Routes of Human Exposure to Per- and Polyfluorinated Compounds (PFCs) in Winnipeg Homes

by

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Abstract

Per and polyfluorinated compounds (PFCs) include a large group of chemicals which are known to be toxic, bioaccumulative and resistant to hydrolysis, photolysis, microbial degradation and metabolism. However, human exposure pathways and toxic effects to humans are still widely unknown and more data is needed over time. The concentrations of 7 PFCs were measured in indoor air from homes in Winnipeg, Manitoba using gas chromatography-mass spectrometry. 16 PFCs were measured in house dust from Winnipeg, Manitoba using on-line solid phase extraction coupled with liquid chromatography mass spectrometry. For commonly detected PFCs in indoor air and dust, concentrations were found at pg/m^3 and ng/g levels, respectively, similar to that observed in other recent studies. Appropriate statistical tests and principal component analysis were used to evaluate possible associations between PFC concentrations and home characteristics. PFCs in indoor air and dust were associated with each other and home characteristics but not with indoor ambient temperature nor type of room (child room or the most used room). Furthermore PFCs did not show significant association with infant wheezing. None of the neutral PFCs in indoor air showed an association with seasonal temperature variation, except 8:2 FTOH and MeFOSE that had significantly higher concentrations in winter than summer.

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Last but not least I would like to thank my family with unconditional support and love.

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2. Figure 1.2. Atmospheric reaction pathway of FTOHs to produce PFCAs. Reprinted (adapted) with permission from Ellis et al., 2004. Copyright (2010) American Chemical Society. Ellis, D. A; Martin, J. W.; De Silva, A. O.; Mabury, S. A; Hurley, M. D.; Sulbaek Andersen, M. P.; Wallington, T. J. Degradation of fluorotelomer alcohols: a likely atmospheric source of perfluorinated carboxylic acids. *Environmental Science & Technology,* 2004, *38*, 3316–21, Figure 1.

1 INTRODUCTION TO PFCs-BACKGROUND, PRODUCTION, USE AND ENVIRONMENTAL BEHAVIOUR

1.1 Introduction for this Thesis

Per- and polyfluorinated compounds (PFCs) are found in indoor air and dust, with concentrations that can be correlated with home characteristics. However, the associations, exposure routes and toxic effects to humans are not well understood. Both production of PFCs and occurrence of asthma have followed a similar temporal pattern, and PFCs have been shown to have toxic effects to mouse lungs in laboratory studies.

The overall objectives of this thesis was to determine the concentrations of PFCs in indoor air and house dust, as well as correlations between PFC levels and home characteristics; and also to investigate whether or not a correlation may exist between concentrations in indoor air and dust and occurrence of symptoms of asthma such as wheezing in infants in Winnipeg, Manitoba. No other study in Canada has investigated this particular aspect of PFCs in relation to symptoms of asthma up to now. Additionally, this study aimed to investigate possible correlations among PFCs with each other as well as correlations of PFCs and both seasonal variations and room ambient temperature.

This study is part of the Canadian Healthy Infant Longitudinal Development (CHILD) study. CHILD is an extensive study which follows infants and their parents from pregnancy through the first five years of the child's life and aims to investigate the impact of environmental factors on health. The house dust and air samples as well as extensive surveys regarding home characteristics which were filled out by people who live

in homes collected by the CHILD organisation. Additionally, young children at one year of age were evaluated by a physician to assess wheezing.

The methods developed to analyze indoor air and dust samples, using gas chromatography mass spectrometry (GC/MS) and online solid phase extraction (SPE) with liquid chromatography tandem mass spectrometry (LC-MS/MS) respectively, are discussed in Chapter 2. The results of the concentrations of PFCs found using the developed methods and correlations between PFC concentrations in indoor air and dust are discussed in Chapter 2. Conclusions of this study and future directions are discussed in Chapter 3.

1.2 Per- and Poly-Fluorinated Compounds (PFCs)-Background

PFCs include a large group of chemicals, consisting of a fully or partially fluorinated hydrocarbon chain: typically C_4-C_{16}, and a hydrophilic functional group. The chemical structures of some important PFCs are given in Table 1. As a result of their unique physical-chemical properties such as chemical resistance and surfactant properties, PFCs have been used in a wide variety of industrial and consumer applications since the 1950s such as water, oil, repellents for fabric, leather, rugs, carpets and tile, fire fighting foams, alkaline cleaners, floor polish, packaging and paper products (Lau et al., 2007; Paul et al., 2009).

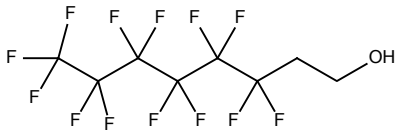
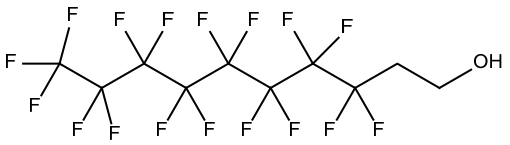
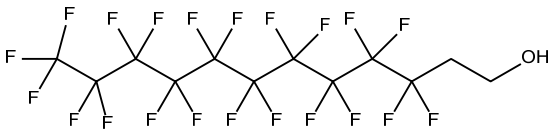
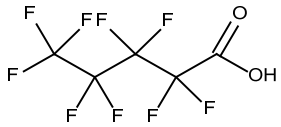
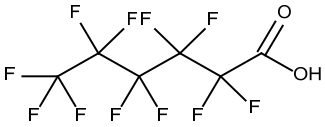
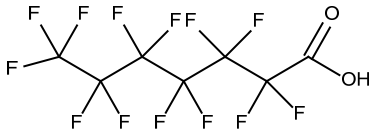
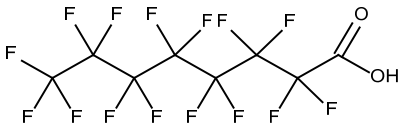
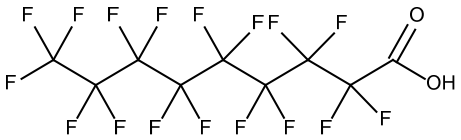
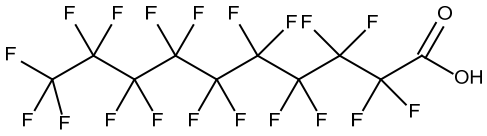
As a result of worldwide use of the chemicals over many decades, these compounds are now distributed widely in the environment and have been detected in wildlife, humans, water, air, and soil (Stock et al., 2007; Kannan et al., 2004; Rosenberg et al., 2008 ; Karrman et al., 2006; Lau et al., 2007; Martin et al., 2003; Olsen et al., 2003;

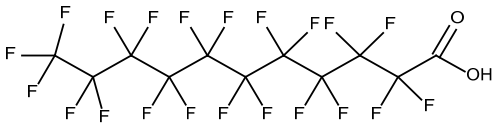
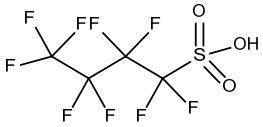
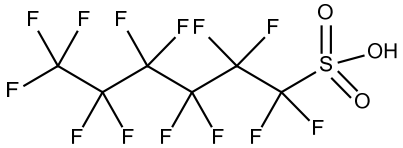
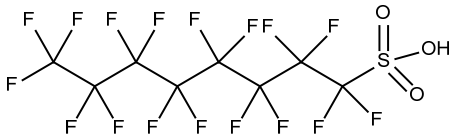
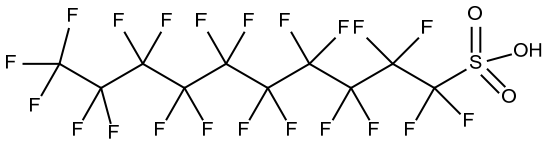
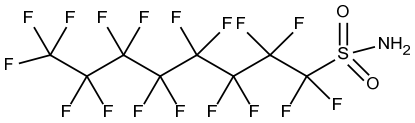
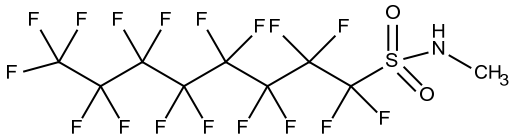
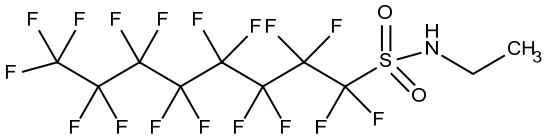
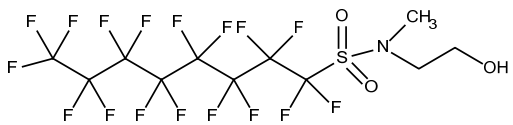
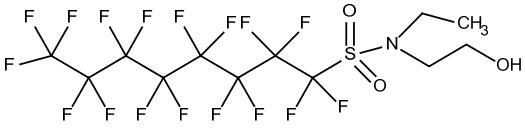
Prevedouros et al., 2006). Concern has come from the fact that some PFCs are resistant to hydrolysis, photolysis, microbial degradation and metabolism (Rosen et al., 2008); they also have a tendency to bioaccumulate and induce toxic effects. These chemicals bind to blood proteins and accumulate mainly in the liver, kidneys and bile secretions (Jones et al., 2003).

Between 1970 - 2002, 96,000 tons of perfluorooctyl-based compounds was estimated to have been manufactured globally, dominated by the 3M Company, a major global manufacturer of PFOS (Paul et al., 2009). On May 2000, 3M voluntaily phased out their perfluorooctyl-based chemistries and issued reducing PFOA emissions (Haug et al., 2009). PFOS and PFOA production has been replaced by production of shorter-chain analogs, e.g. C_4-production (Shelton et al., 2009; Shin-ya et al., 2009; Butt et al., 2010) which are believed to bioaccumulate to a lesser extent and have much less toxicity (Conder et al., 2008).

Even though 3M stopped its production in 2002, production continues in some other companies all around the world with an estimated 1000 tons per year since 2002 (Paul et al et., 2009). Production of fluorotelomer alcohols as intermediates in the production of polymers and surface coatings have been estimated to be 11,000-13,000 tonnes per year (Lindstrom et al., 2011). PFOS has been added in 2009 to the list of Persistent Organic Pollutants by the Stockholm Convention (Stockholm Convention on Persistent Organic Pollutants, 2009).

Table 1.1. List of PFCs used in this project, their abbreviations, and structures.

Compound	Abbreviation	Structure
6:2 Fluorotelomer alcohol	6:2 FTOH	
8:2 Fluorotelomer alcohol	8:2 FTOH	
10:2 Fluorotelomer alcohol	10:2 FTOH	
Perfluoro-*n*-pentanoic acid	PFPA	
Perfluoro-*n*-hexanoic acid	PFHxA	
Perfluoro-*n*-heptanoic acid	PFHpA	
Perfluoro-*n*-octanoic acid	PFOA	
Perfluoro-*n*-nonanoic acid	PFNA	
Perfluoro-*n*-decanoic acid	PFDA	

Compound	Abbreviation	Structure
Perfluoro-*n*-undecanoic acid	PFUA	
Perfluoro-1-butanesulfonic acid	PFBS	
Perfluoro-1-hexanesulfonic acid	PFHxS	
Perfluoro-1-octanesulfonic acid	PFOS	
Perfluoro-1-decanesulfonic acid	PFDS	
Perfluoro-1-octanesulfonamide	FOSA	
N-methylperfluoro-1-octanesulfonamide	MeFOSA	
N-ethylperfluoro-1-octanesulfonamide	EtFOSA	
2-(*N*-methylperfluoro-1-octanesulfonamido)-ethanol	MeFOSE	
2-(ethylperfluoro-1-octanesulfoamido)-ethanol	EtFOSE	

Recently national and international regulatory agencies prompted some government regulations that have limited or restricted the production of some PFCs in favour of shorter chain-length compounds (Shelton et al., 2009; Shin-ya et al., 2009; Butt et al., 2010) as the longer chain size is the most likely to bioaccumulate in living organisms (Conder et al., 2008).

According to the information presented in the Workshop on Managing Perfluorinated Chemicals and Transitioning to Safer Alternatives in Geneva, Switzerland in February 2009, substitution of perfluorooctyl-based products with perfluorinated chains of four and six carbons in length (Shelton et al., 2009; Shin-ya et al., 2009) has begun in most manufacturers (Santoro et al., 2009). One example of four carbon chains is perfluorobutane sulfonate (PFBS) with lower toxicity than corresponding longer chains (Lieder et al., 2009) and higher rate of elimination in rodents (Olsen et al., 2009). PFBS has been detected in children (Holzer et al., 2008), however there is largely a lack of information regarding its effects on humans.

1.3 Physical-Chemical Properties of PFCs

PFCs consist of a fully or partially fluorinated hydrocarbon chain, typically C_4 to C_{16}, attached to a polar group which can be one or more different functional groups. They called perfluorinated compounds if all C-H bonds are replaced with C-F bonds (Thibodeaux et al., 2003), and called polyfluorinated compounds if an alkyl chain is partially fluorinated (Jahnke & Berger, 2009). A perfluorinated compound presents lipopho-

bic character due to presence and properties of the polar functional group; and also presents hydrophobic character due to the presence of non polar polyfluorinated carbon chain in its structure. Fluorotelomer alcohols (FTOHs) contain hydroxide groups as the functional groups, perfluorocarboxylic acids (PFCAs) contain carboxylic, perfluorosulfonates (PFSAs) contain sulfonates, and FOSEs and FOSAs contain sulfonamides as the functional group. These functional groups can be anionic, cationic or neutral.

Due to strong covalent bonds present between the fluorine and carbon atoms which make PFCs stable in the environment, the compounds are resistant against various kind of degradation such as reaction with acids and bases, oxidation, and reduction (Kissa et al., 2001; Hekster et al., 2003; Olsen et al., 2007). Because of the unique properties of PFCs, they have been used for various applications. Their physical and chemical properties made them ideal as surfactants (Lau et al., 2007). The length of the fluorinated chain could affect the properties and behavior of PFCs, such as volatility of the compounds, as well as the rate of elimination (in monkey and rat) (Upham et al., 2009; Lei et al., 2004). Generally, the rate of elimination for PFCAs and PFSAs was found to decrease with increasing chain length (Upham et al., 2009; Kim et al., 2011). Volatility increases with decreasing chain-length in fluorotelomer alcohols (Table 2) (Lei et al., 2004; Stock et al., 2004a).

The FTOHs are a group of perfluorinated compounds consisting of an even number of fully fluorinated aliphatic carbon atoms along with two non-fluorinated carbon atoms adjacent to the hydroxyl group. Their general chemical formula is CF_3-$(CF_2)_n$-CH_2-CH_2OH, where n = 3, 5, 7, 9. Their names are based on the ratio of fluorinated to non-fluorinated carbon atoms, e.g. 8:2 FTOH (Stock et al. 2004a).

There are some limitations to determine physical properties of PFCs such as partitioning coefficients and vapour pressure. For example it is difficult to measure the vapour pressure of PFOA due to its low volatility and high stability; hence, experimentally it takes a long time (several days) to determine the vapour pressure (Barton et al., 2008). Using models to predict K_{ow} values of PFCs is not appropriate because of their unique properties (Houde et al., 2006); PFCs as surfactants tend to be present at interfaces of different solutions. This makes estimation of K_{ow} difficult because ionic surfactants will present in an aqueous phase as ions, but will need to become neutral to partition to an organic phase; this results in inaccurate estimations (Tolls & Sijm, 1995). Some physical-chemical properties of PFCs are given in Table 2.

Table 1.2. Molecular weights (MW), vapor pressures (VP) partitioning coefficient* of seven PFCs.

Compound	MW [g/mol]	VP [Pa] at 25 °C	log K_{oa}	log K_{aw}	log K_{ow}	pK_a
6:2 FTOH	364	876 [a] / 713 [b]	3.6 [a]	1.7 [a]	NA	NA
8:2 FTOH	464	227 [a] / 254 [b]	4.2 [a] / 5.5 [d]	1.3 [a]	1.8 [d]	NA
10:2 FTOH	564	53 [a] / 144 [b]	4.8 [e]	NA	NA	NA
NMeFOSE	557	0.7 [a] / 0.002 [c]	6.8 [a]	NA	4.8 [d]	NA
NEtFOSE	571	0.35 [a] / 0.009 [c]	7.1 [a]	NA	NA	NA
PFOS	500	0.000331 [g] /3.5 [d]	7.8 [d]	-2.40 [d] < $2x10^{-6}$ [g]	NA	NA
PFOA	414	0.32 [d]	6.8 [d]	-2.37 [d]	4.3 [d]	0.7 [c]

NA = Not available.

a: Lei et al., 2004, VPs at 25 °C

b: Stock et al., 2004a, VPs at 25 °C

c: Shoeib et al., 2004, VPs at 23 °C

d: Arp et al., 2006

e: Goss et al., 2006

g: 3M Company, 2003

* The partition coefficients (the equilibrium ratio of solute concentrations in the two phases) are as below:

K_{ow}: Octanol-water partition coefficient

K_{oa}: Octanol-air partition coefficient

K_{aw}: Air-water partition coefficient

pK_a : Logarithmic measure of the acid dissociation constant

1.4 Production

There are both direct and indirect sources of PFCs emissions to the environment. Direct sources result from the manufacture of PFCs, whereas PFCs indirectly may originate from abiotic and biotic transformation of commercially synthesized precursors (Prevedouros et al., 2006). PFCs are produced by two major manufacturing processes. One of the processes is electrochemical fluorination (ECF), applied since the 1970s to yield mixtures of linear and branched isomers (Giesy et al., 2002). In ECF, hydrocarbon alkylsulfonyl halides and alkylcarbonyl halides using hydrogen fluoride in presence of an electric current are converted to the corresponding perfluorinated acid halide analogue (3M Company, 1999). This process, which is based upon the electrolysis of a hydrocarbon analogue of the target PFC in liquid hydrogen fluoride, replaces the C-H bonds of the alkyl chains with C-F bonds. Breakage and rearrangements of the C–C bond is not always complete. Therefore, homologues and branched isomers are formed as by-products along with the linear products. The branched isomer content is batch-dependent and varies between 10% to 30% (Karrman et al., 2007; De Silva and Mabury., 2004). An example for electrochemical fluorination process (reaction 1-1) is the production of POSF (perfluorooctane sulphonyl $C_8F_{17}SO_2F$) from 1-octanesulphonyl fluoride ($C_8H_{17}SO_2F$) (3M Company, 1999):

$$2C_8H_{17}SO_2F + 34HF \longrightarrow 2C_8F_{17}SO_2F + 17H_2 \qquad \text{(reaction 1-1)}$$

POSF is produced and used as the initial material for further PFOS-related chemicals.

Manufacturing sulfonyl-based fluorochemicals using the ECF process was started by the 3M Company in the 1940s and lasted until 2002, when 3200 tons of POSF-derived chemicals were produced (EPA, 2000).

The other manufacturing process is telomerization which has been used since the 1950s for production (Prevedouros et al., 2006; De Silva & Mabury, 2006; Kissa, 2001). Telomerization products are mostly straight chains rather than branched isomers. Hence products obtained by telomerization have generally higher purity than products obtained by ECF (Karrman et al., 2007). Telomerization of tetrafluorethylene with pentafluoro-ethyl iodide produces a mixture of telomeres with different even carbon numbers in their overall carbon chain lengths (Figure 1) (Schultz., 2003; Kissa., 1994).

Example reaction of telomerisation manufacturing process (reactions 1-2, 1-3, 1-4) (Hekster et al., 2003):

$$5C_2H_4 + IF_5 + 2I_2 \longrightarrow 5C_2F_5I \qquad \text{(reaction 1-2)}$$

$$C_2F_5I + nC_2F_4 \longrightarrow C_2F_5(C_2F_4)_nI \qquad \text{(reaction 1-3)}$$

$$C_2F_5(C_2F_4)_nI + C_2H_4 \longrightarrow C_2F_5(C_2F_4)_nC_2H_4I \qquad \text{(reaction 1-4)}$$

The total direct and indirect globally mass production of PFCAs was estimated to be 3200-7300 tons from 1951 to 2004 (EPA, 2000). The reported production of the chemicals by 3M, the major manufacturer of sulfonyl-based PFAS, was 3×10^6 kg in 2000 (EPA, 2000). The total fluoropolymer manufacturing capacity for 33 fluoropolymer

manufacturing sites worldwide located in North America, Japan, China, Europe, Russia and India was estimated to be 144,000 tonnes in 2002 (Ring et al., 2002).

3M produced 6.5 million pounds in 2000 of carboxylated and sulfonated PFC (Karrman et al., 2007). Because of environmental concerns, 3M completely phased out their C_8-production by the end of 2002. Production has been substituted by shorter analogs, e.g. C_4-production, which is believed to bioaccumulate to a less extent and have much less toxicity than longer-chain analogs (Betts et al., 2003). However the telomerization process has been used by various companies since the 1970s for the production of FTOHs. Production of FTOHs was estimated to be 5×10^6 kg/year in various products. (EPA, 2002). FTOHs have been shown to degrade via abiotic (Ellis et al., 2004; Hurley et al., 2004a) and biotic (Hagen et al., 1981) mechanisms to PFCA.

1.5 Uses and Consumer Applications

The properties such as lipophobicity, hydrophobicity and surfactant character of PFCs allow for their multiple uses. Applications of the perfluorocompounds can be classified into three main categories: surface treatments, paper protectors and performance chemicals.

The purpose of surface treatment is to provide resistance to soil, oil and water for carpet, fabric and leather. The US EPA reported that 37% of the production of fluorinated surfactants is devoted for surface treatment applications (US EPA 2000). One of the by-products of the telomerization process is perfluorooctanoate, i.e., the anion of PFOA, which may be present in treated items such as carpets (Washburn et al., 2005).

For paper protection, sulfonated fluorochemicals were used to provide grease, oil

and water resistance for paper products, such as plates, food containers and bags. It has been reported that 42% of sulfonated fluorochemicals were applied for coatings on paper consumptions in 2000 (US EPA 2000). EtFOSE has been reported as the initial material in the production of paper protection products, such as food packaging (Stock et al., 2004). In the production of some products such as adhesives, inks, polymers, waxes, polishes and paints, FTOHs are produced as intermediates (Stock et al., 2004).

Performance chemicals are used for many different industrial, commercial and consumer applications including fire-fighting foams, surfactants, alkaline cleaners, floor polishes, photographic films, denture cleaners, and insecticides used to control cockroaches, termites and ants (Kannan et al., 2002). Home carpet stain repellent products such as Scotchgard contain MeFOSE (Dinglasan-Panlilio & Mabury, 2006). One of the applications of PFOA is in the production of fluoropolymers and fluoroelastomers such as polytetrafluoroethylene (PTFE) (Lau et al., 2007). In recent decades, FOSAs have been used in manufacture of surfactants, or as polymer components (Wang et al., 2009). It has been reported that part of the release of PFCs into the environment results from non-bound residual PFCs such as FOSEs and FOSAs in consumer products (3M Company, 1999; Stock et al., 2004b).

1.6 PFCs in the Environment

The presence of PFCs in the environment has been reported since the 1980s (Begley et al. 2005), but improvement of detection techniques provided more evidence for global contamination. As a result, PFCs were detected in various samples distributed around the world, both near and far from manufacturing sources and populated regions.

The presence of PFCs in remote areas indicates that the compounds are being transported and distributed globally. The wide distribution of PFCs in the environment gained more attention when they were detected in human blood from Arctic regions (Jons et al., 2003; Weihe et al., 2008) and in top predators such as seabirds, ringed seals and polar bears from polar regions (Begley et al 2005). The presence of PFCs in wildlife in remote or polar regions indicates that the chemicals are being transported and distributed globally. Therefore, levels of PFCs were measured in a variety of environmental media all around the world such as remote, industrial, urban and Arctic regions, as noted below.

PFCs have been widely detected in the arctic environment. Transport mechanisms of PFCs to the Arctic are complex. The major potential transport pathway has been suggested to be an indirect pathway, which is the transportation of volatile precursors including FTOHs and FOSEs via the atmosphere, followed by degradation (atmospheric oxidation to PFCAs and PFSAs) and subsequent wet and dry deposition (Butt et al., 2010). PFCAs and PFSAs have been reported as degradation products of FTOHs and FOSEs, respectively, in the atmosphere (Ellis et al., 2004; Hurley et al., 2004a; D'eon et al., 2006; Martin et al., 2006). The other major pathway is direct transportation of PFCs via oceanic currents to the polar environment (Armitage et al., 2006; Wania et al., 2007). Indirect transport of PFCs to Arctic has been supported by some evidence as below:

- Volatile and semi volatile PFCs (FTOHs, FOSEs and FOSAs) can undergo long-range transport having sufficient atmospheric lifetimes (20 – 50 days) (Martin et al., 2006; D'eon et al., 2006; Ellis et al., 2003).
- Detection of degradation intermediates (8:2 and 10:2 fluorotelomer unsaturated carboxylate (FTUCA)) and final products (PFOA and long-chain PFCAs) in the atmospheric

particles in the Arctic. (Stock et al., 2007).

- Detection of PFOS and PFCAs in snow from remote regions in Arctic. Also PFCs have been detected in various compartments surface waters, sediment and fish in Arctic that they have been influenced by atmospheric deposition. (Stock et al., 2007).
- The time-scale of oceanic transport is too short to explain the observed variations in PFOS levels in some species in Arctic. If these variations are resulted from emission changes, ocean currents could not reflex these changes due to long delays (~ 30 years) (Butt et al., 2010).

The direct transport pathway (i.e. transport through oceans) is supported by results obtained from global transport models. The models show that produced PFOA from atmospheric degradation of FTOHs is not enough to make the current seawater concentrations (Hart et al., 2009., Wania et al., 2007). The net flux of PFOA to the northern polar zone was estimated to be 8–23 tonnes per year (Armitage et al., 2006). This flux was reported as 20 to 60-fold greater than that predicted from FTOH degradation (Butt et al., 2010;Wallington et al., 2006). Therefore these modeling results support the direct emissions PFOA in the arctic surface water. Direct transport of the chemicals to the Arctic is less important than indirect transport in circumstances that production of PFOA and PFOS decreases in source regions.

1.6.1 Air

The distribution and presence of PFCs in the atmosphere is influenced by the atmospheric half-life and volatility of the PFC. Volatile PFCs (FTOHs) and semivolatile PFCs (FOSAs and FOSEs) are common in atmospheric air in urban regions and industrial

areas (Dreyer et al., 2010; Prevedouros et al., 2006). Their concentrations decreased with increasing distance from source regions (Dreyer et al., 2010). Lifetimes (residence times) of FTOHs in the atmosphere are assessed to be 10–80 days (Ellis et al., 2003; Piekarz et al., 2007; Dreyer et al., 2009). For gas–phase alkyl–FBSA/Es (*N*-ethyl perfluorobutane-sulfonamide and *N*-methyl perfluorobutane sulfonamidoethanol), lifetimes are 2–50 days (D'eon et al., 2006; Martin et al., 2006). The relatively long atmospheric lifetimes of FTOHs and FOSAs/FOSEs mean that they are capable of long-range atmospheric transport, including from temperature regions where they are most heavily used to colder regions. It is believed that FTOHs could reach to the Arctic before degradation (Shoeib et al., 2006). However, it has been suggested that they undergo wet or dry deposition (Dreyer et al., 2010). The deposition is considered to be too low and insignificant for removal of FTOHs in the atmosphere (Dreyer et al., 2010; Hurley et al., 2004a). Of the PFCs which are removed from the air by deposition, the more volatile ones could be revolatilised again and thus move further. Revolatilisation is not common for compounds such as PFASs and PFCAs, which have high water solubility and low vapor pressure (Arp & Goss, 2009). Table 3 shows concentrations of FTOHs, FOSAs and FOSEs in outdoor air from Europe and North America. Differences between the data from different studies may be due to a diversity of sampling approaches to air and methodology of analysis (Harrad et al., 2010).

Table 1.3. Median outdoor air concentrations of selected PFCs (pg/m^3).

Country	6:2 FTOH	8:2 FTOH	10:2 FTOH	Me-FOSA	Et-FOSA	Me-FOSE	Et-FOSE
Canada, urban[a]	85	117	43	3.2	1.6	7.6	2.4
UK, rural[b]	81	100	75	5.5	7.9	24	9.2
Germany, rural[c]	29	85	28	7.4	3.2	6	8.4
Canada, rural[d]	29	32	17	NA	NA	35	76
USA, remote[e]	4.6	24	15	NA	< 3.2	< 11	< 3.7
Norway, suburban[f]	12	34	17	5.5	7.9	24	9.2
Canada, urban[g]	87	55	29	NA	14	100	210
Germay, urban[h]	26	65	20	2.6	2.8	2.6	1.2
Canada, urban[i]	18	41	22	NA	NA	12	3.3
Germay urban[j]	56	110	29	7	2.6	18	3.5
Canadian Arctic[k]	< 14	9	< 1.5	20	11	31	< 27
Germany marine[l]	8	38	9.6	2.9	1.1	1.6	0.7

NA = Not available

a. Shoeib et al., 2011

b. Barber et al., 2007

c. Jahnke et al., 2007

d. Martin et al., 2002

e. Piekarz et al., 2007

f. Barber et al., 2007

g. Martin et al., 2002

h. Dreyer & Ebinghaus., 2009

i. Shoeib et al., 2006

j. Jahnke et al., 2007

k. Stock et al., 2007

l. Dreyer & Ebinghaus., 2009

1.6.2 Water

Concentrations of PFCs in the environment are highly variable and depend on the parameters such as environmental compartment, location, sources, sinks, and volatility of the compounds (Lohmann et al., 2007, Giesy & Kannan, 2001). It is believed that oceanic transportation of PFCs is the greater part of transportation to the Arctic (Yamashita et al., 2005). Sources of PFCs in the ocean are atmospheric PFCs that undergo wet and dry deposition as well as wastewater and discharges to rivers (Simcik & Dorweiler, 2005). The presence of PFCs in the water can cause contamination of drinking water and food web (Schuetze et al., 2010).

Table 4 shows typical concentrations of these chemicals in various aqueous environments around the world. This data indicates that contamination by PFCs could happen even in regions where there is no industrial or other activities involving PFC production. PFC concentrations in the rivers located close to industrial regions are generally higher than regions where there is no industrial activity (Pan & You, 2010).

Large amounts of PFCs are transferred to waters from urban regions via their direct deposition to rivers from populated areas. They come through wastewaters of fluorotelomer manufacture (Davis et al., 2007), as well as sewer wash-off from buildings and streets, surface runoff, and rainfall in urban regions (Loewen et al., 2005, Murakami et al., 2008; Scott et al., 2006, Dreyer et al., 2010). As can be seen in Table 4, relatively higher concentrations of PFCs in German rivers compared to other places (exept the ground water at the training base contaminated by AFFF spillage) comes from industrial plants located at the river bank. Atmospheric deposition of PFCAs and atmospheric deg-

radation of FTOHs are contamination sources for rural regional waters other than discharge from wastewaters and other human-use waters (Scott et al., 2006).

Table 1.4. Concentrations of selected PFCs in water samples (ng/L).

Type of water (Country)	PFOS	PFOA
River (Japan)[a]	6.5	59
River (Germany)[b]	220	1600
River (China)[c]	4.7	5.4
Sea (Japan)[d]	2600	64
Surface ocean water (S. Atlantic)[e]	< 0.01	< 0.004
Lake & Pond (China)[c]	4	3.9
Ground & river, rural (China)[c]	0.4	0.1
Ground & river, urban (China)[c]	5.7	4.1
Contaminated area,153 days after release of 22000 L AFFF (Canada)[f]	320	20
Ground water at fire-training base (USA)[g]	32000	33000
Drinking water (USA)[h]	1.2	< 5
Tap water (China)[c]	1.8	3.4

AFFF = Aqueous Film Forming Foam

a. Senthilkumar et al., 2007

b. Skutlarek et al., 2006

c. Jin et al., et al., 2009

d.Taniyasu et al., 2005

e. Ahrens et al., 2009

f. Moody et al., 2002

g. Moody et al. 2003

h. Quinones et al., 2009

Contamination from wastewater arises from urban wash-off, industrial releases, and the washing of clothes, utensils and other indoor materials. It has been shown that after washing of PFC-coated clothing, about 73% of the PFCs had been washed off over almost 2 years (3M Speciality Materials, 2000). In waste water treatment plants mainly PFSAs and PFCAs remain within the effluent, however, some semi-volatile compounds are released into the atmosphere and are discharged into rivers (Becker et al., 2008; Sinclair & Kannan, 2006).

Tap water is also polluted by PFCs since the treatment processes are not able to remove the compounds, particularly PFOS and PFOA. As shown in Table 4, concentrations of these compounds in tap water can be comparable to those in river water (Jin et al., 2009; Loos et al., 2007) and can lead to considerable human exposure compared to potential exposure from ground and river waters in Table 4 (Hölzer et al., 2008).

Results of a study from Canada indicates that surface water from four lakes on Cornwallis Island in the Canadian Arctic (Amituk Lake, Char Lake, Resolute Lake and Meretta Lake) had PFOS and $C_7 - C_{12}$ PFCAs persent in all lakes at 0.3-10 ng/L (Stock et al., 2007). The mean value of PFOS and PFOA concentrations ranged from 23-69 ng/L and 5.6-14 ng/L, respectively (Stock et al., 2007). Two lakes (Resolute Lake and Meretta Lake) showed significant differences in concentrations of some PFCs such as PFHxS,

PFOS, PFHpA and PFOA. The levels were up to 60-fold higher compared to the other lakes (Amituk Lake and Char Lake) in the study. The high levels of the mentioned compounds in the lakes resulted from AFFF contamination from the local airport and from sewage runoff. Intermediate FTOHs degradation products, 2H-perfluoro-2-decenoic acid (8:2 FTUCA) and 2H-perfluoro-2-dodecenoic acid (10:2 FTUCA), were also detected in all lakes with mean concentrations of up to 1.9 ng/L for 8:2 FTUCA and up to 6.4 ng/L for 10:2 FTUCA. Quantification of PFCs in surface seawater samples from the Greenland Sea indicates that PFOA (30–111 pg/L) and PFOS (10–90 pg/L) were the major PFCs in the system (Theobald et al., 2007).

1.6.3 Biota and Humans

PFCs have been detected widely in biota due to their high volume of use and the widespread environmental distribution of the chemicals. They have been detected from biota at the bottom of the food web to the top predators (Martin et al. 2004). Some detected values of the PFCs in biota including birds, mammals, and aquatic animals from various countries are given in Table 5.

From laboratory studies results perfluorinated compounds, particularly PFOS and other PFCs, can be acutely toxic to fresh water organisms (Boudreau et al., 2003; Latala et al., 2009). Considering the fact that fish eat algae, microorganisms and small bottom feeders, hence the presence of pollutants in fish can be a criterion for levels of the pollutants in the aquatic environment (Butt et al., 2010). Detection of these chemicals in oceanic biota and fish indicates capability of the chemicals for long range environmental transport. Tomy et al. proved this by measuring the concentrations of the chemicals in

various fishes from the Arctic Ocean (Tomy et al., 2004).

Table 1.5. Concentrations of PFOS and PFOA in some biota (ng/g wet weight).

Location	Animal Liver	PFOS	PFOA
Japan[a]	Cattle	33	< LOD
	Chicken	67	< LOD
	Pig	54	0.04
Alaska[b]	Sea Otter	2.8	ND
Japan[c]	Cormorants	130	2.7
	Eagle	43	2.1
	Crow	6.6	0.31
Souhtern Sea[d]	Sea Otter	55	60
Eastern Arctic[e]	Walrus	2.4	0.34
	Beluga	13	1.6
	Glaucous gull	20	0.14
	Narwhal (Monodon monoceros)	11	0.90

NA = Not available

LOD = Limit of detection

a. Guruge et al., 2008

b. Hart et al., 2009

c. Senthilkumar et al., 2007

d. Kannan et al., 2006

e. Tomy et al., 2004b

Furthermore, availability of the compounds in fish highlights a potential human exposure pathway via fish consumption, particularly for coastal populations that consume fish more than the average population (Berger et al., 2009). PFOS, PFOSA, PFOA and *N*-EtFOSA were detected in zooplankton, shrimp and clams from the eastern Canadian Arctic (Tomy et al., 2004). Mean concentrations of PFOS were found to be 1.8, 0.35 and 0.28 ng/g in the zooplankton, shrimp and clams, respectively, while PFOA levels were 2.6 and 0.17 ng/g in zooplankton and shrimp. Therefore, human dietary exposure in coastal populations is likely due to either bioaccumulation of the compounds in the food chain, or direct consumption of clam and shrimp by humans.

Marine mammals were analysed for PFCs (Tomy et al. 2004). PFOS was found to be approximately similar and in the range of 11-12.5 ng/g in narwhal and beluga from the eastern Canadian Arctic. Analysis of fishes from Quebec, Canada showed mean concentrations of 5.7 ng/g in white sucker to 39 ng/g in brook trout, while PFOA was below the method detection limit (Martin et al., 2004). PFCs in freshwater ecosystems have been studied in a few studies mostly from the Canadian Arctic. PFOS has been reported widely in freshwater fish but other PFSAs such as PFHxS and PFHpS have been detected in very few reports (Butt et al., 2010). PFNA and PFDA are commonly found in freshwater fish (Stern and Tomy, 2007; Martin et al., 2004; Stern et al., 2007; Kallenborn et al., 2004; Muir et al., 2008).

Besides PFOS, PFOSA and C_9-C_{11} PFCAs which were generally detected in terrestrial wildlife, some of the longer-chain length PFCAs such as perfluorododecanoate PFDoA has been also detected in these species (e.g. at 10.8 ng/g ww in Rangifer tarandus) (Martin et al., 2004; Tittlemier et al., 2005).

There are limited data available on PFC levels in humans. It has been suggested that precursor compounds to PFOS and PFOA have a limited contribution to total exposure (Fromme et al., 2009; Vestergren et al., 2008). It was reported that 5% of FTOHs and 20% of FOSAs and FOSEs convert to PFOS and PFOA in humans, and hence the total contribution towards degradation products of precursors such as these will be minimal (Fromme et al., 2009).

1.6.4 Soil and Sediment

Sources of PFCs to sediment are suggested to be atmospheric deposition, the spreading of sludge on agricultural land, and PFC-containing pesticides (Martin et al., 2004; Ahrens et al., 2010). Different studies have shown adsorption of PFC to soil or sediment could be through at least two different types of interactions: first, a hydrophobic interaction of the perfluoronated chain (e.g., with the organic carbon fraction of the soil or sediment), and second, an electrostatic interaction of the functional group (e.g., to the charged clay fraction of the soil or sediment). Although partition coefficients such as the K_d or the K_{OC} are used to express the sorption behavior of PFC (Pan et al., 2009), there are other factors that could also affect sorption behavior and distribution of PFC, such as the fluorinated chain length. It was reported that short-chain PFCAs ($C < 7$) were found exclusively in pore water while long-chain PFCAs ($C > 11$) were found only in sediment (Ahrens et al., 2009). This suggests that the longer the perfluorinated carbon chain, the higher the tendency to adsorb to solid matrices. Therefore, it is more likely to find short chain PFC in aqueous matrices, while long chain PFCs are predominantly in solid matrices. Besides the effects of K_{ow} and K_d on sorption, strong effects of Ca^{2+} concentration

and pH of soil matrices on sorption have been reported (Pan et al., 2009, Becker et al., 2008, Higgins and Luthy, 2006). Reports showed a decrease in adsorption with increasing pH (Johnson et al., 2007). An explanation for these effects can be variations of the electrostatic Coulomb potential of a sorbent as a function of pH and of cation concentrations (Higgins and Luthy, 2006; You et al., 2010). PFCs are detected mainly in upper sediment layers and in top soil, which could be due to either the presence of higher contents of organic materials and protein in the layers, or the fact that recently deposited soils and sediments are more contaminated with PFCs. Reported concentrations of PFCs in sediment from various locations around the world are summarized in Table 6.

Table 1.6. Concentrations of selected PFOS and PFOA in sediment (ng/kg).

Sediment type and location	PFOS	PFOA
Austria, Lake Constance[a]	< 940	380
China, Hun River site (0 - 10cm)[b]	170	130
Canada, Resolute Lake, (0 - 1cm) (associated with a spillage of AFFF from the airport)[c]	85000	7500
USA, Bolinas Lagoon[d]	380	290

ND = Not detected

a. Bao et al., 2009

b. Higgins et al., 2005

c. Stock et al., 2007

d. Beach et al., 2006

1.6.5 Indoor Air

Indoor air measurements of PFCs are approximately one to two orders of magnitude higher than that of outdoor air (Harrad et al., 2010; Shoeib et al., 2011). This is because indoor air is exposed to various PFCs in items such as carpeting and furnishings (Vestergren et al., 2008; Trudel et al., 2008; Yamada et al., 2005). Indoor air PFC concentrations from some studies are given in Table 7. Fluorotelomer alcohols were determined in the gas phase of indoor air (Barber et al., 2007) for the first time. The geometric mean sum concentration of fluorotelomer alcohols was determined to be 11,075 pg/m^3; that of FOSEs/FOSAs was observed to be 14,900 pg/m^3. A study in 2011 from private homes in Vancouver, Canada showed that indoor air was dominated by 8:2 FTOH with a geometric median concentration (pg/m^3) of 2,720 followed with 6:2 and 10:2 FTOHs which had concentrations of 1040 and 980 respectively whereas FOSEs/FOSAs had a range of 20-300 pg/m^3 (Shoeib et al., 2011).

Jahnke for the first time compared levels of PFCs in indoor air homes with indoor air offices. Lower levels of the compounds detected in offices which was reported to be due to presence of some typical sources such as commercial products and PFC coated furniture at those homes (Jahnke et al., 2007).

Up to now only one study has measured indoor air concentrations of PFOS and PFOA in the gas phase. The report shows a mean PFOA concentration of 113 pg/m^3, but PFOS levels were all below the detection limit of 0.02 pg/m^3 (Shoeib et al., 2011). While the indoor air PFCA and PFSA levels were not significantly elevated, volatile polyfluorinated compounds (FTOHs, FOSA/Es) appear to be considerably higher in indoor air. This could result from higher application of the compounds in indoor environment as

well as higher vapour pressure of the chemicals (Barber et al., 2007; Shoeib et al., 2011).

Table 1.7. Median indoor air concentrations (pg/m^3).

Country	6:2 FTOH	8:2 FTOH	10:2 FTOH	EtFOSA	MeFOSA	MeFOSE	EtFOSE
Canada [a]	1040	2720	980	19	21	320	56
Norway[b]	933	5173	2822	0.5	8.3	265	78
Japan[c]	290	5840	1120	NA	NA	NA	NA
Spain[d] (range)	3 - 47	7.5 - 170	< 0.6 - 47	< 0.52 - 6.1	< 1.2 - 14	< 0.52 - 6.1	< 0.52 - 6.1

NA= Not available

a. Shoeib et al., 2011

b. Haug et al., 2011

c. Liu et al., 2013

d. Ericson Jogsten et al., 2012

1.6.6 Dust

It has been indicated that there is an association between dust exposure and allergies in humans, therefore, dust could be considered as an exposure pathway for various man-made chemicals (Roberts et al., 2009). Dust particles can be defined as particles with an approximate diameter of < 500 μm (Lewis et al., 1999). The particles can be produced from micro-fibres abraded from textiles, fabric furnishings, wear and tear of products, degradation of materials and deposition of airborne components, such as pollens, soil minerals and volatile organic compounds (VOCs).

In the last decade there have been several studies on measuring PFC concentrations in house dust (Strynar and Lindstorm., 2008; Fromme et al., 2008; Björklund et al., 2009; Shoeib et al., 2011; D'Hollander et al., 2010; Haug et al., 2011; Knobeloch et al., 2012). The reports that measured PFC concentrations in house dust often focused on measuring the less volatile PFCAs and PFASs with a few exceptions (Shoeib et al., 2011; Xu et al., 2013; Strynar and Lindstrom, 2008) that have measured also volatile PFCs (FTOHs). Dust samples from Japanese houses, analysed in 2003, had ranges of 11 to 2500 ng/g and 69 to 3700 ng/g for PFOS and PFOA respectively (Moriwaki et al., 2003). Higher concentrations of PFOA than PFOS were detected in all the samples in this study. Dust samples from Canada were found to have higher concentrations (ng/g) of PFOA (19.72), PFOS (37.8) and PFHxS (23.1) compare to FOSA which not detected in 90% of the samples (Kubwabo et al., 2005). PFBS was not found in any of the samples; an explanation for that could be that PFBS had just come to the market at the time of this study. Strynar and Lindstrom analysed dust samples from both homes and day-care centers in North Carolina and Ohio (Strynar and Lindstrom, 2008). This was the first study

that PFHxA, PFHpA, PFNA, PFDA, PFUA, and perfluorododecanoic acid (PFDoA) were analysed. PFOS was the dominant compound with a concentration of 35,700 ng/g. PFBS was found in 33% of the samples. 6:2, 8:2, and 10:2 FTOHs were found in 50% of the samples in range of 24 and 33 ng/g (Strynar and Lindstrom, 2008). MeFOSE, Et-FOSE, EtFOSA, and MeFOSEA in indoor dust samples were analysed for the first time by Shoeib (Shoeib et al., 2005). MeFOSE and EtFOSE showed the highest concentrations i.e 113 and 138 ng/g, respectively.

A wide variety of PFCAs and PFAS have been studies in a few studies in North America and Europe (Haug et al., 2011; Shoeib et al., 2011; Kato et al., 2011; Strynar et al., 2008). In all these studies, PFOA and PFOS in house dust have been detected as the dominant compounds compare to other detected PFCs (PFCAs, PFSAs, FOSAs, FOSEs) in the studies. The concentrations of PFOS and PFOA in house dust from some studies are given in Table 8.

Investigating correlations of PFCs and home characteristics by Kubwabo shows that the chemical concentrations in dust were significantly associated with the age of the houses and the floor covering (Kubwabo et al., 2005). It was reported that older houses have significantly lower concentrations of PFOS and PFOA compare to houses built more recently; the two compounds were correlated to each other and with the carpeted floor area.

Because indoor air contains significantly higher levels of PFCs than outdoor air (Harrad et al., 2010), it is reasonable to hypothesize that PFCs will enter the body through regular contact in the home via ingestion, inhalation, and/or dermal contact of household dust. Because the majority of people spend more than 90% of their time indoors (Shoeib

et al., 2011), exposure to PFCs in indoor air and dust could be a significant exposure pathway. This is particularly true for children, who typically spend a large proportion of their time indoors (Osborne et al., 2006). Furthermore, children are also exposed to PFCs via hand-to-mouth contact with PFC-bearing consumer products in the home such as carpet and furniture (Harrad et al., 2010).

Table 1.8. Concentrations of PFOS and PFOA in house dust around the word (ng/g).

Country	PFOS	PFOA
Japan[a]	24.5	165
Japan[b]	37.8	19.7
North America[c]	201	142
Germany[d]	16	11
Sweden[e] (houses)	39	54
Sweden[e] (apartments)	85	93
Sweden[e] (offices)	110	70
Canada[f]	71	30
Belgium[g]	0.5	0.7
Norway[h]	3	18
USA[i]	47	44

a. Moriwaki et al., 2003

b. Kubwabo et al., 2005

c. Strynar and Lindstorm., 2008

d. Fromme et al., 2008

e. Björklund et al., 2009

f. Shoeib et al., 2011

g. D'Hollander et al., 2010

h. Haug et al., 2011

i. Knobeloch et al., 2012

1.6.7 Degradation

The strong C–F bond in PFCs is resistant to various type of degradation including reaction with acids and bases, oxidation, and reduction (Kissa et al., 2001). Perfluorocarboxylates (PFCAs) and perfluorosulfonates (PFSAs), in particularl PFOA and PFOS are resistant against any environmental biotic or abiotic reactions (Fromme et al., 2009). However some volatile precursors such as FTOHs and FOSA/FOSEs can be degraded to PFCAs and PFOS respectively through biotic and abiotic reactions (Vestergren et al., 2009).

Reactions of FTOHs with OH radicals are considered as the major reactions of FTOHs in the atmosphere. The degradation of FTOHs occurs via oxidation, which is initiated by OH radicals, followed by oxidation by O_2 and formation of fluorotelomer aldehydes. Subsequent reactions with peroxy radicals and O_2 produce perfluorinated aldehydes or perfluorinated alkoxy radicals, which produce PFCAs after degradation (Ellis et al. 2004).

Degradation of the FTOHs is affected by the levels of free OH radicals in the atmosphere. Hence, there is a relation between the degradation of FTOH to PFCAs and

seasonal differences in the concentration of free OH radicals. In summer, there is an increase in the rate of FTOH degradation due to increased photolysis and higher levels of free radicals in the atmosphere which impact the production of atmospheric PFCA. Production yield of PFCAs from FTOHs in summer and winter time have been reported 8% and 1%, respectively (Yarwood et al., 2007).

FOSAs/FOSEs have been suggested as precursors of PFCA and PFOS (Martin et al., 2006). However due to the lower volatility of FOSAs/FOSEs compared to C_4 analogues, there has been some difficulties experimentally to show the degradation pathways of the C_8 analogues (Martin et al., 2006). Whereas degradation pathways of the C_4 analogues such as *N*-alkylated fluorobutane sulfonamides (FBSAs) and *N*-methyl fluorobutane sulfonamidoethanol (NMeFBSE) have been investigated by Martin (Martin et al., 2006) and D´Eon (D´Eon et al., 2006) respectively.

The pathway of reaction for FBSAs showed a ketone ($C_4F_9SO_2N(H)C(O)CH_3$) and an aldehyde ($C_4F_9SO_2N(H)CH_2CHO$) as primary products, while the formed aldehyde ($C_4F_9SO_2N(H)CHO$) was presumed to be a secondary oxidation product (Martin et al., 2006). PFBS was not observed among products while C_2-C_4 PFCAs were detected (Figure 1.1).

The degradation reaction of *N*-methyl fluorobutane sulfonamidoethanol (NMeFBSE) was found to be again initiated with OH radicals (D´Eon et al. 2006). The main perfluorinated degradation products were detected to be NMeFBSA, PFBA, perfluoropropanoic acid, and PFBS.

It has been shown that 8:2 FTOH can be atmospherically degraded to produce persistent PFCAs (Ellis et al., 2004). This study proposed that reactions of FTOHs with

OH radicals lead to fluorotelomer aldehyde (FTAL, $C_8F_{17}CH_2CHO$). Subsequent reactions result in the formation of fluorotelomer carboxylate FTCAs ($C_8F_{17}CH_2COOH$) and perfluoroaldehydes (PFALs, $C_8F_{17}CHO$) and finally PFCAs (PFOA and PFNA) (Figure 1.2).

Figure 1.1. Atmospheric reaction pathway for EtFBSA to produce PFCAs. Reprinted (adapted) with permission from Martin et al., 2006. Copyright (2010) American Chemical Society.

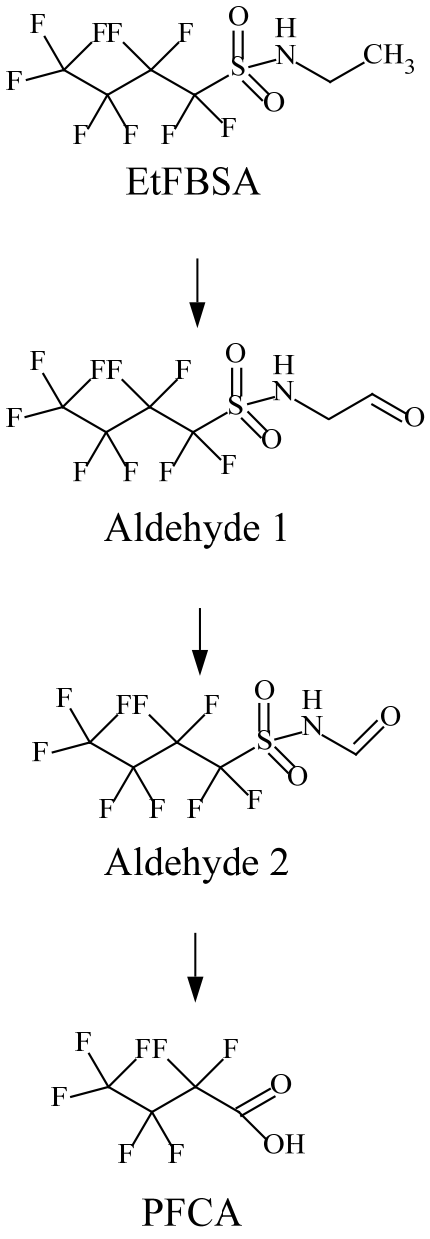
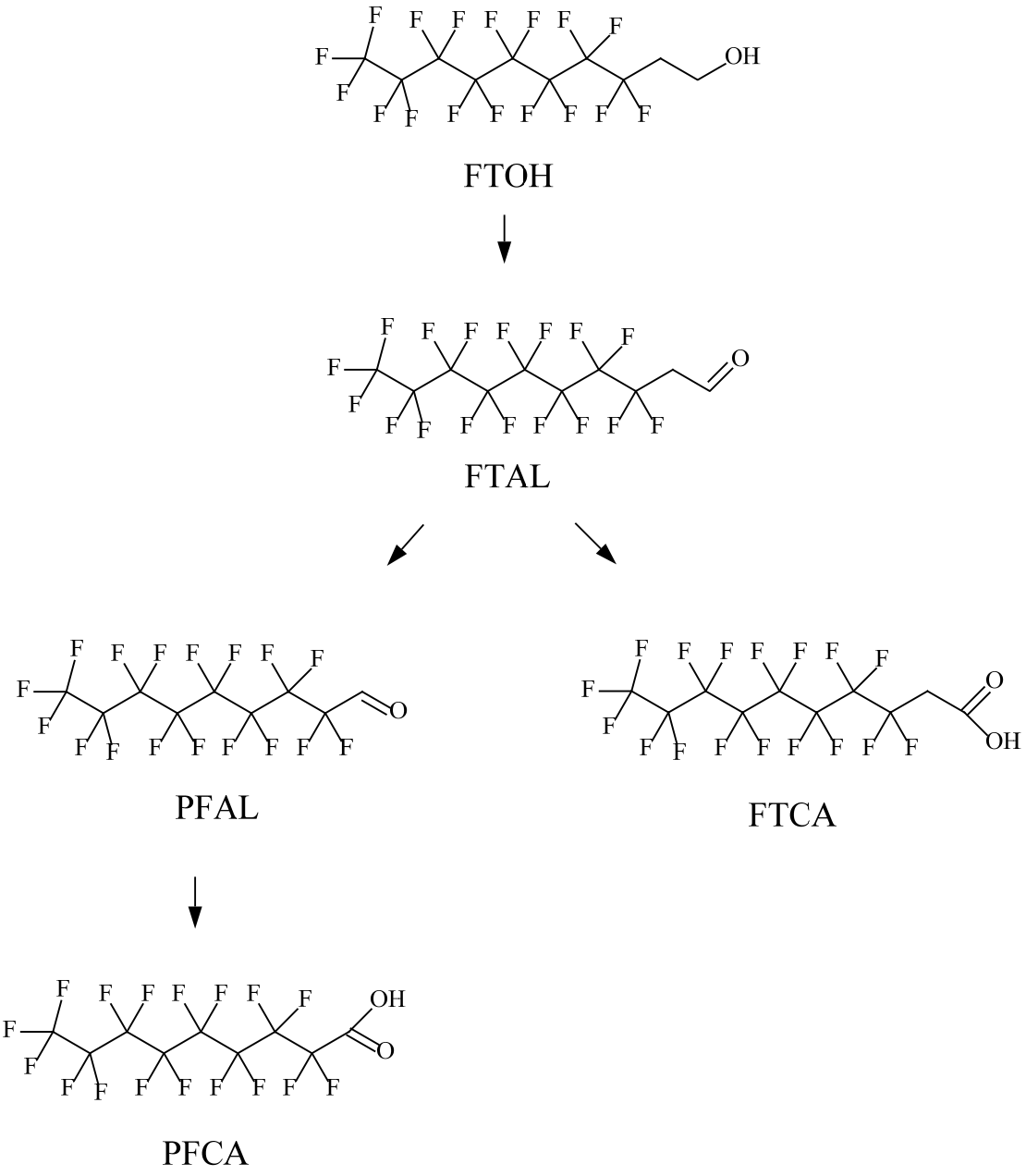


Figure 1.2. Atmospheric reaction pathway of FTOHs to produce PFCAs. Reprinted (adapted) with permission from Ellis et al., 2004. Copyright (2010) American Chemical Society.



8:2 FTOH can be biodegraded to PFOA and PFNA via the intermediates 2-perfluorooctyl ethanoic acid (8:2 FTCA) and 8:2 FTUCA in rats (Butt et al., 2010; Hagen et al., 1981; Martin et al., 2005). Cytochrome P450 enzymes are responsible for oxidation of FTOHs in the first step of the production mechanism of PFCAs. FTOHs can be conjugated to form glucuronide or sulfate metabolites (Butt et al., 2010; Fasano et al., 2006; Martin et al., 2005).

Possible biotransformation of EtFOSE has been investigated by Xu (Xu et al., 2004). The degradation pathway includes deethylation to produce FOSA, which can be subsequently transformed to PFOS.

Hydrolysis is another mode of degradation that large PFCs can undergo. The products have been detected to be PFOS and PFOS precursors. Laboratory examinations showed that NMeFOSE acrylate and NEtFOSE acrylate can undergo hyrolysis in sulfonamide, alkene or ester bonds (Martin et al., 2010).

PFCs such as perfluoroalkyl sulfonamide, amido alcohols, and fluorotelomer olefins can be degraded to PFCAs also (Nakayama et al., 2007). Although atmospheric half-lives of perfluorinated acids considering their reactions with OH radicals have been estimated to be about 130 days, however, the atmospheric presence of these compounds is much shorter than this (approximately 10 days) which is due to wet and dry deposition of the compounds (Hurley et al., 2004a).

1.6.8 Toxicity

After almost one decade, it became apparent that these chemicals have the potential for endocrine disruption (Peden-Adams et al., 2007). Despite no observed effect concentrations (NOECs) of PFCs on the order of mg/L for aquatic organisms (Bossi et al., 2005), PFOS has been shown to bioaccumulate in fish and continue to biomagnify throughout the food web. This suggests that top predators e.g. polar bears and humans are more likely to have greater exposure and therefore potential for toxicity (Bossi et al., 2005). PFCs have been detected in blood serum, bound to proteins (Karrman et al., 2005). That is why the liver is the organ which is typically used for quantification of PFCs. PFOS and PFOA accumulate in major body organs such as liver, kidneys and bile secretions (Jones et al., 2003) and can therein produce hepatotoxicity and endocrine disruption. PFOA has been reported to induce carcinogenic effects in breast cancer bioassays (Rosen et al., 2008).

Rat and mouse studies indicated that fetuses and pups are significantly influenced by PFCs exposure which could lead to serious effects such as reduced birth weights (Hines et al., 2009). Female mouse that were dosed with 5 mg/kg/day of PFOA during gestation had pups that had significantly lower body weight than control mice and mice dosed with lower concentrations of PFOA (Hines et al., 2009).

Because toxicokinetics of different PFCs can be significantly different among animal species and even between different genders, using laboratory animal studies to estimate potential human health effects is complicated (Lau et al., 2007).

The serum half-lives of PFOS and PFOA in humans were determined to be 8.7 and 4.4 years respectively (Burris et al. 2002). It has been reported that PFAS half-lives for other species are significantly shorter than for humans. Elimination half-lives of PFOS in rats and monkeys were estimated to be in the range of 100 - 150 days (OECD 2002). PFCs bind to blood proteins (Chen & Guo, 2009; Salvalaglio et al., 2010) while classical POPs such as polychlorinated biphenyls (PCBs) bind to fatty tissues (Jones et al., 2003).

The no observed adverse effect level – NOAEL of PFOS and PFOA have been estimated to be 6 and 13 mg/L in *Daphnia magna* (Sanderson et al., 2004). Exposure of rats to PFOS in air for one hour yielded an inhalation LC50 of 5.2 mg/L (Rusch et al., 1979). The LC50 value for oral exposure to rats is 251 mg/kg b.w. (Dean et al., 1978). Results of a study showed that workers in the manufacture of perfluorooctanesulphonyl fluoride, which would involve high exposure to PFOS, had 13 times increased risk for bladder cancer mortality compared with the general population of Alabama (Alexander et al., 2003). The lethal concentration (LC50) upon inhalation of PFOA for 4 hours in male rats was reported to be 980 mg/m³ and the no observed effect level (NOEL) was 1 mg/m³ (Kennedy et al., 1986), while oral LD50 values in rats were about 500 mg/kg body weight (Dean and Jessup, 1978).

1.6.9 Human Exposure

The long half-lives of PFCs in humans make it difficult to determine how changes in lifestyle, diet, or other exposure-related factors influence blood levels. It has been

found that age has little influence on circulating PFC levels in humans whereas gender and ethnicity are more likely to influence the accumulation of PFCs (PFOA, PFOS, PFHxS) in humans (Kato et al., 2011). This shows the importance of life style and genetic factors in the uptake and retention of PFCs in humans (Saito et al., 2004; Emmett et al., 2006).

The average level of daily exposure is including all potential routes; indoor and ambient air, house dust, drinking water and food. A number of modeling studies suggested that low-level PFC contamination of food can be responsible for most nonoccupational exposures in industrialized nations (Lindsrtom et al., 2011). Food and breast milk were found to be the dominating sources of exposure to PFOS and PFOA in adults and infants respectively (Haug et al., 2011).

The tolerable daily intakes (TDI) for PFOS and PFOA have been estimated by several scientific institutions. The Scientific Panel on Contaminants in the Food Chain (CONTAM) found values of 150 and 1.5 μg/kg bwt/d for PFOA and PFOS respectively (EFSA, 2008). Furthermore, TDI values for PFOS and PFOA were suggested to be 300 and 3,000 ng/kg bwt/d respectively by the UK Committee on Toxicity of Chemicals in Food, Consumer Products and the Environment (COT, 2006a,b). Another estimation was proposed by the German Federal Institute for Risk Assessment which was 100 ng/kg bwt/d for both PFOS and PFOA (BfR., 2006); the TDIs are significantly higher than the intake values derived from actual human exposure through diet. For example, reported median dietary intakes by Fromme et al. were 1.4 ng PFOS/kg bwt/d and of 2.9 ng/PFOA kg bwt/d (Fromme et al., 2007). Kärrman et al. also calculated similar values for median daily intake of PFOS and PFOA in Japan, which were 1.1 to 1.5 ng/kg bwt/d and from

0.72 to 1.3 ng/g bwt/d respectively (Kärrman et al., 2009).

Haug et al. estimated that the total exposure to PFOS and PFOA for infants (ng/kg bw/day) were significantly (13–16 times) higher than the corresponding estimates for adults. The study showed that maximum intakes of PFOS and PFOA for infants 6 months of age were about 5 (PFOS) and 18 (PFOA) times below this TDI. This indicates that the intakes were relatively close to the present TDIs (Haug et al., 2011).

Fish is the food which has been examined the most frequently among food items. There have been several reports indicate high concentrations of PFOS and possibly other long-chain PFCAs in fish from contaminated waters (Haug et al., 2010; Holzer et al., 2011). For example, median PFOS concentrations in bluegill fillets were found to be between 50 and 100 ng/g of fillet (Delinsky et al., 2009). Consumption of this fish in meal size (195g) results in exposures in the range of 150-330 ng/kg/day. This is approximately 100 times higher than the daily intake predicted in the study by Fromme (Fromme et al., 2009) which means fish is highly potent in human exposure to PFCs. Some of other PFC human exposure routes are as below:

- It has been reported that crops grown on contaminated soils can accumulate PFCs which is likely to be a source of human exposure (Stahl et al., 2008). Hence, agricultural areas that receive amendments of biosolids from wastewater treatment plants can cause contamination of crops, due to the presence of PFCs in sewage (Yoo et al., 2011; Sepulvado et al., 2011). It is obvious that consumption of contaminated drinking water can be a significant source of exposure in such contaminated areas.
- Volatile PFC precursors such as telomers and FOSEs can be present in the indoor environment at pg/m^3 - ng/m^3 levels (Shoeib et al., 2011). After inhalation, they can be me-

tabolized by normal enzymatic processes, and the products may accumulate in vivo (Fromme et al., 2009).

- House dust concentrations of 10-100 ng/g have commonly been reported in some studies (Shoeib et al., 2005; Strynar et al., 2008). This implies that inhalation of airborne material or ingestion of dust from hand-to-mouth contact (particularly for children) is likely to impact human exposure.
- Another exposure pathway for humans is direct contact with commercial products that have been treated with PFCs, as these products contain PFC residuals from manufacturing processes (Guo et al., 2009).

1.6.10 PFCs and Incidence of Asthma in Infants

Asthma is a respiratory disease resulting from narrow airways. Wheezing, coughing, tightness in the chest and shortness of breath are symptoms of asthma (Holgate, 2011). Wheezing or whistling in the chest is the most crucial symptom of asthma. The wheezing can range from mild to severe (Beasley et al., 1998). Patients with severe wheezing may require special care in hospital and the use of inhaled corticosteroids to control symptoms (Herr et al., 2012). From the 1970s to the early 2000s, an increase of asthma incidence in children has been observed all over the word (Boner et al., 2002). Since then, many countries including Canada have seen a decrease in occurrence of asthma (Thomas, 2010). The same temporal pattern, i.e. an increase from 1970 to 2000s and a decrease after that, has also been reported in the use of many PFCs (Wang et al., 2011). This incidence leads to the thought that indoor allergens may be a cause of

asthma (Duffy et al., 1998). Hence, the incidence of asthma until around the time of PFOS phase-out suggests that PFCs could impact the occurrence of asthma in children. There are many uncertainties about what the cause of asthma in children is, however it is believed that certain allergens such as those found on dust mites, dogs, and cats are the most prominent environmental risk factors for childhood asthma (Boner et al., 2002) and these factors are in close association with incidence of asthma in children (Boner et al., 2002).

Inhalation of volatile PFAS such as fluortelomer alcohols (FTOHs) and per-fluorooctanesulfanamido alcohols (FOSEs) which has been detected in high concentrations in indoor homes could be an important exposure pathway for a long time (Shoeib et al., 2008; Barber et al., 2007). PFAS have surfactant-like capacity and can disrupt the natural surfactant-like nature of lungs. PFAS have been reported to directly modify lung function through disruption of lung surfactant activity (Hu et al., 2003). It has been demonstrated that modifying lung function cells in exposed lung cells to PFAS may occur due to the increased potential for cells that have incorporated these common substances into their phospholipid bilayers (Hu et al., 2003).

Histological changes in the lungs were observed in rat pups that were prenatally exposed to PFOS (Grasty et al., 2005). In another study Chen et al. (2012) also found histological changes in the rat pup's lungs that were exposed to PFOS prenatally. Another observation in rat pups in the study was an abundance of cells which had undergone apoptosis (programmed cell death) in a 2.0 mg/kg/day group (Chen et al., 2012).

Yahia reported that a portion of rat pups that were prenatally exposed to PFOS at a high dosage (10 mg/kg bw) showed collapse of the lungs (Yahia et al., 2008). Further-

more, changes in gene expression that were dose-dependent were observed in mice pups that were prenatally exposed to PFOA (Rosen et al., 2007).

This evidence suggests that allergens move more easily across inhalation airway increase risk for sensitization and development of respiratory problems and asthma. Up to now, only two studies in humans have investigated associations between wheezing in infants and concentrations of PFCs (Okada et al., 2012; McConkey, 2012). Both studies reported no correlation between wheezing in infants and the levels of PFOS or PFOA in maternal serum.

1.6.11 Analytical Methods for Analysing PFCs in Indoor Air and Dust

The first method for trace analysis of volatile (FTOHs) and semivolatile (FOSA/Es) perfluorinated compounds in air was developed by Martin (Martin et al., 2002). In this study PFCs were determined in high-volume air samplers. Polyurethane foam (PUF)/XAD-2 cartridges were used to enrich analytes. For sample collection, XAD-2 resin was sandwiched between two sections of PUF inside a clean glass holder with a support (Martin et al., 2002). The addition of XAD, significantly increases the sorption capacity of PUFs for FTOHs and makes it possible to determine concentrations of very volatile PFCs (FTOHs) along with less volatile PFCs (FOSAs/FOSEs) simultaneously. The next studies based on the analyte of interest used either PUF/XAD-2/PUF (Shoeib et al., 2006) or PUF alone (Shoeib, 2005) for air sampling. Haug used polyurethane foam PUF-XAD2-PUF tubes using low volume active air samplers (Haug et al., 2011).

Indoor air sampling methods were improved using passive air samplers contain-

ing PUF disks for sequestering volatile PFCs (Shoeib et al., 2005). Compared to low-volume air samplers, passive air samplers have several advantages: they are easier to deploy, are relatively inexpensive, require no electrical power, make no noise, and can be used both indoors and outdoors. However. the passive sampling device is not able to adsorb volatile PFCs (FTOHs) in air. To overcome the third limitation, Shoeib used polyurethane foam (PUF) disks impregnated with XAD-4 powder (Shoeib et al., 2008). Impregnation of PUFs with 0.4 g XAD doubled the sorptive capacity of the sampler for FTOHs compared to bare PUFs (Shoeib et al., 2008).

Jahnke used SPE cartridges for sampling neutral PFAS in indoor air (Jahnke et al., 2007). These cartridges were easy to handle, and required markedly less time- and solvent-consuming than using PUF/XAD-2 columns. However the disadvantage of this method was that the high back-pressure associated with use of the SPE columns prevented high-volume air sampling.

Soxhlet extraction with petroleum ether or methanol is common for extraction of analytes from air sampling media (Martin et al., 2002; Shoeib et al., 2005; Shoeib et al., 2006; Shoeib et al., 2008; Shoeib et al., 2011). Accelerated solvent extraction (ASE) has been also used as an extraction method in some other studies (Barber et al., 2007, Huag et al., 2011).

Loss of analyte, which generally occurs during the whole analytical procedure, and matrix effects can be corrected by labelled PFC standards, which have not been readily available until recently. A range of isotope-labelled PFCs were used by Martin (Martin et al., 2002) and subsequent studies (Barber et al., 2007; Shoeib et al., 2011). Use of a long 60m GC capillary column was suggested by Dreyer as a solution in order to separate

interfering impurities from the target analytes (Dreyer et al., 2008).

Gas chromatography based-methodology has been used often for analysis of volatile and semivolatile PFCs in air (Jahnke et al., 2007; Barber et al., 2007; Shoeib et al., 2005; Shoeib et al., 2006). However, HPLC-based methods have also been used for the analysis of NEtFOSA and NEtFOSE in air (Boulanger et al. 2004).

For the first time Moriwaki determined PFOS and PFOA in dust house samples using a HPLC-MS/MS methodology in Japan. Moriwaki et al. (2003) and Shoeib et al. (2005) analyzed neutral, volatile PFAS (FOSA/Es) in indoor dust samples using a GC-MS technique.

Because dust is a complex matrix, pre-treatment is often required before analysis. Previous studies have used off-line extraction clean-up procedures prior to analysis by HPLC-MS/MS (Haug et al., 2011; Shoeib et al., 2011). Recently, online extraction and clean-up methods have been improved significantly to reduce matrix effects and interferences. Kato improved the method of analyzing PFCs using on-line solid phase extraction (SPE) (Kato et al., 2009). In on-line SPE, the SPE step is automated and integrated into instrumental analysis via high-performance liquid chromatography–tandem mass spectrometry (on-line SPE–HPLC–MS/MS). This method eliminated the time-consuming step of preparing samples with off-line SPE and was enough efficient to simultaneously determine a wide range of PFCs (17 PFCs) (Kato et al., 2009).

Ultrasonic extraction using methanol is a common technique for extraction of PFCs from dust in several studies (Haug et al., 2011; Kato et al., 2009; Shoeib et al., 2011; Moriwiki et al., 2003).

In order to optimize sample storage of both air and dust, the samples should be re-

frigerated or frozen to prevent degradation of the precursor compounds (Martin et al., 2004; van Leeuwen et al., 2009).

It has been reported that some PFCs can adsorb to surfaces due to their surfactant properties. For example, PFOS concentrations were decreased by 25% when they were stored in glass vials, but no decrease was observed storage was in polypropylene vials (Holm et al., 2004). Thus, standards and sample processing is done using polypropylene containers. Furthermore, using aluminium foil lined lids is essential to avoid absorption of FTOHs into vial septa from solutions (Szostek et al., 2006). Applying direct online extraction techniques can significantly minimize losses of PFCs during the analysis procedure (Kuklenyik et al., 2005).

Because there are many instrumental parts which are made of polytetrafluoroethylene (PTFE) such as tubing, seals etc., it is likely that sample contamination can occur (Szostek et al., 2006; Martin et al., 2004). Hence it is essential to replace fluoropolymer components with stainless steel polyetheretherketone, or polypropylene parts in HPLC systems when possible (Martin et al., 2004). In this study, we were unable to replace Teflon-bearing parts of the pump. Therefore, a C_{18} trap column immediately downstream of the pump heads was used so that PFCs leaching from these parts were pushed away from the analytes eluting from the sample. Hence, they would not coelute and thus interfere.

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2. EVALUATION OF PFC CONCENTRATIONS IN INDOOR AIR AND HOUSE DUST AND ASSOCIATIONS AMONG THOSE CONCENTRATIONS AND HOME CHARACTERISTICS

2.1 Methods and Materials

2.1.1 Chemicals and Reagents

Native standards perfluorohexyl-ethanol (6:2 FTOH), perfluorooctyl-ethanol (8:2 FTOH), perfluorodecyl-ethanol (10:2 FTOH), perfluoro-*n*-pentanoic acid (PFPA), perfluoro-*n*-hexanoic acid (PFHxA), perfluoro-*n*-heptanoic acid (PFHpA), perfluoro-*n*-octanoic acid (PFOA), perfluoro-*n*-nonanoic acid (PFNA), perfluoro-*n*-decanoic acid (PFDA), perfluoro-*n*-undecanoic acid (PFUA), potassium perfluoro-1-butanesulfonate (PFBS), sodium perfluoro-1-hexanesulfonate (PFHxS), sodium perfluoro-1-octanesulfonate (PFOS), sodium perfluoro-1-decanesulfonate (PFDS), perfluoro-1-octanesulfonamide (FOSA), *N*-methylperfluoro-1-octanesulfonamide (MeFOSA), *N*-ethylperfluoro-1-octanesulfonamide (EtFOSA), 2-(*N*-methylperfluoro-1-octanesulfonamido)-ethanol (MeFOSE), 2-(ethylperfluoro-1-octanesulfoamido)-ethanol (EtFOSE) and corresponding isotropically labeled standards 2-perfluorohexyl-[1,1-d_2,1,2-$^{13}C_2$]ethanol (6:2 FTOH-$^{13}C_2$), 2-perfluorooctyl-[1,1-d_2-1,2-$^{13}C_2$]ethanol (8:2 FTOH-$^{13}C_2$), 2-perfluorodecyl-[1,1-d_2-1,2-$^{13}C_2$]ethanol (10:2 FTOH-$^{13}C_2$), perfluoro-*n*-[1,2-$^{13}C_2$]hexanoic acid (PFHxA-$^{13}C_2$), perfluoro-*n*-[1,2,3,4-$^{13}C_4$]octanoic acid (PFOA-$^{13}C_4$), perfluoro-*n*-[1,2,3,4,5-$^{13}C_5$]nonanoic acid (PFNA-$^{13}C_5$), perfluoro-*n*-[1,2-$^{13}C_2$]decanoic acid (PFDA-$^{13}C_2$), perfluoro-*n*-[1,2-$^{13}C_2$]undecanoic acid (PFUA-$^{13}C_2$),

sodium perfluoro-1-hexane[$^{18}O_2$]sulfonate (PFHxS-$^{18}O_2$), sodium perfluoro-1-[1,2,3,4-$^{13}C_4$]octanesulfonate (PFOS-$^{13}C_4$), perfluoro-1-[$^{13}C_8$]octanesulfonamide (FOSA-$^{13}C_8$), *N*-methyl-d_3-perfluoro-1-octanesulfonamide (MeFOSA-d_3), *N*-ethyl-d_5-perfluoro-1-octanesulfonamide (EtFOSA-d_5), 2-(*N*-deuteriomethylperfluoro-1-octanesulfonamido)-1,1,2,2-tetradeuterio-ethanol (MeFOSE-d_7), and 2-(*N*-deuterioethylperfluoro-1-octanesulfonamido)-1,1,2,2-tetradeuterioethanol (EtFOSE-d_9) were purchased from Wellington Laboratories (Guelph, ON, Canada). The chemical purity of all standards was 98% or greater and the isotopic purity of labeled standards was 94% or greater. HPLC-grade acetonitrile, methanol, hexanes and isopropanol were obtained from Fisher Scientific (Ottawa, ON, Canada). HPLC grade isooctane, petroleum ether, acetone, dichloromethane were purchased from EMD (USA). Reagent grade formic acid was purchased from Sigma-Aldrich (Oakville, ON, Canada). De-ionized water was purified as MilliQ water (18 MΩ-cm) using a Millipore Synergy System (Billerica, MA, U.S.A.). HPLC-grade ammonium acetate was purchased from Sigma Aldrich (St. Louis, MO, U.S.A.). BD Vacutainers™ Plus made from a formulation of polyethylene terephthalate were obtained from Becton, Dickinson, and Company (Mississauga, ON, Canada). Sea sand was purchased from EMD (USA).

2.1.2 Air Sample Collection and Extraction

Air and dust samples from homes in or close to Winnipeg, Manitoba were collected by CHILD. Air samples (n=77) and dust samples (n=85) were collected between August 2011 and August 2012. However, presampling steps such as cleaning and prepar-

ing sampling equipment were done in our laburatory. Sorbent impregnated PUF (SIP) disk passive samplers were used to collect air samples, using a previously described procedure (Shoeib et al., 2008). Briefly, PUF disks (Tisch Environmental Inc, Cleves, Ohio, U.S.A.) were precleaned by Soxhlet extraction with acetone for 24 h, then with petroleum ether for another 24 h. The disks were dried in vacuum desiccators for 18 h and stored in solvent-rinsed glass jars until use. Amberlite XAD-4 (Acros Organic, USA) was ground to a powder by mortar and pestle, cleaned by Soxhlet with methanol, dichloromethane, and hexane each for 18h, then dried first by rotary evaporation and then in a vacuum desiccator for 1 day; these materials were used to prepare SIP disks (Shoeib et al., 2008). To prepare sorbent impregnated PUFs (SIPs), i.e. PUFs impregnated with XAD-4, a slurry of XAD-4 in hexane (6.4 g/L) was prepared. The slurry was sonicated for 30 min and was stirred by a glass magnetic bar to suspend XAD powder. PUF discs were dipped into the slurry for 40 s, then PUFs were lifted to allow excess solution to drain for 10 s above the slurry. This was repeated 3 times for each PUF. To make a uniform coating, the solvent was allowed to drain from different sides of the disc. The discs were then placed on solvent-rinsed ashed aluminum foil for 5 min before being transformed to vacuum desiccators and be dried for 3 days. Each batch of slurry was used to prepare 10 SIPs (Shoeib et al., 2008).

The SIPs were deployed in homes for three weeks, within the upper half of the most heavily used rooms, at locations at which there was the least chance of disturbance (e.g. on top of a bookshelf). Temperature loggers (LogTag recorders, USA) were deployed along with air samplers to record the room temperature during the sampling period. After deployment, SIPs were stored in sealed clean glass jars with aluminum-lined

caps at -20°C until processing. Field blanks (n = 10) were prepared by placing separate clean SIP disks in homes and then removing them after 1 min. Other clean SIPs (n=10) were analysed with each batch of samples as procedural blanks.

For extraction, SIP disks were spiked with labelled neutral PFCs (FTOHs, FOSAs, and FOSEs), then extracted via Soxhlet for 18 hours using 1:1 petroleum ether/acetone. Extracts were reduced to ~5 mL by rotary evaporation and reduced further to 0.5 mL by nitrogen blowdown.

2.1.3 Preparations of Standard Solutions for Air Samples

Calibration standards were prepared from PFC standard solutions by diluting PFC standard stock solutions to 1000 µL in isooctane to concentrations of 20, 50, 100, 200, 400, 800 and 1000 pg/µL for 6:2 FTOH, 10:2 FTOH, MeFOSA, EtFOSA, MeFOSE, EtFOSE and concentrations of 40, 100, 200, 400, 800, 1600, and 2000 pg/µL for 8:2 FTOH. All standards were stored at approximately -20^{o}C in polypropylene vials to minimize degradation and volatilization. In order to prevent degradation of analytes, standard solutions were only used if prepared in the previous 60 days. The solutions analyzed within six days of preparation. To prepare calibration curves, relative concentrations (peak area of native compound divided by peak area of internal standard) were plotted versus concentration. Stable-isotope labelled analogues of the analytes were used as the internal standard for all the analytes in air samples.

2.1.4 Gass Chromatography-Mass Spectrometry Analysis of Air Samples

Analysis of neutral PFCs (FTOHs, FOSAs, and FOSEs) in extracts of air samples was done by gas chromatography-mass spectrometry (GC/MS) using a Thermo Scientific (Waltham, MA) DSQ II in positive chemical ionization (PCI) mode chemical mode using methane as reagent gas and an ion source temperature of 240°C. Analytes were separated using a polar CP-Wax 57 CB capillary column (25 m × 0.25 mm i.d. × 0.2 μm d_f; Varian) fitted with a HP-INNOWAX guard column (5 m × 0.25 mm × 0.1 μm, Varian). The temperature program used was 50°C (held 1 min), 3°C/min to 70°C, 10°C/min to 130°C, 20°C/min to 225°C (held 11.4 min). Injection volumes were 1 μL, in splitless mode at 200°C was used. Helium was used as the carrier gas at 1 mL/min (Barber et al., 2007). PCI ions and internal standards for each analyte are listed in Table 1 (Shoeib et al., 2011). However, FOSA was not among the analytes in the literature from which the method was adopted (Shoeib et al., 2011, Barber et al., 2007), and GC/MS full-scan results of FOSA performed in our lab were not effective for development of an analysis method. Therefore, the compound was not analysed in indoor air samples.

2.1.5 Dust Sample Collection and Extraction

Nylon filters used to collect dust samples were placed in 20 mL glass bottles with aluminum lined caps before they were transported to the laboratory. Procedures for sample preparation were adopted from a previous study (Shoeib et al., 2005). Briefly, dust samples were sieved through a 80-mesh sieve (<180 μm), and any visible hair were removed with tweezers. The sieved dust was weighed, then stored at -20°C until being

processed. Sea sand (after ashing for 3 h in 450 °C) was used as a procedural blank because it has a relatively low concentration of PFCs; and was sieved and stored analogously. Field blanks (n=12) were collected by placing separate filters in homes and removing them after 1 min.

In order to extract dust samples, 100 mg of sieved dust in a 15 mL polypropylene test tube was spiked with internal standards, homogenized with a vortex mixer and allowed to settle prior to the addition of 1000 μL each of 0.1 M formic acid and 1 MeOH. Dust was extracted by sonication in an ultrasonic bath for 10 min. Extracts were filtered through a empty SPE cartridge (3mL, Varian) fitted with two consecutive frits (20 μm polyethylene), with further filtration with 0.2 μm nylon syringe filters (Chromatographic Specialties, Inc.) prior to instrumental analysis (Kato et al., 2009).

2.1.6 Preparation of Standard Solutions for Dust Samples

Calibration curve standards were prepared from PFC standard solutions by diluting PFC standard solutions to 150 μL in methanol in polypropylene autosampler vials, then 150 μL 0.1% formic acid was added to provide concentrations of 0.01, 0.1, 1, 20, 100, 200, 500 and 1000 pg/μL. Storage and use of standards followed for air samples (Section 2.2.3). Samples were analyzed within six days of preparation. For most analytes isotopically labelled analogues of the analytes were used as the internal standard. Because isotopic internal standards were not available for a few analytes, stable isotope-labelled PFCs with analogous structures to the analytes were used as internal standards. A linear regression weighting of 1/x was used for all analytes. Samples for method valida-

tion were prepared by spiking sea sand with a range of PFC concentrations and extracted as for dust samples (Kato et al., 2009).

2.1.7 Instrumental Analysis of Dust Samples

Concentrations of PFCs in dust were determined by online solid phase extraction-liquid chromatography-tandem mass spectrometry (SPE-LC-MS/MS), consisting of a HTC PAL autosampler (CTC Analysis, Zwingen, Switzerland), two Agilent LC pumps (Agilent Technologies, Mississauga, ON, Canada), and an Agilent 6410 triple quadrupole MS/MS. The CTC autosampler contained a 6-port switching valve, and the binary LC pump contained a 10-port switching valve. The quaternary pump (model G1311C) was responsible for loading the analytes onto the SPE column, and the binary pump (model G1312B) was responsible for back flushing the analytes off the SPE column onto the analytical column. An EXP trap and in-line holder (C18-ES 2.7 μm, 10 × 4 mm) from Optimize Technologies (Oregon City, OR, U.S.A.) was used as an online SPE column. A ZORBAX Eclipse Plus C18 column (1.8 μm, 50 mm × 2.1 mm) from Agilent Technologies (Mississauga, ON, Canada) was used as an analytical column with a SecurityGuard Cartridge C18 column (3 μm, 4 mm × 2.0 mm) from Phenomenex (Torrance, CA, U.S.A.) as the analytical guard column. For sample cleanup by online SPE, the sample was loaded into a 500 μL sample loop, and then was flashed to the SPE column. The sample was retained on the SPE column by 5:95 (v:v) methanol:0.1% formic acid flowing from the quaternary pump. After 1.4 minutes the sample was back flushed off the SPE column onto the analytical column by 2 mM ammonium acetate in water and acetonitrile flowing from the binary pump. The analyte was then separated on the analytical

column using a flow rate of 0.5 mL per minute at 40°C. The mobile phase gradient from the quaternary pump started at 5% MeOH and 95% formic acid (0.1% in water), then increased to 50% MeOH and 50% acetonitrile after 1.5 min, was held 3 min then 100% 50:50 isopropyl alcohol:water for 4 min and finally changed to 50:50 acetonitrile:MeOH for 1 min. The binary pump mobile phase gradient for analytical separation was initially 2 mM ammonium acetate in water and held for 1.5 min, then changed to 60:40 acetonitrile:2 mM ammonium acetate during 3 min, held on for 30 s, increased to 67:33 acetonitrile:2 mM ammonium acetate in 0.6 min, finally raised to 90:10 (2 mM acetonitrile: ammonium acetate) and was held for 2 min, decreased to 15:85 acetonitrile:2 mM ammonium acetate in 1 min held for 1 min (McConkey, 2012; Gosetti et al., 2010; Haug et al., 2009). Mass spectrometry was performed using dynamic multiple reaction monitoring (MRM) using electrospray ionization in negative mode. Ion transitions and internal standards for each analyte are listed in Table 2. The curtain gas temperature was set to 300°C at a flow of 10 L/min. The nebulizer pressure was set to 55 psi and the capillary voltage was -2000 V. An optimized fragmentor voltage 72V and collision energies 16 and 4 V (for qualifier and quantifier respectively) were applied in the method (McConkey, 2012).

2.1.8 Statistics

Statistical analysis was performed using SPSS Statistics software (version 19.0.0) and GraphPad PRISM software (version 5.01). A significance level of $\alpha = 0.05$ was used. Statistical comparison tests were only performed on analytes which had median values above the limit of quantification (LOQ). Comparisons of PFCs in indoor air and in dust,

correlations between PFCs in indoor air and dust, correlations between PFCs in either indoor air or dust and home characteristics, comparisons of target and control homes, comparisons between seasons and incidence of wheezing, were performed on the measured concentrations of PFCs, using appropriate nonparametric tests (Spearman rank, Kruskal-Wallis test, Mann-Whitney test, Wilcoxon paired *t*-test). No statistical test was used for exploring correlations between wheezing and PFCs, as the eventual sample size was sufficiently small that it was unlikely to have sufficient power to relate these variables.

Principal component analysis (PCA) was used to assess exposure and trends with indoor air and dust PFC concentrations. The proportions of each PFC relative to the sum of total PFCs were used as the variables for PCA. Random values between zero and the LOQ were substituted for concentrations under the LOQ, the proportions were calculated, and then natural log transformed. Survey data regarding home characteristics was used to evaluate exposure and trends with the PCA data. Because the proportions of PFCs were natural log transformed parametric statistical tests (one-way ANOVA, unpaired *t*-test) were used.

2.2 Results and Discussion

2.2.1 Method Validation

In order to minimize potential contamination of both indoor air and dust samples during analysis procedure, parts and/or materials on instruments or laboratory vessels which likely contained polytetrafluoroethylene (PTFE) (tubing, seals in LC-MS/MS, vi-

als, etc) were replaced with Teflon®-free material such as stainless steel or poly propylene when possible (Martin et al., 2004). Clean glassware was baked at 450°C for 3h and was rinsed with solvent (methanol or 1:1 petroleum ether/acetone) before use.

2.2.1.1 Method Validation of Indoor Air Samples

Details of gas chromatography analysis in positive chemical ionization (PCI) mode for each analyte and the corresponding internal standard used for indoor air sample analysis (Barber et al., 2007; Shoeib et al., 2011) are given in Table 2.1. The limit of detection (LOD) for air samples was defined as a value corresponding to the average of 10 SIP blank values + 3 standard deviations (Shoeib et al., 2011). LOD values for SIP disks ranged from 1.2 to 16.9 pg/m^3 (Table 2.2). Total masses of the analytes on each SIP ranged from 0.10 to 1.77 ng (considering the sampling rate and sampling duration).

Table 2.1. Details for analytes and internal standards in positive chemical ionization (PCI) mode in indoor air samples.

Analyte	Monitored Ions (*m/z*) of Analyte (PCI Mode)	Internal Standard	Monitored ions (*m/z*) of Internal Standard Ions (PCI Mode)
6:2 FTOH	365/327	6:2 FTOH-$^{13}C_2$	369/331
8:2 FTOH	465/427	8:2 FTOH-$^{13}C_2$	469/431
10:2 FTOH	565/527	10:2 FTOH-$^{13}C_2$	569/531
EtFOSA	528	EtFOSA-d_5	533
MeFOSA	514	MeFOSA-d_3	517
MeFOSE	540/558	MeFOSE-d_7	547/565
EtFOSE	554/572	EtFOSE-d_9	563/581

Table 2.2. Limit of detection (LOD) for 7 PFCs in indoor air samples (pg/m^3).

Analyte	LOD
6:2 FTOH	8.2
8:2 FTOH	16.9
10:2 FTOH	9.1
EtFOSA	2.7
MeFOSA	1.2
MeFOSE	4.9
EtFOSE	1.8

None of the procedural or field blanks (n = 10) had PFC concentrations greater than the LOD for any analyte, indicating no significant contamination during the sample deployment, retrieval, extraction, or analytical procedures (Table 2.3).

Table 2.3. Field and procedural blank values of PFCs for indoor air samples (pg/m^3).

Analyte	Field Blank	Procedural Blank
6:2 FTOH	3.1	2.4
8:2 FTOH	14	10
10:2 FTOH	6.7	4.8
EtFOSA	1.3	1.9
MeFOSA	ND	ND
MeFOSE	3.0	2.9
EtFOSE	ND	ND

ND = Not detected

Method recoveries were determined by spiking separate clean SIPs (n = 3) with native neutral PFCs (6:2 FTOH, 8:2 FTOH, 10:2 FTOH, MeFOSA, EtFOSA, MeFOSE, EtFOSE), which were extracted and analysed using the same methods as for experimental samples. Recoveries for air samples ranged from 52-110% (Table 2.4), which is close to the range of recoveries reported by Haug et al., 2011.

The coefficient of variance (CV) was estimated by randomly selecting five un-known samples and conducting triplicate analysis over a period of 20 days. The CVs ranged 4.8 to 17.3% (Table 2.5) which were sufficiently narrow; according to Zhou

(2011), CVs below 20% are statitically sufficiently narrow.

Table 2.4. Mean recovery percentages and relative standard deviations (RSD) for 7 PFCs in indoor air samples.

Analyte	Recovery % (RSD %)
6:2 FTOH	52.3 (5.9)
8:2 FTOH	56.1 (7.7)
10:2 FTOH	54.2 (10.9)
EtFOSA	103.8 (7.6)
MeFOSA	110.4 (8.2)
MeFOSE	96.6 (4.3)
EtFOSE	99.5 (6.1)

Table 2.5. Percent coefficients of variance for 7 PFCs in indoor air samples.

Analyte	Coefficient of Variance (%)
6:2 FTOH	15.1
8:2 FTOH	17.3
10:2 FTOH	12.7
EtFOSA	7.1
MeFOSA	11
MeFOSE	4.8
EtFOSE	5.2

2.2.1.2 Method Validation of House Dust Samples

Multiple reaction monitoring transitions and internal standards for each analyte in house dust are listed in Table 2.6. The background concentrations of PFCs were lower than the respective LOQ values (Table 2.7). However, it was not possible to replace all fluoropolymer-containing parts of the LC-MS/MS system. Non-replaceable parts such as solvent bottle tubing, autosampler injection syringe plunger, and frits in the MS, had low background levels of PFCs that were generally below the LOQs. Therefore, background concentrations of PFCs in solvents were measured on each day of analysis by using solvent blanks spiked with internal standards in triplicate. If background PFCs were above the LOQ, the background concentrations were subtracted from all samples analyzed in that batch of samples. In general, no detectable background levels were observed for most PFCs, except PFHpA, which had a mean background level of 6.21 ng/g (with a standard deviation of 2.32 ng/g) in less than 40% of the samples. The largest blank background value subtracted from the smallest measured peak which resulted in a peak observed for PFHpA in sample. Typically, variability of the background concentrations for PFHpA in each set of data was low (standard deviation <5% of the mean values).

Table 2.6. Multiple reaction monitoring (MRM) transitions and internal standards for each analyte in house dust samples.

Analyte	MRM transition	Internal Standard	Internal Standard MRM Transition
PFPA	263 → 219	PFHxA-$^{13}C_2$	315 → 270
PFHxA	313 → 269 313 → 119		
PFHpA	362.9 → 319 362.9 → 169		
PFOA	413 → 368.9 413 → 168.9	PFOA-$^{13}C_4$	417 → 371.9
PFNA	463 → 418.9 463 → 168.9	PFNA-$^{13}C_5$	468 → 422.9
PFDA	513 → 468.9 513 → 269	PFDA-$^{13}C_2$	515 → 469.9
PFUA	563 → 519 563 → 269	PFUA-$^{13}C_2$	565 → 520
PFBS	299 → 80 299 → 98.9	PFHxS-$^{18}O_2$	402.9 → 84
PFHxS	398.9 → 80 398.9 → 99		
PFOS	498.9 → 99 498.9 → 80	PFOS-$^{13}C_4$	502.9 → 99
PFDS	598.9 → 80 598.9 → 99		502.9 → 80
FOSA	497.9 → 78.2	FOSA-$^{13}C_8$	505.9 → 78.2
MeFOSA	512 → 219.1 512 → 169.1	MeFOSA-d_3	515 → 219.1
EtFOSA	526 → 169 526 → 218.5	EtFOSA-d_5	531 → 169
MeFOSE	616 → 59	MeFOSE-d_7	623 → 59
EtFOSE	630 → 59.1	EtFOSE-d_9	639 → 59.1

Table 2.7. Field and procedural blank and LOQ values of PFCs for dust samples (ng/g).

Analyte	Field Blank	Procedural Blank	LOQ (ng/g)
PFPA	1.9	2.0	2.3
PFHxA	2.4	3.1	3.3
PFHpA	1.5	1.1	1.9
PFOA	1.4	1.0	2.9
PFNA	0.8	0.8	2.0
PFDA	1.5	1.2	1.8
PFUA	1.7	1.6	1.9
PFBS	1.8	2.1	2.1
PFHXS	1.6	1.3	1.9
PFOS	2.9	2.5	3.2
PFDS	2.1	2.2	2.3
FOSA	0.6	0.7	2.1
MeFOSA	1.8	1.6	1.8
EtFOSA	1.5	1.6	1.8
MeFOSE	2.2	2.0	2.4
EtFOSE	1.5	1.0	2.0

In order to include matrix effects to the calibration curve, matrix-matched calibration curves were made by spiking the matrix with a range of PFC concentrations. For analysis of PFCs in dust, the matrix can be blank dust, such as sea sand (Kato et al.,

2009).

Matched calibration curves are not necessary if matrix effects are small enough that non-matrix-matched calibration curves behave in the same manner as matrix-matched calibration curves. In this study, eight-point calibration curves from 0.01-1000 pg/mL were derived in both solvent and in sea sand for PFCs analysed by LC-MS/MS. The slopes of the sea sand calibration curves were compared to the slopes of the solvent calibration curves for each analyte (paired *t*-test, n = 3). No significant differences were observed between slopes of the solvent calibration curves and the matrix-matched sea sand calibration curves over the range of 0.01-1000 pg/mL. Therefore, solvent calibration curves were used for analysis. The solvent calibration curves do not account for matrix effects; however, use of isotopic internal standards in all samples would correct for effects of matrix components. These internal standards were added to the dust samples before extraction. Hence, matrix effects and possible loss during the extraction procedure for both internal standards and native analytes were in the same manner.

Isotopic internal standards were not commercially available for all analytes. As a result, slightly different matrix effects in analytes with non-isotopic internal standards (i.e., standards that were not isotopes of that analyte, e.g. PFDS) were possible. However, the different matrix effects due to non-isotopic internal standards are unlikely to have affected the results because, as mentioned above, the method validation results were satisfactory. No significant differences were observed between slopes of the solvent calibration curves and the matrix-matched sea sand calibration curves; good linearity was observed (R^2>0.97) for all calibration curves.

To determine recoveries of PFCs in dust, dust sample were spiked at a concentra-

tion of 30 ng/g for 16 PFCs (n = 3). Recoveries were estimated as $C_{observed}/C_{spike}$, where $C_{observed}$ is the difference between the quantified concentration in the spiked sample and the quantified concentration in the native sample and C_{spike} is the spiked concentration (Kato et al., 2009). The mean recoveries and relative standard deviations (RSD) are given in Table 2.8. The mean recoveries ranged from 74.6 to 121.7%, with RSDs of 5.9 to 14.2%. These results are consistent with recoveries reported in other studies e.g., 47.9 to 107% recovery with RSDs of 3 to 10% (Strynar et al., 2008); 91.8-103.5% with RSDs of 2.8 to 8.4% (Kato et al., 2009).

Fourteen unknown samples (10% of the total number of dust samples) were randomly selected and analysed 3 times over two weeks to assess inter-day measurement variability. Inter-day variability was calculated by determining the percent coefficient of variances for the measured analyte values for the two weeks. The resultant coefficients of variance ranged from 4.1 to 19.3% (Table 2.9). According to Zhou (2011), CVs below 20% statistically are sufficiently narrow.

Table 2.8. Mean percent recovery and relative standard deviations (RSDs) of PFCs in house dust samples.

Analyte	Recovery % (RSD%)
PFPA	100.5 (9.3)
PFHxA	104.2 (10.7)
PFHpA	102.4 (14.0)
PFOA	121.7 (10.4)
PFNA	100.9 (9.3)
PFDA	103.2 (6.2)
PFUA	88.1 (12.2)
PFBS	115.8 (10.3)
PFHxS	110.2 (5.9)
PFOS	107.5 (12.7)
PFDS	106.4 (5.0)
FOSA	79.1 (9.2)
MeFOSA	78.7 (10.4)
EtFOSA	77.1 (14.2)
MeFOSE	74.6 (9.0)
EtFOSE	76.3 (6.7)

Table 2.9. Percent coefficients of variance for 7 PFCs in dust samples.

Analyte	Coefficient of Variance (%)
PFPA	12.1
PFHxA	5.2
PFHpA	9.1
PFOA	4.1
PFNA	19.3
PFDA	14.3
PFUA	15.1
PFBS	11.2
PFHxS	9.4
PFOS	12.1
PFDS	17.1
FOSA	7.4
MeFOSA	7.8
EtFOSA	11.4
MeFOSE	10.2
EtFOSE	13.9

2.2.2 Concentrations of PFCs in Indoor Air

To derive air concentrations from the masses of chemicals accumulated in the SIP disks, sampling rates from a previous study with the same sampling methodology as present study were used (Shoeib et al., 2011). The SIP disk sampling rates for FTOHs and FOSA/FOSEs were 5 m^3/day and 4 m^3/day respectively. In Table 2.10, descriptive statistics for the seven PFCs detected in air are given. The FTOH with the highest concentration was 8:2 FTOH, with a median concentration of 1.34×10^3 pg/m^3, followed by 10:2 FTOH at 560 pg/m^3. The median concentrations of 8:2 FTOH and 10:2 FTOH were determined to be 64% and 24% of the total PFC concentrations (i.e. FTOHs and FOSA/Es) respectively in indoor air.

Table 2.10. Indoor air concentrations (pg/m^3), summary statistics, and detection frequencies of neutral PFCs (FTOHs, FOSAs, FOSEs) in homes located in Winnipeg, Canada (n = 77).

Analyte	Median	Mean	Range	Frequency of Detection(%)
6:2 FTOH	267	572	$47\text{-}2.56\times10^{-3}$	38
8:2 FTOH	1336	1954	$173\text{-}9.08\times10^{-3}$	96
10:2 FTOH	560	920	$55\text{-}7.51\times10^{-3}$	82
EtFOSA	60	88	$<$LOQ-1.07×10^{-3}	88
MeFOSA	47	79	$2.4\text{-}5.94\times10^{-2}$	95
MeFOSE	539	1067	$29\text{-}8.78\times10^{-3}$	96
6:2 FTOH	142	419	$8.5\text{-}5.25\times10^{-3}$	62

Frequency of Detection = The ratio of number of samples in which analyte detected to all analysed samples

The analyte 8:2 FTOH was detected above its LOD in 96% of the samples. All other PFCs, except EtFOSE, were detected in more than 80% of the indoor air samples. Previously, reported indoor air concentrations of 6:2 FTOH have been 2-200 times lower than those of 8:2 FTOH (Barber et al., 2007; Shoeib et al., 2011) (Table 2.10). Therefore, it is not surprising that 6:2 FTOH showed the lowest concentration among FTOHs in this study.

MeFOSE, with a median concentration of 539 pg/m^3 (approximately 4 times higher than the median concentration of EtFOSE), was the dominant compound among FOSA/FOSEs (Table 2.10). The median concentration of MeFOSE was estimated to constitute 68% of the total median concentration of FOSA/FOSEs. The dominance of MeFOSE has also been reported in other studies (Shoeib et al., 2011; Huag et al., 2011; Huber et al., 2008) (Table 2.11). MeFOSA and EtFOSA, with median concentrations of 47 pg/m^3 and 60 pg/m^3, respectively, each represented 7% of the total FOSA/FOSEs.

Table 2.11. Summary of median concentrations (or concentration ranges) of PFCs (pg/m^3) in indoor air samples reported in the present study and related studies.

Year	N	6:2 FTOH	8:2 FTOH	10:2 FTOH	EtFOSA	MeFOSA	MeFOSE	EtFOSE
2011/12	77	267	1336	560	60	47	539	142
2007/8	59	1040	2720	980	19	21	320	56
2008	40	933	5173	2822	0.5	8.3	265	78
2009	10	3-47	7.5-170	<0.6-47	<0.52-6.1	<1.2 to 14	<0.52-6.1	<0.52-6.1

N = Number of samples

[a]. Present study

[b]. Shoeib et al., 2011

[c]. Haug et al., 2011

[d]. Jogsten et al., 2012

Concentrations of neutral PFCs in indoor air from various studies are given in Table 2.11. A diversity of sampling approaches for indoor air and extraction methods complicates comparisons among the studies. Also, transportation of samples to the laboratory has been suggested as another factor affecting results. Transportation of samples under controlled conditions (in sealed containers and at sufficiently low temperatures) minimizes loss of PFCs in air samples (Harrad et al., 2010a).

2.2.3 Correlations Among PFCs in Indoor Air

Nonparametric Spearman rank correlations were used to explore relationships between concentrations of the various PFCs because the indoor air data was not normally distributed. Significant correlations between 8:2 FTOH and 10:2 FTOH, MeFOSE and MeFOSA, EtFOSE and EtFOSA were observed in indoor air (Table 2.12). These were all positively correlated, suggesting that, as the concentration of one compound varied, the other varied in the same direction. This may indicate the presence of common sources for the correlated compounds, e.g., formulation mixtures of PFCs in household products. Correlation between 8:2 FTOH and 10:2 FTOH, which both are from the same class of perfluorinated compounds (i.e., fluorotelomer alcohols) was also reported in a previous study (Liu et al., 2013). Furthermore, the correlation between MeFOSE and MeFOSA was suggested by Haug et al. (2011), while the correlation between EtFOSE and EtFOSA is reported for the first time, to the best of our knowledge, in this study.

Table 2.12. Spearman's rank correlations of perfluorinated compounds in indoor samples from homes in Winnipeg, Canada (n = 77). (*) indicates significant correlation (correlation was significant at the 0.05 level or less).

	8:2 FTOH	10:2 FTOH	Et-FOSA	Me-FOSA	Me-FOSE	Et-FOSE
6:2 FTOH	0.02	0.08	0.01	0	0.55*	-0.58
8:2 FTOH		0.73*	0.14	0.3	0.34	-0.19
10:2 FTOH			0.23	0.39	0.34	-0.12
Et-FOSA				0.37	0.31	0.82*
Me-FOSA					0.86*	0.23
Me-FOSE						0.52

2.2.4 Concentrations of PFCs in House Dust

PFOA (81 ng/g) and PFOS (78 ng/g) had the highest concentrations of the PFCs measured in house dust and were detected in all samples (Table 4). This observation in dust samples has been reported in other studies around the world (Knobeloch et al., 2012; Shoeib et al., 2011; Kubwabo et al., 2005; Strynar et al., 2008, Björklund et al., 2009; Goosey et al., 2012; Moriwaki., 2003) (Table 2.13).

PFHxA was detected in all samples (Table 2.13), with a median concentration of 57 ng/g, and was one of the analytes with high concentrations compared to other detected PFCs in house dust in this study. The ubiquity of PFHxA is not surprising because PFHxA is present in stain and grease-proof coatings on food packaging, furniture, and household products (Shoeib et al., 2011; Kubwabo et al., 2005). Other PFCAs were frequently detected in dust samples. More than 70% of the samples contained PFHpA, PFHxS, PFHxA, PFNA, PFDA, PFDS, MeFOSE, and EtFOSE at concentrations above their LOQ values (Table 2.13).

Among the sixteen PFCs, median concentrations for four PFCs (FOSA, MeFOSA, EtFOSA, PFUA) were lower than their respective LOQs and they were detected in fewer than 30% of samples. Of the PFSAs, the highest median concentrations were seen for PFOS (78 ng/g) and PFHxS (20 ng/g). Measured median and maximum total PFC concentrations (487 ng/g and 44,582 ng/g, respectively) indicate that typical loading of perfluorinated compounds would be 0.5 μg per 1 g dust, but loadings could reach about 45 μg per 1 g of dust. Specifically, the range for PFSAs and PFCAs was 0.1-22 and 0.3-17 μg per 1 g dust respectively, indicating a comparable loading of the two classes of perfluorinated compounds. PFC median values in the present study and other studies are

given in Table 2.14.

Table 2.13. Dust concentrations (ng/g), summary statistics, and detection frequencies of neutral (FTOHs, FOSAs, FOSEs) and acidic PFCs (PFCAs, PFSAs) from homes in Winnipeg, Canada (n = 132).

Analyte	Median	Mean	Range	Frequency of Detection (%)
PFPA	16	210	<LOQ-413	28.6
PFHxA	57	140	20-2214	100
PFHpA	27	207	8.1-2896	96.2
PFOA	81	360	12-3336	100
PFNA	28	153	6.9-2164	97.7
PFDA	28	133	8.2-2182	93.1
PFUA	<LOQ	2.4	<LOQ-12	21.2
PFBS	15	85	4.4-838	53.0
PFHxS	20	249	<LOQ-8476	88.6
PFOS	78	654	6.4-11398	100
PFDS	16	114	2.4-1766	83.3
FOSA	<LOQ	24	<LOQ-1966	18.9
MeFOSA	<LOQ	6.9	<LOQ -21.4	25.0
EtFOSA	<LOQ	2.9	<LOQ-21.1	28.0
MeFOSE	39	115	3.8-1388	55.3
EtFOSE	15	61	<LOQ-4032	82.6

Table 2.14. Summary of median concentrations of PFCs (ng/g) in dust samples reported in the present and related studies.

	Country	Year	N	PFHpA	PFOA	PFBS	PFHxS	PFOS
1	Winnipeg, Canada[a]	2011/12	132 rooms from 86 homes	27	81	15	20	78
2	Vancouver, Canada[b]	2007/8	132 homes	69	30	NA	NA	71
3	Ottawa, Canada[c]	2003/3	67 homes	NA	20	<4.39	23.1	38
4	North Carolina, USA[d]	2000/1	112 homes and daycares	50	142	9	46	201
5	Wisconsin, USA[e]	2008	39 homes	17	44	1.8	16	47
6	UK, AU, DE, US[f]	2004	39 homes	97.3	96.5	359	185.5	479.6
7	Stockholm, Sweden[g]	2006/7	38 apartments	NA	93	NA	NA	85
8	Oslo, Norway[h]	2008	41 homes	10	18	0.4	0.6	3
9	Japan[i]	2003	16 homes	NA	165	NA	NA	25

N = Number of samples

NA = Not available

a. Present study

b. Shoeib et al., 2011

c. Kubwabo et al., 2005

d. Strynar et al., 2008

e. Knobeloch et al., 2012

f. Kato et al., 2009

g. Björklund et al., 2009

h. Haug et al., 2011

i. Moriwaki et al., 2003

In contrast with the Ottawa study in which PFBS was not detected (Kubwabo et al., 2005), this compound was detected in the present study in 53% of the samples (Table 2.14). That may have resulted from application of the perfluorobutyl (PFB) chemical component as a building block molecule for surfactants, which has only occurred in the last few years. Another explanation would be the recent replacements for the longer chain PFCs, such as C-6 and C-8 surfactant analogs (Ehresman et al., 2007), by materials that result in PFBS as a final degradation product. Variation in dust concentrations of PFCs in different geographical locations could result from different application of PFCs, different PFC contents in treated goods within homes, and other parameters related to occupants and home characteristics. As pointed out previously (Harrad et al., 2010b), differences among studies in terms of both sampling approaches for house dust and extraction methods can complicate comparisons among studies. The dust sampling approach could affect results because vacuuming the entire room may oversample less-frequented parts of a room or sampling one specific area of a room may not be a representative sample to assess contamination within the room. The specific area where a sample is collected may have either higher potential for the presence of PFCs (e.g., spots closer to source of PFCs) or lower potential. Moreover, some vacuum cleaners used for sampling may well be treated with PFCs. To minimize the mentioned effects in this study, dust samples were collected by vacuuming in the corner, under the child's bed and inside the bedroom closet; and vacuuming the entire floor in cases where the floor was completely carpeted. Furthermore, parts which likely contained polytetrafluoroethylene (PTFE) tubing, seals in vacuum cleaner were replaced with Teflon®-free material such as stainless steel or polypropylene (Martin et al., 2004).

Transportation of samples under controlled conditions (in sealed containers and at sufficiently low temperatures) minimizes loss of PFCs in air samples (Harrad et al., 2010a). Therefore, transportation of samples to the laboratory has been suggested as another factor affecting results.

2.2.4.1 Correlations Among PFCs in House Dust

A significant association in house dust was seen between PFOA and PFOS (Table 2.15), which was consistent with other studies (Knobeloch et al., 2012; D'Hollander et al., 2010; Haug et al., 2011; Shoeib et al., 2011, Kato et al., 2009). This observation may suggest a common source of these residues in house dust samples. There was also a positive and significant correlation between PFOS and PFHxS concentrations in dust (Table 2.15). That is in line with other reports (Knobeloch et al 2012; Kato et al., 2009; Haug et al., 2011) suggesting that these compounds may originate from a common source. PFOS and PFHxS are likely to originate from floor coverings, draperies, and upholstered furnishings treated with these compounds, whereas home construction materials are not likely to be a significant source (Knobeloch 2012). Furthermore, PFHxS in house dust can originate from products used for coating treatment of furniture and food packaging (Olsen et al., 2003a; Olsen et al., 2003b).

In house dust, significant associations were seen among nearly all the PFCAs, whereas correlations within the PFSAs, as well as between PFCAs and PFSAs, were less observed (Table 2.15). In agreement with this finding, a European study reported less frequent correlations between PFCAs and PFSAs in Norway (Haug, 2011). Less frequent correlations between PFCAs and PFSAs suggests that when concentrations of PFCAs

vary, other compounds do not necessarily vary in the same way. This suggests that the majority of PFCAs and PFSAs are unlikely to come from a common source.

Table 2.15. Spearman's rank correlations of perfluorinated compounds in dust samples from homes in Winnipeg, Canada (n = 77). (*) indicates significant correlation (correlation was significant at the 0.05 level or less).

	MeFOSE	PFPA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFBS	PFHxS	PFOS	PFDS
EtFOSE	0.34	0.12	0.47	0.42	0.6*	0.26	0.3	0.32	0.24	0.44	-0.03
MeFOSE		-0.12	0.2	0.27	0.22	0.14	-0.03	0.13	-0.1	0.29	-0.11
PFPA			0.67*	0.61*	0.64*	0.3*	0.34	0.09	0.08	0.34	-0.23
PFHxA				0.84*	0.78	0.58	0.67	0.19	0.06	0.47	-0.03
PFHpA					0.88*	0.83	0.78*	0.12	0.02	0.69*	0.27
PFOA						0.71	0.83*	0.28	0.14	0.7*	0.3
PFNA							0.58*	0.15	0.06	0.57	0.24
PFDA								0.06	0.06	0.47	0.33
PFBS									0.5	0.56	-0.18
PFHxS										0.57*	0.15
PFOS											0.4

2.2.4.2 PFC Correlations Between House Dust and Indoor Air

Significant correlations between air and dust for MeFOSE in paired dust and air concentrations from the same homes in this study (Table 2.16) indicate that either the compound originated from a similar source or a close coupling between air and dust. Positive associations between FTOHs in air and PFCAs in dust were anticipated based on suggested transformation of FTOHs to PFCAs in the atmosphere (Chapter 1.6) observed by monitoring atmospheric oxidation of FTOHs in a smog chamber (Ellis et al., 2004). The significant associations among all the PFCAs in dust (except PFOA and PFHxA) and FTOHs in air in this study support this hypothesis. No correlation between FTOHs or PFOA indicates that concentrations of PFOA in house dust samples may be driven by sources other than degradation of 8:2 FTOH to PFOA. PFOA and PFNA are known degradation products of 8:2 FTOH (Ellis et al., 2004). In agreement with this hypothesis, a significant correlation between PFNA and 8:2 FTOH was found in the present study (Table 2.16). It has been suggested that FOSA/FOSEs may be transformed to PFOS and PFCAs in the atmosphere (Martin et al., 2006, D'Eon et al., 2006) to form more stable PFCs. MeFOSA and MeFOSE in indoor air were significantly associated with PFOS and PFCAs (Table 2.16), which is consistent with the hypothesis.

Table 2.16. Spearman's rank correlations of perfluorinated compounds in indoor air and dust samples from homes in Winnipeg, Canada (n = 77). (*) indicates significant correlation (correlation was significant at the 0.05 level or less).

		Dust											
		EtFOSE	MeFOSE	PFPA	PFHxA	PFHpA	PFOA	PFNA	PFDA	PFBS	PFHxS	PFOS	PFDS
Indoor Air	6:2 FTOH	-0.04	0.42*	-0.43	0.21	0.55*	0.18	0.33*	0.36	-0.2	0.44	0.14	-0.1
	8:2 FTOH	-0.24	0.29	0.14	0.3	0.32	0.15	0.36*	0.27	-0.05	-0.14	0.13	-0.02
	10:2 FTOH	-0.26	0.13	0.3*	0.37	0.28*	0.18	0.36*	0.35*	-0.06	-0.05	0.04	-0.02
	Et-FOSA	0.11	0.2	0.39	0.13	0.22	0.12	0.24	0.07	0.84*	-0.17	0.27	-0.1
	Me-FOSA	-0.04	0.45	0.09	0.12	0.44*	0.61*	0.37	0.17	0.27	0.71*	0.56*	0.14
	Me-FOSE	0.04	0.86*	0.02	0.19	0.42*	0.57*	0.24	0.11	0.29	0.66*	0.50*	-0.01
	Et-FOSE	0.29	0.18	0.32	0.15	0.17	0.4	0.14	0.16	0.85*	0.36	0.43	-0.31

2.2.5 Correlation of PFCs and Home Characteristics

Extensive surveys regarding home characteristics were performed to investigate impacts of home characteristics on the presence of PFCs in indoor air and dust, and the likelihood that these characteristics contribute to human exposure. The surveys covered various aspects of volunteers' habits, housekeeping practices, and the building materials in their homes. Correlations between PFCs and home characteristics were performed on the measured concentrations of PFCs using appropriate nonparametric tests; Spearman rank and Mann-Whitney tests. Among the various explored variables, those that showed association with PFCs are listed in Table 2.17.

Median values of PFHpA, PFOA, PFOS, and PFHxS in dust were significantly higher in homes built after 1990 than those built in 1940 or earlier. These observations indicate higher concentrations of these PFCs in homes constructed recently compared to much older homes, which could result from higher applications of PFCs in recent years for floor coverings and upholstered furnishings. This is consistent with previous research, which reported a significant negative association between the mentioned PFCs (except for PFHxS) and age of the houses built after 1990 (Knobeloch et al., 2012). The lack of association between PFCs and age of homes built between 1904 and 1990 suggests that the presence of PFCs in those homes may mostly result from other sources, such as commercial products used by occupants. Homes with added or renovated space showed significantly higher levels of MeFOSE in indoor air and PFPA in dust. Higher occurrence of PFPA in renovated homes may result from higher application of PFPA in recent years as a replacement for longer chain PFCs (Conder et al., 2008; Prevedouros et al., 2006) in

building and coating material. This suggests that renovation results in more recent PFC and PFC-bearing materials being brought into the home.

Fully carpeted homes had significantly higher concentrations of 8:2 FTOH in indoor air and MeFOSE in house dust than homes that were not carpeted, whereas homes with laminate flooring had significantly higher concentrations of PFDA. PFDS was the only PFC detected in a borderline significant ($p = 0.055$) higher concentration in homes with carpeted living rooms than uncarpeted ones. Interestingly, homes painted in the 12 months prior to the completion of the questionnaire showed significantly elevated concentrations of 8:2 FTOH in indoor air. MeFOSE and FTOHs are residual compounds produced in the manufacture of PFCs during production of polymeric and surfactant solutions used for carpet cleaning and surfactants for paints and lacquers (Dinglasan – Panlilio & Mabury, 2006). Because more volatile compounds have a greater tendency than less volatile compounds to be present in indoor air, 8:2 FTOH as a volatile PFC is believed to occur from the freshness of the paint in homes.

PFHxA and PFHxS are present in stain and greaseproof coatings on food packaging, furniture, and household products (Olsen et al., 2003a; Olsen et al., 2003b). Levels of PFHxA were significantly lower in homes with upgraded ventilation systems while homes with any major pieces of plastic or vinyl covered furniture showed significantly higher concentration of PFHxS. An explanation for lower levels of PFHxA in the homes with upgraded ventilations could be based on this theory that upgraded ventilation may lead to increasing the ventilation rate. However, this increase does not effectively reduce the overall dust level because the dust production rate would increase with an increase of ventilation rate (Wang et al., 2002). Therefore, better ventilation means more dust which

then dilutes the concentration which can lead to a lower PFHxA loading within the sampled dust (Harrad *et al.*, 2009; Kubwabo *et al.,* 2005). The higher concentration of PFHxS in homes with major pieces of plastic or vinyl-covered furniture is likely, possibly due to a higher leaching rate of the compound from the treated objects, or even just due to the presence of those compounds at all because of the furniture.

Concentrations of EtFOSE and PFHpA were influenced by proximity to a highway/artery road or major/prolonged construction activity, respectively. Significantly higher levels of EtFOSE and PFHpA were detected near those areas. This observation is inconsistent with a previous study that found that outdoor levels of PFCs were typically much less than indoor air (Harrad et al., 2010); however, it is possible that increased concentrations of PFHpA are driven by elevated degradation rates of FTOHs (Ellis et al., 2004) in air created by 'street canyons' (Yarwood et al., 2007). Factors such as vehicle emissions and the presence of free radicals, along with the proposed presence of PFCs in vehicle oils and other surfactants (Drobny et al. 2005) could increase the degradation rates of FTOHs.

Homes with regular use of kitchen stove fans were found to have significantly lower concentrations of 10:2 FTOH in indoor air. This may be explained that because the kitchen stove fan would vent directly out of the house, hence, it should reduce levels of indoor air compounds. Therefore, 10:2 FTOH levels would correlate negatively with stove fan use. However no clear explanation was found that why only 10:2 FTOH is affected by stove fan.

PFHxS was found to be associated with the frequency of cleaning (vacuuming). Homes cleaned more than 15 times per month showed significantly lower concentrations

of PFHxS than those cleaned fewer than 3 times. PFHxA was detected at borderline significantly ($p = 0.057$) lower concentrations in houses using wet dust-attracting devices. In general, greater cleaning frequency can lead to less dust, and finer material in the overall total weight, which can lead to a greater PFC loading within the sampled dust (Harrad *et al.*, 2009 and Kubwabo *et al.,* 2005). However, the lower levels of PFHxS and PFHxA observed in home with greater cleaning frequency in this study suggests that in parallel with increase in cleaning frequency, another factor may vary as well that led to a decrease in the leaching rate of the compounds from treated objects in those homes. The latter overcomes the impact of the first factor (i.e., decreasing dust levels) and the overall trend would be a decrease in concentration of the compounds. Positive significant correlations were found between concentrations of PFHpA PFOA, PFNA, and PFDA in house dust and total volume of the house (excluding the basement) with Spearman's rank correlation coefficients of 0.31, 0.30, 0.23, and 0.45 respectively. This may indicate an increase in concentrations of the PFCs in dust with increasing house volume, which may result from an increase in surface area containing the PFC-coated materials. The positive correlations between PFCs in dust (PFHxS, PFDS, PFOS) and volume of homes have been reported in a previous study (Haug et al., 2011). However, the correlated PFCs are different than those in the current study. Additionally, the considered volume in the previous study was the volume of the living room while in the present study, the total volume of the house (excluding the basement) was considered. The home characteristics that may influence the indoor air and dust concentrations of PFCs are listed in Table 2.17. Other home characteristics that were tested for correlations had no significant correlation with concentrations of PFCs and are listed in Appendix F.

Table 2.17. Home characteristics that may influence the indoor air and dust concentrations of PFCs; they all were tested for bivariate correlations by Mann-Whitney test except "Total volume of the house" which was tested by Spearman rank.

Home characteristic (Variable)	**Unit (range)**
Home originally built	Before 1939-1990 and later (Yes/No)
Added or renovated space to home	(Yes/No)
Upgraded ventilation	(Yes/No)
Painting	(Yes/No)
Any plastic or vinyl covered furniture (major pieces)	(Yes/No)
Laminate (floor)	(Yes/No)
Home near highway/artery	(Yes/No)
Near major/prolonged construction activity	(Yes/No)
Regular use of stove fan in the kitchen	(Yes/No)
Frequency of home cleaning per month	(1-30 times)
Using wet dust-attracting device	(Yes/No)
Full carpet flooring	(Yes/No)
Total volume of the house	(130-650 m^3)

2.2.6 Indoor Ambient Temperature Dependence of Neutral PFCs

For the first time in indoor studies of PFCs, the ambient temperature was logged every 4 minutes during the entire deployment of SIP disks. Temperature variation range was not more than 12^oC in homes (from 15 to 27^oC). Indoor ambient temperatures are typically adjusted by occupants to more or less similar temperatures in different homes. Various ambient temperatures between homes could result from window ventilation

frequency, indoor heating systems, and building characteristics such as applied isolation systems. Not surprisingly, no significant association was found between the ambient temperature and concentrations of PFCs. This suggests that the ambient temperature variations among homes are not sufficiently large to impact the vaporisation rate of neutral PFCs. No previous data has been reported for vaporization temperatures of neutral PFCs hence this represents a unique element of the current study.

2.2.7 Seasonal Dependence of PFC Concentrations

To explore the seasonal variation of indoor air concentrations of PFCs, summer (May to August, n = 14) and winter (September to April, n = 63) concentrations from different homes for each individual compound were compared using the Mann-Whitney test. The percent change in median concentration from summer to winter was found to be 20, 554, -10, 10, -23, 111, and -22% for 6:2 FTOH, 8:2 FTOH, 10:2 FTOH, MeFOSA, EtFOSA, MeFOSE, and EtFOSE, respectively (Table 2.18). Apart from insignificant differences in the majority of the compounds, significant differences between the two seasons were found for 8:2 FTOH (554% increase) and MeFOSE (111% increase). This observation contradicts a Japanese study by Liu et al. (2013) that found higher concentrations of 8:2 FTOH in the summer months compared to those in winter. This disagreement in the seasonal trend of 8:2 FTOH between two geographical places could result from the influence of other factors on the presence of 8:2 FTOH in indoor air, such as home conditions and consumer habits. The higher winter levels of 8:2 FTOH and MeFOSE in indoor air in Winnipeg may be due to lower window ventilation during extremely cold weather

in winter.

Table 2.18. Summer (n = 14) and winter (n = 63) median concentrations (pg/m^3) of PFCs in indoor air. (*) indicates significant differences between the two seasons.

Analyte	Summer	Winter
6:2 FTOH	10.0	12.0
8:2 FTOH*	195	1280
10:2 FTOH	576	520
EtFOSA	36.1	39.7
MeFOSA	70.6	54.4
MeFOSE*	277	583
EtFOSE	77.9	60.7

There has only been one previous publication related to the seasonal variations of PFCs in indoor air (Goosey et al., 2012), and this reported no significant seasonal variation in indoor air concentrations. In the present study, seasonal variation was explored by comparing different homes in different seasons, i.e., PFC data collected in summer time were not from the same homes as winter data. Hence, there are many parameters within a home that may vary besides season, including home characteristics and activities of occupants. These can lead to differences in source emissions of PFCs. While this was not done in the current study, it would be interesting to monitor seasonal variation of PFCs in the same homes in different seasons. By minimizing the number of other interacting variables, one could achieve more relevant results and provide a more definitive characteriza-

tion of the effects of seasonal variability on indoor concentrations of PFCs.

2.2.8 Distribution of PFCs in House Dust Between Rooms in the Same House

No significant differences were observed between concentrations of PFCs in the child's bedroom and the most used room within the same house (Mann-Whitney test). This observation suggests that dust was distributed uniformly in rooms within the same house, probably as a result of indoor circulation, convection, or movement of people between rooms. Furthermore, house dust samples from the child's room and the most used room had significant correlation coefficients for all PFCs with median concentrations higher than their respective LOQ values, except PFPA, i.e., MeFOSE, EtFOSE, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFBS, PFHxS, PFOS, and PFDS (Figure 2.1 and 2.2). All slopes were found to be significantly different than zero. Greater slopes indicate greater dependency of the most used room concentrations on the concentrations in the child's room.

Figure 2.1. Correlations between concentrations of PFCs in dust from the most used room (MR) and child's room (CR) for MeFOSE, EtFOSE, PFBS, PFHxS, PFOS, and PFDS in the same home.

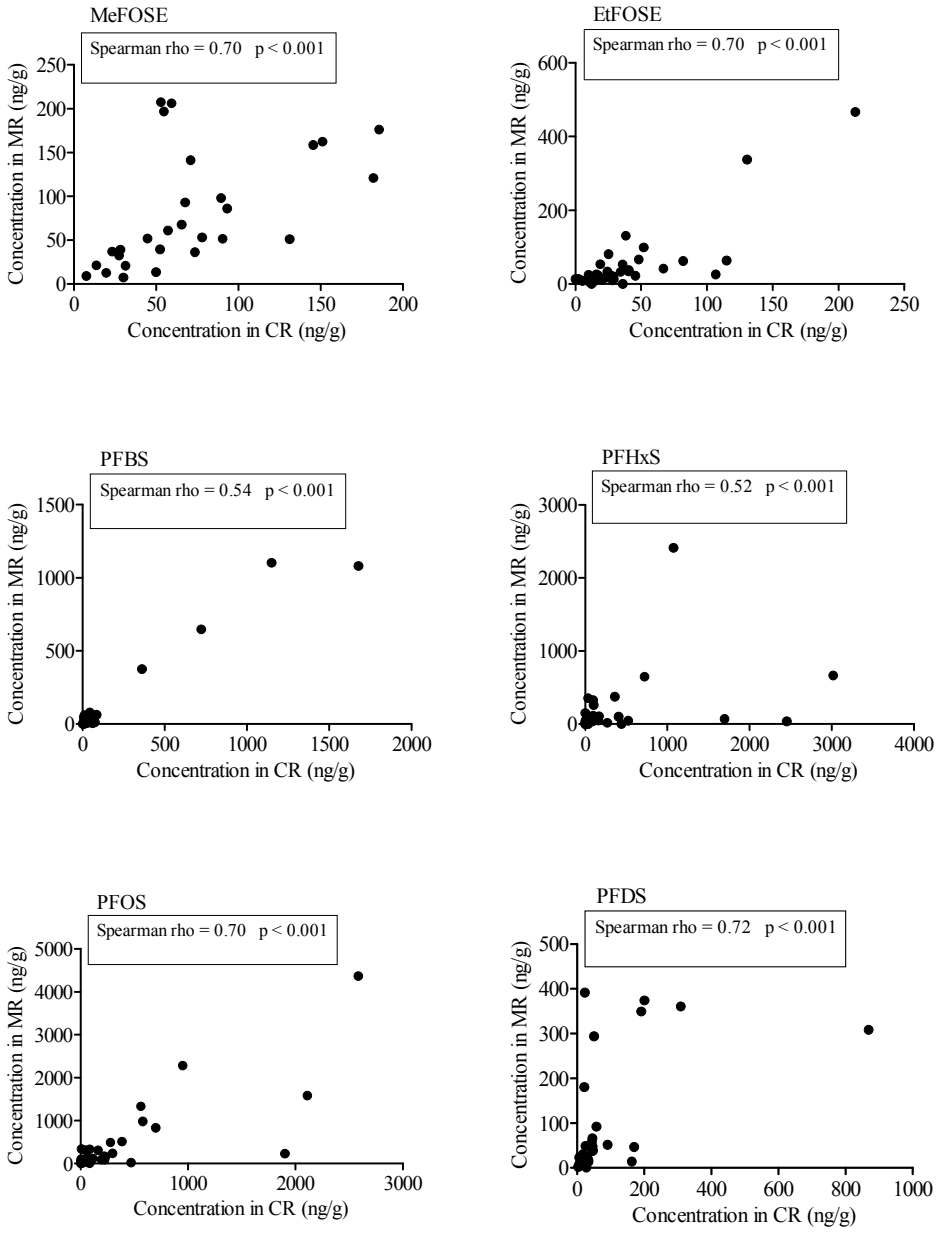
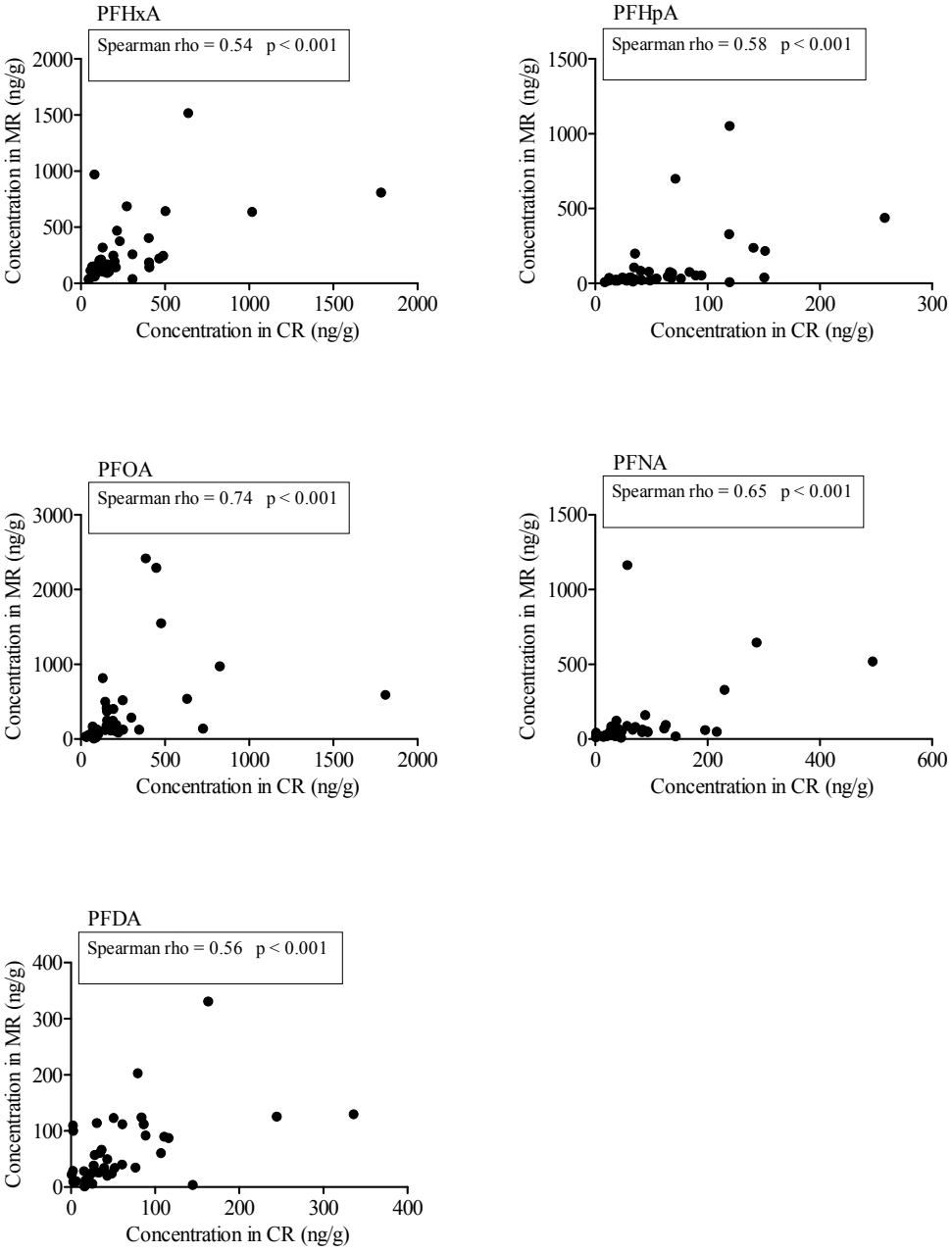


Figure 2.2. Correlations between concentrations of PFCs in dust from the most used room (MR) and child's room (CR) for PFHxA, PFHpA, PFOA, PFNA, and PFDA in the same home.

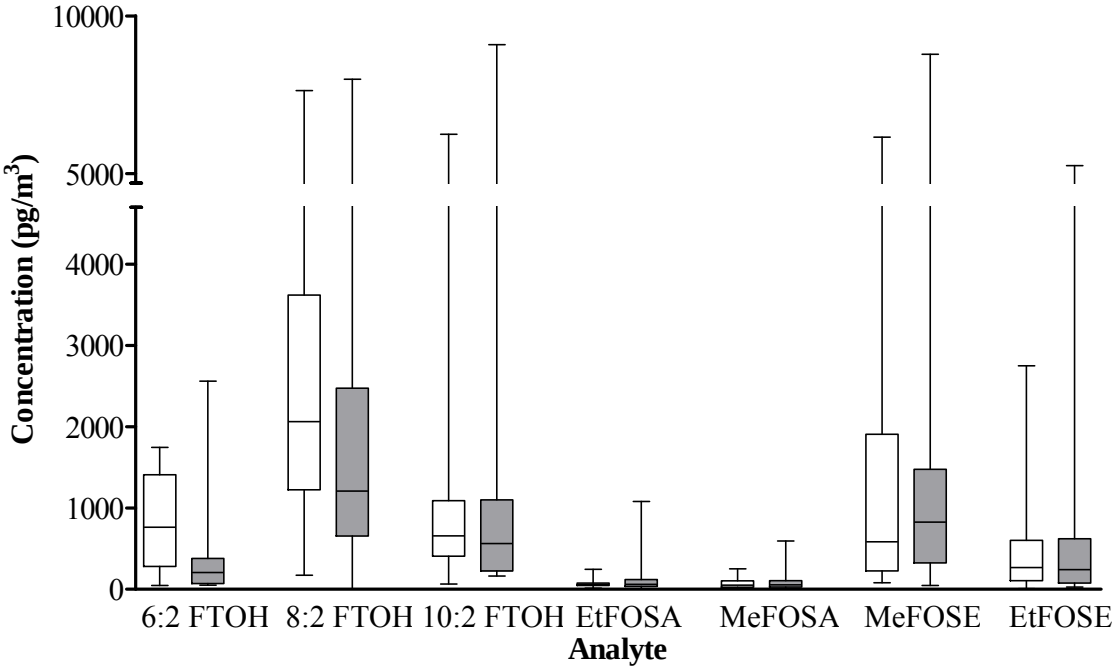


One possible explanation for the lack of correlation between the two rooms for concentrations of PFPA is that PFPA was not detected in as many samples as other PFCs; the frequency of detection was only 29% (Table 2.13). This is the first time that house dust PFC concentrations have been compared between rooms within the same home. However, in a previous study, the effect of room conditions on indoor air concentration of 8:2 FTOH was explored by comparing the concentrations between living rooms and bedrooms; the results showed significantly higher concentrations in the bedrooms in comparison to the living rooms (Liu et al., 2013).

2.2.9 PFC Concentrations in Indoor Air and Dust and Incidence of Wheezing in Infants

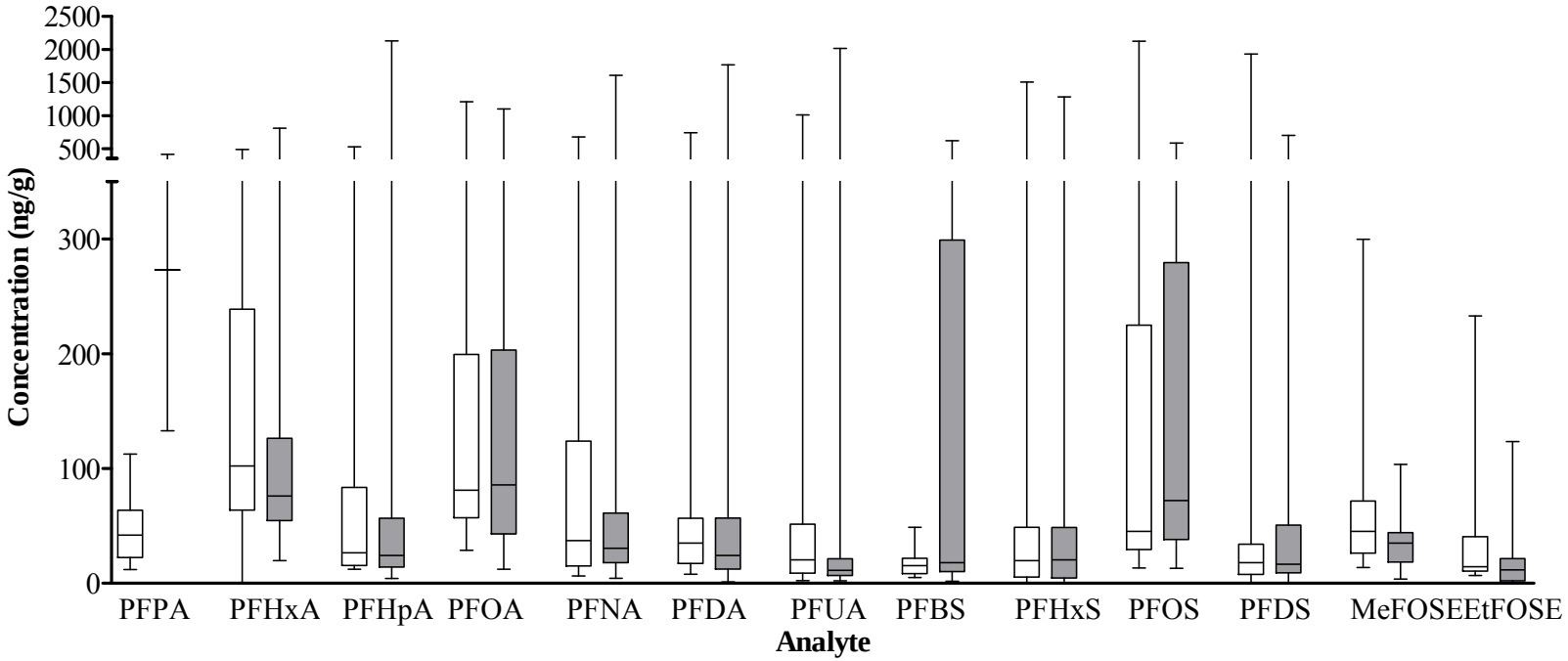
To determine whether or not relationships exist between PFCs in indoor air and dust and incidence of wheezing in infants, indoor air and dust concentrations of PFCs in target and control homes in Winnipeg were compared. Homes in which there was an infant with recurrent wheezing confirmed by a physician were considered target homes, while children in control homes had no diagnosed episodes of wheezing. Target and control homes were selected whenever possible as close to each other in the city to minimize differences in PFCs due to geographical factors. Comparisons of 28 pairs of indoor air samples (control and target) and 24 pairs of house dust samples (control and target) using the Wilcoxon paired *t*-test showed no significant differences between control and target homes for either type of sample. Figure 2.3 and 2.4 compare median concentrations of control and target homes in indoor air and dust, respectively.

Figure 2.3. Box and whisker plot of matched target and control samples in indoor air (n = 28 matched samples). Whiskers show the minimum to maximum values. Grey boxes display target home samples and white boxes display control home samples.



This observation indicates that the specific analytes and concentration levels of PFCs measured in this set of indoor air and house dust did not affect the occurrence of wheezing. However, the sample numbers for these comparisons were relatively low and, therefore, further investigation with a larger sample size may yield different results. This is the first study to assess associations between PFCs present in indoor air and dust and the incidence of wheezing. Only a few reports have been published that explored potential associations between PFCs in plasma and wheezing (Okada et al., 2012; McConkey, 2012); none were found in all cases. It should be noted that FTOHs, which

Figure 2.4. Box and whisker plot of matched target and control samples in house dust (n = 24 matched samples). Whiskers show the minimum to maximum values. Grey boxes display target home samples and white boxes display control home samples.



were explored in present work, were not analysed in previous studies. The lack of association of PFOA with wheezing observed in this study is inconsistent with laboratory studies that have found PFOA exposure can cause changes in the lungs, such as increased levels of activated receptors (Abbott et al., 2012; Rosen et al., 2007), which may cause developmental toxicity (Abbott et al., 2009). It should be noted that in the latter studies, mice were exposed to higher concentrations (5 mg/kg/day) than dust concentrations in the present study (median of 81 ng/g).

2.2.10 Principal Component Analysis

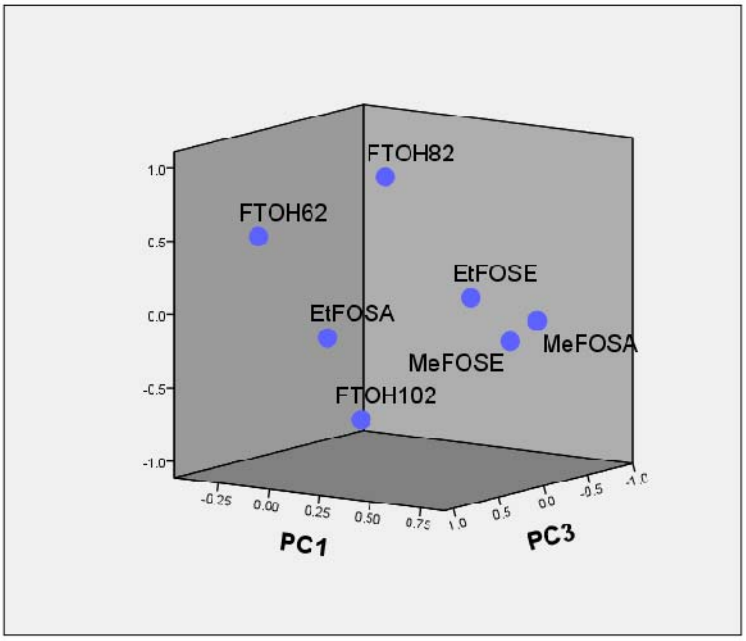
Principal component analysis (PCA) was used to assess exposure and trends with indoor air and dust PFC concentrations. The proportions of each PFC relative to the sum of total PFCs were used as the variables for PCA. For concentrations under the LOQ, random values between zero and the LOQ were substituted, the proportions were calculated, and then natural log transformed. Questionnaire data regarding home characteristics was used to assess exposure and trends with the PCA data. Parametric statistical tests (one-way ANOVA, unpaired *t*-test, Pearson correlation) were used because the proportions of PFCs were natural log transformed and the natural log was normally distributed.

2.2.10.1 Exposure and Trends of PFCs From Home Characteristics

For both indoor air and dust three principal components (PCs) were retained based on the eigenvalue-one criterion (Hatcher, 1994). Because the eigenvalue for each

of the three PCs had a value above unity, the variance described by the PC is greater than the contribution of the variance from one variable. The percent of variability accounted for in the indoor air data by each PC was 26% for PC1, 22% for PC2, and 17% for PC3. The loadings plot of the three PCs is shown in Figure 2.5.

Figure 2.5. Loadings plot of PC1, PC2, and PC3 for PFCs in indoor air.



This plot shows the weight of the proportion of each PFC on the PCs. Because it is difficult to interpret PCA results from a three-dimensional plot, the three-dimensional loadings plot was divided into two-dimensional plots (Figures 2.6 and 2.7). There was a trend that PFCs with lower vapour pressure loaded lower on PC1 (Figure 2.6). The 6:2 FTOH with the highest vapour pressure (876 Pa at 25°C, Lei et al., 2004) loaded around zero, FOSA/FOSEs with lower vapour pressure than FTOHs loaded high on PC1.

Figure 2.6. Loadings plot of PC1 for PFCs in indoor air.

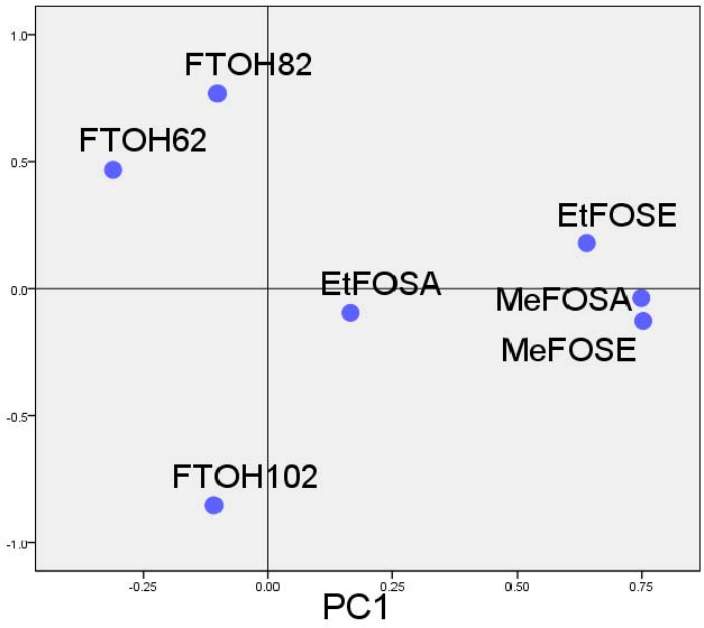
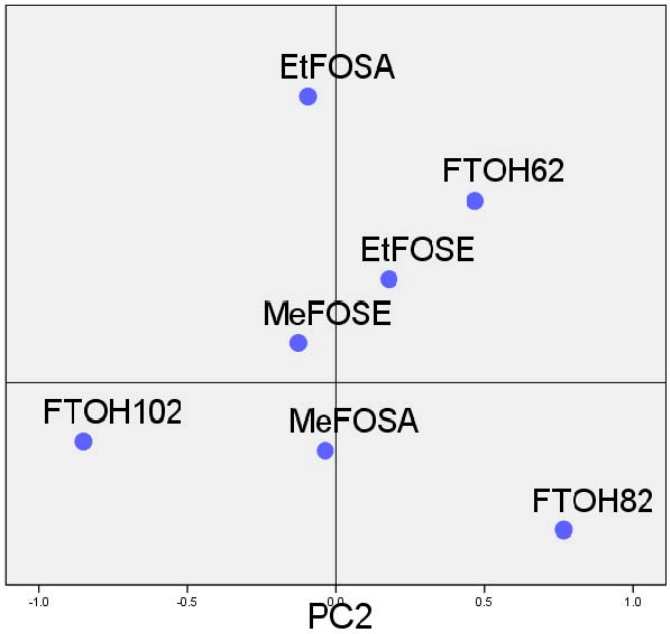


Figure 2.7. Loadings plot of PC2 for PFCs in indoor air.



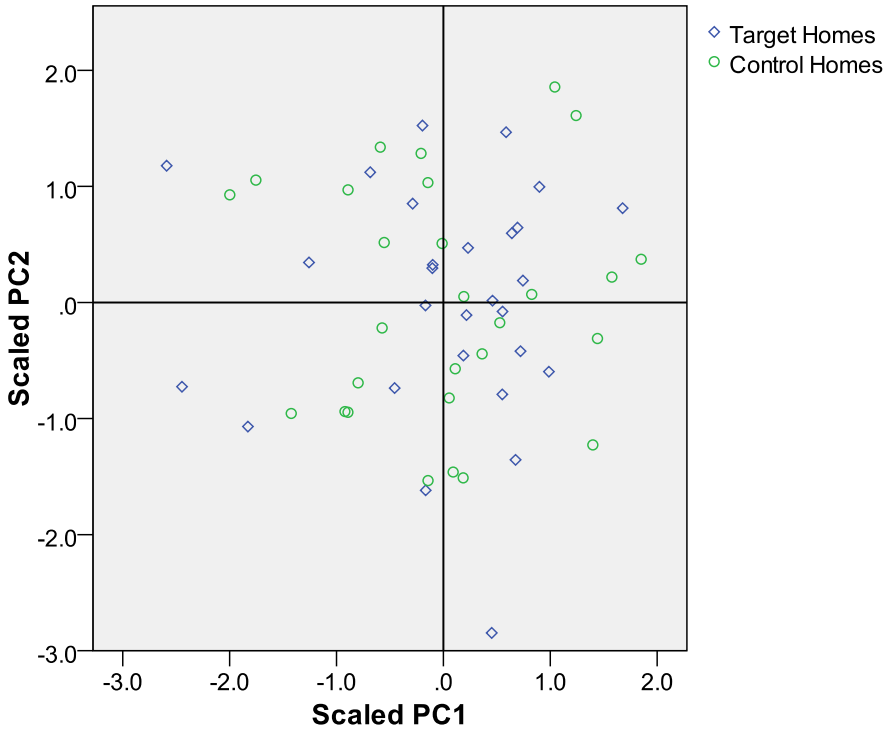
The loadings plot for PC2 in Figure 2.7 shows a high loading for 8:2 FTOH . All other PFCs loaded close to zero or lower than zero on PC2. Although there was no clear trend, however this difference in loading suggests that 8:2 FTOH, the PFC with the highest abundance in indoor air (Haug et al., 2011; Shoeib et al., 2011; Jogsten et al., 2012) loaded higher on PC2. Although 8:2 FTOH is loaded higher, all the six PFCs have approximately similar loadings on PC2 because they may have been used in many different products and often in mixtures so there would be exposure to all of these PFCs combined.

2.2.10.2 PCA Results

2.2.10.2.1 PCA Results of Indoor Air

The scores plots for PC1 and PC2 in indoor air shown in Figure 2.8 show that homes were influenced differently by PC1 and PC2 as most data points were spread out. No significant difference was observed between the control homes and target homes in terms of loading on PC1 (paired *t*-test), indicating no significant difference between loading volatile PFCs between the two groups of homes (because more volatile PFCs were loaded lower on PC1). This observation suggests no correlation between wheezing and presence of the PFCs.

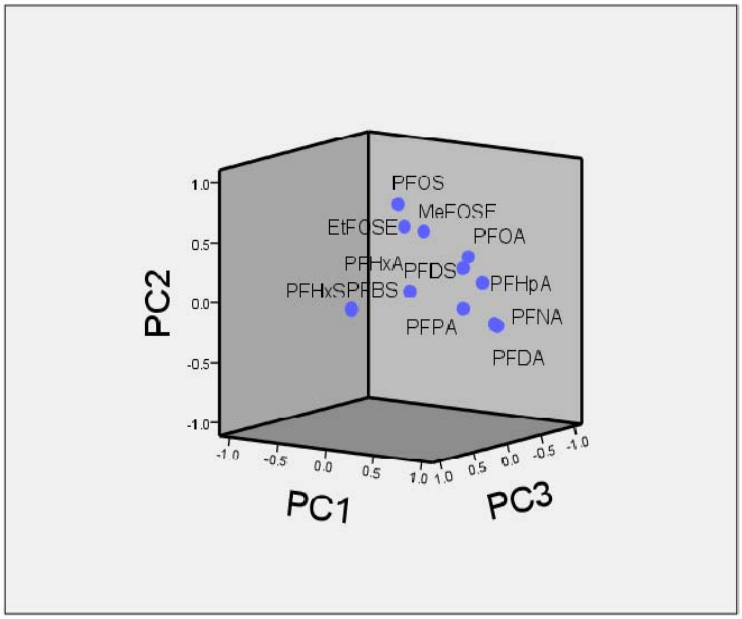
Figure 2.8. Scores plot of PC2 versus PC1 for PFCs in indoor air.



2.2.10.2.2 PCA Results of House Dust

PCA results for house dust showed that the percent of variability the concentrations of house dust data accounted for by each PC was 37% from PC1, 13% from PC2, and 10% from PC3. The loadings plot of the three PCs is shown in Figure 2.9.

Figure 2.9. Loadings plot of PC1, PC2, and PC3 for PFCs in house dust.



The loadings plots for PC1 are shown in the plots in Figure 2.10. There was a trend that perfuorocarboxylic acids (PFCAs) loaded higher than PFSAs and FOSEs. PFCAs loaded closely to each other on PC1; this may indicate mixtures of the PFCAs used in many products such as metal cleaners, electrolytic plating baths, self-shine floor polishes, fire-fighting formulations, varnishes, lubricants, gasoline, and paper, leather, textile treatments, copier toner, electronic, and semiconductor applications (Prevedouros., 2006). A concentration range between 100 and 5000 ppm of PFCAs is used in industrial formulations (Prevedouros, 2006).

Figure 2.10. Loadings plot of PC1 for PFCs in house dust.

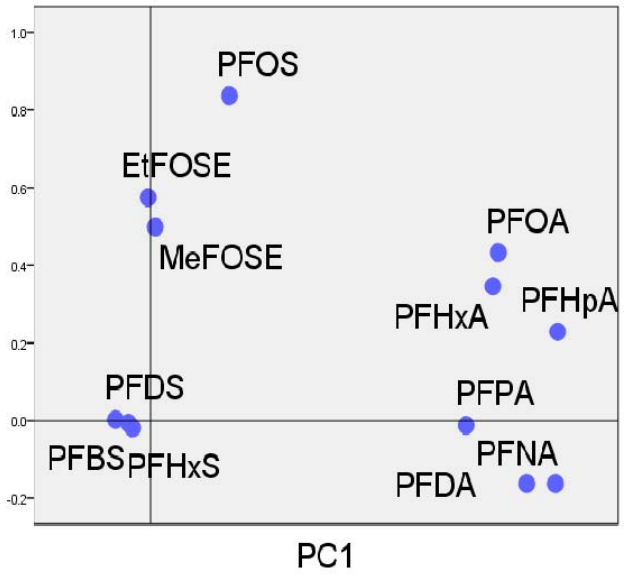
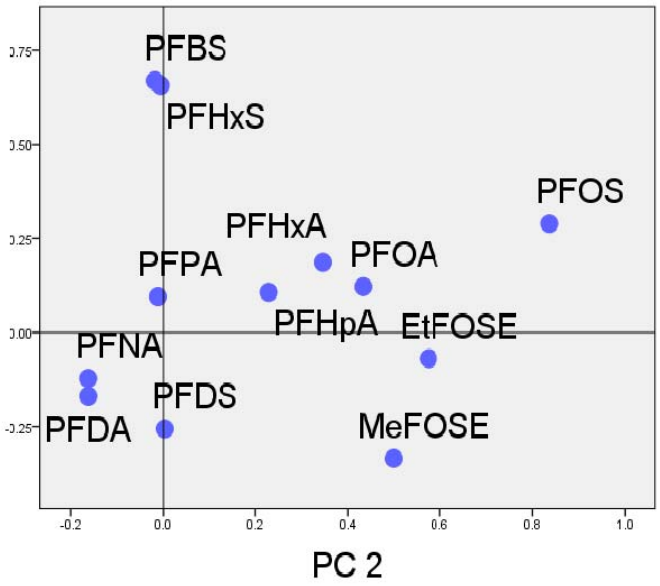


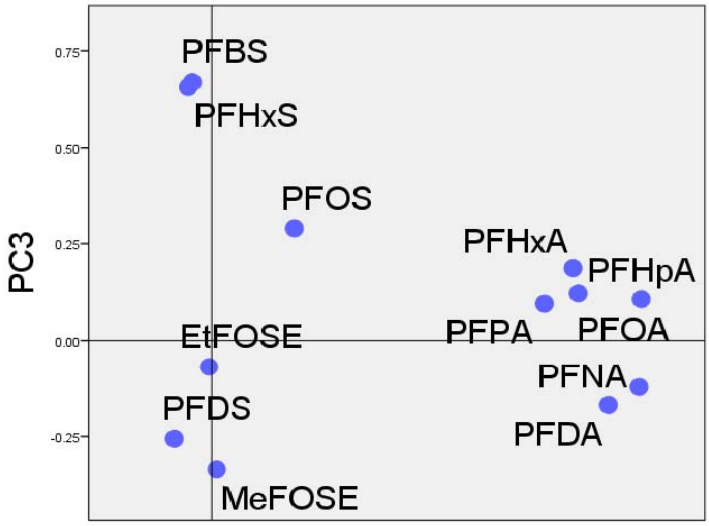
Figure 2.11. Loadings plot of PC2 for PFCs in house dust.



The loadings plots for PC2 are shown in the plots in Figure 2.11. There was a trend that the PFCs which are produced in larger quantities, i.e. PFCs with 8 carbon chains loaded higher. Physical properties of PFCs have made them as ideal surfactants. The eight

carbon chain PFCs are the best surfactants among the various perflurocarbon chain lengths (Lau et al., 2007), which is why the eight carbon chain PFCs were produced in large quantities.

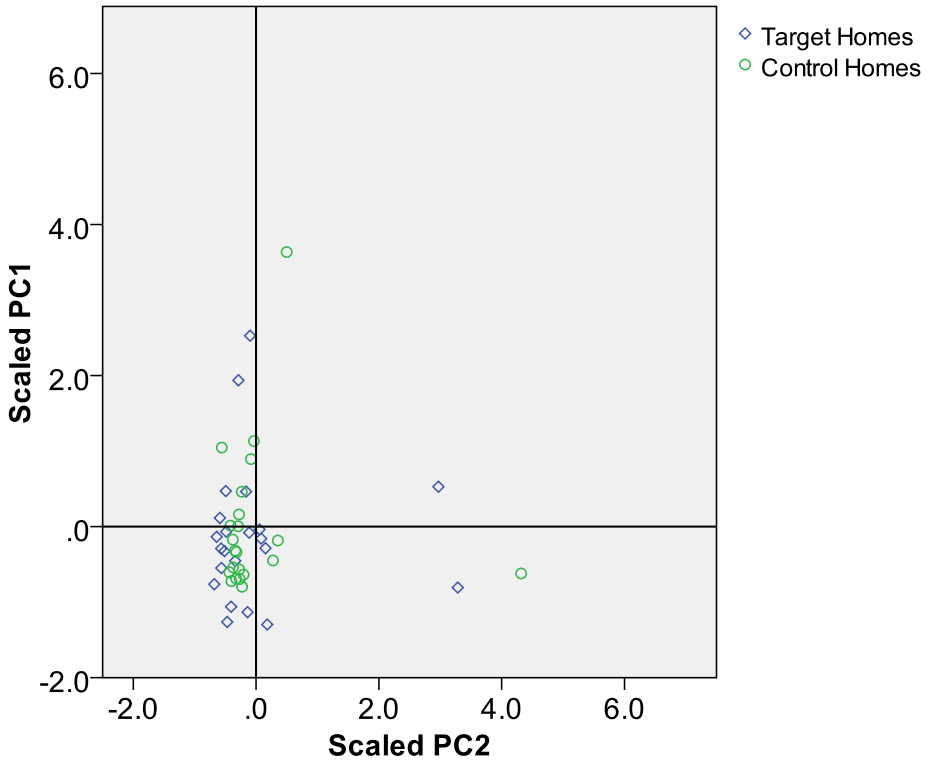
Figure 2.12. Loadings plot of PC3 for PFCs in house dust.



The loadings plots for PC3 are shown in the plots in Figure 2.12. There was a trend based on chain length that the PFCs with shorter chains loaded higher. The loadings plots in Figure 2.12 shows that for PC3, the order of increasing loadings in PFSAs is PFBS (four carbon chain-length) > PFHxS (six carbon chain-length) > PFOS (eight carbon chain-length) > PFDS (ten carbon chain-length) and in PFCAs, the shorter chains, four, six, seven and eight carbon chain-length i.e. PFPA, PFHxA, PFHpA and PFOA loaded higher than longer chain length i.e. nine and ten chain-length PFNA and PFDA. PFPA loaded close to PFOA, probably because shorter chain-length PFCs are being used as replacements for longer chain-length PFCs in recently manufactured products. It

would be interesting for future studies to analyse dust samples for short chain-length PFCs such as PFPA as well as the four carbon chain-length PFBA to see how these levels in dust change in the future.

Figure 2.13. Scores plot of PC2 versus PC1 for PFCs in house dust.



The majority of homes were influenced similarly by PC1 and PC2, as most data points were grouped together and loaded lower in PC1 and PC2 (Figure 2.13). No significant difference was observed for PC1 and PC2 between control and target homes (paired *t*-test), suggesting loading PFCAs, PFSAs and PFCs with 8 carbon chains does not have significant difference between control and target homes which indicate no association between wheezing and distributions of PFCAs, PFSAs and PFCs with 8 carbon chains.

Furthermore, no significant difference was observed for PC1 and PC2 between

child room and the most used room (paired *t*-test), suggesting that distributions of PFCAs, PFSAs and PFCs with 8 carbon chains are approximately similar between the two rooms in the same home. This indicates dust was distributed uniformly in rooms within the same house which could result from indoor circulation, convection or movement of people between rooms.

Home characteristics listed in Appendix F were assessed for association with PC1, PC2, and PC3 (for both indoor air and dust). No association were observed (one-way ANOVA, unpaired *t*-test) between PCs and home characteristics with the exception of a positive significant Pearson correlation between PC2 and volume of homes. This correlation indicates that an increasing in the volume of the homes leads to an increase in PFCA loading on PC2. A similar correlation was observed between the volume of homes and individual concentrations PFHpA, PFOA, PFNA and PFDA. No significant differences were seen between the other home characteristics and either of the other two PCs (unpaired *t*-test). These results suggest that either too small an amount of PFCs was released from items in the homes, or that the contribution of the released PFCs in comparison to other PFC sources was not large enough to influence the loadings of PCs in indoor air or dust.

The limitation of this study is that we are not certain that a specific home characteristic is the only cause of the relevant variation in PFC levels because simultaneously, the other home characteristics and items at homes can have similar effects on the PFC levels. Furthermore, each home characteristic associated mostly with only one PFC. Hence, the associations of home characteristics with either individual concentrations of PFCs or PCs could be accidental.

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3 CONCLUSIONS AND FUTURE DIRECTIONS

3.1 Conclusions

The work reported in this thesis has provided insight into the presence of PFCs in indoor air and in house dust from Winnipeg, Manitoba. As part of this research, the presence and concentrations of these compounds were compared with various aspects of the home characteristics to investigate possible correlations.

With regards to the dust samples, measurement of these compounds was achieved by adapting existing methods involving on-line SPE coupled with LC-MS/MS (McConkey, 2012; Gosetti et al., 2010; Haug et al., 2009), while air samples were analysed by adapting existing methods involving GC-MS (Barber et al., 2007; Shoeib et al., 2011). Correlations with home characteristics were determined using survey results from participants. The methods used to measure PFCs had low detection limits, minimal sample preparation, and required only a small amount of dust. Therefore, the methods were suitable for large-scale quantification of indoor air and dust. In total, indoor air samples were tested from 77 homes and dust samples from 86 homes (132 dust samples total). Numerous PFCs were present in all indoor air samples and dust samples analysed, demonstrating that indoor environment is likely to be an exposure pathway of PFCs for residents in Winnipeg homes.

8:2 FTOH was the most frequently detected PFC in indoor air and was detected in the highest concentration among neutral PFCs. Significant correlations were found between concentrations of 8:2 FTOH and 10:2 FTOH; MeFOSE and MeFOSA; and EtFOSE and EtFOSA in indoor air. PFOA and PFOS were the most prominent PFCs in

house dust and were detected in all samples. The prevalence of 8:2 FTOH in indoor air samples and PFOS and PFOA in dust samples have been reported in other studies around the world (Knobeloch et al., 2012; Shoeib et al., 2011; Kubwabo et al., 2005; Strynar et al., 2008, Björklund et al., 2009., Goosey et al., 2012; Moriwaki., 2003). A significant association in house dust between concentrations of PFOA and PFOS is consistent with findings from other studies (Knobeloch et al., 2012; D'Hollander et al., 2010; Haug et al., 2011; Shoeib et al., 2011, Kato et al., 2009). Associations between PFCs in both air and dust may suggest a common source for these residues. In house dust, significant associations were seen among nearly all the PFCAs, whereas correlations within the PFSAs, as well as between PFCAs and PFSAs, were less frequent.

Concentrations of PFCs in indoor air and dust were similar to those reported in other recent studies for commonly detected PFCs (Strynar et al., 2008; Shoeib et al., 2011; Kato et al., 2009). The concentrations and distributions of PFCs in indoor air and house dust were compared to many home characteristics. This provided opportunities for new insights regarding how these characteristics affect PFCs in indoor air and dust. A significant correlation between concentrations of MeFOSE in paired air and dust samples indicates that the compound originates from a common source and/or there exists a close coupling between air and dust. Significant associations among PFCAs in dust and FTOHs in air in this study support the hypothesis of transformation of FTOHs to PFCAs in the atmosphere (Ellis et al., 2004).

Relatively high concentrations of the PFCs commonly found in recently constructed homes (PFHpA, PFOA, PFOS, and PFHxS) were measured in this study, which could result from higher applications of PFCs in recent years for floor coverings and up-

holstered furniture. This is consistent with a previous study, which reported a significant negative association between concentrations of these PFCs and ages of the houses in which they were found (Knobeloch et al., 2012).

Other positive correlations of PFCs with home characteristics were observed. These included higher occurrence of PFCs in renovated homes; fully carpeted homes; homes painted in last 12 months; homes with any major pieces of plastic or vinyl-covered furniture; homes located near either a highway/artery road or major/prolonged construction activity. Such relationships provide insight into how various home characteristics could impact indoor levels of PFCs and demonstrate that the distribution of PFCs in indoor air and dust is affected by various factors.

It was determined that there was no significant difference between concentrations of PFCs in the child's room and the most used room within the same house. This observation suggests that dust was distributed uniformly in rooms within the same house, probably as a result of indoor air circulation, convection, or movement of people between rooms. Additionally, house dust samples from the child's room and the most used room showed significant correlations with all PFCs except PFPA. However, in a previous study comparing indoor air concentrations of 8:2 FTOH between the most used room and child room showed significantly higher concentrations in the child rooms in comparison to the most used rooms (Liu et al., 2013).

Indoor ambient temperature did not appear to affect concentrations of PFCs in indoor air. While exploring seasonal dependence, it was found that 8:2 FTOH and MeFOSE in indoor air were present at significantly higher levels in winter compared to summer. This could be due to reduced window ventilation during the extremely cold

weather in Winnipeg during the winter.

No significant differences were found between control and target homes for both indoor air and dust samples. This indicated that the proportions and concentrations of PFCs measured in this set of indoor air and house dust did not affect the occurrence of wheezing. The lack of correlation may be due to one or more of the following reasons: because PFCs do not cause harmful effects on human lungs as they have in mouse lungs (Grasty et al., 2005), that the concentrations of PFCs were too low to elicit effects, or that while the outcome of wheezing was not observed, other unmeasured effects on the lungs may have occurred. In general, principal component analysis results showed no correlation between principal components (PCs) and home characteristics for both indoor air and dust.

In summary, PFCs in indoor air and dust were associated with each other and home characteristics but not indoor ambient temperature, wheezing, type of room (child room or the most used room). Non of neutral PFCs in indoor air showed association with seasonal temperature variation except 8:2 FTOH and MeFOSE that had significantly higher concentrations in winter than summer.

3.2 Future Directions

In this work to explore seasonal dependence of PFCs, indoor air samples collected in different seasons from different homes were compared to each other. An interesting modification for a future study would be to collect the samples from the same home in different seasons to minimize impacts of other factors (home characteristics) in different

homes on the results.

In this study, home characteristics were explored in an attempt to identify potential indoor sources of PFCs and as a result, significant correlations were found between many of the home characteristics and concentrations of PFCs. An interesting addition to this work would be to explore a wider range of characteristics by collecting samples from other indoor environments such as offices, cars, or places that are thought to have potential for greater exposure to PFCs (e.g., retail stores, hair salons, etc).

Another important insight for designing future work based upon this study is to monitor concentrations of PFCs over a longer period of time (5 years or longer) in the same place, for example, a certain number of homes in the same neighbourhood, to monitor changes in concentrations of PFCs over time and also the use of shorter chain such as PFPA and PFBS as a replacement for longer chain-length.

The levels of PFCs in plasma samples from CHILD participants have been evaluated by another graduate student in our research group (McConkey, 2012). Further research on the exposure routes of PFCs to people in Winnipeg could be conducted. The results of plasma samples could be compared to the results from air and dust samples to investigate how the airborne route of exposure influences levels of PFCs in blood.

Furthermore, human exposure to PFCs through the diet would be another important aspect for future work. Information regarding levels of PFCs in food from Canada is available (Tittlemier et al., 2006; Tittlemier et al., 2007). As in the current study, participants would also need to fill out surveys regarding their lifestyles, specifically their diets.

3.3 References

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Appendix A: Concentrations of FTOHs and FOSA/FOSEs in indoor (pg/m^3). "N.D." means not detected.

Air Sample ID	Analyte						
	6:2 FTOH	**8:2 FTOH**	**10:2 FTOH**	**MeFOSA**	**EtFOSA**	**MeFOSE**	**EtFOSE**
40029	N.D.	1691.4	668.1	36.9	45.5	203.1	N.D.
40034	50.3	2575.1	4061.2	110.6	366.1	1203.5	36.8
40051	N.D.	1481.8	255.5	30.0	N.D.	100.3	N.D.
40057	N.D.	1768.3	243.0	26.9	N.D.	134.8	N.D.
40090	N.D.	4124.9	1074.2	N.D.	N.D.	N.D.	N.D.
40131	60.91	1191.3	336.1	27.6	1.1	47.1	N.D.
40189	N.D.	1256.5	404.0	47.5	46.9	367.9	16.5
40234	N.D.	2442.6	534.5	68.5	3.0	1177.7	N.D.
40269	N.D.	3467.7	1298.0	594.5	105.3	8780.0	612.6

Air Sample ID	Analyte						
	6:2 FTOH	8:2 FTOH	10:2 FTOH	MeFOSA	EtFOSA	MeFOSE	EtFOSE
40602	N.D.	2461.7	817.6	78.9	74.8	598.9	59.0
40714	N.D.	4684.3	1128.3	147.6	64.2	1804.1	107.2
40069	N.D.	1809.0	1174.3	46.5	73.7	1992.2	762.3
40078	2560.3	2851.7	1099.1	296.1	74.6	5565.8	632.5
40187	72.2	356.8	168.6	N.D.	14.5	49.8	134.2
40320	N.D.	1413.0	517.3	N.D.	119.5	698.5	N.D.
40355	805.5	3604.5	1171.5	N.D.	76.0	617.5	N.D.
40449	N.D.	1349.5	932.7	N.D.	55.7	973.4	452.7
40453	1128.9	865.8	415.0	N.D.	70.7	118.7	N.D.
40722	N.D.	9082.9	6246.2	78.1	245.0	2103.6	805.8

Air Sample ID	Analyte						
	6:2 FTOH	**8:2 FTOH**	**10:2 FTOH**	**MeFOSA**	**EtFOSA**	**MeFOSE**	**EtFOSE**
40734	1584.4	1754.1	636.3	191.9	50.9	6154.5	676.6
40793	1747.7	3632.7	781.5	24.2	60.6	660.8	577.4
40439	N.D.	1812.3	422.5	63.6	77.2	375.8	94.8
40292	219.9	1012.5	176.9	34.0	36.5	171.2	33.7
40159	96.7	1778.1	505.1	95.9	46.5	840.9	68.2
40693	343.7	1493.2	410.0	46.2	65.1	308.9	N.D.
40250	200.1	1846.9	481.0	53.7	70.1	362.6	735.0
40372	N.D.	1208.6	255.4	51.2	49.5	361.3	399.0
40394	212.3	706.7	127.0	36.9	29.6	463.0	N.D.
40671	115.5	1031.0	183.0	39.4	47.2	247.9	62.4

Air Sample ID	Analyte						
	6:2 FTOH	**8:2 FTOH**	**10:2 FTOH**	**MeFOSA**	**EtFOSA**	**MeFOSE**	**EtFOSE**
40704	182.1	7518.3	2991.4	39.7	39.6	151.4	36.1
40137	N.D.	603.1	215.0	35.9	38.9	112.3	N.D.
40262	N.D.	1100.0	520.1	26.8	28.3	620.4	60.7
40147	N.D.	1336.3	419.9	253.6	36.0	3429.7	453.6
40182	N.D.	3160.4	576.2	35.2	24.0	1944.4	768.4
40188	N.D.	494.3	1081.1	16.4	34.7	583.0	28.2
40287	47.3	3912.2	1253.9	209.6	207.2	2932.8	144.5
40289	N.D.	2082.5	915.5	65.7	33.6	832.7	37.8
40295	N.D.	1213.9	1318.4	250.1	N.D.	1603.3	N.D.
40328	204.7	599.1	177.4	N.D.	23.8	416.6	102.0

Air Sample ID	Analyte						
	6:2 FTOH	**8:2 FTOH**	**10:2 FTOH**	**MeFOSA**	**EtFOSA**	**MeFOSE**	**EtFOSE**
40481	N.D.	1912.5	2090.8	59.0	40.1	574.3	357.6
40487	176.8	691.3	179.9	27.7	53.7	260.7	66.1
40685	N.D.	3358.3	980.3	24.4	15.2	1037.8	139.7
40085	1277.6	1245.0	654.3	19.2	59.1	290.4	N.D.
40096	N.D.	1275.3	397.8	55.9	92.6	346.8	123.8
40126	N.D.	957.9	7637.4	42.4	57.6	315.9	N.D.
40260	N.D.	173.7	64.1	8.4	48.5	130.3	N.D.
40263	338.7	449.3	N.D.	N.D.	33.1	N.D.	N.D.
40300	N.D.	2804.1	750.4	14.6	54.3	81.2	N.D.
40338	1216.1	3923.8	1229.6	54.4	89.5	511.0	107.2

Air Sample ID	Analyte						
	6:2 FTOH	**8:2 FTOH**	**10:2 FTOH**	**MeFOSA**	**EtFOSA**	**MeFOSE**	**EtFOSE**
40373	N.D.	673.9	163.8	15.2	73.0	76.1	N.D.
40374	1474.9	877.3	290.6	21.5	51.4	370.3	N.D.
40400	N.D.	1000.4	396.6	98.0	171.2	1348.7	489.7
40483	N.D.	1226.5	709.9	57.3	136.2	568.2	241.6
40535	N.D.	2731.5	788.4	40.2	143.1	2813.5	349.1
40611	N.D.	5034.8	1129.4	12.6	50.7	1516.2	90.4
40652	N.D.	1120.4	562.4	284.5	60.1	781.6	N.D.
40800	N.D.	1832.3	N.D.	2.4	139.0	818.9	N.D.
40922	381.1	583.8	224.9	53.1	133.8	860.3	2750.5
40156	718.5	1040.9	275.2	15.1	47.2	133.7	N.D.

Air Sample ID	Analyte						
	6:2 FTOH	**8:2 FTOH**	**10:2 FTOH**	**MeFOSA**	**EtFOSA**	**MeFOSE**	**EtFOSE**
40356	194.0	7992.9	N.D.	424.6	102.4	5286.7	467.6
40512	237.2	743.3	680.1	161.7	60.6	2867.8	328.2
40663	355.2	889.1	218.2	35.3	1079.1	873.0	5253.1
40912	N.D.	697.9	204.5	83.6	56.1	2555.3	N.D.
40886	N.D.	374.2	88.1	N.D.	42.4	N.D.	N.D.
40788	N.D.	934.2	214.9	9.8	111.4	29.8	343.5
40923	N.D.	528.8	285.0	7.7	57.0	96.0	145.8
40908	N.D.	1880.8	560.7	54.0	97.9	1468.1	N.D.
40984	N.D.	342.2	591.8	70.7	71.4	395.6	134.4
40662	N.D.	2117.9	927.2	16.3	32.4	431.3	N.D.

Air Sample ID	Analyte						
	6:2 FTOH	**8:2 FTOH**	**10:2 FTOH**	**MeFOSA**	**EtFOSA**	**MeFOSE**	**EtFOSE**
40436	N.D.	765.8	440.0	49.1	62.6	130.1	94.5
40086	N.D.	5755.4	1382.7	99.2	97.8	404.0	N.D.
40806	N.D.	3279.9	648.9	23.1	32.4	292.9	N.D.
40735	N.D.	2314.1	1601.0	58.8	100.8	235.6	69.7
40897	N.D.	1957.9	5116.3	121.5	273.8	593.0	392.3
40442	321.4	2062.8	483.9	111.5	65.4	971.3	8.5
40197	N.D.	2193.1	942.5	9.3	72.5	260.5	86.1
40980	267.1	1191.5	221.5	18.3	69.9	144.6	210.2

Appendix B: Concentrations of PFCAs and PFSAs in house dust (ng/g). "N.D.", "MR" and "CR" mean "not detected", "the most used room" and "child room" respectively.

Dust Sample ID	Analyte										
	PFPA	**PFHxA**	**PFHpA**	**PFOA**	**PFNA**	**PFDA**	**PFUA**	**PFBS**	**PFHxS**	**PFOS**	**PFDS**
40029 MR	N.D.	71.9	19.4	71.0	31.6	45.9	N.D.	N.D.	332.5	77.2	23.5
40034 MR	N.D.	31.8	95.3	134.2	118.7	159.9	N.D.	N.D.	16.2	54.5	13.1
40051 MR	N.D.	59.1	N.D.	51.0	41.0	28.3	0.8	N.D.	N.D.	86.2	12.0
40131 MR	N.D.	76.2	11.0	96.8	30.5	43.8	N.D.	17.9	N.D.	13.0	15.4
40189 MR	N.D.	50.2	12.3	28.6	13.9	14.0	N.D.	5.0	24.6	34.9	4.7
40234 MR	N.D.	81.5	25.6	112.0	31.2	13.6	N.D.	14.8	1283.3	544.8	72.5
40269 MR	290.9	1766.5	1565.5	2048.8	121.4	41.3	4.1	196.8	2751.5	9982.8	5.2
40602 MR	33.8	283.0	786.9	376.4	743.4	246.6	0.7	14.3	20.8	488.6	96.7
40714 MR	N.D.	39.8	17.2	50.1	15.3	18.7	N.D.	16.6	12.5	34.9	N.D.

Dust Sample ID	Analyte										
	PFPA	**PFHxA**	**PFHpA**	**PFOA**	**PFNA**	**PFDA**	**PFUA**	**PFBS**	**PFHxS**	**PFOS**	**PFDS**
40078 MR	N.D.	69.2	1407.4	934.0	1214.8	649.8	0.5	N.D.	99.1	79.9	N.D.
40449 MR	N.D.	109.6	183.4	423.7	302.6	395.9	N.D.	47.2	40.5	55.4	9.2
40722 MR	N.D.	98.3	16.1	63.2	16.0	17.2	N.D.	N.D.	23.1	78.8	7.1
40734 MR	38.4	88.3	219.3	486.4	80.2	62.1	1.3	N.D.	57.7	791.5	12.2
40793 MR	62.1	31.4	16.6	88.0	41.0	55.9	N.D.	N.D.	24.7	78.2	9.9
40372 CR	105.5	203.3	788.4	1141.9	128.3	175.5	0.8	42.3	42.3	7023.4	79.1
40704 MR	69.6	238.7	532.6	386.6	695.7	385.0	0.6	N.D.	6.6	77.7	30.2
40147 MR	14.1	37.7	35.9	84.7	42.1	20.6	0.5	8.1	5.3	85.7	N.D.
40287 MR	159.6	258.6	164.7	774.3	34.5	50.1	3.6	31.6	4.7	11397.7	N.D.
40295 CR	N.D.	46.0	694.0	502.0	550.3	378.1	0.3	5.3	11.3	82.2	19.8

Dust Sample ID	Analyte										
	PFPA	**PFHxA**	**PFHpA**	**PFOA**	**PFNA**	**PFDA**	**PFUA**	**PFBS**	**PFHxS**	**PFOS**	**PFDS**
40481 MR	N.D.	62.3	12.7	49.6	13.1	20.6	N.D.	N.D.	812.6	74.2	133.5
40487 MR	N.D.	73.4	53.6	250.8	22.1	33.2	N.D.	N.D.	51.2	256.1	28.6
40685 MR	N.D.	41.6	11.8	66.5	11.9	22.0	0.5	N.D.	N.D.	81.2	N.D.
40085 CR	N.D.	121.0	50.4	141.1	34.3	57.2	N.D.	N.D.	4.9	44.2	17.7
40096 MR	N.D.	54.7	9.6	34.7	23.5	12.1	N.D.	N.D.	10.0	46.6	8.9
40126 MR	413.4	207.4	1727.7	1097.7	1405.4	965.1	N.D.	9.9	11.0	28.2	5.3
40260 MR	N.D.	67.6	19.0	125.0	13.2	20.1	N.D.	N.D.	18.1	77.3	1430.4
40263 MR	N.D.	204.6	23.5	296.1	10.5	64.9	N.D.	5.4	131.2	88.9	154.2
40300 CR	211.2	356.7	312.4	123.4	23.3	36.3	N.D.	6.7	4.7	146.7	N.D.
40338 MR	N.D.	129.4	119.0	201.8	260.0	101.4	0.1	N.D.	164.9	165.7	24.5

Dust Sample ID	Analyte										
	PFPA	**PFHxA**	**PFHpA**	**PFOA**	**PFNA**	**PFDA**	**PFUA**	**PFBS**	**PFHxS**	**PFOS**	**PFDS**
40373 MR	N.D.	19.9	8.1	12.1	14.4	10.1	2.0	4.6	1.6	15.5	2.8
40400 MR	N.D.	35.4	18.7	75.8	19.9	11.1	2.3	N.D.	22.3	79.3	23.2
40483 MR	N.D.	42.1	41.3	187.4	31.0	55.8	N.D.	24.9	24.9	152.9	187.1
40652 MR	N.D.	85.2	33.8	123.1	36.0	19.3	N.D.	30.9	30.9	415.3	24.1
40922 MR	112.6	285.6	525.9	1208.4	581.6	697.0	N.D.	13.5	24.4	489.9	36.1
40512 MR	N.D.	124.6	38.0	144.6	14.3	11.9	9.3	N.D.	75.8	118.2	2.4
40663 CR	132.8	433.5	166.1	531.1	60.9	64.3	3.5	618.1	N.D.	584.0	2.7
40886 MR	N.D.	111.5	9.3	17.7	10.9	11.0	0.5	324.2	324.2	83.2	3.5
40908 MR	N.D.	78.9	N.D.	82.1	11.4	12.5	N.D.	N.D.	6.7	29.9	45.4
40662 MR	16.8	45.0	9.3	87.0	9.1	10.1	N.D.	N.D.	21.3	15.0	3.4

Dust Sample ID	Analyte										
	PFPA	**PFHxA**	**PFHpA**	**PFOA**	**PFNA**	**PFDA**	**PFUA**	**PFBS**	**PFHxS**	**PFOS**	**PFDS**
40436 MR	N.D.	102.5	19.8	29.2	10.2	13.0	0.8	N.D.	4.5	27.6	8.5
40086 CR	41.7	122.3	388.5	594.2	676.8	739.5	N.D.	19.2	19.2	2123.5	25.9
40806 MR	64.8	72.5	108.4	208.2	21.7	61.5	N.D.	N.D.	1.2	27.5	6.9
40735 MR	N.D.	30.4	18.1	36.3	24.0	17.4	8.8	18.2	18.2	49.5	11.9
40897 MR	N.D.	77.5	17.9	56.6	30.4	31.4	N.D.	N.D.	3.4	80.2	10.7
40442 MR	N.D.	159.5	99.8	408.1	61.5	57.0	0.3	N.D.	14.7	244.8	1245.2
40197 CR	N.D.	114.3	86.0	38.0	64.1	16.4	0.9	15.6	15.6	23.1	12.9
40980 MR	11.9	73.1	12.1	55.1	10.1	14.3	0.4	N.D.	5.5	28.2	11.6
40057 CR	N.D.	40.1	94.8	79.0	N.D.	N.D.	0.6	N.D.	46.6	72.3	N.D.
40638 MR	N.D.	251.9	48.7	474.1	50.5	176.8	0.8	N.D.	31.6	7.6	1521.3

Dust Sample ID	Analyte										
	PFPA	**PFHxA**	**PFHpA**	**PFOA**	**PFNA**	**PFDA**	**PFUA**	**PFBS**	**PFHxS**	**PFOS**	**PFDS**
40338 CR	51.6	53.3	70.4	95.6	247.0	39.7	2.4	N.D.	47.4	41.9	12.8
40269 CR	N.D.	178.3	54.5	341.9	17.7	27.0	N.D.	N.D.	4802.9	2695.9	22.7
40126 CR	440.5	66.4	2193.8	3248.1	1852.5	1930.3	11.7	8.8	9.1	22.0	5.7
40029 CR	N.D.	203.3	75.3	362.7	42.1	44.4	N.D.	21.5	1508.1	77.1	85.0
40115 MR	N.D.	106.7	39.0	260.7	44.3	165.4	N.D.	N.D.	25.1	58.7	33.0
40115 CR	N.D.	35.4	23.9	124.2	28.2	81.6	N.D.	N.D.	27.6	57.5	22.8
40153 MR	N.D.	106.3	N.D.	23.8	11.9	10.0	0.4	6.3	96.9	100.3	3.0
40836 MR	N.D.	126.5	105.8	518.1	296.2	386.8	4.6	14.0	24.6	58.4	11.3
40793 CR	N.D.	68.2	14.8	75.9	35.6	43.3	0.6	N.D.	21.0	32.9	9.9
40189 CR	N.D.	29.9	13.3	39.0	10.5	14.6	0.3	9.0	19.8	48.5	3.7

Dust Sample ID	Analyte										
	PFPA	**PFHxA**	**PFHpA**	**PFOA**	**PFNA**	**PFDA**	**PFUA**	**PFBS**	**PFHxS**	**PFOS**	**PFDS**
40221MR	N.D.	57.3	13.0	64.6	18.6	13.6	0.7	8.5	1205.4	665.3	N.D.
40097 MR	N.D.	48.7	23.8	63.1	36.1	20.9	N.D.	55.7	N.D.	79.3	13.3
40784 MR	N.D.	31.0	16.2	48.3	6.9	11.0	4.5	N.D.	N.D.	81.7	3.5
40784 CR	N.D.	41.4	12.4	48.6	N.D.	N.D.	N.D.	N.D.	N.D.	75.5	13.7
40734 CR	N.D.	115.4	128.9	412.0	44.5	42.0	4.8	4.8	N.D.	1055.3	8.2
40131 CR	N.D.	36.6	20.6	104.5	97.9	58.1	N.D.	N.D.	N.D.	78.2	9.1
40487 CR	N.D.	42.4	17.3	71.9	15.5	18.3	1.0	N.D.	202.4	192.9	21.8
40221 CR	N.D.	28.3	9.1	48.2	14.3	14.6	0.5	5.5	537.0	280.4	28.7
40117 CR	N.D.	70.7	27.8	69.3	23.7	N.D.	N.D.	N.D.	17.0	6.6	N.D.
40260 CR	N.D.	52.8	15.3	77.4	13.5	30.4	N.D.	N.D.	N.D.	79.8	N.D.

Dust Sample ID	Analyte										
	PFPA	**PFHxA**	**PFHpA**	**PFOA**	**PFNA**	**PFDA**	**PFUA**	**PFBS**	**PFHxS**	**PFOS**	**PFDS**
40336 MR	66.1	344.1	349.9	1146.2	42.3	45.0	N.D.	32.4	4.1	3679.6	N.D.
40858 MR	N.D.	80.7	18.2	58.5	21.5	55.0	N.D.	N.D.	5.3	39.9	N.D.
40336 CR	N.D.	136.0	35.7	223.9	14.2	55.5	0.4	N.D.	4.4	1292.9	N.D.
40246 MR	3891.4	2214.3	2895.7	3224.0	2163.6	1212.9	N.D.	N.D.	4.8	114.3	N.D.
40287 CR	N.D.	318.8	59.6	238.1	20.4	N.D.	6.6	N.D.	18.0	2975.4	232.6
40551 CR	524.8	912.5	2221.0	3170.4	199.8	49.5	4.0	308.4	76.2	7336.1	10.5
40725 CR	N.D.	67.0	12.4	34.5	13.2	14.1	0.1	N.D.	196.8	79.2	45.4
40725 MR	N.D.	87.7	17.5	83.8	26.0	28.5	0.4	27.6	N.D.	169.8	25.9
40551 MR	86.6	186.1	522.6	2396.9	80.5	N.D.	0.6	84.8	5.0	6770.4	31.1
40805 CR	N.D.	83.3	16.8	28.9	62.7	8.7	0.4	N.D.	45.9	76.2	11.5

Dust Sample ID	Analyte										
	PFPA	**PFHxA**	**PFHpA**	**PFOA**	**PFNA**	**PFDA**	**PFUA**	**PFBS**	**PFHxS**	**PFOS**	**PFDS**
40765 CR	62.2	100.8	33.2	72.2	16.1	19.8	N.D.	N.D.	8476.3	42.3	N.D.
40182 MR	N.D.	80.3	N.D.	80.9	24.1	21.5	4.9	N.D.	43.6	78.5	61.8
40765 MR	N.D.	202.0	38.0	60.6	16.9	17.0	5.3	4.8	35.0	29.9	N.D.
40905 CR	N.D.	48.9	N.D.	23.1	10.0	8.2	0.6	574.5	1226.4	32.9	10.6
40905 MR	N.D.	75.7	9.3	28.7	10.5	N.D.	0.6	551.3	18.8	63.2	90.2
40722 CR	N.D.	99.9	27.2	124.5	16.3	26.0	N.D.	5.2	262.2	114.6	81.5
40218 CR	N.D.	45.5	18.8	29.1	11.5	N.D.	4.5	N.D.	72.8	31.8	11.3
40253 CR	N.D.	59.5	32.0	55.5	16.7	13.3	4.1	258.0	N.D.	45.4	N.D.
40512 CR	N.D.	95.9	42.0	150.0	9.6	11.9	N.D.	18.1	5.5	73.2	N.D.
40356 MR	184.9	397.9	1482.8	1826.3	1612.4	1334.3	2.1	4.5	4.9	479.8	N.D.

Dust Sample ID	Analyte										
	PFPA	**PFHxA**	**PFHpA**	**PFOA**	**PFNA**	**PFDA**	**PFUA**	**PFBS**	**PFHxS**	**PFOS**	**PFDS**
40922 CR	N.D.	40.2	59.8	192.6	28.5	45.5	2.9	8.1	80.3	288.4	1766.5
40391 MR	97.8	232.1	780.4	3336.4	114.6	56.6	0.2	30.4	209.2	6381.0	36.6
40983 MR	N.D.	134.9	14.1	62.8	323.1	24.7	0.4	39.9	177.7	75.5	19.2
40056 CR	N.D.	44.5	45.6	268.1	311.0	99.4	3.1	7.4	N.D.	60.6	26.2
40983 CR	30.7	66.8	26.7	43.5	143.8	21.7	3.5	22.0	N.D.	78.2	22.5
40357 CR	N.D.	73.8	16.2	41.7	16.9	18.6	0.5	6.1	180.4	54.4	26.8
40830 CR	105.7	108.2	766.9	3154.0	108.0	53.5	N.D.	37.4	135.5	9510.3	45.3
40878 MR	N.D.	94.2	26.8	62.6	26.0	11.9	2.1	4.4	N.D.	79.2	274.1
40878 CR	N.D.	102.7	47.3	173.3	46.1	72.4	0.8	10.7	11.8	20.2	1286.6
40858 CR	N.D.	65.4	16.1	36.3	N.D.	9.2	N.D.	10.6	221.1	78.4	701.9

Dust Sample ID	Analyte										
	PFPA	**PFHxA**	**PFHpA**	**PFOA**	**PFNA**	**PFDA**	**PFUA**	**PFBS**	**PFHxS**	**PFOS**	**PFDS**
40805 MR	N.D.	53.3	15.8	18.1	47.4	9.9	4.2	9.1	20.3	8.4	195.6
40830 MR	49.7	118.9	92.3	269.2	24.8	30.3	N.D.	6.5	9.6	115.3	N.D.
40263 CR	N.D.	90.7	32.2	903.9	17.7	167.9	N.D.	30.3	51.4	76.2	433.9
40980 CR	N.D.	92.1	17.9	41.2	9.4	8.9	1.3	N.D.	5.1	13.2	3.0
40981 MR	N.D.	81.3	20.3	57.5	31.0	62.7	3.8	187.7	187.7	37.6	6.1
40923 MR	54.7	227.5	217.9	789.4	445.7	854.2	3.5	N.D.	4.6	86.6	4.0
40862 CR	N.D.	94.2	23.1	49.8	30.1	30.5	2.6	6.1	6.1	28.5	4.1
40436 CR	N.D.	54.9	12.0	40.7	10.1	16.7	3.9	5.6	5.6	81.2	7.3
40491 MR	N.D.	41.1	14.0	14.1	8.9	8.9	0.9	N.D.	4.5	9.9	2.5

Dust Sample ID	Analyte										
	PFPA	**PFHxA**	**PFHpA**	**PFOA**	**PFNA**	**PFDA**	**PFUA**	**PFBS**	**PFHxS**	**PFOS**	**PFDS**
40373 CR	N.D.	53.3	59.8	45.2	22.9	12.7	4.0	N.D.	4.8	77.2	5.7
40806 CR	42.0	151.1	75.6	76.6	20.7	25.4	3.5	N.D.	4.7	33.5	16.7
40629 CR	N.D.	97.6	26.2	95.5	38.6	66.7	3.3	48.8	48.8	78.2	28.3
40746 MR	N.D.	32.4	8.9	17.0	164.5	9.9	4.9	540.1	540.1	71.2	3.1
40442 CR	N.D.	37.8	17.7	64.9	18.7	15.4	N.D.	15.0	15.0	138.8	952.9
40981 CR	N.D.	39.5	15.8	89.4	33.2	122.2	3.8	180.1	180.1	34.3	6.5
40652 CR	N.D.	79.1	34.2	94.5	61.2	13.5	0.8	6.6	6.6	350.0	23.0
40372 MR	N.D.	30.0	40.2	226.3	24.0	33.8	3.6	N.D.	11.0	628.8	30.1
40284 MR	N.D.	64.3	15.7	51.5	26.7	30.6	0.6	N.D.	4.1	42.8	11.9
40096 CR	N.D.	56.8	24.4	51.3	46.7	24.3	N.D.	N.D.	8.5	97.2	16.6

Dust Sample ID	Analyte										
	PFPA	**PFHxA**	**PFHpA**	**PFOA**	**PFNA**	**PFDA**	**PFUA**	**PFBS**	**PFHxS**	**PFOS**	**PFDS**
40491 CR	9.8	41.4	14.2	17.2	17.6	10.6	3.5	N.D.	1.5	6.4	2.9
40086 MR	85.2	196.1	1144.3	2692.4	2013.7	2181.6	3.5	N.D.	11.5	754.7	N.D.
40587 CR	309.2	663.3	984.3	71.0	15.4	15.5	N.D.	6.0	6.0	86.8	33.8
40746 CR	N.D.	29.4	16.1	15.9	115.1	N.D.	N.D.	838.8	838.8	79.2	3.4
40293 CR	N.D.	44.2	44.7	110.9	9.8	N.D.	0.5	N.D.	7.9	110.5	23.8
40886 CR	13.0	232.3	14.0	14.5	7.3	10.3	4.1	360.6	360.6	6.7	2.8
40662 CR	23.3	77.8	20.0	38.2	71.6	21.6	6.7	10.5	10.5	80.2	3.2
40483 CR	N.D.	28.4	20.2	77.1	16.0	30.6	N.D.	N.D.	3.9	80.9	100.5
40735 CR	31.0	52.9	33.8	51.3	41.5	38.4	2.0	N.D.	4.7	7.8	7.8
40790 MR	23.1	55.4	9.9	19.7	186.1	15.1	N.D.	N.D.	7.4	77.2	N.D.

Dust Sample ID	Analyte										
	PFPA	**PFHxA**	**PFHpA**	**PFOA**	**PFNA**	**PFDA**	**PFUA**	**PFBS**	**PFHxS**	**PFOS**	**PFDS**
40293 MR	N.D.	62.5	26.6	44.7	12.5	14.4	N.D.	10.0	10.0	83.6	19.3
40788 MR	N.D.	29.4	10.1	38.4	13.6	15.1	N.D.	12.2	12.2	63.6	4.1
40284 CR	N.D.	46.9	38.2	104.0	23.5	17.4	N.D.	N.D.	5.0	111.9	15.3

Appendix C: Concentrations of FOSA/FOSEs in house dust (ng/g). “N.D.” means not detected. “N.D.”, “MR” and “CR” mean “not detected”, “the most used room” and “child room” respectively.

Dust Sample ID	Analyte				
	FOSA	**MeFOSA**	**EtFOSA**	**MeFOSE**	**EtFOSE**
40029 MR	0.5	N.D.	3.9	25.8	N.D.
40034 MR	1.4	2.7	0.8	37.7	16.5
40051 MR	0.7	N.D.	N.D.	18.5	10.8
40131 MR	0.2	N.D.	0.0	10.5	2.1
40189 MR	N.D.	N.D.	N.D.	25.6	N.D.
40234 MR	1.1	N.D.	0.3	N.D.	2.1
40269 MR	57.5	10.0	0.6	N.D.	50.4
40602 MR	0.7	6.1	4.7	32.4	N.D.
40714 MR	1.6	N.D.	N.D.	N.D.	2.1
40078 MR	27.1	13.9	6.0	378.5	25.9
40449 MR	1.2	2.0	0.4	32.6	21.1
40722 MR	1.7	N.D.	0.6	88.1	19.1
40734 MR	13.0	8.3	0.4	771.1	33.4
40793 MR	1.2	3.7	N.D.	26.6	17.0
40372 CR	1966.5	N.D.	21.1	167.8	4032.4

Dust Sample ID	Analyte				
	FOSA	**MeFOSA**	**EtFOSA**	**MeFOSE**	**EtFOSE**
40704 MR	1.4	N.D.	3.9	37.6	10.6
40147 MR	2.5	14.3	0.3	299.8	34.7
40287 MR	13.2	6.8	4.7	424.8	17.1
40295 CR	N.D.	N.D.	0.4	N.D.	7.9
40481 MR	N.D.	3.2	3.6	33.5	7.4
40487 MR	7.7	N.D.	N.D.	103.7	65.5
40685 MR	1.6	N.D.	0.6	N.D.	6.9
40085 CR	1.8	2.6	3.6	23.4	18.5
40096 MR	2.1	N.D.	0.5	N.D.	2.1
40126 MR	0.5	4.9	0.7	39.0	15.3
40260 MR	1.0	N.D.	N.D.	16.3	10.5
40263 MR	17.1	N.D.	5.7	18.1	31.9
40300 CR	0.9	N.D.	0.3	46.4	42.0
40338 MR	1.5	5.2	1.4	70.5	26.9
40373 MR	N.D.	N.D.	N.D.	N.D.	2.6
40400 MR	2.0	N.D.	N.D.	48.7	2.3
40483 MR	1.1	N.D.	0.9	42.8	12.0

Dust Sample ID	Analyte				
	FOSA	**MeFOSA**	**EtFOSA**	**MeFOSE**	**EtFOSE**
40652 MR	3.8	N.D.	0.6	N.D.	N.D.
40922 MR	12.8	10.2	11.6	60.4	168.7
40512 MR	1.7	18.5	0.3	46.5	49.5
40663 CR	8.6	N.D.	4.4	21.1	123.5
40886 MR	0.4	N.D.	5.3	N.D.	N.D.
40908 MR	1.1	N.D.	10.4	N.D.	N.D.
40662 MR	0.6	2.9	3.9	N.D.	13.0
40436 MR	0.3	7.5	0.1	N.D.	11.2
40086 CR	3.1	N.D.	0.2	32.6	10.1
40806 MR	0.6	4.3	0.1	103.1	9.0
40735 MR	1.2	N.D.	N.D.	98.4	16.4
40897 MR	1.5	N.D.	4.1	N.D.	9.5
40442 MR	5.4	N.D.	N.D.	N.D.	40.6
40197 CR	1.0	N.D.	0.0	59.8	28.4
40980 MR	5.1	N.D.	4.0	79.2	233.1
40057 CR	N.D.	N.D.	N.D.	N.D.	N.D.
40638 MR	1.3	21.4	N.D.	N.D.	N.D.

Dust Sample ID	Analyte				
	FOSA	**MeFOSA**	**EtFOSA**	**MeFOSE**	**EtFOSE**
40338 CR	1.0	N.D.	N.D.	35.5	9.5
40269 CR	9.8	17.0	0.8	N.D.	19.9
40126 CR	0.8	N.D.	0.8	66.7	19.7
40029 CR	3.5	N.D.	0.8	45.2	14.1
40115 MR	1.9	5.6	0.8	83.4	26.6
40115 CR	0.9	N.D.	0.8	N.D.	17.9
40153 MR	8.1	N.D.	3.7	38.8	110.3
40836 MR	1.6	4.2	0.7	37.4	6.2
40793 CR	1.9	2.6	0.3	N.D.	12.1
40189 CR	N.D.	N.D.	N.D.	N.D.	N.D.
40221MR	2.3	N.D.	N.D.	N.D.	N.D.
40097 MR	1.3	N.D.	4.3	29.1	16.7
40784 MR	3.2	3.5	N.D.	81.2	N.D.
40784 CR	3.6	N.D.	N.D.	75.6	24.1
40734 CR	16.5	10.8	0.1	1045.4	24.1
40131 CR	0.3	N.D.	3.9	N.D.	N.D.
40487 CR	2.0	2.7	3.6	26.4	19.1

Dust Sample ID	Analyte				
	FOSA	MeFOSA	EtFOSA	MeFOSE	EtFOSE
40221 CR	2.1	N.D.	4.6	N.D.	2.5
40117 CR	0.6	N.D.	N.D.	17.2	N.D.
40260 CR	1.2	N.D.	N.D.	13.8	14.4
40336 MR	25.3	12.4	N.D.	N.D.	13.0
40858 MR	N.D.	4.0	0.0	N.D.	N.D.
40336 CR	5.6	4.0	0.9	N.D.	53.3
40246 MR	N.D.	N.D.	0.5	43.4	15.2
40287 CR	5.4	10.5	4.7	56.7	20.2
40551 CR	33.5	17.8	4.4	N.D.	10.9
40725 CR	N.D.	N.D.	0.5	N.D.	6.9
40725 MR	N.D.	N.D.	5.1	N.D.	N.D.
40551 MR	12.2	11.6	4.6	838.9	6.4
40805 CR	0.7	N.D.	N.D.	32.7	13.3
40765 CR	0.8	3.5	1.6	14.2	6.2
40182 MR	1.6	N.D.	N.D.	169.1	50.6
40765 MR	0.5	N.D.	N.D.	N.D.	7.7
40905 CR	0.8	5.4	6.1	3.8	22.8

Dust Sample ID	Analyte				
	FOSA	**MeFOSA**	**EtFOSA**	**MeFOSE**	**EtFOSE**
40905 MR	0.7	N.D.	N.D.	N.D.	11.3
40722 CR	3.9	N.D.	N.D.	92.8	20.2
40218 CR	2.3	N.D.	3.4	97.6	21.5
40253 CR	3.4	6.9	0.1	64.0	19.0
40512 CR	1.8	6.9	7.9	33.8	26.0
40356 MR	3.7	N.D.	0.6	209.8	23.7
40922 CR	4.8	N.D.	5.3	N.D.	65.2
40391 MR	112.4	8.8	6.9	N.D.	21.3
40983 MR	0.8	3.9	N.D.	N.D.	12.6
40056 CR	0.9	N.D.	N.D.	17.5	8.3
40983 CR	0.5	N.D.	5.8	N.D.	N.D.
40357 CR	1.4	N.D.	0.7	30.7	11.2
40830 CR	96.6	5.2	6.7	N.D.	14.6
40878 MR	N.D.	N.D.	0.1	N.D.	N.D.
40878 CR	1.4	7.3	0.2	N.D.	11.7
40858 CR	2.1	N.D.	6.2	N.D.	33.4
40805 MR	0.9	4.2	4.5	33.9	11.7

Dust Sample ID	Analyte				
	FOSA	MeFOSA	EtFOSA	MeFOSE	EtFOSE
40830 MR	6.6	10.3	0.4	1387.9	N.D.
40263 CR	13.9	N.D.	9.0	36.8	57.5
40980 CR	3.0	3.7	0.7	72.7	106.4
40981 MR	N.D.	6.0	3.2	10.7	5.6
40923 MR	N.D.	3.7	0.6	N.D.	6.0
40862 CR	N.D.	2.7	4.2	20.1	8.3
40436 CR	N.D.	4.5	1.8	26.2	8.7
40491 MR	1.2	N.D.	1.0	N.D.	4.3
40894 MR	1.8	2.1	3.0	N.D.	N.D.
40373 CR	0.6	N.D.	0.3	N.D.	5.4
40806 CR	N.D.	N.D.	3.7	29.7	6.8
40629 CR	0.5	N.D.	3.1	N.D.	N.D.
40746 MR	N.D.	N.D.	4.2	N.D.	6.0
40442 CR	3.3	N.D.	3.8	61.2	12.6
40981 CR	1.0	4.2	3.2	N.D.	10.0
40652 CR	1.6	5.5	N.D.	N.D.	N.D.
40372 MR	14.0	N.D.	N.D.	N.D.	88.0

Dust Sample ID	Analyte				
	FOSA	MeFOSA	EtFOSA	MeFOSE	EtFOSE
40284 MR	1.4	8.3	7.2	N.D.	31.3
40096 CR	3.7	5.1	0.1	N.D.	18.0
40491 CR	0.2	N.D.	2.0	N.D.	2.7
40086 MR	8.8	6.7	4.3	N.D.	24.6
40587 CR	1.9	N.D.	N.D.	66.0	7.8
40746 CR	0.3	6.5	3.4	22.4	2.7
40293 CR	1.1	N.D.	N.D.	N.D.	N.D.
40886 CR	0.4	N.D.	N.D.	N.D.	5.3
40662 CR	0.4	3.0	1.3	21.7	8.0
40483 CR	N.D.	N.D.	N.D.	N.D.	7.7
40735 CR	N.D.	N.D.	N.D.	N.D.	17.1
40790 MR	N.D.	N.D.	4.3	N.D.	7.8
40293 MR	0.6	N.D.	N.D.	N.D.	N.D.
40788 MR	4.5	4.5	N.D.	12.7	57.5
40284 CR	1.3	7.4	4.8	N.D.	40.9
40284 MR	1.4	8.3	7.2	N.D.	31.3
40096 CR	3.7	5.1	0.1	N.D.	18.0

Appendix D: PCA factor scores for indoor air.

Air Sample ID	PCA Factor Score		
	PC1	**PC2**	**PC3**
40069	0.75	0.02	-0.31
40078	0.68	1.00	0.06
40085	-1.02	-0.90	1.29
40086	-0.99	-0.33	-1.45
40090	-3.04	0.12	-2.00
40096	0.63	0.26	0.10
40126	-1.30	-2.64	-0.73
40131	-0.93	0.53	-1.91
40137	0.34	-0.10	-0.40
40147	1.49	0.61	-0.78
40159	0.53	0.53	-0.18
40182	0.77	0.53	-1.12
40187	-1.33	0.49	0.99
40188	0.13	-0.75	-0.42
40189	0.56	-0.22	-0.91

Air Sample ID	PCA Factor Score		
	PC1	**PC2**	**PC3**
40197	0.11	-1.60	1.03
40234	-0.03	0.25	-2.37
40250	0.32	0.83	0.37
40260	0.67	-0.05	0.38
40262	0.43	0.05	-0.47
40263	-1.53	2.49	1.18
40269	1.44	0.32	-0.95
40287	0.73	0.26	-0.17
40289	0.42	-0.17	-1.10
40292	0.01	0.96	0.38
40295	0.59	-1.71	-1.94
40300	-1.25	0.07	-0.61
40320	-1.17	-1.97	1.30
40355	-2.41	0.24	0.39
40372	0.83	0.64	-0.05

Air Sample ID	PCA Factor Score		
	PC1	PC2	PC3
40373	-0.49	-0.02	-0.02
40374	-0.78	0.84	0.58
40394	0.23	0.95	0.28
40400	1.44	0.13	0.00
40436	0.86	-1.77	1.31
40439	0.57	0.27	-0.32
40442	0.08	0.63	-0.05
40449	-0.07	-0.29	-0.18
40453	-2.68	0.37	1.13
40483	0.77	-0.12	0.06
40487	0.34	0.80	0.74
40512	1.10	-0.64	0.92
40535	1.03	-1.11	0.79
40611	-0.20	0.11	-0.95
40662	-0.12	-2.25	0.66

Air Sample ID	PCA Factor Score		
	PC1	**PC2**	**PC3**
40663	0.39	1.41	1.98
40685	0.08	0.22	-1.27
40693	-0.35	0.56	0.27
40734	0.81	1.15	0.06
40788	0.15	-0.75	1.78
40793	-0.45	1.10	0.38
40800	0.57	0.69	1.69
40897	0.28	-2.08	0.57

Appendix E: PCA factor scores for house dust.

Dust Sample ID	PCA Factor Score		
	PC1	PC2	PC3
40029	-0.33	0.33	1.70
40034	1.00	0.35	0.24
40051	0.17	1.36	0.32
40131	0.42	0.72	0.33
40189	0.25	0.91	-0.14
40234	-1.83	-1.65	0.68
40269	-0.91	-2.02	-0.97
40602	0.89	-0.85	0.42
40714	0.43	-0.39	0.55
40078	1.06	-1.00	1.70
40449	0.83	0.15	0.65
40722	0.10	1.08	-0.72
40734	0.32	0.08	-1.20
40793	0.78	0.65	1.59
40372	-0.34	0.19	-2.00

Dust Sample ID	PCA Factor Score		
	PC1	PC2	PC3
40704	1.55	-1.11	1.66
40147	0.61	0.56	-1.25
40287	-0.86	-1.70	-2.53
40295	1.09	-1.57	1.78
40481	-1.19	-0.37	1.21
40487	0.13	0.90	-1.19
40685	0.82	0.68	-0.17
40085	1.12	0.29	-0.59
40096	0.34	-0.55	0.56
40126	1.60	-1.87	1.22
40260	-1.19	-0.11	0.50
40263	-0.14	0.54	0.34
40300	0.61	-1.02	-1.09
40338	0.47	0.56	0.19
40373	0.28	0.93	-0.58
40400	0.47	0.06	1.23

Dust Sample ID	PCA Factor Score		
	PC1	PC2	PC3
40483	-0.04	1.01	-0.41
40652	-0.25	-0.59	-0.13
40922	1.24	-0.06	-0.29
40512	0.23	0.28	-1.19
40663	0.41	-0.25	-1.28
40886	-1.83	-0.18	1.12
40908	-0.38	0.67	0.20
40662	0.26	1.08	0.05
40436	0.57	0.34	-0.41
40086	0.62	-0.93	-0.17
40806	1.14	-0.27	-0.50
40735	-0.14	1.53	-0.38
40897	0.81	0.58	0.31
40442	-0.31	-0.33	-0.19
40197	0.21	0.68	0.22
40980	-0.06	1.32	-0.96

Dust Sample ID	PCA Factor Score		
	PC1	PC2	PC3
40057	-0.58	-0.06	0.96
40638	-0.46	-0.93	1.53
40338	0.77	0.17	0.67
40269	-2.45	-1.33	-0.84
40126	1.73	-2.15	1.24
40029	-0.93	0.10	1.51
40115	0.61	0.96	-0.18
40115	0.27	0.80	-0.18
40153	-0.94	1.14	-1.11
40836	0.91	-0.23	0.65
40793	0.60	0.79	0.12
40189	0.37	0.68	-0.32
40221	-2.05	-0.96	-0.44
40097	0.73	1.01	0.06
40784	0.71	0.28	-0.62
40734	-0.24	-0.15	-1.83

Dust Sample ID	PCA Factor Score		
	PC1	PC2	PC3
40131	1.05	-0.11	0.88
40487	-0.99	0.52	-0.73
40221	-1.91	0.12	-0.15
40117	0.67	0.53	0.31
40260	0.98	0.79	-0.34
40336	0.33	-2.89	-2.48
40858	1.01	-0.02	-0.35
40336	-0.41	-0.77	-2.19
40246	1.80	-2.82	0.59
40287	-1.60	-0.52	-1.87
40551	-0.08	-2.35	-1.03
40725	-0.56	0.47	0.79
40725	0.14	0.19	-0.55
40551	-0.46	-1.51	-2.43
40805	0.29	0.76	1.24
40765	-2.90	-2.10	1.04

Dust Sample ID	PCA Factor Score		
	PC1	PC2	PC3
40182	-0.88	1.94	-0.50
40765	0.30	-0.05	0.07
40905	-2.86	0.08	0.77
40905	-1.71	0.65	-0.04
40722	-0.78	1.19	-0.42
40218	-0.48	1.25	-0.60
40253	0.01	0.79	-0.79
40512	0.87	0.56	-0.80
40356	1.75	-1.63	-0.22
40922	-1.31	-0.33	-0.04
40391	-0.39	-1.67	-1.22
40983	-0.57	0.43	1.72
40056	0.52	0.20	0.39
40983	0.69	0.18	1.34
40357	-0.43	1.00	0.03
40830	-0.65	-1.96	-1.30

Dust Sample ID	PCA Factor Score		
	PC1	PC2	PC3
40878	-0.20	-0.44	0.93
40878	-0.42	-0.12	0.79
40858	-2.24	0.29	0.72
40805	-0.66	1.23	0.81
40830	0.13	-1.12	-1.06
40263	-0.22	0.58	0.50
40980	0.19	0.96	-0.90
40981	-0.53	0.63	0.71
40923	1.49	-2.26	1.29
40862	0.67	0.68	-0.09
40436	0.39	1.18	-0.08
40491	0.74	1.05	-0.35
40894	0.93	0.17	0.30
40373	0.69	-0.09	0.39
40806	0.99	-0.18	-0.46
40629	0.43	0.73	1.40

Dust Sample ID	PCA Factor Score		
	PC1	PC2	PC3
40746	-2.41	-0.29	2.24
40442	-1.38	0.68	-0.39
40981	-0.91	0.16	0.91
40652	0.11	-0.06	-0.61
40372	-0.15	0.10	-1.22
40284	0.59	1.05	-0.35
40096	0.50	0.70	-0.28
40491	1.09	0.40	0.46
40086	1.37	-2.36	0.18
40587	-0.07	-1.65	-0.41
40746	-2.86	-0.63	2.01
40293	-0.33	-0.63	-1.28
40886	-1.89	-0.29	1.47
40662	0.67	0.65	0.92
40483	0.16	0.39	-0.39
40735	1.05	0.37	1.02

Dust Sample ID	PCA Factor Score		
	PC1	**PC2**	**PC3**
40790	0.37	0.17	1.53
40293	-0.17	0.75	-0.50
40788	-0.07	1.44	-0.80
40284	0.55	0.60	-0.92

Appendix F: Home characteristics used for evaluating associations between PFCs and home characteristics. Units and notes for columns are described at the bottom of the table.

Sample ID	Home Characteristics				
	Stove Fan	Vinyl Covered Furniture	Using Wet Dust-Attracting Device	Home Built	Renovation
40085	Yes	No	Yes	1980-1989	No
40086	No	No	Yes	1970-1979	Yes
40096	Yes	Yes	No	NA	NA
40115	No	Yes	No	1940-1949	No
40126	No	No	Yes	1939 or earlier	No
40131	Yes	No	Yes	1990 or later	Yes
40147	No	No	No	NA	No
40153	Yes	No	No	1940-1949	No

Sample ID	Home Characteristics				
	Stove Fan	**Vinyl Covered Furniture**	**Using Wet Dust-Attracting Device**	**Home Built**	**Renovation**
40182	Yes	No	No	1990 or later	No
40189	No	No	Yes	1950-1959	No
40197	Yes	No	Yes	1960-1969	No
40218	No	No	Yes	1970-1979	No
40221	Yes	No	No	NA	No
40234	Yes	No	Yes	1990 or later	No
40246	No	No	No	1990 or later	No
40253	Yes	No	Yes	1950-1959	No
40260	Yes	No	No	NA	No
40269	Yes	No	No	NA	No

Sample ID	Home Characteristics				
	Stove Fan	Vinyl Covered Furniture	Using Wet Dust-Attracting Device	Home Built	Renovation
40284	No	No	Yes	1980-1989	No
40295	Yes	No	Yes	1939 or earlier	No
40300	Yes	No	Yes	1970-1979	Yes
40336	No	No	Yes	1990 or later	No
40372	Yes	No	No	1960-1969	No
40373	Yes	No	No	1940-1949	No
40391	Yes	No	No	1939 or earlier	No
40400	Yes	No	No	1990 or later	No
40422	Yes	No	No	1940-1949	No
40436	Yes	No	No	NA	Yes

Sample ID	Home Characteristics				
	Stove Fan	**Vinyl Covered Furniture**	**Using Wet Dust-Attracting Device**	**Home Built**	**Renovation**
40449	Yes	No	Yes	1940-1949	Yes
40483	No	No	Yes	1970-1979	No
40487	Yes	No	No	1939 or earlier	No
40491	Yes	No	Yes	NA	No
40512	No	No	No	1940-1949	No
40551	No	No	No	1950-1959	No
40587	No	No	Yes	1970-1979	No
40629	Yes	No	Yes	1939 or earlier	No
40638	No	No	No	1990 or later	Yes
40662	Yes	No	Yes	NA	No

Sample ID	Home Characteristics				
	Stove Fan	**Vinyl Covered Furniture**	**Using Wet Dust-Attracting Device**	**Home Built**	**Renovation**
40663	No	No	Yes	1990 or later	No
40793	Yes	No	No	1939 or earlier	Yes
40878	NA	No	Yes	1960-1969	No

Sample ID	Home Characteristics				
	Ventilation	**Painting**	**Near High-Way/Artery**	**Volume of Home**	**Full Carpet Flooring**
40085	No	Yes	Yes	285.8	No
40086	No	Yes	No	246.4	No
40096	NA	Yes	Yes	176.3	No
40115	No	No	Yes	305.4	No
40126	Yes	No	Yes	290.8	No
40131	Yes	Yes	Yes	415.1	No
40147	No	No	No	207.3	No
40153	Yes	Yes	No	300.75	No
40182	No	No	No	174.2	Yes
40189	No	Yes	Yes	130.7	No

Sample ID	Home Characteristics				
	Ventilation	**Painting**	**Near High-Way/Artery**	**Volume of Home**	**Full Carpet Flooring**
40197	No	No	Yes	253	No
40218	No	Yes	No	177.8	No
40221	No	No	No	150.4	Yes
40234	No	No	No	335.7	No
40246	No	No	No	245.2	Yes
40253	No	Yes	No	213.9	No
40260	No	No	No	186.1	Yes
40269	No	Yes	No	173.3	No
40284	No	Yes	No	203.8	Yes
40295	Yes	No	Yes	346.2	No

Sample ID	Home Characteristics				
	Ventilation	**Painting**	**Near High-Way/Artery**	**Volume of Home**	**Full Carpet Flooring**
40300	No	Yes	No	386.9	No
40336	No	Yes	No	506.1	No
40372	No	Yes	No	226.4	Yes
40373	No	Yes	No	290.8	Yes
40391	No	Yes	Yes	540.4	No
40400	No	Yes	No	465.6	Yes
40422	No	Yes	Yes	327.9	No
40436	NA	Yes	Yes	249.9	No
40449	Yes	Yes	No	621.3	Yes
40483	No	Yes	No	269.3	No

Sample ID	Home Characteristics				
	Ventilation	**Painting**	**Near High-Way/Artery**	**Volume of Home**	**Full Carpet Flooring**
40487	No	No	Yes	387.7	No
40491	No	No	No	160.1	Yes
40512	No	No	Yes	286.4	No
40551	No	No	Yes	286.1	No
40587	No	No	No	174.4	Yes
40629	No	Yes	No	423.6	Yes
40638	No	Yes	No	467	Yes
40662	No	No	No	199	No
40663	No	Yes	No	421.9	Yes
40793	No	Yes	Yes	325.9	No

Sample ID	Home Characteristics		
	Laminate	Area Rug	Home Cleaning Per Month
40085	No	Yes	3
40086	Yes	No	4
40096	No	No	4
40115	No	No	4
40126	No	No	2
40131	No	Yes	2
40147	No	No	2
40153	Yes	Yes	1
40182	No	No	20
40189	No	Yes	1
40197	No	No	8
40218	No	Yes	3
40221	No	No	4
40234	Yes	No	4
40246	No	No	5

Sample ID	Home Characteristics		
	Laminate	Area Rug	Home Cleaning Per Month
40253	Yes	No	4
40260	No	No	5
40269	No	No	4
40284	No	No	1
40295	No	Yes	6
40300	Yes	No	5
40336	Yes	No	5
40372	No	No	3
40373	No	No	4
40391	No	Yes	4
40400	No	No	10
40422	No	No	7
40436	No	Yes	1
40449	No	No	3
40483	Yes	No	2

Sample ID	Home Characteristics		
	Laminate	Area Rug	Home Cleaning Per Month
40487	No	Yes	5
40491	No	No	2
40512	No	No	2
40551	No	No	4
40587	No	No	4
40629	No	Yes	4
40638	No	No	3
40662	No	Yes	1
40663	No	No	8
40793	No	No	2
40878	No	No	4

Sample ID	Home Characteristics				
	Near Gas Station	Near Parking	Fuel Use (Electric)	Remove Their Shoes	Vacuum to Clean
40085	No	No	Yes	Yes	Yes
40086	No	No	No	Yes	Yes
40096	No	Yes	No	Yes	No
40115	No	Yes	No	Yes	Yes
40126	No	Yes	Yes	Yes	Yes
40131	No	No	No	Yes	Yes
40147	No	No	No	Yes	Yes
40153	No	Yes	Yes	Yes	Yes
40182	No	Yes	Yes	Yes	Yes

Sample ID	Home Characteristics				
	Near Gas Station	**Near Parking**	**Fuel Use (Electric)**	**Remove Their Shoes**	**Vacuum to Clean**
40189	No	No	Yes	Yes	Yes
40197	No	No	No	Yes	No
40218	No	No	Yes	Yes	No
40221	No	No	Yes	No	Yes
40234	No	No	No	Yes	No
40246	No	Yes	No	Yes	Yes
40253	No	Yes	No	Yes	Yes
40260	No	No	No	Yes	Yes
40269	No	Yes	No	Yes	Yes

Sample ID	Home Characteristics				
	Near Gas Station	**Near Parking**	**Fuel Use (Electric)**	**Remove Their Shoes**	**Vacuum to Clean**
40284	No	No	No	Yes	Yes
40295	No	Yes	No	Yes	Yes
40300	No	No	No	Yes	Yes
40336	No	No	No	Yes	Yes
40372	No	No	No	Yes	Yes
40373	No	Yes	No	Yes	Yes
40391	No	Yes	No	Yes	Yes
40400	No	Yes	No	Yes	No
40422	Yes	No	No	Yes	No

Sample ID	Home Characteristics				
	Near Gas Station	**Near Parking**	**Fuel Use (Electric)**	**Remove Their Shoes**	**Vacuum to Clean**
40436	No	Yes	No	Yes	Yes
40449	No	Yes	No	Yes	No
40483	No	Yes	No	Yes	Yes
40487	No	No	No	Yes	Yes
40491	No	Yes	Yes	Yes	Yes
40512	No	No	No	Yes	Yes
40551	No	No	Yes	No	Yes
40587	No	No	Yes	Yes	Yes
40629	No	No	No	Yes	Yes

Sample ID	Home Characteristics				
	Near Gas Station	**Near Parking**	**Fuel Use (Electric)**	**Remove Their Shoes**	**Vacuum to Clean**
40638	No	Yes	Yes	Yes	No
40662	No	No	No	Yes	Yes
40663	No	Yes	No	Yes	No
40793	No	No	No	Yes	Yes
40878	No	Yes	Yes	Yes	Yes

Sample ID	Home Characteristics	
	Soft Vinyl Floor	**Home Heated by Forced air**
40085	NA	NA
40086	No	Yes
40096	NA	NA
40115	No	Yes
40126	Yes	Yes
40131	No	Yes
40147	No	Yes
40153	No	Yes
40182	No	Yes
40189	No	Yes
40197	Yes	Yes
40218	NA	Yes
40221	No	No
40234	No	Yes
40246	No	Yes
40253	NA	Yes

Sample ID	Home Characteristics	
	Soft Vinyl Floor	Home Heated by Forced air
40260	NA	NA
40269	Yes	Yes
40284	No	NA
40295	No	Yes
40300	No	Yes
40336	Yes	Yes
40372	No	Yes
40373	No	NA
40391	No	NA
40400	No	Yes
40422	No	No
40436	No	Yes
40449	No	Yes
40483	No	Yes
40487	NA	No
40491	No	Yes

Sample ID	Home Characteristics	
	Soft Vinyl Floor	**Home Heated by Forced air**
40512	No	Yes
40551	NA	NA
40587	No	NA
40629	No	Yes
40638	No	NA
40662	No	Yes
40663	No	NA
40793	No	Yes
40878	NA	NA

Sample ID	Home Characteristics			
	New Heating System	Added Carpeting	Remove Carpeting	New Furniture
40085	No	No	No	Yes
40086	NA	NA	NA	No
40096	No	No	No	Yes
40115	NA	NA	NA	No
40126	No	No	No	No
40131	NA	NA	NA	No
40147	NA	NA	NA	NA
40153	Yes	Yes	No	Yes
40182	NA	NA	NA	No

Sample ID	Home Characteristics			
	New Heating System	Added Carpeting	Remove Carpeting	New Furniture
40189	No	No	No	No
40197	No	No	No	No
40218	NA	NA	NA	No
40221	NA	NA	NA	No
40234	NA	NA	NA	No
40246	NA	NA	NA	No
40253	NA	NA	NA	No
40260	NA	NA	NA	No
40269	NA	NA	NA	No

Sample ID	Home Characteristics			
	New Heating System	Added Carpeting	Remove Carpeting	New Furniture
40284	NA	NA	NA	No
40295	NA	NA	NA	No
40336	No	No	No	No
40373	No	No	No	No
40442	No	No	No	Yes
40449	NA	NA	NA	No
40483	Yes	No	No	Yes
40487	NA	NA	NA	NA
40551	No	No	No	No

Sample ID	Home Characteristics			
	New Heating System	Added Carpeting	Remove Carpeting	New Furniture
40629	NA	NA	NA	Yes
40638	No	No	No	No
40662	No	No	No	No
40663	No	No	No	No
40878	NA	NA	NA	No
40894	No	No	No	No
40897	NA	NA	NA	No

Sample ID	Home Characteristics	
	Use A/C in Summer	Hard Wood Floor
40085	Rarely	Yes
40086	Occasionally	No
40096	NA	Yes
40115	Occasionally	Yes
40126	Regularly	Yes
40131	Rarely	Yes
40147	Regularly	No
40153	Occasionally	No
40182	Occasionally	No
40189	NA	Yes
40197	Occasionally	Yes
40218	Occasionally	No
40221	Occasionally	No
40234	Rarely	No
40246	Regularly	No
40253	Occasionally	No

Sample ID	Home Characteristics	
	Use A/C in Summer	**Hard Wood Floor**
40260	Regularly	No
40263	Occasionally	No
40269	NA	No
40284	Regularly	Yes
40295	NA	No
40300	Regularly	No
40336	Regularly	No
40372	Occasionally	No
40373	Regularly	Yes
40391	NA	No
40400	Regularly	Yes
40436	Regularly	Yes
40442	Regularly	No
40449	Rarely	No
40483	Regularly	Yes
40487	NA	No

Sample ID	Home Characteristics	
	Use A/C in Summer	**Hard Wood Floor**
40491	NA	Yes
40512	Occasionally	Yes
40551	Regularly	No
40587	NA	No
40629	Regularly	No
40638	Regularly	No
40662	NA	No
40663	Regularly	Yes
40793	Regularly	No
40878	Regularly	No
40894	Regularly	No
40897	Regularly	No

Stove fan = Whether or not a stove fan is used in the kitchen.

Vinyl covered furniture = Whether or not major vinyl covered furniture existed at the home.

Using wet dust-attracting device = Whether or not a wet dust-attracting device was used for cleaning the home regularly.

Home Built = When was the participant's home built?

Renovations = Whether or not participants did home renovations in the previous 12 months.

Laminate = Whether or not home has a laminate floor.

Area rug = Whether or not there is an area rug in the living room.

Home cleaning per month= What is the frequency of home cleaning per month?

Ventilation = Whether or not participants upgraded home ventilation.

Painting = Whether or not any painting has been done by participants within the last 12 months.

Near highway/artery = Whether or not home is near a highway/artery.

Volume of home = What is the total volume of the home (m^3)?

Full carpet flooring = Whether or not the living room had full carpet flooring.

NA = Not applicable due to missing surveys, missing survey questions, participant did not know answer to the question or not available.

Near gas station = Whether or not home is within 100 metres of any gas station.

Near Parking = Whether or not home is near any large parking lot.

Fuel use (Electric) = Whether or not type of fuel used at home is electric.

Remove their shoes = Whether or not people remove their shoes when entering home.

Vacuum to clean = Whether or not vacuuming was used to clean the home.

Soft vinyl (tiles/sheet) = Whether or not home has vinyl/sheet tile flooring.

Home heated by forced air = Whether or not home is heated completely or partly by forced air through ducted vents.

New heating system = Whether or not home has a new furnace or heating system.

Added carpeting = Whether or not carpet was added to home within the last 12 months.

Remove carpeting = Whether or not carpet was removed from home within the last 12 months.

New furniture = Whether or not new furniture has been used in home within the last 12 months.

Use A/C in summer = How often A/C is used in summer.

Hard Wood floor = Whether or not home has hard wood flooring.

A/C = Air conditioner

Appendix G: Control and target homes for indoor air evaluation. Control and target described at the bottom of the table.

Air Sample ID		Air Sample ID	
Control Home	**Target Home**	**Control Home**	**Target Home**
40481	40069	40260	40373
40355	40131	40338	40126
40029	40187	40535	40263
40714	40034	40980	40289
40453	40078	40374	40295
40051	40320	40685	40188
40287	40057	40287	40289
40722	40096	40922	40800
40189	40328	40984	40400
40300	40090	40086	40483
40734	40449	40806	40652
40182	40085	40442	40922
40793	40234	40156	40611
40602	40487	40512	40912
40685	40269		40663
40147	40295		40356

Appendix H: Control and target homes for dust evaluation. Control and target described at the bottom of the table.

Dust Sample ID		**Dust Sample ID**	
Control	**Target**	**Control**	**Target**
40980 MR	40652 MR	40922 MR	40858 MR
40980 CR	40652 CR	40983 CR	40858 CR
40086 CR	40373 MR	40983 MR	40905 CR
40442 CR	40373 CR	40029 CR	40905 MR
40442 MR	40663 CR	40793 CR	40400 MR
40806 CR	40483 MR	40189 CR	40836 MR
40806 MR	40483 CR	40189 MR	
40338 CR	40126 MR	40147 MR	
40338 MR	40131MR	40260 CR	
40029MR	40131CR	40260 MR	
40029 CR	40449 MR	40197 CR	
40793 CR	40234 MR	40629 CR	
40793 MR	40487 CR	40922 CR	
40189 CR	40487 MR	40922 MR	
40189 MR	40096 MR	40983 CR	
40147 MR	40096 CR	40983 MR	

Target home = Home in which there was an infant with recurrent wheezing confirmed by a physician.

Control home = Children in control home had no episode of wheezing

Dust Sample ID		Dust Sample ID	
Control	**Target**	**Control**	**Target**
40980 MR	40652 MR	40922 MR	40858 MR
40980 CR	40652 CR	40983 CR	40858 CR
40086 CR	40373 MR	40983 MR	40905 CR
40442 CR	40373 CR	40029 CR	40905 MR
40442 MR	40663 CR	40793 CR	40400 MR
40806 CR	40483 MR	40189 CR	40836 MR
40806 MR	40483 CR	40189 MR	
40338 CR	40126 MR	40147 MR	
40338 MR	40131MR	40260 CR	
40029MR	40131CR	40260 MR	
40029 CR	40449 MR	40197 CR	
40793 CR	40234 MR	40629 CR	
40793 MR	40487 CR	40922 CR	
40189 CR	40487 MR	40922 MR	
40189 MR	40096 MR	40983 CR	

Appendix I: Questionnaires regarding home characteristics

1. Please provide the postal code of the home you are reporting on here.

1.1 Have you moved since you completed the Home Environment Questionnaire during your pregnancy? Yes No

2. What type of home do you now live in? Single family Multi-family

2.1 Choose the type of dwelling that best describes your home:

A single family detached house

A single family house attached to 1 or more houses (i.e. townhouse/semi-detached)

Manufactured home/mobile home or trailer

Low rise apartment/condo/co-op (1-3 floors)

Multi-family home (more than one family in a converted single home, but with separate cooking areas)

High rise apartment/condo/co-op (4 floors or more)

Other

2.1a If Other, specify:

3. When was your present home originally built? (Check one only)

1939 or earlier 1950-1959 1970-1979 1990 or later

1940-1949 1960-1969 1980-1989 Don't know

3.1 If your present home was built in 1990

or later, was it built within the last year?

Yes No N/A

4. How long have YOU lived in this home? Months

5. How old is the ROOF COVERING on the building?

< 5 years 5 - 15 year > 15 years Don't know

6. Does your home have a basement? Yes No

(A basement is a space where part or all of the floor is below the outside ground level)

6.1 If yes, is the basement used as your living

space? Yes No

(Living space is any space in your home that you enter and occupy continuously for at least 30 minutes on a regular basis, multiple days per week)

7. What floor is your main living area on? Basement Ground floor Other

7.1 If Other floor, specify: (Basement=00, ground floor=01, all other floors input actual floor number)

8. On what floor does your baby spend the

majority of time during waking hours?

9. What floor does your baby usually sleep on?

10. How many square feet is your home? (including basement if it is your living space; see Q 6.1 for definition of living space) < 1000 1001-1500 1501-2000 2001-2500 > 2500

11. How many bedrooms are there in your home? (including in basement if it is your living space)

12. How many people currently live in your home? Adults Children

(Adult is 18 years or older; include tenants if you are renting space in your home or basement)

13. How many weeks since birth did your baby NOT sleep (at night) in your home?

weeks (e.g., vacation, cottage, travel)

14. Is your home within 100 metres of any of the following? (100 metres is the length of a football field) Check all that apply.

Major Highway/Artery

Body of water

Factory Farm

Gas station Large parking lot

None of these choices Major/prolonged construction activity (e.g., building of houses or other buildings, road work, typically involving heavy machinery

and/or generation of noticeable amounts of dust in the air)

Other source of pollution

14.1 If near other source of pollution, specify:

14.2 Approximately what percentage of time (including both day and night) since baby came home has one or more windows in your home been kept open (at least partially) to allow exchange with outdoor air?

HOME IMPROVEMENTS

15. Have you done any improvements, renovations or extensions to the home or homes you lived in with your baby since you completed this questionnaire during your pregnancy?

Yes No, go to Q16

15.1 If Yes, indicate which of the following you changed in ANY of the homes that your baby lived in since birth. Check all that apply.

Added to or renovated space in home Added vinyl flooring

Finished basement or added basement insulation Upgraded windows

Added attic or wall insulation Upgraded plumbing

Added wood fireplace or wood stove Painting/finishing

Upgraded ventilation and/or duct work Added carpeting

New furnace or heating system Removed carpeting

New roof Other renovation

15.2 If Other renovation, specify:

(e.g., wallpaper, cabinets, siding, flooring not

mentioned above)

FURNITURE

16. Have you brought any major pieces of NEW furniture into your home since you completed

the Home Environment Questionnaire during your pregnancy?

Yes No, go to Q17

(e.g., sofa, bookcase, cabinet, large armchairs; NEW is less than 1 year old).

16.1 If Yes, how many NEW pieces of furniture have been brought into your home with the following coverings:

(Enter 00 for none; if a piece has 2 types of coverings/exposed surfaces (i.e., upholstery and pressed wood), this piece

is counted in both the Upholstery and Pressed Wood categories)

Solid wood Leather Pressed wood Vinyl

Plastic Upholstery Metal Other

16.2 If Other covering, specify:

16.3 What rooms are these pieces of furniture in?

Check all that apply.

Mother's bedroom Living room

Baby's bedroom (not shared with others) Family room

Other bedrooms Kitchen

Other room not mentioned Basement

16.4 If Other room, specify:

PETS

17. Have you had any FURRY PETS living in your home since you completed this questionnaire

during your pregnancy? Yes No, go to Q18

17.1 How many FURRY pets have you had living in your home during this time period? (Do not include furless pets like birds, reptiles or fish; If you have tenants living in your home, count their furry pets along with your own; Enter 00 for none) Dog(s) Cat(s) Other FURRY pet(s)

17.2 If Other FURRY pets, specify:

18. If you have moved homes since you completed the Home Environment Questionnaire during

pregnancy, did the previous owner of your current home have a FURRY pet?

Check all that apply. Select N/A if you did not move residences, or no previous owner (i.e., newly built home).

N/A Don't know No Dog(s) Cat(s) Other FURRY pet(s)

18.1 If Other FURRY pet(s), specify:

BABY'S TIME / LOCATION

19. Since birth, how much time (in HOURS) during a typical day/night did your baby spend (e.g., sleeping, playing, eating) in the following rooms or places?

Mother's bedroom

Baby's bedroom (not shared with others)

Bedroom shared with others

Family room

Living room

Kitchen

Basement

Other rooms

Outside but near home

Away from home

20. Has baby EVER slept in your bedroom (including naps)? Yes No, go to Q25

20.1 If yes, what percentage of baby's total sleep time was spent in your bedroom since birth?

(Estimate to the nearest 10%; Enter 000 if none)

21. What percentage of time does your baby sleep in the following beds in your room?

(Enter 000 in each field if no time was spent in a given bed)

21.1 Your bed: %

21.1a Does the mattress on your bed have an allergy control cover?

Yes No Don't know N/A

21.2 Other bed for baby alone (e.g., crib,

bassinette, safety bed):

21.2a Does the mattress on this bed have

an allergy control cover?

Yes No Don't know N/A

21.3 Other type of bed:

21.3a If Other type of bed, specify:

21.3b Does the mattress on this bed have

an allergy control cover?

Yes No Don't know N/A

22. On average, how often do you open the windows in your bedroom for longer than an hour during

mid-winter?

Never 1-2 times/month 1-2 times/week >2 times/week N/A

23. On average, how often do you open the windows in your bedroom for longer than an hour during

mid-summer?

Never 1-2 times/month 1-2 times/week >2 times/week N/A

24. Your bedroom is:

Completely carpeted Partly carpeted Not carpeted

BABY'S OWN BEDROOM (not shared with others)

25. Has baby EVER slept in this bedroom? Yes No, go to Q30 N/A, go to Q30

25.1 If Yes, what percentage of baby's total sleep

time was spent in this bedroom since birth?

(Estimate to the nearest 10%; Enter 000 if none)

26. Does the mattress for the bed baby mainly sleeps on in this room have an allergy control cover? Yes No Don't know N/A

27. On average, how often do you open the windows for longer than an hour in this room during mid-winter? Never 1-2 times/month 1-2 times/week >2 times/week N/A

28. On average, how often do you open the windows in this room for longer than an hour during mid-summer? Never 1-2 times/month 1-2 times/week >2 times/week N/A

29. This bedroom is:

Completely carpeted Partly carpeted Not carpeted

30. Has baby EVER slept in this bedroom? Yes No, go to Q36 N/A, go to Q36

30.1 If Yes, what percentage of baby's total sleep

time was spent in this bedroom since birth? (Estimate to the nearest 10%; Enter 000 if none)

31. How many other people typically sleep in this room?

32. Does the mattress for the bed baby mainly sleeps on in this room have an allergy control cover? Yes No Don't know N/A

33. On average, how often do you open the windows in this room for longer than an hour during mid-winter? Never 1-2 times/month 1-2 times/week >2 times/week N/A

34. On average, how often do you open the windows in this room for longer than an hour during mid-summer? Never 1-2 times/month 1-2 times/week >2 times/week N/A

35. This bedroom is: Completely carpeted Partly carpeted Not carpeted

MOST USED LIVING SPACE (Family or Living Room)

Choose either your family room or your living room, whichever room your baby spends the most time in.

36. Has baby EVER slept in this room (i.e., most used living space)?

Yes No, go to Q41 N/A, go to Q41

36.1 If Yes, what percentage of baby's total sleep time was spent in this room since birth? (Estimate to the nearest 10%; Enter 000 if none)

37. What percentage of time that baby slept in this room did s/he sleep in the following:

Playpen - Swing

Baby bed (e.g., crib, bassinette, safety bed) % - Floor Mat

Other, specify below

37.1 If Other, specify:

e.g. arms of adult, sofa, car seat

37.2 Does "Other" have an allergy control cover? Yes No Don't know N/A

37.3 Does "Baby bed" mattress have an allergy control cover?

Yes No Don't know N/A

38. On average, how often do you open the windows in this room for longer than an hour during mid-winter?

Never 1-2 times/month 1-2 times/week >2 times/week N/A

39. On average, how often do you open the windows in this room for longer than an hour during mid-summer?

Never 1-2 times/month 1-2 times/week >2 times/week N/A

40. This room is:

Completely carpeted Partly carpeted Not carpeted

41. How many times per week do you typically use the following products on baby?

41.1 Baby powder, talc or cornstarch: Never 1-5 6-15 >15

41.2 Diaper cream: Never 1-5 6-15 >15

41.3 Baby wipes: Never 1-5 6-15 >15

41.4 Baby shampoo: Never 1-5 6-15 >15

41.5 Baby lotion: Never 1-5 6-15 >15

41.6 Baby soap: Never 1-5 6-15 >15

41.7 Adult soaps, lotions, shampoos: Never 1-5 6-15 >15

41.8 Other skin products: Never 1-5 6-15 >15

(e.g., sunscreens, insect repellents, prescription creams)

41.8a If Other products used, specify type:

42. How many minutes in a typical 24-hour period (day/night) does your baby: (Enter whole numbers only; enter 000 for none)

42.1 Use a pacifier? minutes

42.2 Play with or use soft plastic toys/teething rings? minutes (Include jungle gym toys, stroller toys, etc)

42.3 Play with or use soft plastic toys/teething rings that involves putting them in his/her mouth? minutes (If baby puts these plastic toys/teething rings in his/her
mouth for all of the playing time given in Q42.2, enter the same number of minutes as given in Q42.2)

43. What percentage of time in a typical 24-hour
period are you using cloth diapers on baby? (Enter whole numbers only; enter 000 for none)

44. What percentage of time in a typical 24-hour period are you using disposable diapers on baby? (Enter whole numbers only; enter 000 for none)

WORK ACTIVITIES

45. Are there members of the household who work with hazardous materials on the job? (e.g., cleaners, chemicals, asbestos, batteries, lead, mercury, paint or pesticides)

Yes No, go to Q46 Don't know, go to Q46

45.1 If Yes, what is their job?

45.2 BEFORE ENTERING the home, do they:

45.2a Change work clothes: Yes No Don't know

45.2b Change work shoes: Yes No Don't know

45.2c Shower: Yes No Don't know

45.2d Are their work clothes laundered separately

from the family clothes?

Yes No Don't know

CLEANING AND DUSTING

46. When people come into your home, do they usually remove their shoes?

Yes No

47. How many times per month do you or someone else usually clean your home (i.e., a general overall cleaning)?

48. Do you usually use a vacuum to clean your

floors?

Yes No

48.1 If Yes, what type of vacuum?

Portable vacuum without HEPA filter Portable vacuum with HEPA filter

Central vacuum Don't know

49. Other tools you use to clean your floors: (Check all that apply)

Dry dust-attracting device (e.g., Swiffer Dry) Broom Other tool, specify below

Wet dust-attracting device (e.g., Swiffer Wet Jet) Mop Wheeled sweeper

If Other tool, specify:

50. Which of the following products have been USED in your home since your baby was born? (Day=daily, Wk=weekly, Mth=monthly, <Mth=less than monthly, No=not used)

50.1 Liquid or solid air freshener: Day Wk Mth <Mth No

50.2 Spray air freshener: Day Wk Mth <Mth No

50.3 Plug-in deodorizer/air freshener: Day Wk Mth <Mth No

50.4 Floor Cleaner: Day Wk Mth <Mth No

50.5 Furniture polish: Day Wk Mth <Mth No

50.6 Floor polish: Day Wk Mth <Mth No

50.7 Dusting polish or spray: Day Wk Mth <Mth No

50.8 Drain cleaner: Day Wk Mth <Mth No

50.9 Hand dishwashing detergent: Day Wk Mth <Mth No

50.10 Dishwasher detergent: Day Wk Mth <Mth No

50.11 Bleach: Day Wk Mth <Mth No

50.12 Multi-surface cleaner: Day Wk Mth <Mth No

50.13 Silver or brass polish: Day Wk Mth <Mth No

50.14 Disinfectant in bedrooms: Day Wk Mth <Mth No

50.15 Disinfectant in home in general: Day Wk Mth <Mth No

50.16 Unscented candle: Day Wk Mth <Mth No

50.17 Scented candle: Day Wk Mth <Mth No

50.18 Incense: Day Wk Mth <Mth No

50.19 Eco & organic cleaning product: Day Wk Mth <Mth No

50.20 Glass cleaner: Day Wk Mth <Mth No

50.21 Chemical hand cleaner (e.g., for grease): Day Wk Mth <Mth No

50.22 Purell-type hand cleaner: Day Wk Mth <Mth No

50.23 Unscented laundry detergent: Day Wk Mth <Mth No

50.24 Scented laundry detergent: Day Wk Mth <Mth No

50.25 Fabric softener: Day Wk Mth <Mth No

50.26 Toilet bowl cleaner: Day Wk Mth <Mth No

50.27 Oven cleaner: Day Wk Mth <Mth No

50.28 Bathroom tile cleaner: Day Wk Mth <Mth No

50.29 Solvents (e.g. nail polish/paint remover): Day Wk Mth <Mth No

50.30 Insecticide: Day Wk Mth <Mth No

50.31 Other products not listed above: Day Wk Mth <Mth No

50.31a If Other products, specify type:

51. Since your baby was born, have you operated

a portable air cleaner/purifier in your home?

Yes No, go to Q52

51.1 How often do you use it? Never Rarely Occasionally Regularly

51.2 How does it purify the air? (Check all that apply)

HEPA filter Ozone Water Negative ion Don't know

51.3 What room(s) is it located in?

Check all that apply.

Mother's bedroom Baby's bedroom Other bedrooms Family room

Main living space Kitchen Basement Other room

51.4 If Other room, specify:

PESTS AND BUGS

The following questions ask about any pests and bugs that you have noticed in your home from the time you completed the Home Environment Questionnaire during your pregnancy until now.

This does not include the occasional sighting of a bug that has found its way into your home,

but a recurrent problem that you felt you needed to deal with. For large pests (e.g., mice, rats, other rodents), the sighting of a single animal (or signs of its presence in the home) should be counted.

52. Have you had any PESTS or BUGS in your home during this time period?

Yes No, go to Q59

53. Have you noticed MICE in your home during this time period?

Never, go to Q54 Rarely Occasionally Regularly

53.1 Did you use a pest control company for

the mouse problem during this time period?

Yes No

53.2 Did you personally treat the mouse

problem during this time period?

Yes No

53.2a If Yes, what product did you use? (Check all that apply) Rodent poison Rodent trap Mouse adhesive pad Other

53.2b If Other products used, specify:

54. Have you noticed RATS in your home during this time period?

Never, go to Q55 Rarely Occasionally Regularly

54.1 Did you use a pest control company for

the rat problem during this time period?

54.2 Did you personally treat the rat problem during this time period?

Yes No

54.2a If Yes, what product did you use?

(Check all that apply)

Rodent poison Rodent trap Other

54.2b If Other product used, specify:

55. Have you noticed TERMITES/CARPENTER ANTS at your home during this time period?

Never, go to Q56 Rarely Occasionally Regularly

55.1 Did you use a pest control company for the termite/carpenter ant problem during this time period? Yes No

55.2 Did you personally treat the termite/carpenter ant problem during this time period?

Yes No

55.2a If Yes, what product did you use? (Check all that apply)

Insect powder Ant traps Ant poison liquid Other

55.2b If Other product used, specify:

56. Have you noticed COCKROACHES in your home during this time period?

Never, go to Q57 Rarely Occasionally Regularly

56.1 Did you use a pest control company for the cockroach problem during this time period?

Yes No

56.2 Did you personally treat the cockroach problem during this time period?

Yes No

56.2a If Yes, what product did you use? (Check all that apply) Dry powder Cockroach poison Chinese chalk Trap Other

56.2b If Other product used, specify:

57. Have you noticed MOTHS in your home during this time period?

Yes No

57.1 If yes, did you use moth balls? Yes No

58. Have you noticed OTHER PESTS or BUGS in your home during this time period?

Never, go to Q59 Rarely Occasionally Regularly

58.1 Did you use a pest control company for the other pests or bugs during this time period?

Yes No

58.2 Did you personally treat the other pests or bugs problem during this time period?

Yes No

58.2a If Yes, what product did you use? (Check all that apply) Dry powder Poison Trap Insect spray Other

58.2b If Other, specify type of pest and product used:

SMOKING

For the following, include all forms of tobacco smoking (e.g., cigarettes, cigars and pipe)

59. Since baby's birth, has anyone smoked AT baby's home? Yes No, go to Q60

59.1 Where do the person(s) smoking AT baby's home usually smoke? (Check all that apply)

Inside baby's home

Inside baby's home near an open window or in the attached garage

Outside but near baby's home

59.2 On average, how many cigarettes in total are smoked per day AT baby's home?

Total cigarettes, pipes, cigars for all locations listed in the previous question (e.g., if mother smokes 4, father smokes

5 and other persons smoke 3, total = 12 smokes).

None 1 to 5 6 to 10 > 10

MOISTURE AND VENTILATION

60. Do you see condensation, sweating/dripping on the inside of windows in cooler weather? Yes No

61. Is there a fan/exhaust in the bathroom where baby spends (or will spend) the most time?

Yes No

61.1 If Yes, how often do you turn this bathroom fan on? Never Rarely Occasionally Regularly

62. Does the window or mirror in the bathroom where baby spends the most time stay fogged up for more than 15 minutes after a shower or bath? Yes No

63. Have you used a PORTABLE humidifier/vaporizer in your home since you completed the Home Environment Questionnaire during pregnancy?

Yes No

63.1 If Yes, how often on average do you use the portable humidifier/vaporizer?

Rarely Occasionally Regularly Constantly

64. Do you regularly take other active measures to add humidity to the air inside your home (e.g., boiling pot on stove, tub full of water)? Yes No

65. Have you used a DEHUMIDIFIER in your home

since you completed the Home Environment Questionnaire during your pregnancy? Yes No

65.1 If Yes, how often on average do you use a dehumidifier?

Rarely Occasionally Regularly Constantly

66. Do you have a WASHING MACHINE inside your home?

(Don't include washing machines located in a common area of an apartment building but include shared washing machines located inside multi-family homes) Yes No

66.1 If Yes, how many times per week on average, do you operate a washing machine inside your home? (Indicate number of loads washed per week) loads

67. How do you dry your clothes? (Check all that apply.)

Gas Dryer, vented outdoors Electric Dryer, vented outdoors

Gas Dryer, vented indoors Electric Dryer, vented indoors

Gas Dryer, unknown venting Electric Dryer, unknown venting

Dryer in common area of apartment building Hanging outside the house

Other, specify below Hanging inside the house

67.1 If Other, specify:

WATER LEAKS/INCIDENTS

The following questions ask about any leak(s) that you have become aware of in your home since you completed the Home Environment Questionnaire during your pregnancy. If more than one leak has occurred during this time period, select the WORST one to complete these questions.

68. Are you aware of any water leaks/incidents in your home during this period of time?

Yes No, go to Q69

68.1 Where did the leak(s) occur? (Check all that apply)

Child's bedroom Bathroom(s) Attic

Mother's bedroom Kitchen Basement

Other bedroom Other rooms Crawl space

Living/dining/family rooms

68.2a What type of leak(s) was it? (Check all that apply)

Roof leak Plumbing Window leak Overhead apartment leak

Basement leak Sewer back up Flood Toilet overflow

Other leak

68.2b If Other, specify:

68.3 How much water was there? (Check all that apply)

Dampness Dripping/puddles Standing water

68.4 How many days was this area wet/damp? days

(If don't know, enter 99)

68.5 Is the affected area currently wet? Yes No Don't know

68.6 Indicate which of the following structures were affected: (Check all that apply)

Carpet Raised floor Wall Ceiling

Furniture Subfloor Window Other Nothing was affected

68.7 If Other area affected, specify:

68.8 What was done about the leak(s)? (Check all that apply)

Nothing Removed carpet or flooring

Removed water right away Repaired the problem myself

Removed water within a few days Repaired by professional cleanup company

68.9 How would you best describe the leak? Minor Major

68.10 How many other leaks occurred in your home during this time period? (enter 00 if none)

COOKING INSIDE THE HOME

69. Do you have a cooking appliance(s) that you use to cook food inside your home?

Yes No, go to Q70

69.1 If Yes, what type of cooking appliance(s) do you have? (Check all that apply)

Gas/Propane stove Wood burning stove

Electric stove Other, specify below:

69.2 If gas/propane, is there a pilot light? Yes No Don't know N/A

69.3 How often do fumes from cooking create noticeable cooking odours inside your home (e.g., smoke from frying meat or vegetables)?

Never Rarely Occasionally Regularly N/A

69.4 Is there a stove fan/exhaust in the kitchen? No Yes, recirculated Yes, vented outside Yes, venting unknown

69.5 When you cook, how often do you turn this fan/exhaust on? Never Rarely Occasionally Regularly N/A

69.6 Are there additional cooking facilities in your home (e.g., stove in your basement, basement apartment)? Yes No

69.6a If Yes, how often are these facilities used? Never Rarely Occasionally Regularly N/A

BASEMENT

70. Do you live in an apartment building? Yes, go to Q83 No

71. Does your home have a basement? Yes No, go to Q83

72. Does the basement in your home smell earthy,

mouldy or musty? Yes No Don't know

73. Are the walls around the perimeter of your basement finished?

(e.g. perimeter means the walls in contact with the exterior foundation walls; exclude furnace room and fruit cellar) Unfinished Partially finished Fully finished Don't know

74. Is the basement ceiling finished?

Unfinished Partially finished Fully finished Don't know

75. Is your basement insulated? Yes No Don't know

76. What type of flooring is in the basement? (Check all that apply)

Cement/concrete Laminate Hard vinyl (tiles)

Full carpet Hardwood Soft vinyl (tiles/sheet)

Partial Carpet Ceramic tiles Other

76.1 If Other (cork, unfinished wood), specify:

77. Is there a raised floor? Yes No Don't know

78. Do you have a DEHUMIDIFIER in the basement?

Yes No, go to Q79 Don't know, go to Q79

78.1 How often does this dehumidifier run in winter?

Never Rarely A few times per week Daily Don't know

78.2 How often does this dehumidifier run in summer?

Never Rarely A few times per week Daily Don't know

79. Is there a sump pump? Yes No Don't know

79.1 If Yes, how often does the sump pump turn on?

Never Rarely A few times per week Daily Don't know

80. Is there a portable HUMIDIFIER in the basement?

Yes No, go to Q81 Don't know, go to Q81

80.1 How often does this humidifier run in winter?

Never Rarely A few times per week Daily Don't know

80.2 How often does this humidifier run in summer?

Never Rarely A few times per week Daily Don't know

81. Are there any visible signs of mould in the

basement?

Yes No Don't know

82. Indicate the number of items below stored in your basement:

82.1 Gasoline cans: None 1 2-5 6-10 > 10

82.2 Camping fuels: None 1 2-5 6-10 > 10

82.3 Paints/stains and thinners: None 1 2-5 6-10 > 10

82.4 Solvents not gasoline (e.g., turpentine): None 1 2-5 6-10 > 10

82.5 Lawn/garden products:

(e.g., fertilizers, insecticides, herbicides)

None 1 2-5 6-10 > 10

82.6 Automobile batteries: None 1 2-5 6-10 > 10

82.7 Lubricant sprays (e.g., car care products,

WD40, silicone, engine degreasers):

None 1 2-5 6-10 > 10

82.8 Antifreeze, windshield wiper fluid, coolants: None 1 2-5 6-10 > 10

82.9 Adhesives (e.g., rubber cement, glue): None 1 2-5 6-10 > 10

82.10 Cleaning products:

(e.g., disinfectants, polishes, detergents)

None 1 2-5 6-10 > 10

83. Since baby was born, how often has anyone conducted small projects or hobbies

ANYWHERE

INSIDE your home that generated dust (e.g., sanding or grinding) or fumes (e.g., from

paints,

glues, solvents or finishes)?

Never Rarely Occasionally Regularly Don't know

CRAWL SPACE

84. Does your home have a crawl space? Yes No, go to Q85

84.1 Is the crawl space accessible to the

basement? Yes No

84.2 Is the crawl space damp? Yes No Don't know

COOLING AND HEATING

85. What type of AIR CONDITIONING does your residence have?

Don't have A/C, go to Q86 Central A/C Window, wall or portable unit(s)

85.1 How often do you use air conditioning in the summer?

Never

Rarely

Occasionally (average once or more per week)

Regularly (turned on most of the time with windows kept closed)

85.2 Do you have window, wall or portable A/C units? Yes No, go to Q86

85.2a If Yes, how many units do you have?

85.2b How many times do you change or clean the filter(s) per year in the window, wall or portable

A/C units? (If don't know, enter 99)

86. Does your home have a furnace? Yes No, go to Q88

86.1 What is the age of the furnace?

(If more than one furnace present, indicate age of the oldest working furnace)

≤ 10 years old > 10 years old Don't know

86.2 Where is the furnace located? (Check all that apply)

Main floor Basement Other

If Other, specify:

86.3 Is your home heated all or partly by forced air through ducted vents?

Yes No

86.4 What type of FILTER do you have on your furnace? Fibrous Electrostatic None

Pleated

Electronic (powered) Don’t know Other

86.4a If Other filter, specify:

86.5 How many times per year do you change or clean the furnace filter?

(If N/A, enter 88; If don't know, enter 99)

86.6 Is there a HUMIDIFIER on the furnace?

Yes No, go to Q87 Don't know

86.6a If Yes, do you use the humidifier on the

furnace?

Yes No Don't know

86.6b How many times per year do you clean it? (enter 99 if you don't know)

87. Do you have a heat recovery ventilation (HRV)

system?

Yes No Don't know

88. Does your home have a boiler?

Yes No, go to Q89 Don't know, go to Q89

88.1 Where is the boiler located? Main floor Basement Other

88.1a If Other location, specify:

88.2 What age is the boiler?

≤ 10 years old > 10 years old Don't know

89. What type of fuel heats your home? (Check all that apply)

Gas/Propane Wood/Pellet

Oil Other, specify below

Electric Don't know

89.1 If Other type of fuel, specify:

90. What fuel heats the HOT WATER supply (e.g., for showers, tap water, laundry, etc)?

Gas/propane Electric Solar Oil Wood Don't know

90.1 How old is this water heating system?

(if don't know, enter 999)

years

91. Indicate how often you use the following for heating your home:

91.1 Electric Baseboard:

Main source of heat Supplement regularly Supplement occasionally Not used

91.2 Radiators (portable or part of the boiler system):

Main source of heat Supplement regularly Supplement occasionally Not used

91.2a Radiator covers? Yes No N/A

91.2b Type of radiator covers: (Check all that apply)

Plastic Wood Metal N/A Other

If Other type of covers, specify:

91.3 Gas fireplace:

Main source of heat Supplement regularly Supplement occasionally Not used

91.4 Gas stove/oven (used for heating):

Main source of heat Supplement regularly Supplement occasionally Not used

91.5 Gas space heater:

Main source of heat Supplement regularly Supplement occasionally Not used

91.6 Electric space heater:

Main source of heat Supplement regularly Supplement occasionally Not used

91.7 Wood burning stove or fireplace:

Main source of heat Supplement regularly Supplement occasionally Not used

91.7a How many days supply of wood do you store INSIDE your home?

None < One week supply ≥ One week supply N/A

91.8 Pellet stove:

Main source of heat Supplement regularly Supplement occasionally Not used

91.9 If you use other heating sources, specify:

(e.g., radiant heating in floor, additional

furnace/heating coil)

91.9a How often is this other heating source used:

Main source of heat Supplement regularly Supplement occasionally Not used

92. In winter, do you regularly lower your temperature setting for extended blocks of time?

(e.g., at night or with a programmable thermostat)

Yes No

92.1 If Yes, indicate the temperature that you lower it to?

(If don't know, enter 99)

Degrees Celsius

93. Does your home or the building you live in

have a SWIMMING POOL, HOT TUB or SAUNA?

Yes No, go to Q94

(Include both indoor and outdoor)

93.1 Does your home or building have an outdoor

swimming pool?

Yes No, go to Q93.2

93.1a How often is baby near or in this outdoor

pool?

Once or more per week

Once or more per month

Less than monthly

Never

(Baby is "near" the pool if s/he is within 3 metres

or 10 feet)

93.1b What chemical is used for this outdoor

swimming pool?

Chlorine Bromine Salt Don't know

93.2 Does your home or building have an outdoor

hot tub?

Yes No, go to Q93.3

93.2a How often is baby near this outdoor hot tub? Once or more per week

Once or more per month

Less than monthly

Never

(Baby is "near" the hot tub if s/he is within 3 metres or 10 feet)

93.2b What chemical is used for this hot tub? Chlorine Bromine Salt Don't know

93.3 Does your home or building have an indoor

sauna?

Yes, in my home

Yes, located in my building

No

93.4 Does your home or building have an indoor

swimming pool?

Yes, in my home

Yes, located in my building

No, go to Q93.5

93.4a How often is baby near or in this indoor

pool?

Once or more per week

Once or more per month

Less than monthly

Never

(Baby is "near" the pool if s/he is within 3 metres

or 10 feet)

93.4b What chemical is used for this indoor swimming pool: Chlorine Bromine Salt

Don't know

93.5 Does your home or building have an indoor hot

tub?

Yes, in my home

Yes, located in my building

No, go to Q93.6

93.5a If Yes, how often is baby near this indoor

hot tub?

Once or more per week

Once or more per month

Less than monthly (Baby is "near" the hot tub if s/he is within 3 metres

or 10 feet)

93.5b What chemical is used for this indoor hot

tub?

Chlorine Bromine Salt Don't know

93.6 Do you store chemicals for use in ANY

swimming pool or hot tub?

Yes No Don't know N/A

93.6a If Yes, where do you store these chemicals? In my home In the garage

Check all that apply. In the shed Other

93.6b If Other, specify:

94. Did your baby spend time at a swimming pool

away from your home?

Yes No, go to Q95

If Yes, provide the names and locations of the (public and/or other private) pools. If the pools'

addresses are not known, please describe the locations (neighbourhood, nearby intersection, etc).

94.1 Name of pool:

94.2 Location:

94.3 This pool is: (Check all that apply) Indoor Outdoor

94.4 Name of additional pool:

94.5 Additional location:

94.6 This pool is: (Check all that apply) Indoor Outdoor

95. How many hours has your baby spent in or at any INDOOR swimming pools since birth? hours (Enter 00 if none; count total hours for pool at your home AND away from your home)

96. How many hours has your baby spent in or at any OUTDOOR swimming pools since birth? hours (Enter 00 if none; count total hours for pool at your home AND away from your home)

LAWN

97. Do you have a lawn? Yes No, go to Q98

97.1 Do you or your lawn service use pesticides, herbicides, garden sprays or weed killers on your lawn in summer?

Yes No

97.2 If Yes, number of applications per year:

GARAGE OR PARKING GARAGE

98. Do you live in an apartment building? Yes No, go to Q101

99. Does your building have a parking garage? Yes No, go to Q103

100. Do fumes from the parking garage come into

your apartment or into areas outside your apartment (e.g., interior hallways)?

Yes No, go to Q103

100.1 If Yes, how often do you smell fumes in the hallways just outside your apartment or in your apartment?

Never Occasionally

Rarely Regularly (most days)

Apartment dwellers, go to Q103

101. Does your home have an ATTACHED

GARAGE/CARPORT?

Yes No, go to Q103

101.1 Is it a garage (enclosed) or a carport? Garage Carport

101.2 Is there a door between the home and

garage/carport?

Yes No

101.3 How many cars and trucks are typically

parked in there?

101.3a How many other gasoline-powered vehicles

(e.g., motorcycles, mopeds, etc) are typically parked in there?

101.3b How many recreational vehicles are parked/stored in there?

101.4 Is the garage/carport used for vehicles in the winter?

Yes No

101.4a If Yes, how many days per week do one or more of these vehicles use the garage/carport in winter?

101.4b On an average winter day, how many times does each vehicle leave or return? (enter total number for all vehicles; e.g., 1 car leaves and returns = 02)

101.5 Is the garage/carport used for vehicles in

the summer? Yes No

101.5a If Yes, how many days per week do one or more of these vehicles use the garage/carport in summer?

101.5b On an average summer day, how many times does each vehicle leave or return? (enter total number for all vehicles; e.g., 1 car leaves and returns = 02)

102. Do you keep gasoline-powered equipment

(e.g., lawn mower, snow blower) in the garage/carport?

Yes No N/A

BABY'S ACTIVITIES

103. Has baby been exposed to tobacco smoke? Yes No, go to Q104

This includes all locations where the baby spent time (e.g., home, babysitter) but not encounters such as passing by smokers when entering a building.

103.1 On how many days in the last week was baby exposed to tobacco smoke? days (if exposed every day, answer=7)

103.2 On days when baby was exposed to tobacco

smoke, how long on average was s/he exposed? (Round up to nearest half hour)

104. Does your baby regularly go to a location away

from home (e.g. daycare, babysitter, activities with mom) for at least 1 hour per day on average, or at least 7 hours total in a week? Yes No, go to Q105

104.1 Do you feel comfortable providing a specific address for this location?

Yes No, go to Q104.3

104.2 What is the address of this location? House/building number:

Unit/Apt. Number:

Street name:

Street type (do not abbreviate):

(e.g., Street, Avenue, Crescent, etc.)

City:

Province (do not abbreviate):

Postal code:

104.3 When did your baby start going to this location on a regular basis?

104.4 While baby was at this location, what percentage of his/her time was spent outdoors? (Round answer to nearest 10%; enter 999 if don't know)

104.5 On average, how many days per week did

baby go to this location? days per week

104.6 On average, how many hours per week does your baby spend at this location?

hours per week

104.7 When the baby is taken to/from this location, how much time (in MINUTES) on average is spent in the following modes of transportation?

Walking/Biking

Car/Van/Taxi

Train/Subway

Bus

Other

(Enter 000 if no time spent)

104.8 If Other, describe mode of transportation:

105. Since birth, how much time (in HOURS) per day/night did your baby spend in the following

locations?

105.1 On a typical WEEKEND day: Home indoors (including sleeping)

Outdoors near home (in yard, local park,

walk/bike in neighbourhood)

On roadways (car/van, bus)

Other forms of transit (train, subway)

Other indoor places

Other outdoor places

(Hours must total 24; whole numbers only; enter 00 if no time spent)

105.2 On a typical WEEK day: Home indoors (including sleeping) Outdoors near home (in yard, local park, walk/bike in neighbourhood)

On roadways (car/van, bus)

Other forms of transit (train, subway)

Other indoor places

Other outdoor places

(Hours must total 24; whole numbers only; enter 00 if no time spent)

106. How did you complete this questionnaire? By myself With an interviewer With assistance from partner or other family member

107. What is TODAY'S DATE?