human exposure to ambient PM. As well, many epidemiological studies associating ambient particle exposure and health are based on central site ambient measurements, so variability in residential infiltration over time and between homes may be an important modifying factor in exposure-health outcome relationships (Hodas et al., 2012; Allen et al., 2012). A few Canadian studies have estimated infiltration factors for UFP and FP in multiple homes (e.g. Windsor, Ontario (Kearney et al., 2011; MacNeill et al., 2012), Toronto (Clark et al., 2010), and Prince George (Barn et al., 2008). This paper builds on this work by reporting on continuous measurements of indoor and outdoor FP and UFP as well as the estimated daily infiltration factors and ambient and non-ambient components of indoor FP and UFP concentrations, from an indoor air study carried out in 74 homes in Edmonton, Alberta in winter and summer 2010 (50 homes each season, 26 of which were measured over two seasons).

2. Methods

2.1. Study design

A single stage stratified sample design was used, in which Edmonton neighborhoods were stratified into five groups based on the construction age of the majority of homes in the neighborhood (≤1945, 1946–1960, 1961–1980, 1981–2000, and ≥2001), according to the Edmonton Community Profile of the 2006 Canadian Census. Two neighborhoods were randomly selected for each of the house age groups, and five homes were randomly selected in each of the neighborhoods based on a proposed total target sample size of 50 homes per season.

Home recruitment within each neighborhood stratum was accomplished using door-to-door surveys of home owners on randomly selected streets within the selected neighborhoods. Participants had to be the owner of the home, at least 18 years of age, capable of participation with no language barriers and if possible, available to participate for both sampling phases. As well, all residents of the homes had to be non-smokers. Informed consent was obtained from each participant. Ethics approval was obtained from both Health Canada and the University of Alberta Research Ethics Boards.

The study was carried out in the winter (January–April) and summer (June–September) seasons in 2010. Nine seven-day sampling periods were conducted per season with six homes measured concurrently per sampling period. There were seven 24-h measurements made per home. Participants were asked to continue with their normal activities throughout the sampling period.

The study included baseline and daily questionnaires and measurements of 24-h daily air exchange, continuous indoor UFPs, continuous indoor and outdoor FPs and daily 24-h indoor and outdoor PM_1 and $PM_{2.5-1}$ mass. Ambient measurements were carried out at the National Ambient Pollution Surveillance (NAPS) Edmonton South monitoring site (on 61 Ave NW) operated by the Alberta Environment, including (in duplicate) continuous UFPs, continuous FPs and daily 24-h PM_1 mass. XRF elemental analysis, including sulfur, was carried out on the PM_1 filters.

2.2. Questionnaires

A technician-administered baseline questionnaire was used to obtain information on housing characteristics, such as age and type of home and heating, ventilation and cooking systems. Participants also completed daily questionnaires to obtain information on activities in the home in the previous 24-h, such as cleaning and

cooking, use of kitchen and bath fans, air conditioning and air cleaners.

2.3. Continuous measures of UFPs, FPs, temperature, relative humidity

UFPs were measured inside each home in the main living area using two P-Traks (Model 8525, TSI, Inc.; flow rate 0.7 lpm) to allow for 24-h operation before replenishment of the alcohol. One P-Trak operated from minutes 1 to 15 and the other from minutes 31 to 45 each hour. Ambient measurements were taken at the NAPS site using a P-Trak (15 min/h) which measures particles from 20 nm to about 1 μm. At least 80% of particles are normally in the ultrafine range (<100 nm), so the P-Trak is considered to be mainly a UFP monitor. In winter only, a Scanning Mobility Particle Sizer (SMPS) (Model 3080, TSI, Inc.) was also used to measure the size-specific UFP distribution from 3 to 135 nm.

Indoor and outdoor FPs were continuously measured using two DustTraks (Model 8520, TSI Incorporated, Shoreview, MN, USA) with 2.5 μm inlets to restrict particles to those smaller than 2.5 μm in diameter ($PM_{2.5}$). Air was sampled at a flow rate of 1.7 lpm. The outdoor continuous monitors and pumps were housed in a waterproof enclosure. In the winter this was heated (10 °C); in summer, a fan was used for cooling and reflective insulating bubble wrap was used to reflect the sun and reduce the impact of heat. DustTrak measurements were adjusted by the zero-check values reported by the instrument.

DustTraks are optical monitors and depend on an estimate of particle density to be related to $PM_{2.5}$ mass concentrations; DustTrak values typically overestimate $PM_{2.5}$ (Wallace et al., 2011). Twenty-four hour average DustTrak concentrations were compared to the sum of the co-located $PM_{2.5-1}$ and PM_1 gravimetric samples (from the Harvard Coarse Impactor) using Reduced Major Axis (RMA) regression. The slopes of the regressions were 2.44 (winter indoors), 3.01 (winter outdoors), 2.76 (summer indoor) and 2.75 (summer outdoors) (see Supplemental Information (SI) for further detail). These values are similar to that reported from Windsor (indoor 2.05/outdoor 2.78, Wallace et al., 2011) and slightly higher than those reported from Halifax (indoor 1.88/outdoor 2.05, MacNeill et al., 2014). The concurrent optical and gravimetric measurements were used to test whether the DustTrak F_{inf} was similar to the gravimetric F_{inf} (provided in the SI).

Indoor temperature and relative humidity were recorded continuously at each house at 10 min intervals using a YES-206 Falcon data logger (Young Environmental, Calgary, AB). Daily outdoor temperatures, relative humidity and other meteorological data from the Edmonton South air monitoring station were obtained from www.casadata.org for each sampling season.

2.4. PM_1 and sulfur in PM_1

Filter-based size-fractionated samples of particulate matter were measured indoors and outdoors at each home and at the central site for 24-h using Harvard Coarse Impactors (HCIs) which have 3 stages in series to collect $PM_{10-2.5}$, $PM_{2.5-1}$ and PM_1 (HCI, Harvard School of Public Health, Boston, MA, USA). These were operated at 5 lpm using a BGI personal sampling pump (model # 400-10, BGI Inc., Waltham MA, USA). Flow rates were validated at the start and end of each 24-h period using Dry Cal DC lite (Bios *Int.*, Butler, NJ, USA). In the HCI, polyurethane foam filters were used to collect the $PM_{10-2.5}$ and $PM_{2.5-1}$ stages and a Teflon filter (37 mm, 2 μm pore size, Pall Inc., Port Washington, NY) collected the PM_1 stage. Gravimetric analysis of both the 37 mm Teflon filter and the polyurethane foam was conducted using the method outlined in the Assurance Guidance Document 2.12 by the US EPA (U.S. EPA, 1998). All parts of the HCIs were thoroughly cleaned at the beginning of the study, and the impaction plates, filter holders and body were cleaned when filters were changed out.

Following determination of PM_1 mass, elemental analyses were completed using XRF (RTI Laboratories, Research Triangle Park, NC) following protocols consistent with EPA method IO-3.3 (EPA 625/R-96/010). All valid samples having a PM_1 mass greater than 5 μg were analyzed for sulfur (S), while samples having a mass greater than 10 μg were analyzed for 33 elements, including sulfur.

2.5. Air exchange

Home air exchange rates (AER) were determined for the corresponding 24-h periods by the perfluorocarbon tracer (PFT) technique (Dietz et al., 1986). Four PFT emitters were placed throughout the same floor on which the air monitoring equipment was located (in general, the main floor in the living room). Each day, one capillary absorption tube (CAT) was placed near the center of the sampling floor at head height. CATs were analyzed by gas chromatography with electron capture detection. Air exchange rates were calculated by dividing the infiltration rate by the measured house volume. One home per week was chosen for two-zone sampling. Further details are in the SI.

2.6. QA/QC

P-Traks: A pre- and post-season instrument inter-comparison was carried out on the P-Trak and DustTrak samplers each season to evaluate instrument variability. The laboratory inter-comparison of 14 P-Traks lasted over 17 h. The P-Traks were allowed to run throughout, and the time at which their alcohol reservoir ran out was determined; this time ranged from 7 to 11 h. Two P-Traks recorded a spike about 2 h after the experiment began that greatly reduced their subsequent readings; these were removed from data analysis. The remaining 12 P-Traks showed biases (relative to the median value) ranging from −11% to +14%; 9 instruments had biases ranging from −5% to +8%. The median bias-corrected precision was 4% (range 2%–11%).

The field study employed two indoor P-Traks measuring 15-min periods separated by a half hour (section 2.3). The Spearman rank correlation of these UFP measurements across 322 person-days was 0.91 (see SI for further details). Another estimate of precision was available from field data where the occasional incorrect programming of the timers resulted in 14,677 min during which both P-Traks were operating simultaneously. For these measurements, the median precision was again 4% (IQR 2–7%).

DustTraks: The laboratory inter-comparison of the DustTrak instruments was carried out in pre- and post-winter and summer seasons. A total of 21 monitors were tested at least once, between 14 and 17 monitors at a time, for a total of 59 tests. Three comparisons lasted about a day, the fourth for only a few hours. Twelve of the 59 tests (20%) resulted in monitors showing a bias and precision greater than 20% with 9 different monitors affected at least once. The median absolute bias ranged from 4 to 10%. Median bias-corrected precision was excellent in all tests (3–4% in winter, 8% in summer). The LOD was 3 $\mu g/m^3$ in two tests, about 6 $\mu g/m^3$ in a third and was undetermined in one test due to lack of concentrations below 10 $\mu g/m^3$.

PM_1 and Sulfur There were 87 valid pairs of co-occurring 24-h PM_1 mass measurements from the two HCIs at the NAPS station. There was no significant difference in measurements between the two instruments (paired *t*-test, $p = 0.15$). The median precision was 6% (range <1%–68%, IQR 2%–15%). Similarly, there were 93 co-occurring 24-h sulfur measurements. There was no significant difference in the two sets of measurements (paired *t*-test $p = 0.18$).

When one measurement below the LOD was excluded, the median precision was 1.8% (range 0.01%–10%, IQR 0.9%–3%).

Air exchange rates Duplicate CAT measurements were used to estimate precision for the one zone air exchange measurements; the median precision was 1.2% ($n = 109$ duplicates, IQR: 0.5–2.5%). The median one-zone to two-zone ratio was 0.8 in summer (IQR: 0.64–0.94) and 0.94 in winter (IQR: 0.83–1.0) (Van Ryswyk, personal communication).

2.7. Data analysis

Invalid indoor and outdoor field measurements of FPs and UFPs were identified by inspection of plots or concurrent measurements, and data were omitted when a monitor appeared to be performing poorly (for example, large instantaneous spikes without a subsequent decay, continuous declines to zero and negative values).

Daily infiltration factors for UFPs and FPs were calculated using the half-hourly datasets. For the P-Trak, half-hour indoor averages were based on the measurements from the two instruments. For FPs, outdoor measurements from each home were used in calculating $F_{inf.}$ For UFPs, only ambient measurements at the central site were available for the calculation of F_{inf}, which may introduce uncertainty in the F_{inf} estimates depending on the proximity of the homes to the central site and the degree of spatial variability of UFPs in Edmonton. For both P-Trak and DustTrak data, missing outdoor/ambient values were replaced by imputed values for periods up to 6 h using linear interpolation. Daily infiltration factors were calculated as the ratio of the daily mean of censored indoor values divided by the daily mean of the outdoor or ambient values. The censoring algorithm is described in detail in Kearney et al. (2011). Daily F_{inf} estimates were calculated for sample days that had at least 36 of 48 possible half-hour averages (75% data completeness). This method 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 value, due to the lag of the indoor values behind the outdoor values, over time the mean values will approach the equilibrium indoor/outdoor ratio.

An additional method of estimating FP F_{inf} was based on the indoor/outdoor sulfur ratio. Since sulfur has few indoor sources and is found primarily in ambient fine PM (rather than coarse PM), the sulfur I/O ratio has been used as an estimate of $PM_{2.5}$ F_{inf} (Allen et al., 2012; Clark et al., 2010, Wallace and Williams, 2005; Allen et al., 2003).

The F_{inf} estimates were also used to estimate a) the daily average concentration of ambient UFPs or FPs that has infiltrated indoors, calculated as the product of the daily F_{inf} estimate and the daily mean outdoor UFP or FP concentration (termed the 'ambient' component), and b) the average UFP concentration originating from indoor sources (termed the 'non-ambient' component) calculated by subtracting the infiltrated ambient estimate from the daily mean indoor concentration. In cases where the ambient-component exceeded the indoor concentration then the ambient component was assumed to be equal to the indoor concentration and the percentage contribution for the non-ambient component was assumed to be zero.

Mixed models were used to identify predictors of the daily F_{inf} and non-ambient component estimates for both FPs and UFPs. Given the repeated measures design of the study, a generalized linear mixed model with a variance components covariance structure was used (using PROC MIXED (SAS Inc.)). A list of potential predictors was selected from the baseline and daily questionnaires, and the meteorological data for both F_{inf} and the non-ambient components based on previous knowledge and literature, such as window opening behavior, air cleaning devices, ventilation for the F_{inf} models and possible indoor sources (cooking, candles, heating) for the non-ambient component models. See Table S1 for a list of these predictors and frequency counts/medians. Where possible, missing values were imputed using the median value of the non-missing data. To reduce collinearity within the predictors, all pairs of variables with a correlation coefficient >0.7 or <-0.7 were identified, and for each pair, the variable with the poorest association with the outcome variable (based on a p-value from the univariate mixed model) was dropped from the model selection process. Further collinearity was examined using the variance inflation and collinearity estimates produced in PROC REG (SAS Inc.). As well, variables associated with a variance inflation factor of 10 or greater, or a collinearity index of 30 or greater were removed from the analyses. It is important to consider that the specific types of cooking were correlated. In order to reduce collinearity, some cooking variables were removed from the analysis that were only slightly less associated with the outcome variables. After variable selection was complete, all remaining variables were entered into a backward stepwise selection process (using mixed models). All variables that had a strong univariate association with either F_{inf} or the non-ambient component were later re-entered into the model to see if they were still significant. Final models were chosen based on minimizing the Akaike Information Criteria (AIC) and the significance of each of the independent variables.

Intercept-only mixed models were used to provide estimates of the between and within-subject variance components of the daily F_{inf} and non-ambient component variables. The variance components from the final predictive models were compared with the variance components of the intercept-only model to estimate the percentage of the between and within-variance explained by each model. All models were run separately for each season.

Data cleaning and management was carried out using Excel 2007 (Microsoft Inc.) and SAS (v9.2 in SAS/EG 4.2, SAS Inc.). Statistical analyses were carried out using SAS; graphs were prepared in Statistica v 9.0 (Statsoft Inc.).

3. Results and discussion

Fifty homes were sampled each season. Twenty-four homes participating in the first season did not participate in the second season and were replaced by 24 other homes. Basic statistics of house characteristics are provided in Table S1. All homes but one had forced air heating distribution systems. Three homes had an air exchanger (two were sampled in summer only, the other was sampled in the winter only). Three homes had an electrostatic precipitator on their furnace (one was sampled in both seasons, one in summer only and the other in winter only). Thirteen homes in summer reported using air conditioning. These homes reported using air conditioning on an average of 5.1 days during the 7 day study period (range 2–7 days) (9 used central a/c, 4 used room a/c and 2 reported using both central and room air conditioners). Twelve homes in winter reported running their furnace fan continuously on an average of 5.2 days during the 7 day study period (range 1–7 days). In summer, participants reported opening at least one window on 73% of the 346 home-days with a mean number of 2.47 windows opened (median = 2, range 1–10). In winter, participants reported opening at least one window on 35% of the 348 home-days with a mean number of 0.48 windows opened (median = 0, range 1–5). One participant reported that smoking occurred indoors on all 7 days, both seasons. Two other participants reported indoor smoking events on 1 day each, both in winter.

For indoor and outdoor DustTraks, 1.5% of the indoor and 7.6% of the outdoor continuous measurements were invalidated after inspection of the datasets and plots (e.g. erratic or flat-line

measurements indicating machine malfunction). For indoor and ambient P-Traks, 15% of the indoor data and <1% of the ambient data were invalidated. The UFP monitors were programmed to start at 7 am the day following the set-up so there was generally only about 10 h of data collected on the 1st day. For winter week 4 (16–22 Feb), missing ambient P-Trak data was imputed based on measurements from the 15.4–130 nm size fraction collected by the SMPS (r^2 = 0.9). Extreme values were noted for the outdoor FP measurements starting 19 Aug 2010, attributed to a forest fire, with maximum values reaching 1500 μg/m^3 or greater and levels exceeding 100 μg/m^3 through to the end of the sampling week on 23 Aug 2010. UFP data from the ambient site was missing from 19 Aug 6:15 AM to 22 Aug 7:00 AM during the forest fire event due to equipment malfunction; SMPS data were not available for imputing the missing data. For the gravimetric samples, the first 4 weeks of data in winter were lost because of technical problems in sampler assembly, affecting 24 homes.

3.1. Residential indoor, outdoor and ambient levels and air exchange

Basic summary statistics for the FP and UFP half-hour datasets used in the F_{inf} calculations are provided in Table 1, as well as basic statistics on the daily PM_1, $PM_{1-2.5}$, sulfur (in PM_1), air exchange, temperature and relative humidity measurements. The median outdoor FP (near-home) and ambient (central-site) UFP concentrations were higher than indoors in both winter and summer, but maximum values were measured indoors. Median outdoor FPs and ambient UFPs were both slightly higher in winter than summer but the median indoor FPs and UFPs were higher in summer. The outdoor and indoor median FP concentrations were similar to those found in a multi-season study in Windsor, ON (Wheeler et al., 2011). Median indoor FP concentrations were slightly lower than those found in a multi-home study in Halifax (winter: 5.0 vs. 6.8 μg/m^3; summer: 9.0 vs. 10.1 μg/m^3) while median outdoor concentrations were slightly higher in Edmonton (winter: 14.0 vs. 7.7 μg/m^3; summer: 11 vs. 9.8 μg/m^3) (MacNeill et al., 2014). The median ambient UFP concentrations in winter (5.9 × 10^3 cm^{-3}) and summer (4.8 × 10^3 cm^{-3}) were considerably lower than outdoor concentrations in Windsor (winter: 13.4 × 10^3 cm^{-3}; two summers: 11.8 × 10^3 and 7.6 × 10^3 cm^{-3}). The median indoor concentrations (winter 2.5 × 10^3 cm^{-3}; summer 4.4 × 10^3 cm^{-3}) were also lower than in Windsor homes (winter: 6.9 × 10^3 cm^{-3}; two summers 5.8 × 10^3 cm^{-3} and 6.0 × 10^3 cm^{-3}).

The median AER in summer (0.31 h^{-1}) was about 50% higher than in winter (0.21 h^{-1}). This is opposite of the trend seen in Windsor, ON where the AER geometric means were 0.27 h^{-1} and 0.30 h^{-1} during 2 winter seasons and 0.14 h^{-1} during 2 summer seasons (Wheeler et al., 2011). The higher summer air exchange rates in Edmonton may be explained by the predominance of open windows compared to extensive use of air conditioning in Windsor. The lower median winter AER in Edmonton compared to Windsor may be explained by increased insulation and possibly reduced window opening due to the significantly colder winter

Table 1
Basic statistics – Half-hour UFP and FP, and daily $PM_{2.5}$ and AER measurements.

	Number of half-hours or days*	Min	p10	p25	Median	p75	p90	Max	Mean	Std dev
Winter (n = 50 homes)										
Indoor FP (μg/m^3)	14,984	0.0	1.1	2.7	5.0	10	24	2500	15	56
Outdoor[a] FP (μg/m^3)	15,478	0.0	2.0	5.5	14	34	58	220	24	28
Indoor UFP (×10^3/cm^3)	6565	0.1	0.7	1.3	2.5	6.2	21	440	9.7	25
Ambient[a] UFP (×10^3/cm^3)	1608	0.5	2.3	3.7	5.9	9.7	16	48	7.6	5.8
Indoor PM_1 (μg/m^3)	173*	0.3	0.3	0.8	2.2	6.1	14	66	5.2	8.1
Outdoor[a] PM_1 (μg/m^3)	177*	0.3	0.3	1.5	3.3	6.9	14	35	5.5	6.8
Indoor $PM_{1-2.5}$ (μg/m^3)	174*	0.25	0.27	0.27	0.29	1.6	3.2	12	1.3	1.8
Outdoor[a] $PM_{1-2.5}$ (μg/m^3)	179*	0.26	0.27	0.28	0.31	2.0	3.7	9.0	1.4	1.6
Indoor $PM_{2.5}$ (μg/m^3)	173*	0.54	0.57	1.5	3.1	8.5	17	74	6.5	9.2
Outdoor[a] $PM_{2.5}$ (μg/m^3)	175*	0.52	0.82	2.1	4.3	8.9	17	44	6.9	7.9
Indoor S (μg/m^3)	173*	0.01	0.03	0.05	0.08	0.15	0.25	0.54	0.11	0.10
Outdoor[a] S (ng/m^3)	164*	0.00	0.06	0.10	0.15	0.36	0.47	0.75	0.23	0.17
Air exchange (/h)	338*	0.04	0.13	0.16	0.21	0.29	0.54	1.30	0.27	0.19
Indoor temp (°C)	15,949	13.1	17.9	19.5	20.9	21.9	22.8	34.2	20.6	2.0
Indoor RH (%)	10,637	19.9	25.7	28.4	32.9	35.3	36.7	44.9	31.8	4.4
Ambient[b] temp (°C)	3050	−29.9	−15.4	−10.0	−5.0	0.3	4.0	15.7	−5.2	7.7
Ambient[b] RH (%)	3050	13	46	65	73	80	87	100	70.1	15.9
Summer (n = 50 homes)										
Indoor FP (μg/m^3)	14,946	0.0	3.4	5.6	9.0	17	44	1200	24	61
Outdoor[a] FP (μg/m^3)	15,418	0.0	4.0	6.8	11	21	48	1800	34	110
Indoor UFP (×10^3/cm^3)	8876	0.1	1.5	2.5	4.4	8.3	18	450	9.3	19
Ambient[a] UFP (×10^3/m^3)	2603	1.0	2.4	3.4	4.8	7.5	11	54	6.2	4.6
Indoor PM_1 (μg/m^3)	329*	0.27	1.1	2.4	4.4	8.6	18	110	8.3	13
Outdoor[a] PM_1 (μg/m^3)	319*	0.27	1.2	2.6	4.3	7.4	17	97	9.0	16
Indoor $PM_{1-2.5}$ (μg/m^3)	329*	0.24	0.3	0.3	1.0	2.6	5.2	26	2.2	3.2
Outdoor[a] $PM_{1-2.5}$ (μg/m^3)	321*	0.24	0.3	0.3	0.3	0.7	4.5	25	1.7	3.8
Indoor $PM_{2.5}$ (μg/m^3)	328*	0.54	1.6	3.2	5.9	11.0	23	140	11	15
Outdoor[a] $PM_{2.5}$ (μg/m^3)	318*	0.53	1.5	2.9	4.7	8.5	19	120	11	20
Indoor S (μg/m^3)	329*	0.00	0.07	0.11	0.18	0.27	0.39	0.74	0.21	0.14
Outdoor[a] S (ng/m^3)	269*	0.00	0.11	0.15	0.26	0.36	0.51	0.87	0.28	0.17
Air exchange (/h)	340*	0.04	0.11	0.16	0.31	0.57	1.01	2.60	0.46	0.45
Indoor temp (°C)	16,547	17.1	21.4	22.7	24.1	25.9	28.6	36.6	24.6	2.8
Indoor RH (%)	15,911	21.8	28.2	33.8	36.3	39.9	42.9	55.6	36.4	5.4
Ambient[b] temp (°C)	3054	−0.2	8.4	11	14.1	18.3	22	27.6	14.7	5.2
Ambient[b] RH (%)	3054	26	48	61	80	92	97	100	75.4	18.9

[a] Ambient UFP measurements are from the central site; outdoor FP, PM_1, $PM_{2.5}$, S measurements are from outside the homes.
[b] Ambient temperature and RH measurements from the Edmonton airport.

temperatures in Edmonton. Higher median AER in summer (0.44) compared to winter (0.30) has also been reported for Halifax (MacNeill et al., 2014), where window-opening was reported on 92% of days in summer. Median AER rates in both seasons were considerably lower compared to those measured in U.S. cities across various seasons: Raleigh/Chapel Hill, NC: 0.36–0.81 h^{-1} (Breen et al., 2010); Detroit, MI: 0.92 h^{-1} summer, 1.46 h^{-1} winter, Elizabeth, NJ: 0.63–1.07 h^{-1}, Houston, TX: 0.38–0.63 h^{-1}, Los Angeles, CA: 0.61–1.13 h^{-1} (Isaacs et al., 2013); Ann Arbor & Ypsilanti, MI, 0.35 h^{-1} (Batterman et al., 2007).

3.2. Temporal variability

In winter and summer, FPs were slightly elevated during the morning hours but the diurnal variation was not large (Fig. 1). Higher indoor UFPs were seen during dinner time hours (5–7 pm) during both summer and winter. Median ambient UFPs were somewhat higher during the afternoon and evening hours in the winter. There was little diurnal variation in ambient UFPs in summer.

3.3. F_{inf} estimates

Statistics on the FP and UFP F_{inf} estimates are provided in Table 2; plots of the daily F_{inf} by home are provided in Fig. 2. Considerable variability was seen between homes and between days within some homes.

For both FPs and UFPs, the median F_{inf} estimates are significantly higher in summer than in winter, likely due to the frequent window openings in summer because the summer temperatures in Edmonton are moderate. This seasonal difference has been reported in a number of studies where air conditioning is not widely used (MacNeill et al., 2014; Allen et al., 2003; Hänninen et al., 2005; Allen et al., 2012). In comparison, there were few seasonal differences in the median UFP or FP F_{inf} in a study of 94 homes in Windsor. This was attributed to the use of air conditioning (and fewer window openings) in summer (Kearney et al., 2011; MacNeill et al., 2012).

For FPs, the median daily summertime F_{inf} (0.69; range 0.11–1.05) found in this study is also considerably higher than that found in Windsor (0.35, 0.36) (MacNeill et al., 2012) but slightly lower than Halifax (0.80), where window opening was also very prevalent (MacNeill et al., 2014). The median wintertime FP F_{inf} in Edmonton (0.28; range 0.06–1.02) is similar to that found in Windsor in 2 winter seasons (0.26, 0.34) (MacNeill et al., 2012) but lower than in Halifax (0.55) (MacNeill et al., 2014) which has more moderate winter temperatures than Edmonton. The mean FP F_{inf} estimates in this study (0.34 (winter), 0.68 (summer)) were slightly higher than the mean $PM_{2.5}$ F_{inf} estimates reported by Barn et al. (2008) in Prince George of 0.27 (winter, $n = 21$ homes) and 0.61 (summer,

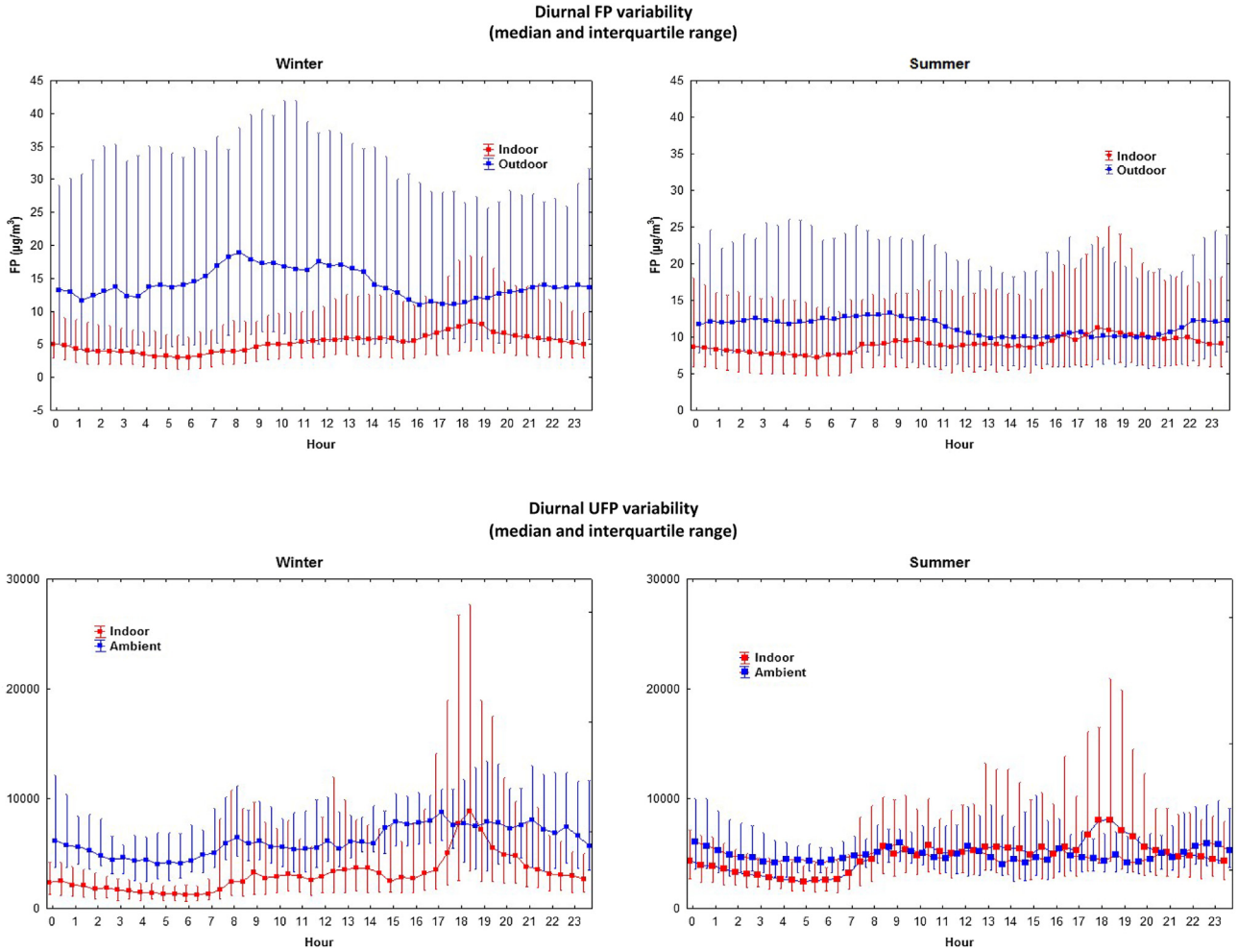

Fig. 1. Diurnal FP and UFP variability by season.

Table 2
Estimates of daily F_{inf} and ambient/non-ambient components.

	Number of days	Min	p10	p25	Median	p75	p90	Max	Mean	Std Dev
Winter (n = 50 homes)										
Fine particles (based on DustTrak)										
F_{inf}	329	0.06	0.14	0.19	0.28	0.44	0.66	1.02	0.34	0.21
Ambient (μg/m^3)	329	0.38	1.3	2.3	4.2	6.9	11	31	5.7	5.2
Non-ambient (μg/m^3)	329	0.00	0.00	0.07	0.98	7.09	27	243	9.3	24
%Ambient	329	0.47	12	38	79	99	100	100	66	34
%Nonambient	329	0	0	1	21	62	88	100	34	34
Indoor FP (μg/m^3)	329	0.8	2.3	3.9	6.8	16	35	245	15	24
Outdoor FP (μg/m^3)	329	0.4	2.7	6.4	16	33	53	112	24	24
Fine particles (based on S)										
F_{inf}	162	0.16	0.25	0.33	0.45	0.58	0.71	2.1	0.49	0.25
Ambient (μg/m^3)	162	0.06	0.27	0.28	0.94	2.0	3.9	17	1.8	2.6
Non-ambient (μg/m^3)	162	0.00	0.00	0.00	0.58	3.32	10	66	3.4	7.6
%Ambient	162	1	8	27	70	100	100	100	62	38
%Nonambient	162	0	0	0	30	73	92	99	38	38
Indoor S (μg/m^3)	162	0.01	0.03	0.04	0.07	0.15	0.23	0.52	0.11	0.09
Outdoor S (μg/m^3)	162	0.04	0.07	0.10	0.15	0.35	0.47	0.75	0.23	0.17
Ultrafine particles										
F_{inf}	152	0.02	0.08	0.13	0.19	0.28	0.38	0.78	0.22	0.13
Ambient (×10^3/cm^3)	152	0.17	0.39	0.7	1.4	2.1	2.9	4.9	1.6	1.0
Non-ambient (×10^3/cm^3)	152	0.00	0.6	1.7	5.0	11	17	64	8.0	9.6
%Ambient	152	0	4	9	22	50	82	100	32	29
%Nonambient	152	0	18	50	78	91	96	100	68	29
Indoor UFP (×10^3/cm^3)	152	0.48	2.20	3.4	6.8	12.9	18.5	64.5	9.6	9.5
Central site UFP (x10^3/cm^3)	152	2.22	2.5	5.8	7.3	9	13	15	7.4	3.2
Summer (n = 50 homes)										
Fine particles (based on DustTrak)										
F_{inf}	323	0.11	0.42	0.53	0.69	0.83	0.96	1.1	0.68	0.20
Ambient (μg/m^3)	323	0.28	3.43	5.30	7.81	12.8	37.0	331	19.5	41.3
Non-ambient (μg/m^3)	323	0.00	0.00	0.18	1.02	3.77	12	86	4.4	9.4
%Ambient	323	7	39	70	91	98	100	100	80	24
%Nonambient	323	0	0	2	9	30	61	93	20	24
Indoor FP (μg/m^3)	323	0.9	4.8	7.0	10	21	47	373	24	46
Outdoor FP (μg/m^3)	323	1.0	5.1	7.7	12	21	63	446	34	75
Fine particles (based on S)										
F_{inf}	265	0.00	0.40	0.57	0.79	0.87	0.97	2.0	0.73	0.24
Ambient (μg/m^3)	265	0.01	0.29	1.25	2.78	5.2	12	70	6.0	11.0
Non-ambient (μg/m^3)	265	0.00	0.00	0.00	1.01	2.42	5	21	2.1	3.4
%Ambient	265	2	20	47	79	100	100	100	70	31
%Nonambient	265	0	0	0	21	53	80	98	30	31
Indoor S (μg/m^3)	265	0.00	0.07	0.11	0.18	0.27	0.39	0.71	0.21	0.14
Outdoor S (μg/m^3)	265	0.04	0.12	0.15	0.26	0.36	0.51	0.87	0.29	0.17
Ultrafine particles										
F_{inf}	239	0.05	0.20	0.35	0.51	0.68	0.79	1.09	0.51	0.22
Ambient (×10^3/cm^3)	239	0.30	1.33	1.9	2.7	3.8	5.0	11.0	3.0	1.6
Non-ambient (×10^3/cm^3)	239	0.00	0.6	1.7	4.1	8	14	30	5.9	5.5
%Ambient	239	4	13	21	37	65	86	100	44	27
%Nonambient	239	0	14	35	63	79	87	96	56	27
Indoor UFP (×10^3/cm^3)	239	1.15	3.46	4.8	7.5	11	17	33	8.9	5.6
Central site UFP (×10^3/cm^3)	239	2.44	3.1	4.7	5.6	7.6	9.3	15	6.2	2.4

$n = 17$). The mean summertime F_{inf} based on sulfur in PM_1 in this study (0.73) is considerably higher than the mean sulfur (in $PM_{2.5}$) F_{inf} reported for 46 homes in Toronto, sampled from July–November (0.52) (Clark et al., 2010). Median F_{inf} estimates for $PM_{2.5}$ in U.S. cities have typically ranged from 0.4 to 0.8 (Allen et al., 2012; Chen and Zhao, 2011; Landis et al., 2001; Liu et al., 2003; Wallace et al., 2006).

For UFPs, the median winter F_{inf} estimate in this study (0.19) is very similar to the median winter F_{inf} estimate in the Windsor study (0.21) (Kearney et al., 2011). The summer median, however, was considerably higher in Edmonton (0.51) compared to the two summer medians in Windsor (0.16 (summer 2005) and 0.26 (summer 2006)), presumably due to the more frequent use of open windows and less frequent use of air-conditioning in Edmonton. The median summertime daily UFP F_{inf} (0.51; range 0.05–1.09) in this study is slightly higher than the F_{inf} estimates ranging from 0.11 to 0.47, reported for 7 northern California homes (particles > 6 nm) (Bhangar et al., 2010). The median wintertime UFP F_{inf} in this study (0.19; range 0.02–0.78) was lower than 6 of the 7 California homes but greater than the 1 home that had the lowest F_{inf} (0.11) (attributed to the use of air filtration device on the furnace). The use of central site data in the calculation of F_{inf} is a limitation of this study, given that considerable spatial variability in outdoor UFPs has been reported in many studies (higher levels in traffic environments) (Boogaard, H. et al., 2011, Mejia, J.F., 2008,Wang et al., 2012). However, moderate correlations and coefficients of divergence of UFPs have been reported in a number of studies of residential sites (Kearney et al., 2011; Cyrys et al., 2008; Puustinen et al., 2007, Buzorius et al., 1999) particularly for the larger ultrafines (suggesting that, in these studies, UFP levels at residential sites were affected by regional factors including regional sources (e.g. photochemical sources) and meteorology. The robustness of the UFP F_{inf} estimates in this study will depend on the degree of influence of background vs. local UFP sources on the central and residential sites.

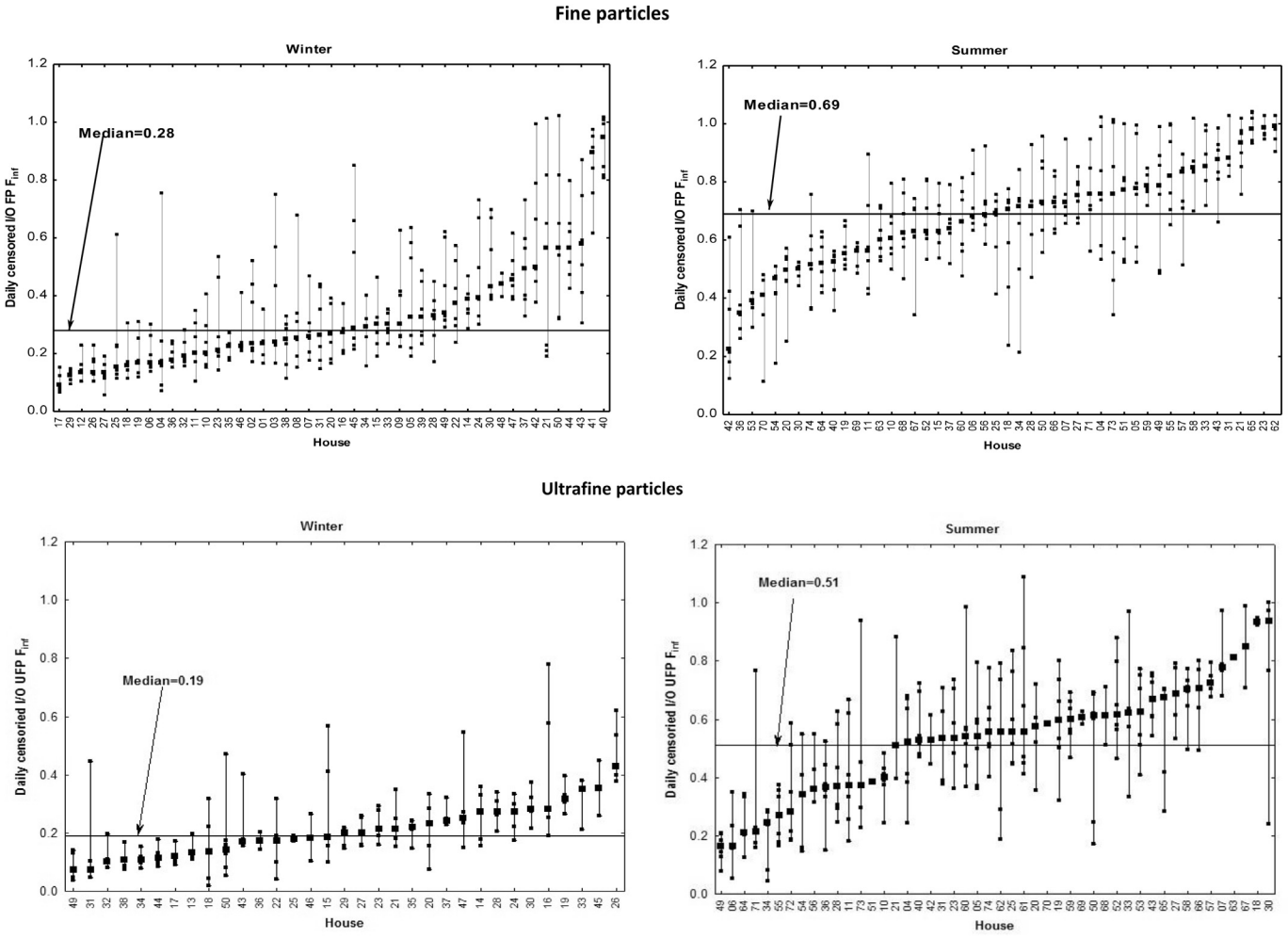

Fig. 2. Daily FP and UFP F_{inf} estimates by home (with median of daily values).

The median F_{inf} estimates are consistently higher for FPs than for UFPs in this study, due to higher penetration efficiencies and lower deposition rates, as has been observed in Windsor (MacNeill et al., 2012), and by others (Long et al., 2001; Sarnat et al., 2006; Hussein et al., 2004).

The median FP F_{inf} estimates based on the DustTrak data were lower than the median sulfur-based F_{inf} estimates particularly in the winter. The median difference between the DustTrak and sulfur-based paired F_{inf} estimates in winter was 0.11 ($n = 152$) and in summer was 0.04 ($n = 246$). As well, the DustTrak and sulfur-based F_{inf} estimates were poorly correlated (Spearman $r = 0.37$ and 0.25 in winter and summer, respectively). The poor correlation, also reported in a recent study in Halifax (MacNeill et al., 2014) may be due to variability in the volatile components of the aerosol that is reflected in the DustTrak measurements but is not reflected in the S measurements. The difference in the F_{inf} estimates using DustTrak and sulfur data may also be explained by the loss of volatile components from the FP aerosol as it enters indoors. This would lead to lower FP as measured by the DustTrak and HCl; however this would not affect the sulfur measurement as S is not a volatile component of FP. Hence the sulfur I/O ratio may not be representative of the infiltration of an outdoor aerosol that has significant amount of semi-volatile components (Allen et al., 2012). Lower infiltration can result when semi-volatile components of PM partition to the gas phase when entering an indoor environment (a process dependent on temperature, humidity and concentrations of the gas-phase components) (Lunden et al., 2003). Sangiorgi et al. (2013) has reported lower F_{inf} estimates for PM components with more semi-volatiles (e.g. PAHs, NO_3^-, NH_4^+) compared with non-volatile ammonium sulfate. Sarnat et al. (2006) also reported lower F_{inf} estimates (median 0.18) for nitrate (NO_3^-) (median 0.18), a volatile PM component, compared with non-volatile black carbon (median 0.84) and $PM_{2.5}$ (median 0.48) in a study of 17 houses in Los Angeles area. It was hypothesized that the lower NO_3^- F_{inf} estimates were due to volatilization of NH_4NO_3 to nitric acid (HNO_3) and ammonium (NH_3), after infiltration indoors.

There was considerable within-house variability for both FPs and UFPs (slightly higher for UFPs) (Table 3), indicating infiltration is related to variables that are changing day to day such as meteorology or participant behavior (e.g. window opening), as much as by factors that vary between homes, such as building characteristics. Similar findings were reported by MacNeill et al. (2012) for homes in Windsor and Halifax (MacNeill et al., 2014).

The variability in F_{inf} between and within residences is one source of heterogeneity in personal exposure to ambient PM. This heterogeneity will introduce measurement error in epidemiological studies that use central site monitoring measurements as the exposure metric in health-exposure models (Baxter et al., 2013; Allen et al., 2012). For UFPs, the seasonal-averaged F_{inf} (the average of the daily F_{inf} estimates) ranged from 0.08 to 0.47 across homes in winter (median = 0.21, $n = 33$ houses) and from 0.16 to 0.94 in summer (median = 0.57, $n = 48$ houses). For 16 homes that

Table 3
Between- and within-subject variance components.

Parameter	Between-subject variance (δ_{BS}) (*% of total*)	Within-subject variance (δ_{WS}) (*% of total*)
F_{inf}		
FP winter	0.028	0.018
	(60%)	*(40%)*
FP summer	0.023	0.017
	(59%)	*(41%)*
UFP winter	0.006	0.010
	(39%)	*(61%)*
UFP summer	0.024	0.027
	(47%)	*(53%)*
Non-ambient component		
FP winter	249.8	313.3
	(44%)	*(56%)*
FP summer	18.3	71.7
	(20%)	*(80%)*
UFP winter	5.0E + 07	4.3E + 07
	(54%)	*(46%)*
UFP summer	1.3E + 07	1.9E + 07
	(41%)	*(59%)*

had both summer and winter F_{inf} estimates, the annual F_{inf} estimates (calculated as the average of the F_{inf} from both seasons) ranged from 0.12 to 0.54 (median = 0.39, n = 16 houses). For FPs, seasonal-averaged F_{inf} ranged from 0.10 to 0.92 in winter (median = 0.30, n = 49) and 0.31 to 0.99 in summer (median = 0.68, n = 48 houses). Information on the range of F_{inf} across homes may provide useful information for future epidemiological studies in order to better characterize ambient PM exposures (Baxter et al., 2013).

3.4. F_{inf} models

The predictive models for F_{inf} are provided in Table 4. The FP winter model explained 57% of the between-subject variance and 12% of within-subject variance. In contrast, the FP summer model explained virtually no between-subject variance and only 9% of the within-subject variance. The summer UFP model explained more variance than the winter model particularly for the between-subject variance (27% vs. 6%).

Home age (strata) was only significantly associated with F_{inf} for FPs in the winter. In that model, older homes were associated with the highest F_{inf}, and the parameter estimates were progressively smaller across the home age strata (lowest for the newest homes). This effect was not seen in the summer for either pollutant, possibly because of the high frequency of open windows in Edmonton (88% of summer days had open windows), which would have an overwhelming effect on air exchange regardless of home age. In summer, the number of window open-hours was associated with an increase in UFP F_{inf}, a finding also reported by MacNeill et al. (2012). In that study of homes in Windsor, ON, home age was significantly associated with FP F_{inf} in one winter season in Windsor and open windows (y/n) and the number of hours that windows were open were associated with an increase in FP and UFP F_{inf} in two summer seasons (MacNeill et al., 2012). MacNeill et al., 2014 also found an association with winter-time F_{inf} and older home age in Halifax. Hystad et al. (2009) did not find a significant association between home age and $PM_{2.5}$ F_{inf} in the heating (p = 0.09) or non-heating season (p = 0.55) in a study of Victoria and Seattle homes.

In all models, wind speed was positively associated with F_{inf}; higher wind speed leads to higher air exchange because of the increased pressure difference between the windward and leeward side of the house. An association with wind speed was also reported by MacNeill et al. (2012) for FP F_{inf} winter models in Windsor. Hystad et al. (2009) did not find a significant relationship between $PM_{2.5}$ F_{inf} and wind speed in either the heating or non-heating season in Victoria and Seattle homes.

The presence of an air exchanger and chimney in the home was associated with increased FP F_{inf} in winter, presumably due to increased air exchange. Having an electrostatic precipitator (ESP) in the home was associated with increased F_{inf}, which is not expected. Only three winter homes had ESPs so this finding may not be robust. Having the furnace fan running continuously was associated with reduced UFP F_{inf} in the winter, presumably due to increased particle deposition in the duct work or removal due to

Table 4
Predictors of FP and UFP daily F_{inf} by season.

Model effects	Parameter estimate	Standard error	p-value	% Variance explained by predictive model	
				Between-subject	Within-subject
FP – winter *(n = 328)*					
Intercept	0.501	0.070	<0.0001	57%	12%
Strata			0.0076		
Built > 2004	−0.237	0.067	0.0011		
Built 1981–2004	−0.234	0.064	0.0008		
Built 1961–1980	−0.129	0.059	0.0363		
Built 1946–1960	−0.090	0.058	0.1258		
Built < 1946	0.000				
Air exchanger in home (yes)	0.347	0.137	0.0152		
Chimney in home (yes)	0.267	0.055	<0.0001		
Electrostatic precipitator in home (yes)	0.287	0.099	0.0061		
Mean wind speed (km/h)	0.005	0.001	0.0002		
Mean absolute I–O temperature difference	−0.009	0.002	<0.0001		
FP – summer *(n = 321)*					
Intercept	0.562	0.033	<0.0001	0%	9%
Mean wind speed (km/h)	0.011	0.002	<0.0001		
UFP – winter *(n = 152)*					
Intercept	0.188	0.026	<0.0001	6%	5%
Furnace fan runs continuously (yes)	−0.067	0.034	0.0506		
Mean wind speed (km/h)	0.004	0.001	0.0142		
UFP – summer *(n = 237)*					
Intercept	0.340	0.046	0.0000	27%	7%
Portable air cleaner (# hours)	−0.025	0.012	0.0382		
Number open window-hours	0.001	0.000	0.0002		
Mean wind speed (km/h)	0.013	0.004	0.0005		

filtration. The use of a portable air cleaner was associated with reduced UFP F_{inf} in summer; this is another factor that would increase deposition resulting in reduced F_{inf}. Allen et al. (2012) also reported lower F_{inf} associated with air cleaner use while MacNeill et al. (2014) reported lower winter F_{inf} associated with the presence of an air exchanger and premium furnace filter. Other variables not measured in this study, such as heating system run time and flow rate, and sizes of window openings (affected by window type) are likely to be factors contributing to the unexplained variability of F_{inf}.

3.5. Ambient and non-ambient components

Summary statistics on the daily ambient and non-ambient components for UFPs and FPs (based on DustTraks and sulfur F_{inf} estimates) and plots of the daily UFP and FP (based on DustTrak F_{inf} estimates) components by house are presented in Table 2 and Fig. 3, respectively. For UFPs, generally the non-ambient sources contributed more to indoor levels; the median percent contribution to total indoor levels was 78% in winter and 63% in summer. For FPs, ambient sources were generally more important, with a median contribution of 70% in winter and 91% in summer. Higher ambient components and lower non-ambient components are seen in summer compared to winter for both UFPs and FPs, despite lower ambient/outdoor concentrations. This is due to the considerably larger infiltration factors in the summer for both pollutants.

These findings are similar to Windsor (Kearney et al., 2011; MacNeill et al., 2012) where median estimates of the percentage of indoor-generated contribution to indoor UFPs ranged from 58 to 69% across 3 sampling seasons (Kearney et al., 2011). For FPs, the ambient sources were more important with median percent ambient contributions between 58 and 68% in Windsor (MacNeill et al., 2012). In Halifax, the median percent ambient contribution was 59% in winter and 84% in summer (MacNeill et al., 2014). Other studies in the U.S. have also found that, on average, a majority of indoor $PM_{2.5}$ has originated from ambient sources (Allen et al., 2003 (Seattle); Wallace et al., 2006 (Research Triangle Park, NC); Meng et al., 2005 (Los Angeles County, CA and Elizabeth, NJ) however that was not the case in approximately 100 Houston, TX homes where the median contribution of ambient sources to indoor PM was much lower (33%) and was attributed to the higher prevalence of closed windows due to air conditioning use (Meng et al., 2005).

When the ambient and non-ambient components were calculated using the sulfur F_{inf} estimates, 31% of the summer ambient component estimates and 38% of the winter ambient component estimates were greater than the actual measured indoor concentration (and hence the calculated non-ambient component was negative). This could be caused by an elevated indoor sulfur measurement or an indoor PM_1 mass measurement that is lower than expected, perhaps due to loss of volatiles. The presence of indoor sulfur sources was tested by regression of the indoor vs. outdoor sulfur across days for each home. The intercepts were generally very low, providing evidence of few or no indoor sulfur sources. As discussed before, an alternative explanation is that the infiltrated PM_1 loses mass when it infiltrates

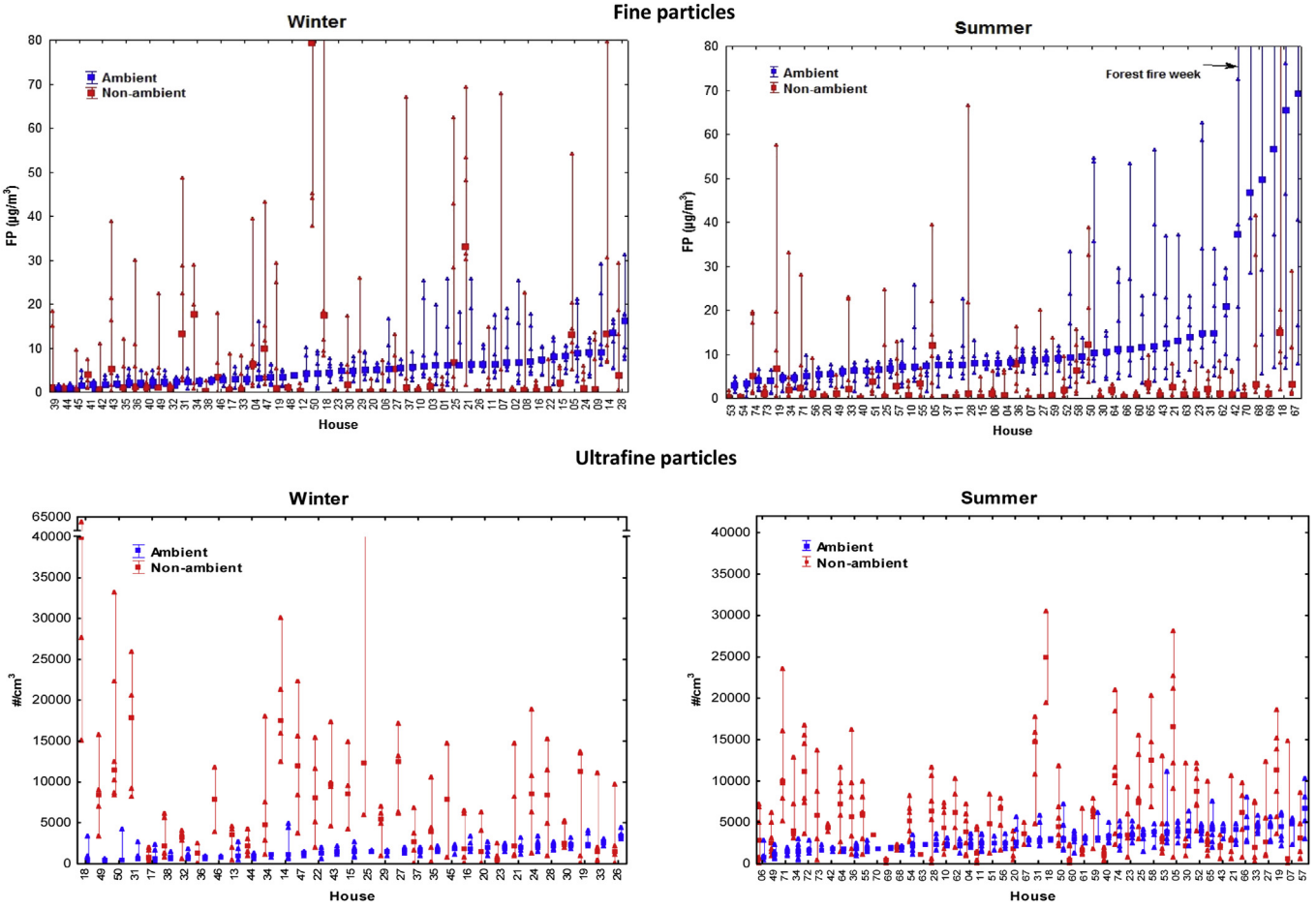

Fig. 3. Non-ambient components by house.

Table 5
Predictors of the non-ambient component of indoor FP and UFP by season.

Model effects	Parameter estimate	Standard error	*p*-value	% Variance explained by predictive model	
				Between-subject	Within-subject
FP − winter *(n = 328)*					
Intercept	−2.3	2.9	0.42	72%	2%
Number of people in home	2.1	0.7	0.003		
Smoking indoors (yes)	57.0	7.9	<0.0001		
Power tool use (yes)	12.0	5.7	0.04		
Stove frying (yes)	5.7	2.3	0.01		
Gas fireplace supplemental heat (yes)	0.028	0.013	0.04		
FP-summer *(n = 321)*					
Intercept	−11.6	4.7	0.015	44%	7%
Smoking indoors (yes)	13.4	4.6	0.006		
Power tool use (yes)	8.3	3.2	0.009		
Stove frying, grilling or sauteing (yes)	2.9	1.1	0.007		
Oven use (yes)	2.4	1.2	0.043		
Number open window-hours	−0.030	0.014	0.028		
Equipment idling in garage (yes)	13.1	5.3	0.013		
Mean indoor relative humidity (%)	0.4	0.1	0.002		
UFP − winter *(n = 152)*					
Intercept	−2.3E + 03	1.7E + 03	0.18	58%	50%
Number cigarettes smoked in home	2.0E + 03	3.9E + 02	<0.0001		
Number of people in home	2.3E + 03	4.0E + 02	<0.0001		
Use of gas stove (yes)	4.7E + 03	2.2E + 03	0.039		
Number of times stove used for grilling	1.3E + 04	2.2E + 03	<0.0001		
Number of times stove used for frying	2.0E + 03	7.7E + 02	0.010		
Oven used for baking (yes)	5.0E + 03	1.0E + 03	<0.0001		
Number of times toaster used	1.6E + 03	6.0E + 02	0.010		
Number of times car moved in/out of garage	−6.9E + 02	2.3E + 02	0.004		
UFP − summer *(n = 228)*					
Intercept	−7.6E + 03	3.3E + 03	0.024	36%	21%
Oven used for baking (yes)	2.7E + 03	7.4E + 02	0.0004		
Food burned (yes)	3.8E + 03	1.3E + 03	0.003		
Number of times any cooking	8.7E + 02	2.0E + 02	<0.0001		
Mean indoor relative humidity (%)	3.0E + 02	8.9E + 01	0.001		

indoors, due to loss of volatile components and the sulfur I/O ratio does not capture this loss of volatile components. This provides further evidence that the lack of agreement between the DustTrak and sulfur-based F_{inf} estimates may be due to semi-volatile components in the outdoor PM aerosol which are lost when the aerosol moves indoors.

As with $F_{inf,}$ for FPs (winter and summer) and UFPs (summer), the within-subject variance estimates were greater than the between-subject variance estimates, ranging between 64% and 87%. For UFPs in winter, the two variance estimates were similar (50% each). This is an indication that the non-ambient component is affected by factors that vary over time, such as particle generating activities within the home. This finding is similar to that reported for homes in Windsor (MacNeill et al., 2012) and Halifax (MacNeill et al., 2014).

3.6. Non-ambient component models

The predictive models for the non-ambient component are provided in Table 5. The percentage of variance explained by the models ranged from 36% to 72% for between-subject variance and 2–50% for within-subject variance (Table 3), indicating the models were better at explaining the between-subject variance.

Smoking indoors was associated with increased non-ambient components for both FPs (summer and winter) and UFPs (winter). While the study intended to recruit only homes with non-smoking residents, smoking was reported in 3 homes in winter and 1 home in summer. Various forms of cooking were strongly associated with both FPs and UFPs in both seasons as has been reported in other studies (MacNeill et al., 2012; 2014). For FPs, the cooking predictors were stove frying in winter and stove frying/grilling/sautéing in summer. For UFPs, a number of cooking variables were significantly associated with the non-ambient component in both seasons. Indoor relative humidity (RH) was significantly associated with a higher non-ambient component in both the FP and UFP summer models but not in winter. Open windows were negatively associated with the non-ambient component of indoor FP concentrations in the summer, similar to results from Halifax (MacNeill et al., 2014).

4. Conclusions

This study provides information about infiltration of FPs and UFPs into homes in Edmonton, AB. Median winter F_{inf} estimates (FP 0.28, UFP 0.19) were relatively low compared to other locations, presumably due to tighter house construction due to cold winter temperatures in Edmonton. Summer F_{inf} estimates were quite high (median FP 0.69, UFP 0.51), presumably due to frequent opening of windows during summer, when temperatures are quite moderate. A main finding is the considerable variability in FP and UFP F_{inf} seen both within and between homes in Edmonton, AB. This variability may cause substantial exposure misclassification in epidemiological studies using only ambient measurements. Epidemiological studies may be improved with incorporation of information regarding the regional and temporal variability of F_{inf}, or predictors of F_{inf}, as reported in this study. Median F_{inf} estimates were slightly higher for FPs than UFPs. On average, the majority of indoor FPs originated outdoors whereas the majority of indoor UFPs originated indoors. In summer, more of the indoor FPs and UFPs were from ambient origin (compared to winter) due to the higher infiltration factors. Estimates of F_{inf} using indoor/outdoor sulfur ratios were problematic; suggesting that volatilization of indoor aerosols

affects this method by reducing indoor PM mass. F_{inf} estimates obtained from the optical DustTrak monitors appeared to be reasonable estimates of the gravimetric F_{inf}. These findings may be helpful in developing risk management strategies to target the most significant sources of exposure.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2014.05.020.

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