1-1-2015

Estimation of bias with the single-zone assumption in measurement of residential air exchange using the perfluorocarbon tracer gas method

K Van Ryswyk

L. Wallace

D. Fugler

M. MacNeill

M. E. Heroux

See next page for additional authors

Authors

K Van Ryswyk, L. Wallace, D. Fugler, M. MacNeill, M. E. Heroux, M. D. Gibson, J. R. Guernsey, W. Kindzierski, and A. J. Wheeler

Estimation of bias with the single-zone assumption in measurement of residential air exchange using the perfluorocarbon tracer gas method

Abstract Residential air exchange rates (AERs) are vital in understanding the temporal and spatial drivers of indoor air quality (IAQ). Several methods to quantify AERs have been used in IAQ research, often with the assumption that the home is a single, well-mixed air zone. Since 2005, Health Canada has conducted IAQ studies across Canada in which AERs were measured using the perfluorocarbon tracer (PFT) gas method. Emitters and detectors of a single PFT gas were placed on the main floor to estimate a single-zone AER (AER_{1z}). In three of these studies, a second set of emitters and detectors were deployed in the basement or second floor in approximately 10% of homes for a two-zone AER estimate (AER_{2z}). In total, 287 daily pairs of AER_{2z} and AER_{1z} estimates were made from 35 homes across three cities. In 87% of the cases, AER_{2z} was higher than AER_{1z}. Overall, the AER_{1z} estimates underestimated AER_{2z} by approximately 16% (IQR: 5–32%). This underestimate occurred in all cities and seasons and varied in magnitude seasonally, between homes, and daily, indicating that when measuring residential air exchange using a single PFT gas, the assumption of a single well-mixed air zone very likely results in an under prediction of the AER.

K. Van Ryswyk[1], L. Wallace[2],
D. Fugler[3], M. MacNeill[1],
M. È. Héroux[1,*], M. D. Gibson[4],
J. R. Guernsey[5], W. Kindzierski[6],
A. J. Wheeler[1,7]

[1]Air Health Science Division, Health Canada, Ottawa, ON, Canada, [2]Consultant, Santa Rosa, CA, USA, [3]Consultant, Ottawa, ON, Canada, [4]Department of Process Engineering and Applied Science, Dalhousie University, Halifax, NS, Canada, [5]Community Health and Epidemiology, Faculty of Medicine, Dalhousie University, Halifax, NS, Canada, [6]School of Public Health, University of Alberta, Edmonton, AB, Canada, [7]School of Natural Sciences, Edith Cowan University, Joondalup, WA, Australia, *Present address: World Health Organization, European Centre for Environment and Health, Bonn, Germany

K. Van Ryswyk
Air Health Science Division
Health Canada
269 Laurier Ave West, AL4903C
Ottawa
ON
Canada K1A 0K9
Tel.: +1-613-952-4582
e-mail: keith.vanryswyk@hc-sc.gc.ca

Practical Implications

The results of this study suggest that the long-standing assumption that a home represents a single well-mixed air zone may result in a substantial negative bias in air exchange estimates. Indoor air quality professionals should take this finding into consideration when developing study designs or making decisions related to the recommendation and installation of residential ventilation systems.

Introduction

The understanding of residential air exchange is a critical element of indoor air quality (IAQ) research. It has been found to be an indication of the ability of a residence to clear contaminants and is inversely related to concentrations of indoor-generated air pollutants such as nitrogen dioxide (NO_2), formaldehyde, acetaldehyde and acrolein (Gilbert et al., 2005, 2006; Heroux et al., 2010; Hun et al., 2010). These results suggest that the attainment of a sufficient degree of air exchange will dilute indoor sourced air pollutants and reduce the health risks associated with their exposures. However, air exchange is also positively associated with the infiltration of outdoor air pollutants and the costs of controlling indoor humidity and temperature levels. Therefore, optimal ranges of air exchange within and between regions can vary as ambient air pollution levels, prevalent fuels and systems, climate, and IAQ issues are considered.

In advancing our understanding of residential air exchange and its relationship with IAQ, we must assess the errors and biases in the methods by which we measure air exchange. Several studies of air exchange rates (AERs) have been undertaken in some thousands of homes (Dodson et al., 2007; Gilbert et al., 2005, 2006; Heroux et al., 2010; Hun et al., 2010; Weisel et al., 2005; Wheeler et al., 2011a). These studies have employed the commonly used tracer gas method which employs inert gases not found in nature [perfluorinated cyclic hydrocarbon tracers (PFTs)] to determine AERs (Dietz and Cote, 1982; Dietz et al., 1986). Typically, the single-zone approach of the PFT tracer gas method has been employed which involves the release and measurement of a single tracer gas in a central location in the home. This approach rests on the assumption that the home is a single, well-mixed air zone. The AER is then calculated assuming perfect instantaneous mixing throughout the volume of interest (sometimes a single room but more often the entire house). However, imperfect airflow between the various rooms and floors of a home may create degrees of separation between its air spaces in which its residents spend large portions of their time. This would result in the creation of 'zones,' each of which may have their own characteristic AER. However, the frequency of 'multizonal' homes is unknown and the task of establishing this for each home in an IAQ study is cumbersome and of an unknown benefit to the understanding of residential AER. This makes the single-zone assumption a convenient one but also introduces an unknown magnitude of error.

In homes where two zones are expected, a unique tracer gas is emitted in each zone. With the measurement of each gas in each zone, a set of six equations with six unknowns can then be solved to provide the interzonal flows as well as the exfiltration and infiltration flows for each zone. The 2-zone whole house AER is then determined by dividing the sum of the exfiltration flows of each zone by the sum of their volumes (the volume of the whole house). A comparison of the two-zone AER with that of the single zone provides an indication of what degree of air exchange is being uncharacterized by the single-zone approach.

Health Canada (HC), in collaboration with local universities, has conducted IAQ studies in several Canadian cities over the past 9 years (Clark et al., 2010; Heroux et al., 2010; MacNeill et al., 2014; Wheeler et al., 2011b). Each study represented an extensive program of IAQ measurements and AER characterization using the perfluorocarbon tracer (PFT) single-zone method. This research typically involved 50–100 recruited homes measured for a period of 5–10 days in summer and winter. Along with IAQ and air exchange, information on house envelope data and daily occupant behavior was collected. The measurement of IAQ and air exchange was conducted in the main living area that was typically located on the main floor. To address the question of the introduction of AER measurement error resulting from the single, well-mixed zone assumption, this study reports on a subsample of homes from each of these studies where a floor adjacent to the main floor was treated as a separate air zone.

The refinement of the residential air exchange measurement is important to IAQ research as well as to the recommendation and installation of mechanical ventilation devices. The primary goal of this study is to produce estimates of error associated with the single-zone assumption by comparing single-zone AER estimates (AER_{1z}) with two-zone AER estimates (AER_{2z}). Secondly, estimates of precision are presented which investigate the error of AER estimation introduced by the choice of the central location in the main floor where the PFT gas is typically released and measured. Finally, sources of error in the tracer gas method in the literature are reviewed in detail. This study builds upon the current body of literature by increasing our knowledge of these errors, reinforcing the importance of reporting the details of AER measurement methodology (Persily and Levin, 2011) when publishing and recommending measures that can be taken to improve the PFT tracer gas method's precision and accuracy.

Methods

Air exchange data used in this analysis were collected in two IAQ studies in Edmonton, Alberta and Halifax, Nova Scotia and one panel study of asthmatic children of Montreal, Quebec. Results of these studies have been published elsewhere (MacNeill et al., 2014;

Smargiassi et al., 2014; Wallace et al., 2013). Each of these studies included daily measurement of residential air exchange using the single-zone PFT gas method. Approximately 10% of the homes from each study were selected to be a 'two-zone home' for this investigation.

Study designs

The Air Health Science Division (AHSD) of HC conducted IAQ studies in Halifax, Nova Scotia (2009) and Edmonton, Alberta (2010) in collaboration with Dalhousie University and the University of Alberta, respectively. Each involved the recruitment of 50 homes in which daily indoor and outdoor concentrations of several pollutants were assessed over periods of seven consecutive days in both the winter (January–April) and summer (June–September). Along with these measurements and questionnaires on home design and daily activities, daily measures of air exchange were made using the PFT method. In both cities, homes that dropped out after the first season were replaced with newly recruited homes to maintain a sample size of 50 homes in each season. Recruitment was stratified for home age. The groups of home age were categorized by the following years of construction: pre-1945, 1946–1960, 1961–1980, 1981–2000 and 2001 and after.

From October 2009 to April 2010, the AHSD, in collaboration with McGill University, conducted a panel study of 72 asthmatic children in Montreal, Quebec. Personal exposures of the children were measured for ten consecutive days along with questionnaires on home design and daily activities. These personal measures were also accompanied with central site ambient measures and residential air exchange using the PFT method. Data from this study have been classed as 'winter' in this study.

Each study sampled several homes simultaneously in groups of up to six homes. In each group, one home was selected to be the 'two-zone home'. This home was selected randomly other than the only exclusion criterion being the lack of the floor designated to be the second air zone. The second air zone was designated as the basement in the Halifax and Montreal studies while the first air zone was all above grade floors. Most basement spaces in Canadian homes are either finished or partially finished and often connected to the heating system of a home. They are considered 'conditioned spaces' in Canada. To provide data for comparisons of AER_{2z} and AER_{1z} in the case where the second zone was a floor other than the basement, the second zone was assigned as the second floor in the Edmonton study. In these cases, the main air zone represented the spaces of the main floor and the basement. All but one Edmonton home had a basement.

Home volume measurement

Home volume was measured using a sonic volumeter (Zircon Corp., Campbell, CA, USA). This device allows for a quick volumetric reading of any space by taking measures of the length width and height of a space. We measured the home spaces within the outer walls, spanning from the floor of the lowest floor (main floor or basement) to the ceiling of the top floor. This approach excluded the interior walls and inner floor spaces and included the volume of the furniture, closets, and cabinets. Volumes of each floor were combined to represent the volume of the home for AER calculation.
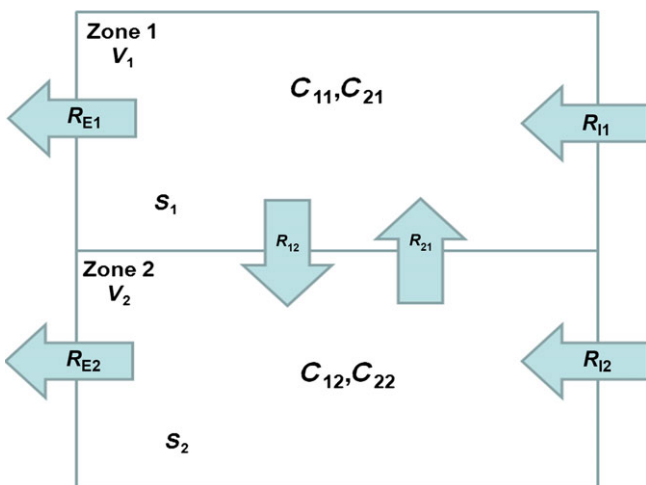
Air exchange

In each of these studies, the PFT gas technique developed by Brookhaven National Laboratory was used to measure residential air exchange using single or multiple air zone approaches (Dietz and Cote, 1982; Dietz et al., 1986). Briefly, the single-zone approach involved the deployment of three to four perfluoromethylcyclohexane (PMCH) emitters along with a tracer gas collection device (capillary adsorption tube or CAT) on the main floor of the home away from doors and windows. The emitters were placed far from the CATs, generally in the corners of the room. Typically, the living room was used. The emitters and CATs were installed at the beginning of each 7- to 10-day sampling period. For each 24-h period, a CAT was used to collect the tracer gas. Each CAT was placed on the main floor of the home in a central area. After exposure, the CATs were shipped to Brookhaven National Laboratory and analyzed by gas chromatograph with electron capture detector (GC/ECD). Results from the GC/ECD analysis for main floor CATs provided the amount of PFT gas collected by the CAT. Calculation of AER_{1z} in exchanges per hour was carried out using several parameters (Figure 1). The concentration of the PFT gas on the main floor (C) was adjusted for the gas-specific sampling rate of the CAT and the duration of its deployment. The emission rate of the PFT (S) was adjusted for the indoor temperature of the home. Dividing S by C gave the exfiltration rate of the home (R_E) in cubic meters per hour. This value is also equal to the infiltration rate (R_I). Dividing the flow rate 'R_E' by the volume of the home resulted in the single-zone air exchange rate estimate (AER_{1z}) in exchanges per hour.

The two-zone approach of the PFT method was used in the subset of homes to provide concurrent one and two-zone AER estimates (Figure 2). A second set of PFT emitters, releasing perfluoro-1,2-dimethylcyclohexane (ocPDCH), and a CAT were deployed on another floor in the home (Zone 2). The main floor and second zone CATs then provided concentrations for both main floor and the second zone PFT gases in each
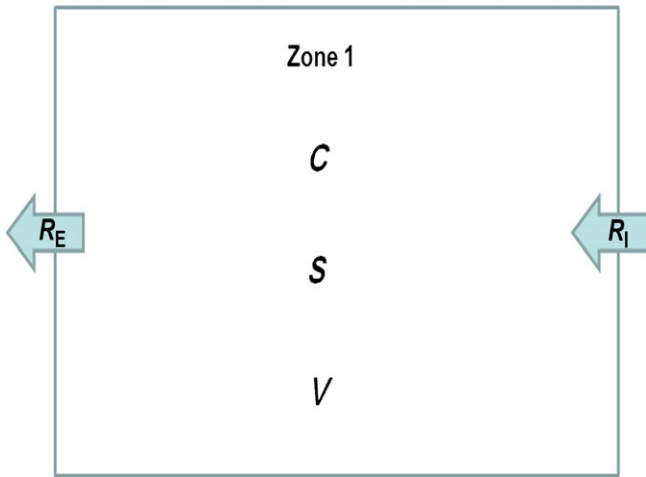
Fig. 1 Single zone air exchange. C, perfluorocarbon tracer (PFT) concentration (pl/l); S, PFT source emission rate (nl/h); V, home volume (m³); R_E, exfiltration rate (m³/h); R_I, infiltration rate (m³/h)

zone (C_{11}, C_{21}, C_{21}, C_{22}). These values, along with the temperature-adjusted PFT emission rates (S_1 and S_2), were then used in calculating two exfiltration flows (R_{E1} and R_{E2}), two infiltration flows (R_{I1} and R_{I2}) and two interzonal flows (R_{12} and R_{21}), all values in cubic meters per hour. AER_{2z} was calculated as the sum of both exfiltration values, divided by the volume of the entire home ($V_1 + V_2$) to provide the number of exchanges per hour (1/h). All PFT source rates were calculated based on the number of emitters deployed and the indoor temperature of the home. Indoor temperatures were measured continuously on the main floor using a HOBO data logger (Onset Computer Corporation, Bourne, MA, USA).

Fig. 2 Two zone air exchange. Cxy, concentration of perfluorocarbon tracer (PFT) emitted in zone x and measured in zone y (pl/l); Sx, PFT source emission rate of zone x (nl/h); Vx, volume of zone x (m³); REx, exfiltration rate of zone x (m³/h); RIx, infiltration rate of zone x (m³/h); Rxy, air flow rate from zone x to zone y (m³/h)

Analysis

The assumption of the single, well-mixed zone was tested in two ways. The first was by a method proposed in an extensive study into the errors and characteristics of tracer gas sampling (Lunden et al., 2012). This approach expressed each PFT tracer gas as a ratio of its concentration in its emission zone (C_{11} and C_{22}) to the adjacent air zone (C_{12} and C_{21}, respectively). In the single-zone assumption scenario, these ratios should be 1. As it is unlikely that the concentration would be higher in the adjacent zone, increases in this ratio indicate the separation of these two air zones. This is a simple approach in testing the single-zone assumption as it involves only the CAT laboratory reports and the calculation of a ratio.

The second method calculates the AER_{2z} to AER_{1z} ratio ($AER_{2z:1z}$) for each participant day. This ratio reflects that AER_{2z} is a more precise measure of air exchange, relative to AER_{1z} and can be interpreted as the factor by which a single-zone estimate must be multiplied in order for it to better represent air exchange. Trends in $AER_{2z:1z}$ were explored by city and season as a function of several home characteristics and meteorological conditions. Variables examined for relationships with $AER_{2z:1z}$ included factors such as home age, ventilation types and settings, wind speed, relative humidity, number of floors in the home, window opening, construction year, home type and connection to basement. Due to the repeated daily measures of the air exchange measurements, the SAS MIXED procedure (SAS Institute, Cary, NC, USA), which adjusts error estimates for autocorrelation and clustering, was used to estimate these relationships individually for categorical and continuous variables.

As our investigation was designed, principally, to test the single, well-mixed zone assumption by comparing AER_{2z} and AER_{1z}, this investigation also included the estimation of other sources of error by the deployment of two types of duplicates. They were each deployed in the main floors of 10% of the homes in each season of each study. The first type of duplicate, a 'traditional' duplicate, was placed in parallel with the CAT on the main floor. Precision estimates from these duplicates represent the accumulation of error from the GC-ECD analysis, the sampling rate of the CATs and field handling. The second type of duplicate was termed the 'second location' duplicate and was designed to estimate error introduced by the technician's choice of location in the main floor. In each of these studies, technicians were instructed to place the CAT samples in a central location of the floor, away from windows and doors to the outdoors and not closed off in a room. As these instructions do not dictate an exact location, technicians were instructed to deploy this duplicate in another location that also

adhered to the protocol of the CAT sample placement. As this type of duplicate also represents the errors of the traditional duplicate, its comparison with them reveal the added imprecision introduced by the technician's choice of a central area. For both duplicates, estimates of precision were calculated as the absolute difference divided by the sum (Equation 1).

$$Precision = \frac{ABS(A - B)}{A + B} \qquad (1)$$

Data management, statistical analyses, and figures were completed using SAS V.9.2 within SAS EG V.4.2 (SAS Institute).

Table 1 Characteristics of homes used in testing the single-zone assumption

Characteristic	Edmonton	Halifax	Montreal
Year tested	2010	2009	2009–2010
Total number of individual houses in both seasons	12	10	13
Number of houses tested in each season			
Winter	8	6	13
Summer	8	9	0
Median no. of valid daily air exchange rate measures per house per season (min/max)			
Winter	5 (4–6)	7 (2–7)	8 (4–10)
Summer	7 (6–7)	5 (5–7)	–
Type of house			
Detached	11	9	6
Other	1	1	7
Heating fuel			
Natural gas	12	3	0
Electricity	0	3	11
Oil	0	3	0
Other	0	1	2
Heat distribution			
Forced air	12	6	1
Baseboards	0	2	9
Radiators	0	2	2
Other	0	0	1
Mean age of home (min/max)	1996 (1910/2007)	1990 (1892/2008)	1966 (1945/2006)
Median house volume in m³ (min/max)	474 (154–1040)	399 (258–572)	379 (144–547)
Mean daily temperature (min/max)			
Winter	−4 (−11/3)	−2 (−5/5)	4 (−9/9)
Summer	15 (12/17)	17 (13/21)	–
Number of homes reporting daily use of air conditioning[a]			
Winter	0	0	0
Summer	3	1	–
Number of homes reporting open windows (min/max n days)[b]			
Winter	2 (1/1)	2 (2/3)	6 (1/6)
Summer	7 (3/7)	9 (1/7)	–
Basement?			
Yes	11	10	13
No	1	0	0
Number of above grade floors			
1	0	3	5
>1	12	7	8
Second zone location	Second floor	Basement	Basement

[a]Central or window ac unit.
[b]Represents count of windows open for at least 6 h of day.

Results

Table 1 shows the characteristics of the houses tested in each of the three cities. Each house had PFT testing 2–10 times in each season. The Montreal houses were only sampled in winter, unlike the Edmonton and Halifax houses that had both winter and summer sampling campaigns. Of the Edmonton and Halifax homes in summer, four of the 17 reported the presence of an air conditioner (be it a central or a window air conditioning unit). All but one reported window opening on at least 1 day during their sampling period. In winter, 6 of the 13 Montreal homes reported at least 1 day in which windows were open in the home. For four of these participants, window opening was reported for only 1 day.

To test the single, well-mixed zone assumption, ratios were calculated for the concentration of the PMCH and ocPDCH between their source zone (main floor for PMCH and basement or second floor for ocPDCH) and the adjacent zone (main floor for ocPDCH and basement or second floor for PMCH). The concentrations in these zones should be identical on the two floors. Table 2 presents the percentiles of these ratios. Of the 287 pairs of measurements for both PFT gases, approximately 95% were found to be lower in the adjacent zone (5th percentiles of 0.93 and 1.1 for PMCH and ocPDCH, respectively). Median values for these ratios ranged from 1.1 to 9.2 (Table 2).

The single, well-mixed assumption was also tested by comparing the single-zone AER (AER_{1z}) and two-zone AER (AER_{2z}) estimates. As in the case of the PFT concentrations in each zone, these two estimates of air exchange should be equal as per the scenario of the complete mixing dictated in the single, well-mixed assumption. In 87% of cases, AER_{2z} was greater than the AER_{1z}. The distributions of AER_{1z} and AER_{2z} are provided for all city-season combinations in Table 3. Mean estimates of AER_{1z} and AER_{2z} with 95% CL error bars for summer (Figure 3) and winter (Figure 4) depict the consistent low bias of the single-zone approach relative to that of the two-zone approach. The most extreme differences were seen in Halifax winter and Edmonton summer data.

To facilitate the comparison between AER_{1z} and AER_{2z} and to provide bias estimates of AER_{1z}, relative to AER_{2z}, a ratio was calculated ($AER_{2z:1z}$) for each participant day ($n = 287$). Table 4 presents the percentiles for $AER_{2z:1z}$ by city and season. Median values of $AER_{2z:1z}$ indicate the highest ratios in Halifax winter (1.67) and Edmonton summer (1.26). The sample from Edmonton winter provided the lowest AER_{1z} underprediction, relative to AER_{2z}, with a 90th percentile of 1.41. While some subsamples of houses, for example Edmonton houses in winter, had very similar results from AER_{1z} and AER_{2z}, overall, the median $AER_{2z:1z}$ estimate indicated that AER_{1z} underestimates AER_{2z} by 16% (ratio of 1.16).

**Table 2** Percentiles of source zone to adjacent air zone perfluorocarbon tracer concentration ratios

Study	Season	n Homes (samples)	PMCH[a] main: 2nd zone					ocPDCH[b] 2nd zone: main				
			5th	25th	50th	75th	95th	5th	25th	50th	75th	95th
Edmonton	Winter	8 (37)	0.9	1.0	1.1	1.3	3.4	1.0	1.2	1.4	2.4	6.1
	Summer	8 (49)	0.8	1.0	1.3	1.7	2.5	1.1	2.3	5.1	9.9	51.6
Halifax	Winter	6 (33)	1.5	2.0	3.4	29.6	202.1	0.8	1.4	1.6	2.1	3.2
	Summer	9 (60)	0.9	2.0	2.8	6.0	11.1	1.7	5.2	9.2	20.7	201.5
Montreal	Winter	13 (108)	1.0	1.5	2.2	3.8	10.7	1.1	1.3	1.5	3.3	12.7
All	All	35 (287)	0.9	1.3	2.0	3.6	16.1	1.1	1.3	2.3	6.6	25.6

[a]Perfluoromethylcyclohexane; released in main floor.
[b]Perfluoro-1,2-dimethylcyclohexane; released in second zone (Edmonton = second floor, Halifax and Montreal = basement).
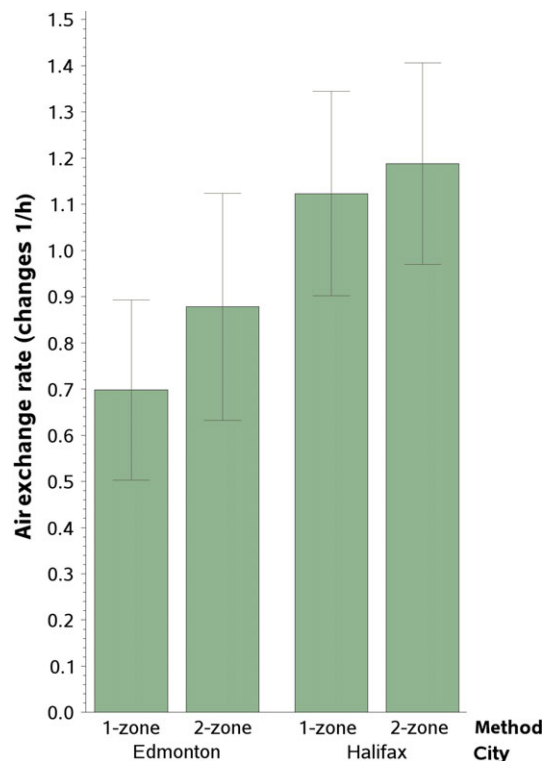
Table 3 Distribution of single zone air exchange (AER_{1z}) and two zone air exchange (AER_{2z}) (1/h) by city and season

City	Season	n Homes (samples)	AER perfluorocarbon tracer Method	Air exchange (AER) (1/h)						
				Mean	s.e.	5th	25th	50th	75th	95th
Edmonton	Summer	8 (49)	Single Zone	0.69	0.23	0.17	0.28	0.47	0.86	2.16
			Two Zone	0.87	0.29	0.18	0.34	0.56	1.15	2.69
	Winter	8 (37)	Single Zone	0.37	0.09	0.15	0.19	0.28	0.53	0.83
			Two Zone	0.42	0.11	0.16	0.20	0.28	0.61	1.02
Halifax	Summer	9 (60)	Single Zone	1.17	0.25	0.20	0.71	1.04	1.45	2.51
			Two Zone	1.23	0.25	0.36	0.73	1.01	1.48	2.55
	Winter	6 (33)	Single Zone	0.35	0.05	0.21	0.28	0.33	0.36	0.59
			Two Zone	0.55	0.10	0.36	0.37	0.47	0.58	1.03
Montreal	Winter	13 (108)	Single Zone	0.44	0.15	0.17	0.23	0.31	0.35	0.40
			Two Zone	0.49	0.15	0.23	0.27	0.37	0.43	0.51
All	All seasons	35 (287)	Single Zone	0.62	0.10	0.17	0.22	0.37	0.72	1.49
			Two Zone	0.70	0.11	0.18	0.27	0.45	0.97	1.83
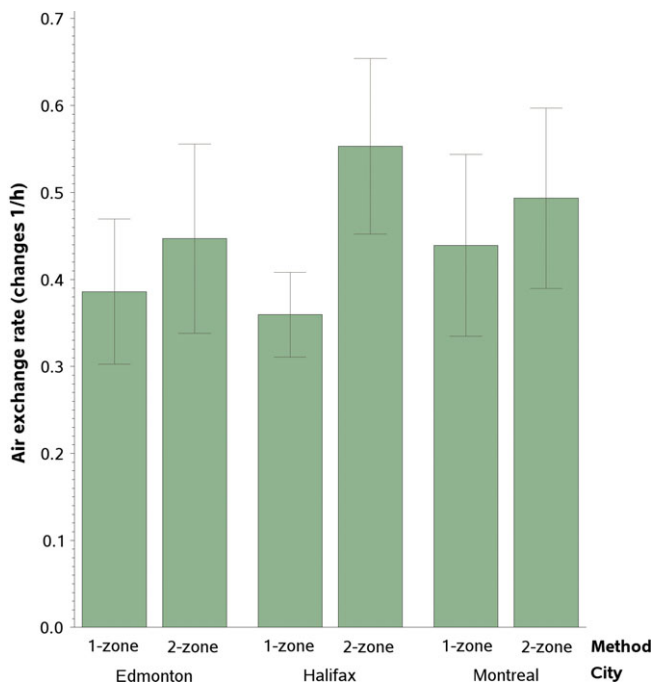
Fig. 3 Winter air exchange rates by city and air zones (single or two zone) (1/h)

Fig. 4 Summer air exchange rates by city and air zones (single or two zone) (1/h)

Figure 5 presents the daily $AER_{2z:1z}$ data in box-plots for each combination of participant and season (*n* = 35). Considerable variability can be seen between

Table 4 Two zone to one zone air exchange rate (AER) ratios ($AER_{2z:1z}$) by city and season

City	Season	n Homes (samples)	10th	25th	50th	75th	90th
Edmonton	Summer	8 (49)	0.97	1.11	1.26	1.40	1.69
	Winter	8 (37)	0.97	1.02	1.07	1.12	1.41
Halifax	Summer	9 (60)	0.99	1.02	1.07	1.11	1.77
	Winter	6 (33)	1.07	1.43	1.67	1.74	1.74
Montreal	Winter	13 (108)	1.01	1.07	1.10	1.30	1.54
All	All	35 (287)	1.00	1.05	1.16	1.32	1.54

seasons, homes, and day to day. The seasonality of $AER_{2z:1z}$ can be seen in Edmonton and Halifax where sampling was completed in summer and winter. In Edmonton, where the second floor of the home was selected for the second air zone, summer was seen to have larger estimates of underprediction and more day to day variability within homes as seen by the larger interquartile ranges. Halifax's seasonality was the opposite of the Edmonton findings as the winter data was seen to have higher values of $AER_{2z:1z}$ and more day to day variability. In the Montreal study, $AER_{2z:1z}$ is seen to vary comparatively little day to day but there is a wide range of $AER_{2z:1z}$ by participant.

The CAT duplicates deployed in parallel with the primary CAT samples (a 'traditional' duplicate) represent the accumulation of error from the laboratory analyses, CAT sampling rate, and field deployment. Similarly, these sources of error are also represented in the precision estimates from the 'second location' duplicates with the added element of the technician's choice of the central site in the main floor, as directed by the deployment protocol. As expected, the precision for the traditional duplicates is better (Table 5). The median precision for the 'traditional' duplicates ranged from 1.2 to 2.5% and 1.9 to 3.9% for the 'second location' duplicates. This suggests that the error introduced by the technician's location selection had limited impact.

Discussion

In this investigation, the assumption that homes consist of a single, well-mixed zone was tested. The internal floors of the tested homes appeared in general to form separate air zones. In the single-zone approximation, the concentration of a PFT is the same everywhere in the house. However, for the PMCH primary

Table 5 Precision estimates using the 'traditional' and 'second location' duplicates (%)

City	Season	n Homes (samples)	Duplicate type	10th	25th	50th	75th	90th
Edmonton	Summer	9 (63)	Traditional	0.2	0.4	1.2	2.0	3.8
		9 (63)	Second location	0.5	0.9	2.3	3.2	4.4
	Winter	7 (46)	Traditional	0.2	0.6	1.3	2.7	5.9
		7 (47)	Second location	0.8	1.8	3.4	7.3	98.7
Halifax	Summer	9 (61)	Traditional	0.4	1.3	2.1	4.2	8.2
		9 (60)	Second location	0.7	1.7	3.9	6.4	12.7
	Winter	6 (38)	Traditional	0.2	0.7	1.7	4.1	7.4
		6 (38)	Second location	0.2	1.0	1.9	5.8	13.9
Montreal	Winter	14 (117)	Traditional	0.4	1.2	2.5	4.3	8.5
		14 (125)	Second location	0.5	1.3	3.1	5.4	13.2
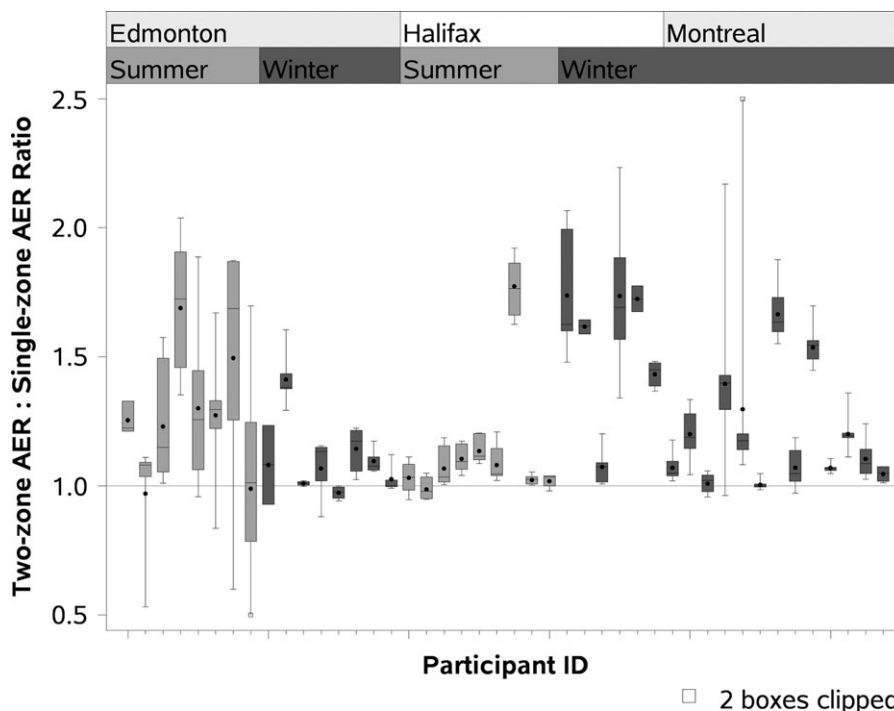
Fig. 5 Two-zone: single-zone air exchange rate ratios by participant. Two boxes clipped

PFT, the concentration in the main living floor was higher than that in the secondary areas in 268 of 287 cases (93%). As this concentration is used to estimate single-zone air exchange, the overestimate of the housewide average concentration leads to an underestimate of the AER. Had the second PFT and its concentration on the floor of its release been used to estimate the AER, once again the estimate would be biased low, since in 278 of the 287 cases (97%), the concentration on the floor of release was greater than that on the main floor.

Assuming that AER_{2z} is more accurate than AER_{1z} (due to the additional information supplied by the second tracer gas), we conclude that the assumption of a single, well-mixed zone inherent in the single gas PFT gas method resulted in an underestimation of air exchange in all three cities and both seasons. This finding occurred in houses from several Canadian cities, in winter and summer, in homes of all ages, and whether the second zone was the basement or the second floor. The median two zone to single-zone air exchange estimate ratio ($AER_{2z:1z}$) was found to be 1.16 (IQR: 1.05–1.32).

In a classic series of papers dating from 1986, Sherman and coworkers studied errors associated with passive measurement of air infiltration. They showed that, due to variability of infiltration over time, there is a bias toward underestimating the AER. Measurements on a test house in Edmonton, Canada indicated that the mean underestimate over a year was on the order of 16% and was highest in the summer months (Sherman and Wilson, 1986). A later paper was concerned with spatial variation across zones or floors in a residence (Sherman, 1989). Ventilation efficiency was defined as the ratio of the amount of gas in a zone compared to the amount that would have been found if the home was a single zone. Using a model of climate factors and housing types in six cities, it was found that using a single-zone estimate of AERs for a colonial style (two story with finished basement) house would lead to underestimates of the AER on the order of 20–30%. However, a multizonal model could improve the estimates by an amount on the order of 10%.

Several studies have used two PFTs to provide AERs for specific rooms in a home, such as bedrooms where children spend a significant proportion of time at home. However, few studies have compared the 1-zone to the 2-zone estimates. In one study of the benefits of air filtration in the bedrooms of 126 Detroit, Michigan homes, the interzonal flows between the main living area and the children's bedroom were quantified as well as the AERs specific to the child's bedroom and that of the rest of the home (Du et al., 2011). The floor level of the child's bedroom was not reported and a housewide 2-zone AER was not calculated or compared with a single-zone estimate; however, the indication that each zone's exchange of air

with the outdoors differed does imply that the single-zone and 2-zone AERs would differ.

The multizonal approach to the tracer gas method has also been used to characterize the amount of air coming from known areas of the home containing indoor pollutant sources. This was carried out in 2004 and 2005 for 45 Boston homes (Dodson et al., 2007). Again, the overall 2-zone AER was not calculated; they treated other air zones in their 45 homes to be apartment hallways (10), attached garages (11), and basements (35). By calculating the interzonal flows, they determined the fraction of air in the main living zone originating from basements to be 26% (s.d. = 34%) and 47% (s.d. = 26%) in winter and summer, respectively. Sinden (1978) provided a particularly influential contribution, with a mathematically elegant discussion of the multizone system.

Table 6 summarizes factors that affect uncertainty of the tracer gas method. These error estimates come from this study and from Lunden et al. (2012), in which the authors investigated various sources of error in the use of the PFT tracer gas method of measuring air exchange. We consider our error of volumetry (a major factor in the calculation of AER) to be dependent on the unit's length measurement error (0.005 m). In a larger home, which can take ten separate measurements, the error is on the order of 1%. However, uncertainty can still remain in a researcher's designation of what represents actual mixing volume. Measuring this space by floor area and ceiling height results in an overestimate as the resulting volume includes the space occupied by all solid objects within each floor. The volumes measured in these HC studies were completed room to room and thus represent all spaces within the outer walls, from the basement floor to top floor ceiling minus the volume of the interior floors and walls but including the volumes of home furniture. This was preferable to using floor area and ceiling height as the exclusion of floor and wall spaces would somewhat compensate for the inclusion of furniture volume. As these estimates can differ significantly, providing details on volume measurement methodology can be considered good practice.

In the investigation by Lunden et al. (2012), the effect of temperature on the emission rate of the PFT gas was quantified. Their result of an emission rate change of 4% per 1°C highlights the value of careful temperature measurement when using the PFT tracer gas method. As the PFT tracer gas method includes the calculation of a temperature adjusted emission rate, emission rate error was reported to be tied to the precision of the dry block heaters used to provide a constant temperature for their emitters (±1°C). This resulted in an emission rate uncertainty of (±4%). The HOBO data logger used in the present study reports to a precision of 0.35°C; therefore, error in this regard is reduced. However, as we measured indoor temperature

Table 6 Error estimates for various elements of the perfluorocarbon tracer (PFT) tracer gas method

Unit	Parameter	Error source	Reference	Error estimate (%)	Description
Zircon DM S50L	Home volumetry	Sonic Volumeter's limit of quantification	This paper	1	0.005 m; 1% for a 10 room home, each with dimensions of 2.2 m $\times$ 5 m $\times$ 10 m
PFT Emitter	Emission rate	Effect of temperature on emission rate	Lunden et al. (2012)	4	4% per 1°C. Resulting error tied to precision of temperature instrument ($\pm$4%)
			This paper	1	HOBO data logger precision $\pm$0.35°C (~2% for the range of 10–30°C)
Capillary absorption tube	CAT sampling rate	Unit to unit bias	BNL[a]	1–3	Reported by Brookhaven National Laboratory
PFT calibration for gas chromatography	CAT PFT result	GC/ECD precision	BNL[a]	1.50	Reported by Brookhaven National Laboratory
Traditional duplicate	Air exchange rate (AER) estimate	GC/ECD precision, field handing, CAT tube sampling variation	This paper	1.6–6.3[b]	CAT duplicates used same data on emission rate, indoor temperature, & home volume
Second central site duplicate		GC/ECD precision, field handing, CAT tube sampling variation + spatial variability in main living area	This paper	2.3–8.6[b]	In comparison to traditional duplicates, a small amount of error is introduced by the selection of the central site in which to deploy the CAT
PFT AER method (single zone vs. two zone)		Single, well-mixed air zone assumption	This paper	16 (IQR: 5–32%)	A negative bias

[a]Brookhaven National Laboratory.

[b]Range of median values across five groups.

centrally, there may be additional unquantified error due to spatial variation of temperature within a home. Direct measurement of emitter temperature is paramount in reducing the uncertainty of the PFT tracer gas method. In relation to these errors, which are centered around zero, the bias of the single-zone assumption appears to represent a key factor in improving the precision of measuring air exchange.

Our attempt to determine factors that affect AER_{1z}'s underprediction of AER_{2z} was weakened by the limited sample size. The available predictors of meteorology and study questionnaire variables were investigated using mixed models for their possible effect on $AER_{2z:1z}$. Most of these variables showed no significant effect. Three variables were significantly associated with $AER_{2z:1z}$ in one or two of the five city-seasons cases. These variables were 2-floor vs. 3-floor homes, open vs. closed windows, and open vs. closed doors at the stairwells to the basement. However, allowing for multiple comparisons and using the Bonferroni correction, none of these achieved statistical significance. We found no evidence in our data that $AER_{2z:1z}$ varies by heat distribution system. A forced air system could create a well-mixed single zone within a home, but this depends on the duty cycle, duct system design, degree of appliance oversizing, severity of climate, and other factors. The use of the second floor as the second zone in the Edmonton homes also proved to be a weakness as comparisons between $AER_{2z:1z}$ by second zone location was impossible due to the fact that all Edmonton homes had forced air ventilation, limiting any conclusions made from such a comparison. A much larger source of error may be the assumption of constant flow rate. This is affected by temperature, pressure, and wind speed variation over the 24-h sampling period, and in some cases, even more highly affected by occupant behavior such as opening

windows and using exhaust fans. As shown by Sherman (1989), this source of error results in a negative bias in the estimated AER. In a subsequent publication looking more carefully at the errors in Lunden et al. (2012), Sherman et al. (2014) estimated that the error under ideal conditions (including, e.g., constant flow rate over the sampling period) could range from 6% to 15%. Under field conditions with excellent instrument calibration and well-trained personnel, the errors could be expected to reach 20–25%. And under typical uncontrolled field studies (e.g., when occupants may open windows or operate fans at will), the error would more likely be on the order of a factor of two. Shinohara et al. (2010) investigated three PFTs and found emission rate errors of about 10% for two of them and 5% for a third. Concentration errors due to variability in recovery were about 7–7.5% for all three PFTs. Using these three PFTs, Shinohara et al. (2011) carried out a 3-zone study of 26 homes in Japan. The calculated uncertainties in the airflow estimates were less than 20% for more than 70% of the data. Uncertainties exceeded 50% for 7% of the data.

Conclusion

The assumption that a residence's air zone is single and well-mixed has been a long-standing one. It has been made in light of the barriers of practicality and the impression that it introduced an acceptable degree of error. However, our findings indicate a robust and substantial bias in this assumption in many of our study homes. The single-zone air exchange estimates were observed to underestimate the two-zone estimates by approximately 16% (IQR: 5–32%). This underprediction was seen to vary seasonally, between homes, and on a daily basis. These AER conclusions apply specifically to the Canadian houses tested in this research. As

such, further comparisons of single-zone and multizonal AERs are desirable to investigate the potential for this underprediction in homes of different housing stock and climates. Underestimates of AERs could result in overestimating the impact of indoor sources and underestimating ambient pollutant infiltration. Furthermore, these results also reveal that there are varying degrees of incomplete mixing within homes, which illustrate the importance of the location of sources (garage, basement, main floor) and where people spend their time indoors. Other sources of error when using the PFT tracer gas method have been reviewed in the literature.

References

Clark, N.A., Allen, R.W., Hystad, P., Wallace, L., Dell, S.D., Foty, R., Dabek-Zlotorzynska, E., Evans, G. and Wheeler, A.J. (2010) Exploring variation and predictors of residential fine particulate matter infiltration, *Int. J. Environ. Res. Public Health*, **7**, 3211–3224.

Dietz, R.N. and Cote, E.A. (1982) Air infiltration measurements in a home using a convenient perfluorocarbon tracer technique, *Environ. Int.*, **8**, 419–433.

Dietz, R.N., Goodrich, R.W., Cote, E.A. and Wieser, R.F. (1986) *Detailed Description and Performance of a Passive Perfluorocarbon Tracer System for Building Ventilation and Air Exchange Measurements*, Philadelphia, PA, ASTM Special Technical Publication, American Society for Testing and Materials, 203–264.

Dodson, R.E., Levy, J.I., Shine, J.P., Spengler, J.D. and Bennett, D.H. (2007) Multi-zonal air flow rates in residences in Boston, Massachusetts, *Atmos. Environ.*, **41**, 3722–3727.

Du, L., Batterman, S., Parker, E., Godwin, C., Chin, J.Y., O'Toole, A., Robins, T., Brakefield-Caldwell, W. and Lewis, T. (2011) Particle concentrations and effectiveness of free-standing air filters in bedrooms of children with asthma in Detroit, Michigan, *Build. Environ.*, **46**, 2303–2313.

Gilbert, N.L., Guay, M., Miller, J.D., Judek, S., Chan, C.C. and Dales, R.E. (2005) Levels and determinants of formaldehyde, acetaldehyde, and acrolein in residential indoor air in Prince Edward Island, Canada, *Environ. Res.*, **99**, 11–17.

Gilbert, N.L., Gauvin, D., Guay, M., Héroux, M.E., Dupuis, G., Legris, M., Chan, C.C., Dietz, R.N. and Lévesque, B. (2006) Housing characteristics and indoor concentrations of nitrogen dioxide and formaldehyde in Quebec City, Canada, *Environ. Res.*, **102**, 1–8.

Heroux, M.E., Clark, N., Van Ryswyk, K., Mallick, R., Gilbert, N.L., Harrison, I., Rispler, K., Wang, D., Anastassopoulos, A., Guay, M., MacNeill, M. and Wheeler, A.J. (2010) Predictors of indoor air concentrations in smoking and non-smoking residences, *Int. J. Environ. Res. Public Health*, **7**, 3080–3099.

Hun, D.E., Corsi, R.L., Morandi, M.T. and Siegel, J.A. (2010) Formaldehyde in residences: long-term indoor concentrations and influencing factors, *Indoor Air*, **20**, 196–203.

Lunden, M., Faulkner, D., Heredia, E., Cohn, S., Dickerhoff, D., Noris, F., Logue, J., Hotchi, T. and Singer, B. (2012) *Experiments to evaluate and implement passive tracer gas methods to measure ventilation rates in homes*, Report LBNL-5984E, Berkeley, CA, Lawrence Berkeley National Laboratory.

MacNeill, M., Kearney, J., Wallace, L., Gibson, M., Héroux, M.E., Kuchta, J., Guernsey, J.R. and Wheeler, A.J. (2014) Quantifying the contribution of ambient and indoor-generated fine particles to indoor air in residential environments, *Indoor Air*, **24**, 362–375.

Persily, A.K. and Levin, H. (2011) Ventilation measurements in IAQ studies: problems and opportunities. In: *Proceedings of Indoor Air 2011, 12th International Conference on Indoor Air Quality and Climate*. Available at: http://www.nist.gov/manuscript-publication-search.cfm?pub_id = 907718.

Sherman, M.H. (1989) Analysis of errors associated with passive ventilation measurement techniques, *Build. Environ.*, **24**, 131–139.

Sherman, M.H. and Wilson, D.J. (1986) Relating actual and effective ventilation in determining indoor air quality, *Build. Environ.*, **21**, 135–144.

Sherman, M.H., Walker, I.S. and Lunden, M.M. (2014) Uncertainties in air exchange using continuous-injection, long-term sampling tracer-gas methods, *Int. J. Vent.*, **13**, 13–28.

Shinohara, N., Kataoka, T., Takamine, K., Butsugan, M., Nishijima, H. and Gamo, M. (2010) Modified perfluorocarbon tracer method for measuring effective multizone air exchange rates, *Int. J. Environ. Res. Public Health*, **7**, 3348–3358.

Shinohara, N., Kataoka, T., Takamine, K. and Gamo, M. (2011) Distribution and variability of the 24-h average air exchange rates and interzonal flow rates in 26 Japanese residences in 5 seasons, *Atmos. Environ.*, **45**, 3548–3552.

Sinden, F.W. (1978) Multi-chamber theory of infiltration, *Build. Environ.*, **13**, 21–28.

Smargiassi, A., Goldberg, M.S., Wheeler, A.J., Plante, C., Valois, M., Mallach, G., Kauri, L.M., Shutt, R., Bartlett, S., Raphoz, M. and Liu, L. (2014) Associations between personal exposure to air pollutants and lung function tests and cardiovascular indices among children with asthma living near an industrial complex and petroleum refineries, *Environ. Res.*, **132**, 38–45.

Wallace, L., Kindzierski, W., Kearney, J., MacNeill, M., Héroux, M. and Wheeler, A.J. (2013) Fine and ultrafine particle decay rates in multiple homes, *Environ. Sci. Technol.*, **47**, 12929–12937.

Weisel, C.P., Zhang, J., Turpin, B.J., Morandi, M.T., Colome, S., Stock, T.H., Spektor, D.M., Korn, L., Winer, A.M., Kwon, J., Meng, Q.Y., Zhang, L., Harrington, R., Liu, W., Reff, A., Lee, J.H., Alimokhtari, S., Mohan, K., Shendell, D., Jones, J., Farrar, L., Maberti, S. and Fan, T. (2005) Relationships of Indoor, Outdoor, and Personal Air (RIOPA). Part I. Collection methods and descriptive analyses, *Res. Rep. Health Eff. Inst.*, **130** (Pt 1), 1–107; discussion 109–127.

Wheeler, A.J., Xu, X., Kulka, R., You, H., Wallace, L., Mallach, G., Van Ryswyk, K., MacNeill, M., Kearney, J., Dabek-Zlotorzynska, E., Wang, D., Poon, R., Williams, R., Stocco, C., Anastassopoulos, A., Miller, J.D., Dales, R. and Brook, J.R. (2011a) Windsor, Ontario exposure assessment study: design and methods validation of personal, indoor, and outdoor air pollution monitoring, *J. Air Waste Manag. Assoc.*, **61**, 142–156.

Wheeler, A.J., Xu, X., Kulka, R., You, H., Wallace, L., Mallach, G., Van Ryswyk, K., MacNeill, M., Kearney, J., Rasmussen, P.E., Dabek-Zlotorzynska, E., Wang, D., Poon, R., Williams, R., Stocco, C., Anastassopoulos, A., Miller, J.D., Dales, R. and Brook, J.R. (2011b) Windsor, Ontario exposure assessment study: design and methods validation of personal, indoor, and outdoor air pollution monitoring, *J. Air Waste Manag. Assoc.*, **61**, 324–338.