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QUANTIFYING THE CONTRIBUTION OF AMBIENT AND INDOOR-GENERATED FINE PARTICLES TO INDOOR AIR IN RESIDENTIAL ENVIRONMENTS

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Abstract

Indoor fine particles (FPs) are a combination of ambient particles that have infiltrated indoors, and particles that have been generated indoors from activities such as cooking. The objective of this paper is to estimate the infiltration factor (F_{inf}) and the ambient/non-ambient components of indoor FPs. To do this, continuous measurements were collected indoors and outdoors for seven consecutive days in 50 non-smoking homes in Halifax, Nova Scotia in both summer and winter using DustTrak (TSI Inc) photometers. Additionally, indoor and outdoor gravimetric measurements were made for each 24-h period in each home, using Harvard Impactors (HI). A computerized algorithm was developed to remove (censor) peaks due to indoor sources. The censored indoor/outdoor ratio was then used to estimate daily F_{infs}, and to determine the ambient and non-ambient components of total indoor concentrations. F_{inf} estimates in Halifax (daily summer median=0.80; daily winter median=0.55) were higher than have been reported in other parts of Canada. In both winter and summer, the majority of FP was of ambient origin (daily winter median=59%; daily summer median=84%). Predictors of the non-ambient component included various cooking variables, combustion sources, relative

humidity and factors influencing ventilation. This work highlights the fact that regional factors can influence the contribution of ambient particles to indoor residential concentrations.

Keywords: Infiltration factor (F_{inf}), fine particulate matter, indoor air quality, ambient component, non-ambient component

Practical Implications

Ambient and non-ambient particles have different risk management approaches, composition and likely toxicity. Therefore, a better understanding of their contribution to the indoor environment is important to manage the health risks associated with fine particles effectively. As well, a better understanding of the factors F_{inf} can help improve exposure assessment and contribute to reduced exposure misclassification in epidemiologic studies.

1. Introduction

In North America, people spend approximately 90% of their time indoors (Klepeis et al., 2001; Leech et al., 2002). Here they are exposed to outdoor-generated particles that have infiltrated indoors, and particles that have been generated indoors from activities such as cooking, cleaning and other combustion sources. As a result, the majority of exposure to fine particulate matter (FP) of ambient origin often occurs indoors. Since ambient and non-ambient particles have different risk management approaches, composition and likely toxicity (Wilson et al., 2006), it is important to distinguish the contribution of ambient and non-ambient particles to indoor air.

The fraction of outdoor particles that penetrate the building envelope and remain suspended is termed the infiltration factor (F_{inf}). F_{inf} is a function of the penetration efficiency (P), the air exchange rate (a) and the decay rate (k) (Equation 1).

$$F_{inf} = (Pa)/ (a+k) \tag{1}$$

F_{inf} has been shown to vary both temporally and spatially (Allen et al., 2003, 2012; Barn et al., 2008; Briggs et al., 2000; MacNeill et al., 2012; Meng et al., 2005; Nazaroff, 2004; Wallace et al., 2003; Wallace and Williams, 2005; Zeger et al., 2000) and to be influenced by factors such as climate, housing stock, air conditioning use and window opening (Allen et al., 2012, Clark et al., 2010., Hystad et al., 2009; MacNeill et al., 2012, Wallace et al., 2002). As a result, there can be significant variation in the contribution of ambient particles to indoor residential concentrations (Hodas et al., 2012; Meng et al., 2005).

This variation has been identified as a potential source of exposure misclassification in epidemiologic studies that use concentrations measured from a central-site monitor as a surrogate for an individuals' exposure to particulate matter of ambient origin (Sarnat et al, 2007). This can result in uncertainty surrounding the true health effects associated with particulate matter. As a result, F_{inf} and the factors that influence F_{inf} should be taken into account in epidemiologic studies in order to reduce exposure misclassification and minimize the uncertainly surrounding exposure-response estimates.

Indoor-generated (non-ambient) particles may also be of concern. It is well recognized that indoor sources such as smoking, cooking and other activities can increase an individual's exposure to particulate matter (Wallace, 1997; Özkaynak et al., 1996; MacNeill et al., 2012).

Studies have found associations between indoors sources such as environmental tobacco smoke and indoor wood combustion with adverse health outcomes (Sherman, 1991, Kocbach Bølling et al., 2009). As well, a monograph on high-temperature frying also found "*limited evidence* in humans for the carcinogenicity of emissions from high-temperature frying" but concluded that these emissions are "*probably carcinogenic to humans"* (IARC, 2010). As a result, many organizations provide advice on how to reduce indoor sources of particulate matter in the home (Health Canada, 2012; US EPA, 2008). A better understanding of the factors that influence non-ambient particle concentrations can help refine advice to homeowners on how to reduce their exposure to FP.

The objective of this paper is to estimate the infiltration factor (F_{inf}) and the ambient/non-ambient components of indoor FPs, as well as to develop predictive models for F_{inf} and the non-ambient component of indoor FPs using questionnaire and meteorological variables for Halifax homes. Similar work has been completed for other parts of Canada, where climate and housing stock may differ (MacNeill et al., 2012; Kearney et al., *submitted*; Clark et al., 2010; Hystad et al., 2009; Pellizzari et al., 1998). This paper builds on previous work by providing a clearer understanding of how regional factors influence F_{inf} and the components of indoor PM.

2. Methods

2.1 Study Design

In 2009, Health Canada, in collaboration with Dalhousie University, conducted a residential indoor air quality study in the Halifax Regional Municipality, Nova Scotia, Canada. A range of air pollutants typically found within residences were measured indoors and outdoors for seven consecutive 24-hour periods in 50 homes in both the winter (January to April) and the summer (June to September), with 42 homes participating in both seasons. Indoor sampling equipment was typically located in either the family or living room where participants spent the majority of their time. Where possible the equipment was located away from combustion sources, heating ducts or televisions. Outdoor sampling equipment was located in the backyard away from sources such as barbeques and motor vehicles. The parameters measured included nitrogen dioxide, ozone, sulphur dioxide, aldehydes, naphthalene, volatile organic compounds, particulate matter (gravimetric and continuous), air exchange rates, and continuous measures of carbon monoxide, carbon dioxide, temperature, and relative humidity. Approval was obtained from Health Canada's and Dalhousie University's Research Ethics Boards to conduct this study. All personal information is protected in accordance with the *Access to Information Act* (Canada, 2011a) and the *Privacy Act* (Canada, 2011b).

2.2 Participant Recruitment

Homes were selected using stratified sampling based on home age and type of cooking fuel used. In each season, a target of 10 homes was identified in each of the construction year categories (1945 and before, 1946-1960, 1961-1980, 1981-2000, and 2001-2008) as well as a target of 10 homes from any age category with a gas or propane stove, for a total of 50 homes. The recruitment region was carefully delineated using GIS methods, and aligned with the 2006 Statistics Canada census boundary files. Participants were contacted using random digit dialing methods using a telephone listing from the Greater Halifax Regional Municipality. Recruitment and enrolment were further controlled using a formalized enrolment protocol. Dwellings where smoking was permitted and multiple unit rental accommodations were excluded. Among the eligible homes, preference was given to households that were spatially distributed across the Halifax Regional Municipality. The location of participant homes in summer and winter can be found in Figure S1.

2.3 Air Exposure Measurements

Continuous measurements (1-min integration) of FPs were collected simultaneously both indoors and outdoors at each home using TSI DustTraks (Model 8520, TSI, St. Paul, MN). Air was sampled at a flow rate of 1.7 Lpm. DustTrak measurements were adjusted using the zero-check values reported by the instrument. The outdoor continuous monitors and pumps were housed in a waterproof enclosure, and heated to 10ºC during the winter sampling session. All monitors were downloaded and cleaned at the start of each sampling week. Since comparisons with concurrent gravimetric $PM_{2.5}$ concentration show that the DustTrak overestimates gravimetric concentrations (Wallace et al., 2011), the DustTrak values reported here should not be interpreted as gravimetric equivalent $PM_{2.5}$ concentrations.

In addition to continuous measurements, 24-hour gravimetric $PM_{2.5}$ concentrations were also collected using MS&T Area samplers (37 mm diameter, 2 µm pore size, ring supported PTFE filters, Air Diagnostics and Engineering Inc. Harrison, ME) operated at a flow rate of 10 L/min (10 LPM model, BGI Inc., Waltham MA, USA). An end flow rate of ± 20% of the initial flow was deemed acceptable. A conditioned and pre-weighed Teflon filter (37mm, 2µm pore size, Pall Inc, Port Washington, NY, USA) was used to capture particles smaller than 2.5 µm. Gravimetric analyses were conducted using the method outlined in the Quality Assurance Guidance Document 2.12 by the US EPA (U.S. EPA, 1998). All samples having $PM_{2.5}$ mass greater than 5µg were also analyzed for sulphur (S). Elemental analyses were conducted using

XRF following protocols consistent with EPA method IO-3.3 (EPA 625/R-96/010). Blank samples were deployed randomly at a rate of 10% across all sampling locations.

Temperature (^{o}C) and relative humidity (%) were measured continuously (1-min integration) indoors for the duration of the 7-day sampling period, in both seasons, using YES-206LH monitors (Yes Environment Technologies Inc., Delta, BC, Canada). Outdoor meteorological data (including temperature (^{o}C), relative humidity (%), atmospheric pressure (kpa), and wind speed) from the Halifax Stanfield International Airport were downloaded from Environment Canada's National Climate Data and Information Archive (http://climate.weatheroffice.gc.ca/climateData/ canada_e.html).

Air exchange rates were measured inside the home by the perflurocarbon tracer (PFT) technique (Dietz et al, 1986). Four PFT emitters were placed throughout the same floor on which the air monitoring equipment was located, preferably on the edge of a door frame, picture frame or shelf. Technicians were instructed to avoid placing PFTs near windows or doorways leading outside. A daily capillary adsorption tube (CAT) was used as the receptor for the PFT tracer gas. For each home, the air exchange rate, expressed as air changes per hour (ACH), was calculated by dividing the air exchange rate by the technician measured house volume.

2.4 Assessment of Housing Parameters and Daily Activities

Information on housing characteristics such as the home age and size, presence of air conditioning and type of ventilation was collected via interviewer-administered questionnaire at the beginning of the study and verified in the second sampling session. In addition, each participant was asked to complete a daily self-administered questionnaire to record activities that occurred during sampling such as cooking, cleaning, and candle use.

2.5 Data Quality

Co-located sampling sessions were conducted to determine the limit of detection (LOD), bias and precision of the DustTrak instruments. Each of the sessions ran for approximately 24 hours, and was conducted after the winter sampling campaign and before and after the summer campaign. A session was not conducted before the winter campaign as the continuous units were calibrated by the manufacturers before the beginning of the study. The methods for quantifying the limit of detection, bias and precision were followed as outlined in Wallace et al. (2011).

2.6 Data Analysis

2.6.1 Creating F_{inf} estimates

Daily and weekly estimates of F_{inf} were calculated using two approaches – 1) The censored indoor/outdoor ratio (censored I/O ratio) and 2) the indoor/outdoor ratio of sulphur (sulphur I/O ratio).

1) Censored I/O ratio: Indoor peaks were removed or 'censored' from the continuous data (half-hour averages) using a previously developed censoring algorithm (Kearney et al., 2011, MacNeill et al., 2012). This was done to remove any particles originating from indoor sources. The start of an indoor peak was identified when the indoor concentration increased by at least 5 μg/m^3 compared to the previous (half-hour) reading or the indoor concentration first exceeded the outdoor concentration. Indoor, outdoor and censored plots were then visually inspected and manually censored, if necessary. This ensured that all indoor peaks were removed and that the algorithm had not over-censored indoor levels that were likely due to outdoor fluctuations. Daily

F_{inf} estimates were calculated as the ratio of the daily mean of censored indoor values divided by the daily mean of the outdoor values. Weekly F_{inf} estimates were calculated as the ratio of the weekly mean of censored indoor values divided by the weekly mean of the outdoor values. In order to calculate a valid estimate of F_{inf}, at least 75% of the data was required to create both the censored indoor mean and the outdoor mean. This method assumes that a daily or weekly mean ratio will be close to the steady-state ratio. Although the hourly ratio is seldom equal to the steady-state value due to the lag of the indoor values behind the outdoor values, over time the mean values will approach the steady-state ratio.

2) Sulphur I/O ratio: The second method involves using sulphur as a tracer of $PM_{2.5}$ of ambient origin. Daily F_{inf} estimates using this method were calculated as the ratio of the daily sulphur concentration found indoors to the sulphur concentrations found outdoors (Sulphur I/O ratio). Weekly F_{inf} estimates were calculated as the ratio of the average weekly sulphur concentration found indoors to the average weekly sulphur concentration found outdoors. This method assumes that sulphur compounds are primarily of ambient origin and that the physical behaviour of sulphur is similar to that of other outdoor $PM_{2.5}$ constituents. Although the first assumption has been examined in several monitoring studies, there remains some uncertainty surrounding the validity of the second assumption given the differences in size range and volatility of sulphur compared with other outdoor constituents (Sarnat et al., 2002; Long et al., 2001; Kearney et al., *submitted*).

For all methods, all estimates of F_{inf} <1.2 were considered in the analysis in order to allow for error in the concentration measurements. Values > 1.2 were considered to indicate indoor sources that were missed in the censoring process.

2.6.2 Calculation of the ambient and non-ambient generated components.

Daily estimates of F_{inf} were used to estimate the non-ambient component ($C_{in(na)}$) and ambient component ($C_{in(a)}$) of the total daily indoor FP concentrations (C_{in}). Both the ambient and non-ambient components were calculated using the uncensored data. In cases were the ambient component exceeded the total indoor concentration, the ambient concentration was set to the total measured concentration, and the indoor-generated component was set to zero.

$$C_{in(a)} = F_{inf} \times C_{out} \tag{2}$$

$$C_{in(na)} = C_{in} - C_{in(a)} \tag{3}$$

2.6.3 Generalized linear mixed models

Statistical analyses were carried out using SAS EG version 4.2 (SAS Institute Inc, NC, USA) and Excel 2007 (Microsoft Inc.). A generalized linear mixed model with a variance components covariance structure was used to identify predictors for F_{inf} and the non-ambient component of FP. Potential predictors were identified from the baseline and daily questionnaires based on *a priori* hypotheses. Air exchange data was not considered for this analysis given its collinearity with many of the factors of interest (i.e. window opening, home age, I-O temperature difference, wind speed) and the non-linear relationship between the two variables. A number of variables were created through the combination of 2 or more questionnaire variables and where possible, missing values were imputed by replacing the missing observations with the median value. Variables with insufficient sample size (<5%) or variability within the distribution were not considered for further analysis. As well, the Multi-collinearity within the data was assessed by examining the tolerance, variance inflation factor (VIF) and condition indices among the full model. Where multi-collinearity was evident, the strongest predictor (defined by the *p-value* and Akaike information criterion or AIC) was chosen going forward.

A generic equation capturing the model used for these analyses is presented below:

$$Y_{ij} = \propto + \sum_{k=1}^{n} \beta_k X_{ijk} + b_i + \varepsilon_{ij} \quad (4)$$

Here Y_{ij} represents a F_{inf} estimate or non-ambient FP concentration for subject i, on day j; α signifies the regression intercept; β_k *rep*resents the parameter estimate for the k^{th} fixed effect; X_{ijk} denotes the value of the k^{th} independent variable for subject i, day j; b_i denotes the random subject effect $\sim N\ (0,\ \sigma^2\ b)$ and ε_{ij} represents the random error. A combination of subject matter knowledge, information criteria and likelihood ratio tests were used to choose an appropriate covariance structure. Since first-order autoregressive (ar(1)) and variance components (vc) structures were rarely significantly different when likelihood ratio tests were applied, the simpler covariance structure (vc) was chosen.

A backwards stepwise model selection was conducted to choose final models. Univariate regressions and a straight backwards selection were also run to inform final model selection. Final models were chosen based on the AIC, and the significance of the predictor variables. To remain in the final model, a variable was required to have a *p-value* <0.05. The effects of influential points were examined in all models, as well as plots of residuals.

3. Results and Discussion

3.1 Household Characteristics

Household characteristics for all homes are presented in Table 1. Most homes were detached single family dwellings (90% in winter; 92% in summer) with electric stoves (80% in winter; 82% in summer). The size of homes ranged from 624-5806 ft^2. Many homes had an attached garage with a connecting door (44% in winter; 40% in summer). Approximately one-third of homes had forced air as the main heat distribution system (34% in winter; 28% in summer). Many homes had some form of air cleaning device on their furnace (36% in winter; 34% in summer).

Window opening was prevalent in the summer sampling session, with windows open on 92% of days. In general, homeowners typically opened multiple windows (median=5) and left them open for that majority of the day (median/window ranging from 16-24 hours). There was very little air conditioning use (< 4% of days). This is in sharp contrast to Windsor, ON where 86% of homes had a central air conditioning (summer 2005) unit or used air conditioning daily (84% of sampling days in summer 2006) (MacNeill et al., 2012). Edmonton had moderate air conditioning use (13 homes reported some air conditioning use), and frequent window opening (participants reported opening at least one window on 73% of days) in summer. The prevalent window opening in Halifax is likely a result of the moderate summer climate. The median outdoor temperature observed during the summer sampling session was 17.1°C. Daily air exchange values ranged from 0.09/h to 4.07/h in summer (median=0.44) and from 0.08/h-1.14/h in winter (median=0.30).

3.2 Data Quality

3.2.1 DustTraks

LODs were calculated for each continuous instrument in each collocation session in which they participated. LODs ranged from 2.2 μg/m^3 (post-summer) to 4.0 μg/m^3 (post-winter). Bias and precision estimates were calculated for each individual unit by collocation session in which they participated. Of the total 46 DustTrak collocation tests across all three sessions, bias for 42 tests was between 0.7 and 1.3 (±30%). The median bias-corrected precision estimates were acceptable across all units, and ranged from 8.5% (post-summer) - 12.7% (post-winter).

3.2.2 Gravimetric $PM_{2.5}$ and Sulphur

Duplicate samples were used to calculate precision for both gravimetric $PM_{2.5}$ and sulphur. Good precision was observed for both metrics. The median precision for $PM_{2.5}$ was 3% in summer based on 57 paired samples (range 0% to 39%) In winter, the median precision for $PM_{2.5}$ was also 3% based on 35 paired samples (range 0% to 66%). In both seasons, sulphur had a median precision of 2% (n=57 in summer, n=35 in winter). In summer, the precision ranged from 0% to 13%. In winter, the precision ranged from 0% to 51%.

3.2.3 Air Exchange

Duplicate AER measurements were collected throughout this study and were used to assess the precision of the AER measures. The median precision found for the air exchange measurements in this study was 2% in summer (n=61; range: 0.0-10.5%) and 2% in winter (n=38; range: 0.0-18.6%).

3.2.4 Comparison of DustTraks and Gravimetric $PM_{2.5}$

Daily average DustTrak values were compared to the gravimetric measurements using Reduced Major Axis (RMA) regression. RMA regression allows for variance in both the dependent and independent variables. The slopes of the regressions were 1.88 (SE 0.19) indoors and 2.05 (SE 0.05) outdoors and were not significantly different (see SI for further details). These values are comparable to RMA slopes observed for Windsor, Canada (Wallace et al., 2011), Edmonton, Canada (Kearney et al., *submitted*) and further confirm the overestimation of the DustTraks compared to gravimetric methods. Since the ratio is similar both indoor and outdoor aerosol mixtures, the magnitude of the F_{inf} ratio when applied to gravimetric data should not be greatly different from the ratio calculated here using optical methods. DustTrak concentrations were not corrected to provide gravimetric estimates of indoor and outdoor concentrations. However, the results suggest that a division of all DustTrak readings by 2 will give a reasonable estimate of the $PM_{2.5}$ mass concentration.

3.3 Comparison of Methods

Daily and weekly F_{inf} estimates generated using the censored I/O ratio and the sulphur I/O ratio showed relatively poor agreement, despite the similarities in the distribution of estimates (Table 2). The two methods were only moderately correlated in both summer (daily: Spearman rho=0.26; weekly: Spearman rho=0.30), and winter (daily: Spearman rho=0.30; weekly: Spearman rho=0.38). The median difference in the daily paired samples was -0.06 in winter and 0.03 in summer. The median difference in the weekly samples was similar (winter=0.03; summer=0.04).

One possible explanation for this observation is that the use of sulphur as a tracer does not account for changes in $PM_{2.5}$ properties that result from indoor-outdoor transport (Meng et al., 2005; Polidori et al., 2006). In particular, sulphur would not reflect losses of volatile or semi-volatile components of PM that tend to partition to the gas phase when entering an indoor environment (Lunden et al., 2003). Therefore, the sulphur I/O ratio will be less likely to be a good tracer of ambient aerosols that contains a significant amount of semi-volatile components (Allen et al., 2012). Recent work in Halifax, Nova Scotia indicates the presence of the semi-volatiles NH_4+ and NO^3- in the total $PM_{2.5}$ mass, originating predominately from long-range secondary inorganic PM and marine aged secondary aerosols, respectively (Gibson et al., 2013). Loss of semi-volatiles during indoor-outdoor transport is supported by other recent studies. Sangiorgi et al., (2013) reported lower F_{inf} estimates for PM components with more semi-volatiles such as PAHs, NO^3-, NH_4+ compared with non-volatile ammonium sulphate. Sarnat et al. (2006) has also reported lower F_{inf} estimates for volatile nitrate (NO^3-) compared to non-volatile black carbon and $PM_{2.5}$. Kearney et al. (*submitted*), also found poor agreement between the censored I/O ratio and the sulphur I/O ratio. This finding highlights the fact that estimating F_{inf} of FP is not straightforward. The various components of the PM mixture will exhibit different behaviours when moving from an outdoor to an indoor environment and therefore, the composition of indoor particles of ambient origin may be different from outdoor particles.

Given the likelihood that sulphur is a poor tracer of ambient fine particles, the censored I/O ratio was used as an estimate of F_{inf} in subsequent analysis.

3.4 F_{inf} Estimates

Median daily and weekly F_{inf} estimates based on the censored I/O ratio can be found in Table 2 and Figure 1. Estimates in Halifax (daily summer median=0.80; daily winter median=0.55) were higher than have been reported in other parts of Canada. For example, median daily FP F_{inf} estimates in Windsor, Ontario ranged from 0.26 to 0.36 across both summer and winter seasons (MacNeill et al., 2012). Estimates for Toronto, Edmonton, and Victoria were higher than Windsor but lower than Halifax. Toronto median F_{inf} estimates ranged from 0.52 in summer/fall (Clark et al., 2010) to 0.59 (annual estimate) (Pellizzari et al., 1998). Median daily FP F_{inf} estimates in Edmonton were 0.69 in summer and 0.28 in winter (Kearney et al., *submitted*). In Victoria, mean seasonal estimates were 0.72 in summer, and 0.49 in winter (Hystad et al., 2009). Window opening behaviour was very different across these regions, with Halifax demonstrating the most window-opening. This is likely a result of the moderate climate experienced by this city. The differences in F_{inf} in these Canadian cities suggests that regional factors such as climate, air conditioning use and window opening can influence the contribution of ambient particles to indoor residential concentrations. Other factors such as housing stock, differences in provincial building codes, and availability of building materials may also play a role.

3.5 Between and within-home variability of F_{inf}

The between-home and within-home variance estimates for F_{inf} can be found in Table 3. In summer, the between-home variance estimates were greater than the within-home variance estimates (Table 3). The greater between-home variability is likely a result of several homes (one of which used air conditioning) having relatively low F_{inf} in the summer (median household F_{inf} <0.40) (Figure 1). In winter, there was a comparable amount of between and within-home variance ($\sigma^2_{BH=}0.026$; $\sigma^2_{WH=}0.022$). This is consistent with findings reported elsewhere (Bennett and Koutrakis (2006); MacNeill et al., 2012).

3.6 Predictive Models of F_{inf}

Predictive models of F_{inf} can be found in Table 4. Predictors of F_{inf} in winter included home age, income, presence of an air exchanger, use of a premium filter on the furnace, and the absolute temperature difference between indoors and outdoors. Winter models explained 57% of the between-home variance and 14% of the within-home variance. Predictors of F_{inf} in summer included the presence of carpets, number of windows open, and atmospheric pressure, and explained 9% and 7% of the between-home and within-home variance, respectively.

In winter, home age was found to be significantly associated with $F_{inf.}$ Homes that were built prior to 1945 were shown to have the highest F_{inf}. This can be attributed to changes in the building code, changes to building materials over time, and deterioration of the building over time. These results are remarkably consistent with results found in Windsor, ON. In the Windsor study, homes built before 1949 were associated with increased $F_{inf,}$ of 0.118 on average (MacNeill et al., 2012). In this study, homes built before 1945 were associated with an average increase in F_{inf} of 0.11. Other studies have also found an effect of home age on F_{inf} (Lachenmyer and Hidy, 2000; Long et al., 2001; Hänninen et al., 2005; Hystad et al., 2009).

In previous work, we also reported that older homes tended to be situated near major roads (MacNeill et al., 2012). In this study, we found a moderate correlation between home age and distance to major roads (spearman rho=0.27, p<0.0001), suggesting that F_{inf} is higher in homes located near major roads. In fact, a statistically significant correlation was observed between F_{inf} and distance to major roads in winter (spearman rho= -0.29, p<0.0001). Although this relationship may be due to differences in particle properties (Nazaroff, 2004) near roadways, it is likely mediated through home age. This variable did not remain significant when tested in combination with other variables, and therefore was not included in the final models.

Few studies have examined the influence of socioeconomic status (SES) on F_{inf}. Previous research related to air pollution and SES has primarily examined the relationship between SES and proximity to sources of exposure to air pollution (Gunier et al., 2003; Jerret et al., 2004, O'Neill et al., 2003), increased smoking rates (Watson et al., 2003) and greater occupational exposures (Rotko et al., 2000). In this study, we found that household income was a significant predictor of $F_{inf,}$ with low income households having lower F_{inf}. This is dissimilar to the findings of Hystad et al. (2009) who found that low improved building value increased F_{inf}. This result may be partially explained by the correlation observed between income and home age (spearman rho=0.25; p<0.000). In this study, people with greater household incomes were more likely to own homes built prior to 1945 than people with lower household incomes. However, income was a better predictor than home age suggesting that SES may be act as a surrogate for other variables influencing F_{inf} as well. More research is required in this area to fully understand the relationship between F_{inf} and SES. The relationship between F_{inf} and SES may have important implications for both epidemiological studies and health equity.

Air exchangers, the use of a premium filter on the furnace, and carpets were associated with reduced F_{inf}, suggesting these devices/characteristics were effective in increasing FP deposition. PM filtration devices have been shown previously to be associated with reduced F_{inf} (Barn et al., 2008; MacNeill et al., 2012) and therefore may provide an effective method of reducing indoor concentrations of ambient generated PM.

Determining factors that influence the variability in F_{inf} in summer was difficult given that windows were open on approximately 92% of days, and there was little air conditioning use (<5% of days). As a result F_{inf} was consistently high in the summer (IQR=0.65-0.89), and the contribution of other factors was difficult to elucidate as they were largely overwhelmed by the increased air exchange. Despite this, number of windows open was shown to increase F_{inf}. The influence of this factor is relatively small (an average increase of 0.01 for each window open). Information regarding window position was available, but did not add any additional information to the model.

3.7 Ambient and Non-ambient component estimates

Descriptive statistics for the ambient and non-ambient component estimates can be found in Table 5. In both summer and winter, the majority of FP was of ambient origin. In summer, there was a higher ambient contribution compared with winter (daily summer median=84%, daily winter median=59%), as a result of higher F_{inf} and higher outdoor FP concentrations in the summer months.

A partial validation of the estimates of the ambient and non-ambient components using the DustTrak F_{inf} values was carried out using the indoor and outdoor gravimetric measurements of the HI monitors. The DustTrak F_{inf} was multiplied by the outdoor HI concentration to give an estimate of the gravimetric ambient component of the total indoor particle mass. This component was then subtracted from the total HI value to give an estimate of the non-ambient gravimetric component. The non-ambient DustTrak component was regressed on the independently determined (because dependent on a separate indoor HI measurement) gravimetric non-ambient component. If the regression parameters were similar, it would indicate that the DustTrak F_{inf} was a reasonable proxy for the gravimetric F_{inf}. In fact, the regression parameters were nearly identical, with the same slope of 1.86. Tables and figures illustrating this method can be found in the S1.

Other studies have also found that the majority of indoor particle concentrations are of ambient origin (Allen et al., 2003, Wallace et al. 2006, Meng et al., 2005). MacNeill et al., (2012) found a median contribution of ambient particles ranging from 57% to 73% in Windsor, Ontario. Kearney et al. (*submitted*) also found that ambient sources were generally more important in Edmonton, Alberta, with a median ambient contribution of 70% in winter and 91% in summer. In both seasons, there was a higher ambient contribution of ambient particles to indoor concentrations in Edmonton compared with Halifax, despite the higher F_{inf} in Halifax. This is likely a result of the higher outdoor concentrations in Edmonton (Edmonton outdoor median: winter=16 $\mu g/m^3$; summer=11.7 $\mu g/m^3$; Halifax outdoor median: winter=9.76 $\mu g/m^3$; summer=7.69 $\mu g/m^3$) (Kearney et al., *submitted*). The high ambient contribution to total indoor FP concentrations in the three Canadian studies (Halifax, Edmonton and Windsor) may partially be due to the recruitment of exclusively non-smoking homes. Homes where smoking is permitted would likely have higher non-ambient contributions than those reported here.

3.8 Correlation between the ambient component and central-site concentrations

In both summer and winter, the ambient component of the indoor FPs was significantly correlated with central-site FP concentrations (summer: spearman rho=0.81, $p<0.0001$; winter: spearman rho=0.68, $p<0.0001$) There was no significant correlation between the non-ambient component and the central-site monitors. Wilson et al. (2006) suggest that if the ambient component is correlated with central-site concentrations then central-site concentrations can serve as a surrogate for exposure to ambient FP. As well, if the non-ambient component is not correlated with central-site concentrations, then the non-ambient component will not confound risk estimates. The correlations observed in this study provide evidence for why recent epidemiological studies have consistently shown health effects associated with ambient PM, despite the poor correlation between ambient PM concentrations and measured personal exposure (Mage et al., 1999). Despite this correlation, the variability in F_{inf} over homes suggests that the relative contribution of ambient particles to indoor air varies widely, and correlations may not be sufficient to assess the true health effects of PM.

3.9 Between and within subject variability of ambient and non-ambient components

The between-home and within-home variance estimates for the ambient and non-ambient component can be found in Table 3. In both summer and winter, the majority of the variability was within-subject for non-ambient FP. This can be explained by the high concentrations observed when indoor sources were present (Figure 2). The within and between-home variance estimates were more comparable for ambient FP. Typically variance estimates were smaller for the ambient component when compared to the non-ambient component, suggesting more homogeneity in ambient FP than non-ambient FP. These results are consistent with other studies (MacNeill et al., 2012; Kearney et al., *submitted*) and suggest a substantial amount of within-home variability in non-ambient FP regardless of geographic location.

3.10 Predictive models of non-ambient FP

Predictive model results for the non-ambient component can be found in Table 6. In both summer and winter models, cooking (including broiling, oven baking, whether food had been burned, number of times frying had occurred, number of times sautéing had occurred) were found to significantly increase the FP non-ambient component. Of the cooking variables, broiling and burning food was found to have the greatest impact. This finding is consistent with several other studies that have identified cooking as a major source of indoor PM (Ozkaynak et al., 1996; Wallace et al., 1996; Janssen et al., 1998; Abt et al., 2000; Wallace et al., 2003, MacNeill et al., 2012). Given the significant contribution of cooking to the non-ambient PM, mitigation strategies may be warranted to reduce exposure.

Other combustion sources (candle use, wood fireplace use, moving the car in/out of the attached garage) were also shown to increase the non-ambient component. Candle burning has previously been shown to be a significant source of indoor PM (Long et al., 2000; Ogulei et al., 2006; MacNeill et al., 2012), as has wood combustion (Traynor et al., 1987; Larson and Koenig, 1994; Naeher et al., 2007). Moving the car in/out of the attached garage was also associated with increased non-ambient FPs. In terms of source and composition, particles originating from a motor vehicle would be better categorized as ambient particles. However, in this instance, they are generated within the building envelope, and could therefore be described as indoor-generated. Given that this emerged as a significant predictor in our models, we chose to report it as a factor contributing to FP non-ambient component.

Indoor relative humidity was associated with increased particle concentrations of non-ambient origin in summer. This finding has not been widely explored in the literature. However, it has been suggested that both low and high relative humidity can increase particle deposition (Litvak et al., 2000; Miguel et al., 2004; Fromme et al., 2007). Relative humidity may also influence secondary aerosol formation, and the composition of particulates in the home. Alternatively, relative humidity may be acting as a surrogate for other activities such as cooking.

Factors influencing ventilation (duration of any exhaust fan used, and window opening behaviour) were shown to decrease the non-ambient component, with window opening showing greater reductions than exhaust fan use. In summer, each window opening decreased the non-ambient component by 0.37 $\mu g/m^3$, on average. In winter, the duration of window opening was found to be a better predictor. An average decrease of 0.11 $\mu g/m^3$ for each minute the windows were open was observed in winter. By both increasing the ambient component, and decreasing the non-ambient component, window opening is an important factor when considering the indoor-outdoor relationship for FP.

The results suggest that these models did not predict any of the between-home variance in winter, or any of the within-home variance in summer. According to Table 3, the between subject variance is a very small percentage of the total variance (2%). Our models were not successful at predicting this small amount of variability. In summer, the within-subject variance is high. However, most of that variability is being driven by a handful of homes, and many homes have very little variability (likely due to the open windows exhausting indoor particle sources) (Figure 2). Therefore, it is likely that this model is not successful at capturing the low variability found in the majority of homes.

Limitations of this study include a lack of sampling in spring or fall when estimates and predictors would likely vary from those presented here. Detailed information was not collected on the type of air exchangers or air filtration devices present in the home, or the MERV rating of the filters. As well, only homeowners were included in this study. This may influence the generalizability of the results as this study as we likely captured higher SES homes. Finally, by excluding smokers, an important source of indoor particles has not been captured and therefore, the non-ambient contribution may be greater in homes influenced by ETS.

This study highlights the fact that there is a significant amount of variability in F_{inf} between homes in a community and between communities. Regional factors such as climate, housing stock, air conditioning use and window opening influence the contribution of ambient particles upon indoor residential concentrations. Given that F_{inf} is an important modifier of the relationship between ambient particles to the indoor environment, this factor should be considered in future health studies to reduce exposure misclassification As well, by increasing our knowledge of non-ambient and ambient exposures, the risks associated with PM can be managed more effectively.

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Table 1: Housing Characteristics

Housing Characteristic	Winter		Summer	
	n	%	n	%
Home Age				
1945 and before	12	24%	12	24%
1946-1960	8	16%	8	16%
1961-1980	10	20%	10	20%
1981-2000	10	20%	10	20%
2001 and later	10	20%	10	20%
Home Type				
Detached	45	90%	46	92%
Row house	1	2%	0	0%
Duplex/triplex	3	6%	2	4%
Modular home	1	2%	1	2%
Missing	0	0%	1	2%
Median Home Size (ft^2) (min-max)	2086 (624-5806)		2093 (624-5806)	
Garage Type				
No garage	21	42%	20	40%
Detached	7	14%	9	18%
Attached with connecting door	22	44%	20	40%
Missing	0	0%	1	2%
Stove Type				
Natural gas or propane	10	20%	8	16%
Electric	40	80%	41	82%
Missing	0	0%	1	2%
Total Household Income				
Prefer not to say	2	4%	2	4%
<35000	5	10%	5	10%
35 000-80 000	17	34%	17	34%
>80 000	26	52%	24	48%
Missing	0	0%	2	4%
Any Air Cleaning Device on Furnace				
Yes	32	64%	16	32%
No	18	36%	33	66%
Missing	0	0%	1	2%
Heating Distribution System				
Forced air	17	34%	14	28%
Steam or hot water radiators	17	34%	18	36%
Baseboard heater	14	28%	18	36%
Other	2	4%	0	0%
Air Cleaning Device on Furnace				
Yes	17	34%	18	36%
No	33	66%	32	64%
Number of Days with Window Opening	84	24.6%	314	91.6%
Number of Days with any Air Conditioning Use	n/a	n/a	12	3.5%
Presence of Central Air Conditioning Unit	n/a	n/a	3	6,1%
Median Air Exhange /h (min, max)	0.30(0.08, 1.14)		0.44 (0.09, 4.07)	
Median Indoor Temperature (°C) (min, max)	20.4 (17.3, 25.2)		23.6 (18.5, 30.1)	
Median Outdoor Temperature (°C) (min, max)	-3.2 (-15.8, 8.8)		17.1 (8.9, 24.6)	
Median Indoor Relative Humidity (%) (min, max)	29.9 (22.2, 39.9)		45.9 (32.7, 57.3)	
Median Outdoor Relative Humidity (%) (min, max)	76.8 (44.8, 99.9)		86.5 (55.2, 98.6)	
Median Atmopheric Pressure (kpa) (min, max)	99.4 (96.4, 101.2)		99.8 (98.5, 101.3)	
Median Wind Speed (km/h) (min, max)	19.2 (7.4, 42.8)		13.9 (4.0, 29.8)	
Median Distance to major roads (m) (min, max)	256.7 (24.8, 2023.6)		255.3 (24.8, 2023.6)	

Table 2: Descriptive statistics for $PM_{2.5}$ F_{inf} estimates

Estimate	n	Percentiles								
		min	p5	p10	Q1	median	Q3	p90	p95	max
Winter										
Daily										
Censored I/O ratio	273	0.08	0.24	0.31	0.41	0.55	0.72	0.89	0.97	1.03
S I/O ratio	260	0.10	0.25	0.30	0.39	0.49	0.63	0.75	0.91	1.17
Weekly										
Censored I/O ratio	43	0.26	0.27	0.32	0.40	0.48	0.65	0.71	0.73	0.96
S I/O ratio	47	0.21	0.25	0.26	0.38	0.47	0.64	0.69	0.80	1.07
Summer										
Daily										
Censored I/O ratio	288	0.00	0.34	0.47	0.65	0.80	0.89	0.96	0.98	1.03
S I/O ratio	293	0.26	0.48	0.61	0.72	0.83	0.94	1.02	1.05	1.14
Weekly										
Censored I/O ratio	46	0.17	0.38	0.48	0.68	0.80	0.85	0.94	0.97	1.16
S I/O ratio	48	0.38	0.58	0.62	0.75	0.83	0.92	0.98	1.07	1.19

Table 3: Variance Components for daily F_{inf}, and indoor FP of ambient and non-ambient origin

	Between-home variance (σ^2_{BH})	Within-home variance (σ^2_{WH})	% Between-home variance of total variance	% Within-home variance of total variance
F_{inf}				
Winter	0.026	0.022	54	46
Summer	0.030	0.012	71	29
Non-Ambient FP				
Winter	320	1600	2	98
Summer	16	49	25	75
Ambient FP				
Winter	4.2	5.5	43	57
Summer	17	21	45	55

Table 4: Predictive models for FP F_{inf} by season

Fixed-effect	Parameter Estimate	Standard Error	p-value	proportion of the σ^2_{BH} explained	proportion of the σ^2_{WH} explained
Winter					
Intercept	0.27	0.06	***<0.0001***	57%	14%
Home Age			***0.002***		
1945 and below	0.11	0.06	0.06		
1946-1960	0.01	0.08	0.9		
1961-1980	-0.06	0.06	0.3		
1981-2000	-0.09	0.06	0.1		
2001 and later	0.00				
Income			***0.002***		
Prefer not to say	-0.07	0.10	0.4		
<35000	-0.20	0.07	0.004		
35 000-80 000	0.08	0.05	0.1		
> 80 000	0.00				
Air Exchanger (yes)	-0.31	0.10	***0.002***		
Premium Filter on Furnace (yes)	-0.23	0.07	***0.0004***		
Absolute I-O Temperature Difference (°C)	0.01	0.00	***<0.0001***		
Summer					
Intercept	-3.23	1.27	***0.01***	7%	9%
Any carpets present in home (yes)	-0.12	0.06	***0.02***		
Imputed number of windows open	0.01	0.00	***0.004***		
Atmospheric pressure (kpa)	0.04	0.01	***0.002***		

Table 5: Descriptive statistics for Daily $PM_{2.5}$ Components (using Cens IO Finf estimate)

Estimate	n	Percentiles								
		min	p5	p10	Q1	median	Q3	p90	p95	max
Winter										
Daily										
Ambient $PM_{2.5}$ ($\mu g/m^3$)	273	0.15	1.41	1.74	2.63	3.92	5.81	8.82	11	22
Non-Ambient $PM_{2.5}$ ($\mu g/m^3$)	273	0.00	0.05	0.10	0.61	2.31	7.43	17.54	28	2100
Indoor $PM_{2.5}$ ($\mu g/m^3$)	295	0.93	2.57	3.06	4.55	6.78	13.41	24.23	36	2100
Outdoor $PM_{2.5}$ ($\mu g/m^3$)	324	0.18	2.03	2.97	4.73	7.69	11.96	17.71	21	50
Percent Ambient (%)	273	1	11	17	38	59	86	97	99	100
Weekly										
Ambient $PM_{2.5}$ ($\mu g/m^3$)	43	1.95	2.41	2.77	3.15	3.93	5.35	7.17	10	12
Non-Ambient $PM_{2.5}$ ($\mu g/m^3$)	43	0.36	0.90	1.36	2.19	4.10	8.68	16.86	20	320
Indoor $PM_{2.5}$ ($\mu g/m^3$)	44	3.99	4.42	5.04	6.13	8.51	12.41	24.94	26	330
Outdoor $PM_{2.5}$ ($\mu g/m^3$)	49	3.46	4.38	4.62	6.68	9.27	11.68	14.85	17	23
Percent Ambient (%)	43	3	20	28	33	47	64	77	79	91
Summer										
Daily										
Ambient $PM_{2.5}$ ($\mu g/m^3$)	288	0.00	2.04	2.81	4.56	7.19	11.30	17.04	23	33
Non-Ambient $PM_{2.5}$ ($\mu g/m^3$)	288	0.00	0.04	0.14	0.47	1.46	3.59	7.41	15	95
Indoor $PM_{2.5}$ ($\mu g/m^3$)	329	0.04	3.28	4.65	6.71	10.10	15.53	25.02	29	100
Outdoor $PM_{2.5}$ ($\mu g/m^3$)	318	1.49	3.39	4.41	6.17	9.76	14.58	21.95	26	36
Percent Ambient (%)	288	1	23	39	63	84	94	98	100	100
Weekly										
Ambient $PM_{2.5}$ ($\mu g/m^3$)	46	1.98	2.97	3.32	5.67	7.79	12.49	15.08	19	22
Non-Ambient $PM_{2.5}$ ($\mu g/m^3$)	46	0.12	0.37	0.43	1.00	1.77	5.45	7.56	12	28
Indoor $PM_{2.5}$ ($\mu g/m^3$)	48	3.80	4.14	5.74	8.10	11.71	16.06	20.78	25	39
Outdoor $PM_{2.5}$ ($\mu g/m^3$)	48	4.71	5.00	6.45	8.01	10.94	15.34	20.66	22	23
Percent Ambient (%)	46	17	29	35	71	80	90	95	97	99

Table 6: Predictive Models for Non-Ambient FP by Season

Fixed-effect	Parameter Estimate	Standard Error	p-value	proportion of the σ_{BH}^2 explained	proportion of the σ_{WH}^2 explained
Winter					
Intercept	1.60	1.22	0.2	0.0	30%
Anyone broil food today (yes)	6.23	2.68	0.02		
Total fan time (minutes)	-0.02	0.01	0.02		
Duration oven used for baking (minutes)	0.04	0.01	0.002		
Duration windows open (minutes)	-0.11	0.03	0.0005		
Anyone burn food today (yes)	8.42	2.45	0.0007		
Imputed number of times frying	2.68	0.77	0.0006		
Imputed number of times sauteing	4.28	1.08	0.0001		
Candles burned (yes)	7.95	1.99	<0.0001		
Wood fireplace used today (yes)	14.33	3.04	<0.0001		
Summer					
Intercept	-9.73	4.50	0.04	70%	0.0
Duration stove used for frying (minutes)	0.11	0.03	0.001		
Duration stove used for sauteing(minutes)	0.12	0.04	0.005		
Indoor relative humidity (%)	0.28	0.10	0.004		
Duration oven used for baking (minutes)	0.03	0.01	0.03		
Car moved in and/or out of garage today (yes)	5.60	1.49	0.0002		
Imputed number of windows open	-0.37	0.16	0.02		

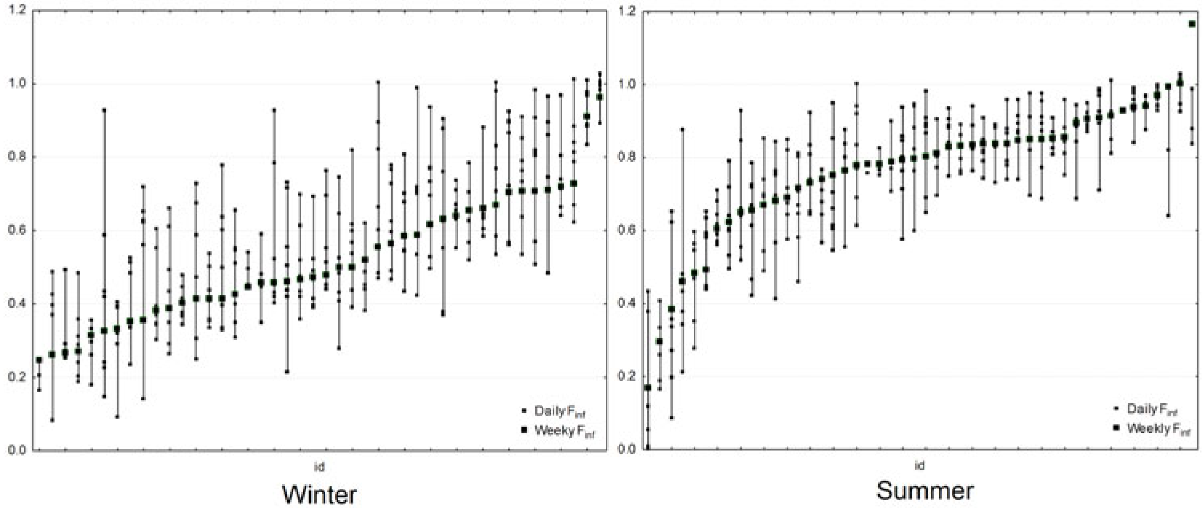

Figure 1: Daily and weekly F_{inf} estimates by id/season (each bar represents the full range of daily values for each household)

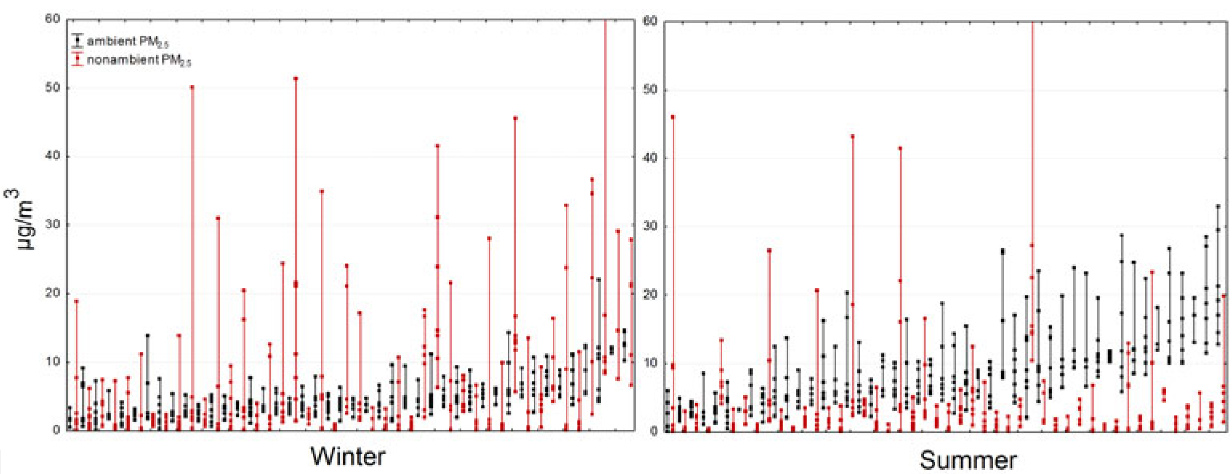

Figure 2: Ambient and non-ambient components by id/season (each bar represents the full range of daily values for each household)