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Publisher’s version / Version de l'éditeur:

Proceedings of Indoor Air 2020, 2020-11-01

The 16th Conference of the International Society of Indoor Air Quality & Climate
ONLINE | From November 1, 2020
Paper ID ABS-0199

Determining inter-zonal flow rates using passive sampling of tracer gases

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SUMMARY
Whole-house and inter-zonal ventilation rates are important parameters to understand energy efficiency and indoor air quality. Under occupancy, passive sampling using thermal desorption (TD) tubes is more desirable due to its simple and less obtrusive nature. In this research, the uptake rates of six perfluoro-carbon tracer (PFT) gases were determined in a laboratory setting for 4 to 7-day passive sampling on two types of TD tubes. The sources of PFTs were manufactured and their stability were investigated for 4 months. The passive method was validated against the SF_6 decay method and applied to determine inter-zonal air flows in a research house. The air change rates measured with the passive release and sampling of PFT gases were comparable to those from the SF_6 method. This study shows that the passive uptake rates determined in a chamber and PFT sources manufactured in-house can be useful to measure the inter-zonal air flows in homes under occupancy.

KEYWORDS
Perfluoro-carbon tracer gas; PFT; ventilation rate; thermal desorption

1 INTRODUCTION

Inter-zonal air flows are a critical parameter to understand the transport of indoor pollutants between different zones in a building. The information is typically obtained by deploying multiple tracer gases and sampling them actively or passively. Under occupancy, passive sampling using TD tubes is more desirable due to its simple and less obtrusive nature. Knowledge of passive uptake rates for different tracer gases is a prerequisite to a successful application of the passive sampling method for inter-zonal air flow investigation.

A passive method based on passive release of PFT and passive sampling on TD tubes has been developed (Batterman et al. 2006; Lunden et al. 2012). However, the passive uptake rate of different PFT gases on TD tubes is not readily available, preventing a wide application of the method. In response to the research need, the purposes of this research are 1) to determine passive uptake rates of six PFTs in a laboratory setting for 4 to 7-day passive sampling on two types of TD tubes; 2) to validate the passive PFT method, including the measured uptake rate, against the SF_6 decay method; 3) to apply the method to measure the whole-house and inter-zonal air flows in a research house.

2 MATERIALS/METHODS

PFT sources

The PFT chemicals tested include hexafluorobenzene (HFB), octafluorotoluene (OFT), perfluoromethyl cyclohexane (PMCH), perfluorodimethyl cyclobutane (PDCB), perfluorotrimethyl cyclohexane (PTCH) and m-perfluorodimethyl cyclohexane (mPDCH). The sources of PFT chemicals were fabricated using a glass vial and a septum (Lunden et al. 2012). In addition to vial size (2 or 7 mL), different septa, including silicone rubber (SR, 1.6 mm thickness) and polydimethyl silicone membrane (PDMS, 0.13, 0.33 and 0.62 mm thickness), were used to vary the PFT emission rates (ER). The PFT sources were tested with different temperatures (10 to 36 ℃) in an environmental chamber to determine the ER as a function of temperature. The long-term stability of emitters were tested over 4 months at 23±0.5 ℃ with periodical weight measurements.

Measuring passive uptake rates in a chamber

The passive uptake rates of PFT gases were determined in a 31 m^3 stainless steel chamber for 4 to 8 days by passive sampling with two types of TD tubes (6.4 mm O.D. x 89 mm length) filled with Carbopack B (CB) or Carbograph 5TD (CG5). The TD tubes were selected with a goal of sampling PFT gases as well as volatile organic compounds (VOCs) indoors. The uptake rates of typical VOCs on the two tubes are reported in a companion paper of this conference (Won et al. 2020).

The PFT emitters were placed inside the chamber maintained at 23 ±1 ℃, 50±5 % and 0.5 or 1 air change rate per hour (ACH). Multiple CB and CG5 with a diffusion cap on the sampling side were placed on top of a stainless steel table (~1 m high) in the middle of the chamber for passive sampling. A subset of 3 - 5 tubes were removed from the chamber at 4, 6, 7 or 8 days to determine the uptake rate over time. The 4 to 8-day sampling durations were chosen since it is typical to characterize ventilation and indoor air quality over a week in a field study. Daily active sampling with CB and CG5 tubes was conducted in duplicates to determine the chamber concentrations. The passive uptake rate (UR, mL min^{-1}) was calculated by dividing the mass collected on a TD tube (ng) with the chamber concentration (μg m^{-3}) and the passive sampling time (Time, min).

The TD tubes were analysed with thermal desorption and gas chromatography/mass spectrometry (TD/GC/MS), including a Markes' Unity Series 2 thermal desorber, a multi-tube autosampler ULTRA, an Agilent 6890A GC equipped with a DB-624 capillary column (30 m × 0.25 mm I.D. × 1.4 µm thickness), and a 5973N Mass Selective Detector. The calibration mixture of PFT gases (~1 ppm for each chemical) was purchased from Air Liquide. A series of calibration tubes (5 to 400 ng) were made using gastight syringes (0.5 – 25 mL).

Validation in a research house

The passive sampling method with the measured UR was validated in a research house against the SF_6 decay method. The research house is part of the Canadian Centre for Housing Technology (CCHT) facilities in Ottawa, Canada. It is a 2–storey single-detached house with 210 m^2 liveable area and a mechanical ventilation system, representing the current housing stock in Canada (Ouazia et al. 2019). Six PFTs (3 – 4 emitters for each PFT) were placed inside the house (PDCB in basement, OFT and mPDCH on 1st floor, HFB and PMCH on 2nd floor, and PTCH on all 3 floors). The size and septum thickness of PFT emitters are the same as those summarized in Table 1. The emission rate of PFT sources was determined by weighing vials before and after the test. The interior doors were left open during the test.
CB and CG5 tubes were placed in duplicates at four locations (basement, kitchen on the 1st floor, main bedroom and bedroom 2 on the 2nd floor) for 4 and 7 days. The PFT concentrations were determined by diving the mass collected on each tube with UR and Time. Consequently, the air change rate was determined by dividing the PFT emission rate by the concentration.

The SF_6 was injected to the return air duct of the house at a rate of 40 mL min^{-1} for 12 min every

8 hours for 7 days. The injection rate was regulated with a mass flow controller and a timer. The SF_6 cylinder was located in the garage and Tygon tubing was used to deliver the SF_6 to the return air duct in the basement. SF_6 concentrations were measured with an INNOVA 1412i (LumaSense Technologies), located on top of the kitchen island.

Application to measure inter-zonal air flows in a semi-detached research house

A similar set-up for passive emitters and TD tubes was used to measure the inter-zonal air flows between two units (Unit E and F) of a duplex, which is part of the National Research Council's net-zero energy Semi-Detached Twin Test House facility. Table 1 shows the PFT emitters placed in each unit. CB tubes were placed on three floors in duplicates for 7 days.

Table 1. PFT emitters (vial volume, septum material, septum thickness) placed in two units of a semi-detached research house

	Unit E	Unit F
Basement (BM)	PDCB (7 mL, PDMS, 0.62 mm)	PTCH (7 mL, PDMS, 0.13 mm)
1st floor (1st)	mPDCH (7 mL, PDMS, 0.33 mm)	PMCH (7 mL, PDMS, 0.33 mm)
2nd floor (2nd)	OFT (7 mL, SR, 1.66 mm)	HFB (2 mL, SR, 1.66 mm)

3 RESULTS

Characteristics of PFT sources

Figure 1 (a) presents the emission rates (ER) of selected PFT emitters (7 mL vial and PDMS septum), measured at 6 different temperatures from 10 to 36 °C. It can be seen that the ER varies exponentially with the reciprocal of the absolute temperature (T) in the Arrhenius equation form. Figure 1 (b) shows the ER measured over 4 months at 23±0.5 °C. The relative standard deviation was < 1% for PDCB, PMCH and mPDCH and 2% for PTCH. Overall, the emitter showed stable ERs over 4 months.

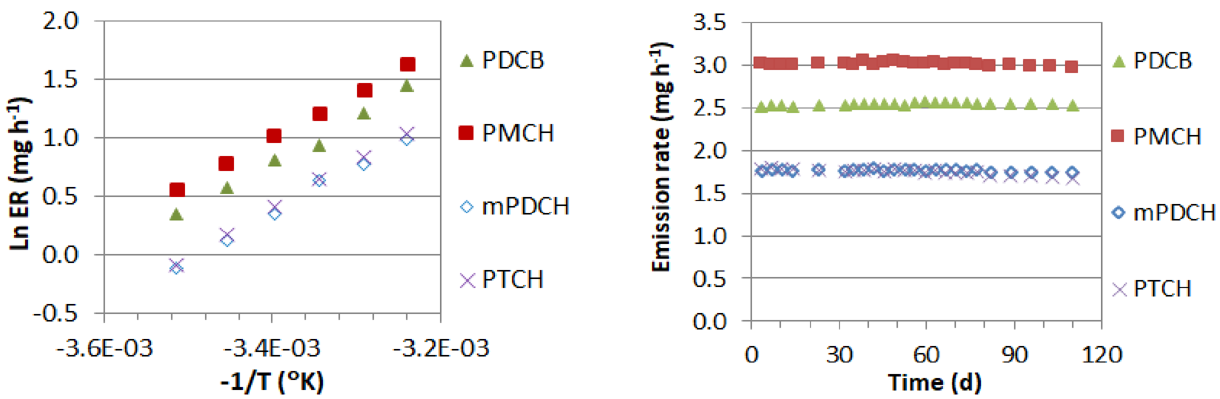

Figure 1. The emission rate (ER) of PFT emitters as a function of temperature (a) and the ER measured over 4 months at 23 °C (b).

Chamber concentrations and passive uptake rates

Figure 2 presents one example of the PFT concentrations measured daily in the 31 m^3 chamber with active sampling (a) and mass collected on CB tubes for 3 passive sampling durations (b). Similar graphs were obtained for CG5 tubes. Figure 2 (a) shows that the chamber concentrations of PFTs emitted from PFT vials reached steady state within 1 day and stayed relatively constant over 8 days. Figure 2 (b) shows that the mass collected on TD tubes was proportional to the sampling duration. Duplicate samples agreed well for both active and passive TD tubes with a difference less than 10%.

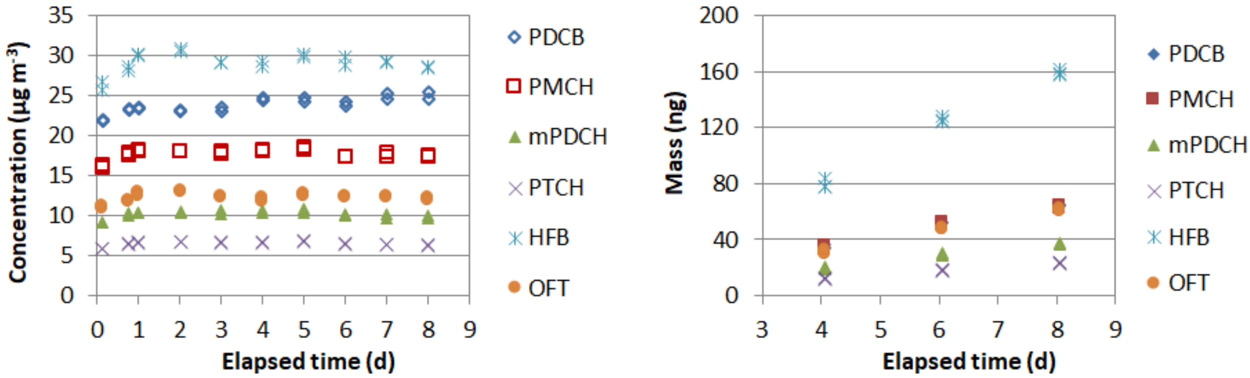

Figure 2. Chamber concentrations measured with active sampling (a) and mass passively collected on CB tubes (b).

Table 2 presents the URs measured in the chamber for CB tubes. Slightly higher URs were observed for CG5 tubes, but they are not presented here due to space constraints. The standard deviations were calculated from repeated tests. The URs were similar for 4 different sampling durations, indicating UR does not change over time from 4 to 8-day sampling. Therefore, the mean value of all tests with four sampling durations was used in subsequent studies.

Table 2. Mean and standard deviation of passive uptake rates (mL min^{-1}) measured for CB.

PFT	4 d (n = 6)	6 d (n = 3)	7 d (n = 3)	8 d (n = 3)	Mean (n = 15)
PDCB	0.26 (±0.03)	0.26 (±0.0003)	0.24 (±0.01)	0.24 (±0.01)	0.25 (±0.03)
PMCH	0.33 (±0.02)	0.33 (±0.004)	0.32 (±0.01)	0.31 (±0.01)	0.33 (±0.02)
mPDCH	0.33 (±0.01)	0.33 (±0.004)	0.33 (±0.01)	0.33 (±0.01)	0.33 (±0.01)
PTCH	0.31 (±0.02)	0.32 (±0.003)	0.32 (±0.01)	0.32 (±0.01)	0.32 (±0.01)
HFB	0.46 (±0.05)	0.50 (±0.01)	0.47 (±0.02)	0.50 (±0.03)	0.48 (±0.03)
OFT	0.42 (±0.03)	0.44 (±0.01)	0.43 (±0.02)	0.45 (±0.02)	0.43 (±0.02)

Research house test

Figure 3 (a) shows a daily pattern of SF_6 concentrations, which was repeated similarly for 7 days. The air change rate was calculated from each decay curve, leading to 21 measurements from the SF_6 decay method. The PFT concentrations from the 7-day sampling on CB tubes are also presented in Figure 3 (b). The concentrations were similar at different locations except where a particular PFT was placed. For example, the concentration of HFB was highest in main bedroom where HFB emitters were located. The concentrations of all four locations were averaged to represent the house concentrations. The concentrations at different locations were not different enough to determine the inter-zonal air flows. The total emission rates of each PFT ranged from 6.1 to 13.1 mg h^{-1}.

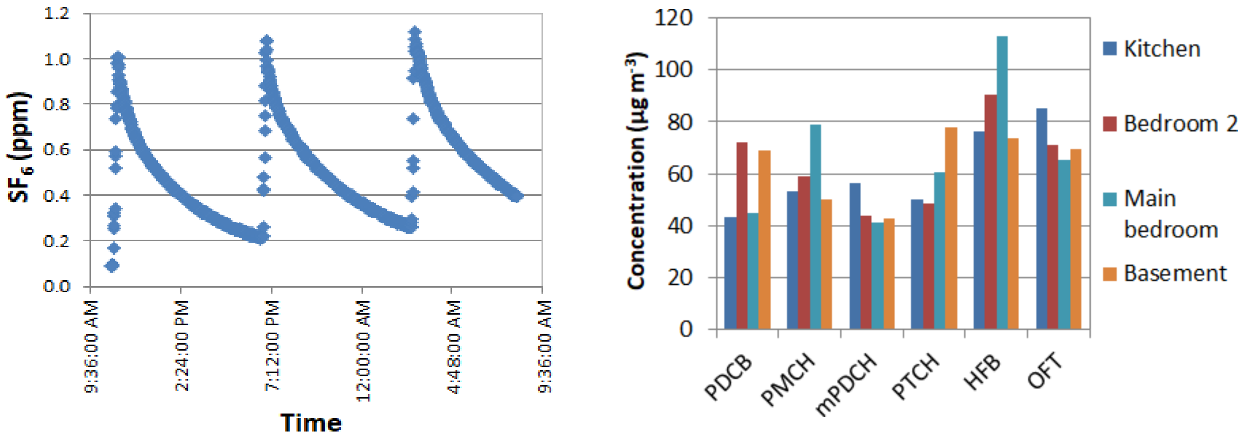

Figure 3. SF_6 concentrations at Day 1 (a) and concentrations of PFT gases measured with 7- day passive sampling on CB tubes (b).

Figure 4 compares the ACHs measured with passive sampling of PFT gases on CB and CG5 tubes with those from the SF_6 decay method. The error bars show the standard deviation of different temporal measurements (SF_6) and spatial measurements (PFTs). The passive sampling was done for 4 and 7 days. The ACHs from the SF_6 and PFT method agreed within 20% for most cases. Cases with a difference greater than 20% are highlighted in red in Figure 4, including PDCB (-27%), HFB (28%) and OFT (27%).

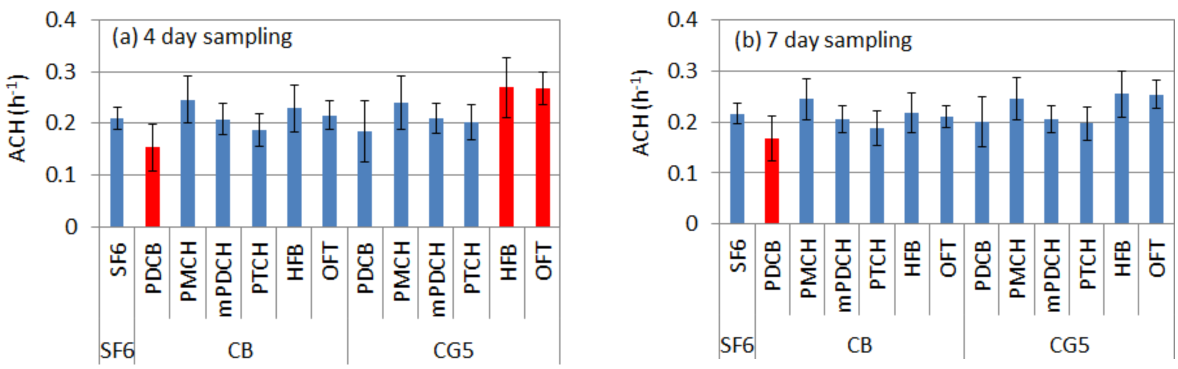

Figure 4. Air change rates measured with SF_6 and PFT gases with CB and CG5 tubes

Inter-zonal air flows in a semi-detached research house

Table 3 presents the PFT concentrations measured in two units of the duplex. Since the HRV was broken in Unit F, the concentrations were much higher in Unit F.

Table 3. PFT concentrations in NRC's semi-detached research house ($\mu g\ m^{-3}$)

PFT deployment location			Unit E			Unit F		
Unit	PFT	Location	BM	1st	2nd	BM	1st	2nd
Unit E	mPDCH	1st	22.4	30.3	20.1	78.0	68.8	63.5
	OFT	2nd	22.1	22.0	69.8	2.5	2.9	3.3
	PDCB	BM	57.6	42.0	42.7	0.0	0.0	0.0
Unit F	PMCH	1st	9.9	11.0	10.1	357.3	419.6	367.5
	HFB	2nd	10.4	10.8	11.3	478.6	598.4	728.7
	PTCH	BM	3.6	3.9	3.9	272.8	236.7	219.2

The concentrations at different locations were similar for most PFTs except for OFT. PFTs placed in Unit E were measured in Unit F and vice versa. Therefore, each unit was considered as a well-mixed zone and the inter-zonal flows between Unit E and F were calculated based on the mass balance equations (Du et al. 2012). The total concentration and emission rate of 3 PFTs in one unit were used for the calculation, treating 3 PFTs as one. The resulting inter- zonal flows were 15 $m^3 h^{-1}$ (Unit E to F) and 4 $m^3 h^{-1}$ (Unit F to E). The air flows from outdoors were 210 (Unit E) and 7 $m^3 h^{-1}$ (Unit F), leading to a whole-house ventilation rate of 0.5 (Unit E) and 0.052 h^{-1} (Unit F). The air flows to outdoors were 200 (E) and 18 $m^3 h^{-1}$ (F).

4 DISCUSSION

This study showed that the emission rates of PFT sources made of glass vials and silicone-based septa are temperature-dependent and stable over time. This indicates that the ERs can be estimated based on indoor air temperature and emitters can be used over a long-term period. These features can be helpful for field trials in remote locations where the weight measurement of PFT emitters is not feasible and the emitters need to be used for an extended period of time. The air change rates measured with the PFT method agreed with those from the SF_6 decay method within 20% for 4-day and 7-day sampling period. This supports the validity of using the passive method based on the laboratory test results in field applications.

In addition to the application example presented here, this passive method is being used to determine the whole-house ACH and inter-zonal air flows in research houses. One example of the application is presented in Ouazia et al. (2019), where the passive method was used to determine the impacts of the balanced and exhaust-only ventilation on ventilation rates and IAQ. The method is also being used to measure the inter-zonal air flows between a living space and an attic in one of CCHT's research houses from energy aspects. Although the examples show that this passive method can support various research on ventilation and IAQ studies, they are mainly associated with research houses. More work is recommended to validate and understand the inter-zonal air flows in field settings under occupancy.

5 CONCLUSIONS

A passive method, involving passive release of PFT gases and passive sampling on TD tubes such as CB and CG5, was developed and validated to determine whole-house and inter-zonal ventilation rates. The information about 6 PFT gases obtained can provide an opportunity to measure inter-zonal flows passively up to 6 zones. Due to its passive nature, this method is useful in buildings under occupancy. Since CB and CG5 tubes can also measure VOCs indoors, the method can provide information about IAQ as well as ventilation.

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