Sveriges lantbruksuniversitet
Swedish University of Agricultural Sciences

Faculty of Natural Resources and Agricultural Sciences

Perfluoroalkyl substances (PFASs), flame retardants and cyclic volatile methylsiloxanes in indoor air in Uppsala, Sweden

– occurrence and human exposure assessment

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Abstract

Indoor air samples were collected from three buildings located at the Swedish University of Agricultural Science (SLU) campus in Ultuna, Uppsala, Sweden and from residences of nine volunteers working in the three buildings. Sorbent-impregnated polyurethane foam (SIP) disk passive air samplers were deployed in different types of rooms in the three buildings including computer room (n = 1), labs (n = 3), lecture rooms (n = 3), offices (n = 8) and dining areas (n = 3) and homes (n = 9) of the volunteers between September and November in 2016. In addition, fingernail samples were collected from the nine volunteers. The samples were analyzed for three fluorotelomer alcohols (FTOHs), eight brominated flame retardants (BFRs), five organophosphorus flame retardants (OPFRs) and three cyclic volatile methyl siloxanes (cVMSs) to investigate their concentrations in indoor air, the influence of building and room types on the concentration level and human daily exposure dose (DED) via inhalation.

Average concentrations of ΣFTOHs, ΣBFRs, ΣOPFRs and ΣcVMSs in in-door air were 5100 pg m^{-3}, 110 pg m^{-3}, 430 pg m^{-3} and 1700 ng m^{-3}, respectively, and varied greatly both within each building and across buildings. The most abundant compounds were 8:2 FTOH for the FTOHs, decamethyl cyclopentasiloxane (D5) for the cVMSs, and 2,4,6-tribromophenol (2,4,6-TBP) for the FRs. Variations in the OPFRs composition were observed among different types of rooms. Home samples had a higher average concentration of Tris(2-chloroethyl) phosphate (TCEP), while office samples had higher average concentration of Tris(2-ethylhexyl) phosphate (TEHP) and tributyl phosphate (TNBP). Distribution of cVMSs and FTOHs followed a similar pattern in the three buildings that concentration in offices and dining areas was higher than in lecture rooms and labs, and significant correlation was found between the two compound groups in all air samples ($R = 0.51$, $p<0.05$). BFRs were found significantly correlated with the age of the buildings ($R = 0.60$, $p<0.05$) and with the number of electronic equipment ($R = 0.50$, $p<0.05$) at the sampling sites. Average inhalation DED of ΣFTOHs, ΣBFRs, ΣOPFRs and ΣcVMSs were 1200, 17, 94 and 340000 pg day^{-1} kg body weight $(BW)^{-1}$, respectively. Generally, the higher average concentration of the analytes in samples from the homes of the volunteers and longer exposure duration time at home resulted in 5 times on average higher DED at homes compared to offices. No correlation was observed between 2,4,6-TBP in fingernail samples and its DEDs, suggesting inhalation may be a less important pathway of human exposure to this compound. DEDs of all the four compound groups were much lower than the reference dose values.

Keywords: passive air sampling, SIPs, indoor air, PFAS, flame retardants, siloxanes, human exposure

Summary

Indoor air samples were collected using passive air samplers (PAS) from three buildings on the Ultuna campus area of the Swedish University of Agricultural Sciences (SLU), Uppsala, Sweden and from homes of nine volunteers working in the three building. Sampled rooms including one computer room, three laboratory rooms, three lecture rooms, eight offices, three dining areas and nine homes. The air samples were analyzed for three fluorotelomer alcohols (FTOHs), eight brominated flame retardants (BFRs), five organophosphorus flame retardants (OPFRs) and three cyclic volatile methyl siloxanes (cVMSs) to investigate their presence in indoor air, potential influencing factors and human daily exposure via inhalation (gaseous phase). Fingernail samples were also collected from these nine volunteers to estimate the level of the target chemicals in human body.

Concentrations of the four groups of target chemicals varied greatly both within each building and across buildings. Office and dining area air samples contained higher concentration of cVMSs and FTOHs on average than in lecture rooms and labs. The most abundant compounds were 8:2 FTOH for the FTOHs, decamethyl cyclopentasiloxane (D5) for the cVMSs, and 2,4,6-tribromophenol (2,4,6-TBP) for the FRs. Variations in the OPFRs composition were observed among different types of rooms. Home samples had a higher average concentration of Tris(2-chloroethyl) phosphate (TCEP), while office samples had higher average concentration of Tris(2-ethylhexyl) phosphate (TEHP) and tributyl phosphate (TNBP). Significant correlation ($p<0.05$) was found between cVMSs and FTOHs in all air samples. The concentrations of BFRs significantly correlated ($p<0.05$) with the age of the buildings and with the number of electronic equipment at the sampling sites.

Estimated daily exposure dose (DED) via inhalation of the four compound groups were around 3 to 3 orders of magnitudes lower than their references values. Generally, the higher average concentration of the target compounds in home samples and longer exposure duration time at home resulted in 5 times on average higher DED at homes compared to offices. No correlation was observed between 2,4,6-TBP in fingernail samples and its DEDs, suggesting inhalation may be a less important pathway of human exposure to this compound. DEDs of all the four compound groups were much lower than the reference dose values.

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Abbreviations

10:2 FTOH	10:2 fluorotelomer alcohol
2,4,6-TBP	2,4,6-tribromophenol
2,4-DBP	2,4-dibromophenol
2,6-DBP	2,6-dibromophenol
6:2 FTOH	6:2 fluorotelomer alcohol
8:2 FTOH	8:2 fluorotelomer alcohol
ANOVA	Analysis of variation
BDE-100	2,2',4,4',6-pentabromodiphenyl ether
BDE-153	2,2',4,4',5,5'-heptabromodiphenyl ether
BDE-183	2,2',3,4,4',5',6-heptabromodiphenyl ether
BDE-47	2,2',4,4'-tetrabromodiphenyl ether
BDE-99	2,2',4,4',5-pentabromodiphenyl ether
BFR	Brominated flame retardant
CR	Computer room
cVMS	Cyclic volatile methyl siloxane
D4	Octamethyl cyclotetrasilocane
D5	Decamethyl cyclopentasiloxane
D6	Dodecamethyl cyclohexasiloxane
DA	Dining area
DED	Daily exposure dose
EC	Eco-center
FOSA	Perfluorooctane sulfonamide
FOSAA	Perfluorooctane sulfonamidoacetic acid
FOSE	Perfluorooctane sulfonamidoethanol
FR	Flame retardant
FTOH	Fluorotelomer alcohol
H	Home
IS	Internal standards

LR	Lecture room
LRTP	Long-range transport potential
MDL	Method detection limit
MQL	Method quantification limit
MVM	Soil-Water-Environment center
O	Office
OPFR	Organophosphorus flame retardant
PAS	Passive air samplering
PBDE	Polybrominated diphenyl ethers
PBT	Persistence, bioaccumulation, toxicity
PFAS	Per- and polyfluoroalkyl substance
PFCA	Perfluoroalkyl carboxylate
PFOS	Perfluorooctanesulfonic acid
PFSA	Perfluoroalkane sulfonate
POP	Persistent organic pollutant
PUF	Polyurethane foam
RfD	Reference daily exposure dose
r_p	Pearson product-moment correlation coefficient
RS	Recovery standards
r_s	Spearman's rank correlation coefficient
S/N	Signal to noise ratio
SIP	Sorbent impregnated polyurethane foam
TCEP	tris(2-chloroethyl) phosphate
TCIPP	tri(1-chloro-2-propyl) phosphate
TEHP	tris(2-ethylhexyl) phosphate
TNBP	tributyl phosphate
TPeP	tripentyl phosphate
VHC	Veterinary Medicine and Animal Science Centre
vPvB	very persistent and very bioaccumulative
x:2 FTSA	x:2 fluorotelomer sulfonic acid

1 Introduction

A great variety of chemicals and materials, especially organic compounds, have been synthesized and used in consumer products to make people's life convenient and comfortable. While such a massive use has greatly improved our living standards, it also raises environmental and health concerns. In re-cent years, chemicals such as per- and polyfluoroalkyl substances (PFASs), flame retardants (FRs) and cyclic volatile methylsiloxanes (cVMSs) have increasingly attracted public attention due to their persistence and bioaccumulative behavior in the environment, and potential adverse effect on human health (Birnbaum & Staskal, 2004; Jensen & Leffers, 2008; Wang *et al.*, 2013). These chemicals have been widely used as additives in electrical equipment, construction materials and daily care products etc. Significantly higher levels of PFASs, FRs and cVMSs have been reported in indoor air than outdoor, indicating that indoor environments could be important sources for those compounds to the air and human exposure (Wilford *et al.*, 2004; Covaci *et al.*, 2011; Wang *et al.*, 2013; Hou *et al.*, 2016). Considering that people spend 90% of their times indoors (Klepeis *et al.*, 2001), investigating the levels of PFASs, FRs and cVMSs in indoor air is crucial for the assessment of human exposure to these chemicals and for health risk management.

In this project, indoor air samples were collected by passive air samplers (PAS) from three buildings on the Ultuna campus area of the Swedish University of Agricultural Sciences, Uppsala, Sweden and from homes of nine volunteers living in Uppsala, Sweden. The air samples were analyzed to determine the concentration of PFASs, FRs and cVMSs in indoor environments. Meanwhile, fingernail samples were collected from these nine volunteers to estimate the level of the target chemicals in human body. Finally, human exposure to PFASs, FRs and cVMSs were assessed based on the results from both air and fingernail samples.

1.1 Organic micropollutants

1.1.1 Per- and polyfluoroalkyl substances (PFASs)

Per- and polyfluoroalkyl substances (PFASs) is a group of anthropogenic chemicals (Lehmler, 2005) consisting of a fully fluorinated (per-FASs) or a partially fluorinated (poly-FASs) carbon chain and a functional group. Based on the functional group attached, PFASs could be categorized into different subgroups, for example perfluoroalkane sulfonates (PFSAs), perfluoroalkyl carboxylates (PFCAs), perfluorooctane sulfonamides (FOSAs), perfluorooctane sulfonamidoethanols (FOSEs), perfluorooctane sulfonamidoacetic acids (FOSAAs), fluorotelomer alcohol (FTOHs) and x:2 fluorotelomer sulfonic acid (x:2 FTSAs). Because of their unique water-repellent and grease-repellent properties, PFASs are widely used in metal plating, fire-fighting foam products as well as in surface treatment to provide water and oil resistance for paper, furniture, carpet etc. (OECD, 2002, 2006).

PFASs could bioaccumulate in the environment and are very persistent to natural degradation process (Jensen & Leffers, 2008) They can undergo long range transport in the atmosphere and oceans (Ahrens *et al.*, 2011). As a result, trace levels of PFASs have been detected globally in water, in the atmosphere, as well as in wildlife and in human blood samples (Giesy & Kannan, 2001; Barber *et al.*, 2007; Calafat *et al.*, 2007; Ahrens *et al.*, 2010; Goosey & Harrad, 2012). Toxicity studies indicate that human exposure to PFASs may lead to endocrine disruption, immune-related problems and increased carcinogenic risk (Jensen & Leffers, 2008; Chang *et al.*, 2014, 2016). According to the environmental and health concerns, perfluorooctanesulfonic acid (PFOS) was prohibited in EU in 2008 and listed as a persistent organic pollutant (POP) in Annex B under the Stockholm Convention in 2009 (Stockholm Convention, 2009; Vierke *et al.*, 2012). Another PFAS, perfluorooctanoic acid (PFOA), was proposed to for listing under the convention (UNEP, 2015).

Waste water treatment plants (WWTPs) and landfills have acted as important environmental pathways of PFASs to the atmosphere (Ahrens *et al.*, 2011). The high levels of PFASs in indoor air makes the indoor microenvironments possible diffusive sources of PFASs to the outdoor air (Barber *et al.*, 2007; Langer *et al.*, 2010). Percentage of home carpeting and age of the residence seem to be factors that affect the levels of PFASs in indoor environments (Gewurtz *et al.*, 2009). The variety of composition of PFASs across the world suggest a difference in PFASs use patterns (Goosey & Harrad, 2011). However, several studies found that FTOHs are the predominant class in most of the indoor air samples, though which

kind of FTOHs that dominated may vary (Barber *et al.*, 2007; Langer *et al.*, 2010; Haug *et al.*, 2011; Huber *et al.*, 2011; Shoeib *et al.*, 2011; Goosey & Harrad, 2012). Levels of FTOHs in indoor air from a few recent studies are presented in Table 1.

Tabel 1. *Concentration range (mean) of 6:2 FTOH, 8:2 FTOH and 10:2 FTOH in indoor air from recent studies (pg m^{-3})*

6:2 FTOH	8:2 FTOH	10:2 FTOH	Location	Reference
(3000)	(3400)	(3600)	Norway	Barber et al., 2007
63 – 9400 (1500)	920 – 25000 (6400)	380 – 29000 (4100)	Norway	Haug et al., 2011
100 – 37000	1100 – 209000	100 – 54000	Germany	Langer et al., 2010
n.d. – 23000 (2400)	660 – 16000 (3800)	220 – 8200 (1400)	Canada	Shoeib et al., 2011

n.d. – not detected.

1.1.2 Flame retardants (FRs)

Flame retardants (FRs) are a diverse group of industrially produced organic compounds applied to a wide range of commercial products, such as furniture, carpets, construction materials and electronics, to provide fire protection (Papachlimitzou *et al.*, 2012). Brominated flame retardants (BFRs) are one of the most used FRs due to their low cost and good performance (Birnbaum & Staskal, 2004). Although BFRs have played an important role in reducing fire risk and saving lives, great concern has been raised because of their ubiquitous presence in the environment, bioaccumulation potential and possible adverse effects on wildlife and human health (de Wit, 2002; Birnbaum & Staskal, 2004). In fact, three kinds of the most widely used BFRs, hexabromocyclododecane (HBCD), octabromodiphenyl ether (octa-BDE) and pentabromodiphenyl ether (penta-BDE), have been listed as persistent organic pollutants (POPs) under Stockholm Convention Annex A, since they fulfil the persistence, bioaccumulation, toxicity (PBT) and long-range transport potential (LRTP) criteria of POPs. Another commercial mixture of polybrominated diphenyl ethers (PBDEs), decabromodiphenyl ether (deca-BDE), was recently proposed to be included in Annex A of the Stock-holm Convention (UNEP, 2013).

The ban on production and use of PBDEs and HBCD have led to a shift towards use of alternative FRs (AFRs) such as organophosphorus FRs (OPFRs), to meet market demands (Birnbaum & Staskal, 2004). Chlorinated alkyl organophosphates and aryl phosphates are widely used in plastics, textiles, electronic equipment and furniture as FRs, while non-chlorinated alkyl phosphates are mainly used

as plasticizers (Reemtsma *et al.*, 2008). For example, tris(2-chloroethyl) phosphate (TCEP) and tris (1-chloro-2-propyl) phosphate (TCIPP) are usually added to polyurethane foams as substitutes for penta-BDE (Cequier *et al.*, 2014), and tri-n-butyl phosphate (TNBP) is used as a primary plasticizer in the manufacture of plastics and vinyl resin (WHO, 1991). Though OPFRs such as TCEP usually do not meet the PBT criteria, it is recognized that many OPFRs are carcinogenic, highly toxic to organisms and environmentally persistent (Reemtsma *et al.*, 2008).

Most BFRs and OPFRs are used as additives in production of materials and commercial products, and may over time slowly be released from the material. For example, 2,4,6-tribromophenol (2,4,6-TBP) is usually used as a reactive flame retardant intermediate for brominated epoxy resin or as wood preservative, and both 2,4,6-TBP treated wood and plastics containing BFRs derived from 2,4,6-TBP are possible sources for its occurrence in indoor environments (WHO, 2005).

A number of studies have observed high concentrations of FRs in indoor environments (Wilford *et al.*, 2004; Saito *et al.*, 2007; Abdallah *et al.*, 2008; Toms *et al.*, 2009; Bergh *et al.*, 2011b; Cequier *et al.*, 2014), indicating in-door air as a source of FRs to the environment and an important pathway for hu-man exposure (Table 2 and Table 3).

Tabel 2. *Concentration range (mean) of OPFRs in indoor air from recent studies (pg m^{-3})*

TCEP	TCIPP	TNBP	TPHP	Location	Type	Reference
(3200)	(83000)	(9700)	(380)	Norway	Home	Cequier *et al.*, 2014
(7000)	(12000)	(3100)	(79)	Norway	Classroom	
n.d.– 86 (10000)	<0.5 – 1200000 (59000)	n.a.	n.a.	Sweden	Home	Bergh *et al.*, 2011a
n.d. - 28000 (8.3)	2400 – 64000 (15000)	n.a.	n.a.	Sweden	Home	
7800 – 230000 (47000)	1300 – 72000 (19000)	n.a.	n.a.	Sweden	Day care	Bergh *et al.*, 2011b
n.d. – 100000 (21000)	16000 – 240000 (110000)	n.a.	n.a.	Sweden	Work	

n.d. – not detected.
n.a. – not analyzed.

Tabel 3. *Concentration range (mean) of BFRs in indoor air from recent studies (pg m^{-3})*

BDE-47	BDE-99	BDE-100	BDE-153	2,4,6-TBP	Location	Type	Reference
n.d. - 1600 (160)	n.d. - 890 (42)	n.d. - 160 (10)	n.d. – 74 (1.6)	n.a.	Canada	Home	Wilford *et al.*, 2004
58 – 7100 (1669)	9 – 6500 (852)	4.1 – 1500 (217)	n.d. – 180 (22)	n.a.	UK	Office	Harrad *et al.*, 2004
45 – 1300 (424)	8.7 – 210 (70)	2.6 – 82 (27)	n.d. – 6.1 (1.9)	n.a.	UK	Home	
n.d. – 1700	n.d. – 3200	n.d. – 1000	n.a.	n.d. – 6800	Japan	Home	Saito *et al.*, 2007
n.d. – 1400	n.a.	n.a.	n.a.	n.d. – 2800	Japan	Office	
4.1 - 280 (55)	n.d. - 53 (33)	n.d. - 7.7 (1.3)	n.d.	n.a.	Australia	Home	Toms *et al.*, 2009
(180)	(41)	(11)	(7.6)	n.a.	Norway	Home	Cequier *et al.*, 2014
(180)	(26)	(9.1)	(0.74)	n.a.	Norway	Class room	

n.d. – not detected.
n.a. – not analyzed.

1.1.3 Cyclic volatile methylsiloxanes (cVMSs)

Cyclic volatile methylsiloxanes (cVMS) are a group of organosilicon compounds that have a ring structure consisting of alternating silicon-oxygen bonds (Si-O), with each silicon atom bearing two methyl groups. cVMSs has been widely used in the production of silicone polymers, in coatings (e.g. paints, varnishes, lacquers and furniture polishes etc.) as well as in personal care products (e.g. cosmetic, shampoo, etc.) (Lu *et al.*, 2010; Wang *et al.*, 2013). The three mostly used cVMSs are octamethylcyclotera siloxane (D4), decamethylcyclopenta siloxane (D5) and dodecamethylcyclohexa siloxane (D6). In 2004, approximately 9500, 19000 and 2000 tons of D4, D5 and D6 were used in the European Union for the production of silicone polymers and personal care products (Wang *et al.*, 2013).

Due to the very low water solubility and high vapor pressure, cVMSs favor to partition to air, while their relatively long half-life time in air makes it possible for them to undergo long range atmospheric transport and therefore distribute regionally and globally (Wang *et al.*, 2013). A global passive air sampling study has reported the ubiquitous presence of cVMSs in the atmosphere, even in remote regions such as the Arctic (Genualdi *et al.*, 2011).

Currently, there is no restriction on the use of cVMSs, however several regulatory jurisdictions have prioritized D4, D5 and D6 due to their persistent and bioaccumulative potential (Wang *et al.*, 2013; Gobas *et al.*, 2015). A risk assessment of cVMSs conducted by the UK Environment Agency has classified D4 as very persistent and very bioaccumulative (vPvB) and persistent, bioaccumulative and toxic (PBT), and D5 as vPvB (Brooke *et al.*, 2009b, a; c). Mammalian toxicity studies indicate that exposure to D4 could lead to impaired fertility, estrogen mimicry and liver damage (McKim *et al.*, 2001; Meeks *et al.*, 2007; Quinn *et al.*, 2007b; a; Siddiqui *et al.*, 2007) and long-term inhalation exposure to D5 may have potential carcinogenic effect (US EPA, 2009). German Working Group on Indoor Guidelines of the Federal Environment Agency and the States' Supreme Health Authorities suggested a health hazard guide value of 4 mg m^{-3} and a health precaution guide value of 0.4 mg m^{-3} for the sum of cVMSs (D3 to D6) in indoor air (German Working Group on Indoor Guidelines, 2011).

In a recent review of the occurrence and fate of cVMSs (Wang *et al.*, 2013), landfill gas and sewage gas were proved to be important sources of cVMSs in outdoor air, while personal care products were major sources of indoor air. Horii and Kannan (2008) reported cVMSs in 76 consumer products sampled in Albany, NY. Capela *et al*. (2016) estimated that the amount of total cVMSs released from personal care products to air was on average 1607 μg day^{-1} and was predominated by D5 and D6. Tang *et al*. (2015) found D5 to be the dominant volatile organic compounds from direct human emission, which is associated with use of personal care products. Siloxane concentrations in dust have also been found to be correlated with the number of occu-pants, number of electrical/electronic appliances and smokers living in the house (Lu *et al.*, 2010).

A few studies have focused on the cVMSs in indoor air; Yucuis *et al*. (2013) sampled both outdoor and indoor air in Chicago and found the levels of cVMSs in indoor air to be significantly higher than in outdoor air; Pieri *et al*. (2013) measured cVMSs in different indoor environments in UK and Italy and observed differences between the two countries and various types of rooms; Tran and Kannan (2015) determined the concentration of siloxanes in 60 indoor air samples in USA; Meng and Wu (2015) examined cVMSs in household and automobile settings and found concentrations in rooms that being renovating/redecorating to be slightly higher than in ordinary rooms. A summary of recent studies focusing on cVMSs in indoor air is presented in Table 4.

Table 4. *Concentration range of D4, D5 and D6 in indoor air from recent studies (ng m^{-3})*

D4	D5	D6	Location	Reference
23 – 500	970 – 56000	<59 – 2800	USA	Yucuis *et al*. 2013
0.06 – 720	6.4 – 3700	3.1 – 890	USA	Tran and Kannan 2015
n.d. – 73000	n.d. – 510000	n.d. – 180000	Italy	Pieri *et al*. 2013
n.d. – 270000	2400 – 440 ×10^3	40 – 79000	UK	

n.d. – not detected.

1.2 Passive air sampling

Passive air sampling (PAS) is an easy and low cost method for time-integrated sampling (Bohlin *et al.*, 2007). Compared to active air samplers, PAS does not require a pump during the sampling period, which means it is noise-free and easy to handle. These advantages make PAS much less intrusive when deployed in indoor environments (e.g. office and home) and therefore an ideal technique for indoor air sampling. Once deployed, PAS can gradually capture airborne pollutants via gaseous diffusion and sorption (Bohlin *et al.*, 2007).

Polyurethane foam (PUF) is a world-widely used PAS for collecting time integrated samples of airborne persistent organic pollutants (POPs) in both outdoor and indoor environments (Abdallah *et al.*, 2008; Pozo *et al.*, 2009; Shoeib *et al.*, 2011). However, one disadvantage of PUF is its relatively low sorptive capacity. Sorbent impregnated PUF (SIP) is a new type of PAS developed by Shoeib *et al.* (2008). By coating powdered XAD on PUF disks, SIPs have shown a significant greater capacity and longer linear uptake phase than PUFs. Several studies indicate that SIPs have a linear uptake phase of 30-90 days for PFASs (Ahrens *et al.*, 2013), 9-24 days for different type of VMSs (Ahrens *et al.*, 2014) and more than 49 days for most BFRs (Saini *et al.*, 2015).

PAS like SIP and PUF mainly captures gas phase pollutants. However, it could also collect a small portion of particles since fine particles behave like gas-phase chemicals and could enter the sampling chamber. It is estimated that about 10% of the ambient particles was sampled by PUF disk when housed in a fully sheltered double-doom chamber (Klánová *et al.*, 2008).

Several studies on SIP and PUF uptake characteristics of various chemicals in outdoor environment have shown similar uptake rates (R-values) (Shoeib & Harner, 2002; Wilford *et al.*, 2004; Ahrens *et al.*, 2013, 2014; Liu *et al.*, 2016) as classical POPs (4 m^3 d^{-1}) (Pozo *et al.*, 2009), indicating a similar uptake rate. For indoor environments, where the air condition is more static, the R-value can be affected by the type of housing chamber used (Figure 1). Wilford *et al.* (2004)

housed indoor samplers with a tripod support that allows air to flow through all directions and obtained a R-value of 2.5 m^3 d^{-1}, while Bohlin *et al.* (2014) housed SIP disks with a fully sheltered double-dome chamber only obtained a R-value of 1.4 m^3 d^{-1}. Saini *et al*. (2015) compared the partially sheltered chamber and fully sheltered chamber and recommended a R-value of 3.5 ± 0.9 m^3 d^{-1} for partially sheltered ones and 1.0 ± 0.4 m^3 d^{-1} for fully sheltered housing chambers. A short summary of R-values from recent studies is presented in Table 5.

Table 5. *A summary of R-value (m^3 d^{-1}) for PFASs, BFRs, OPFRs and cVMSs from recent studies*

Compounds	R-value	Type	Chamber design	Reference
BFRs	2.5	indoor	partially sheltered	Wilford *et al.*, 2004
BFRs	1.12-1.95	indoor	fully sheltered	Hazrati & Harrad, 2007
BFRs	1.4	indoor	fully sheltered	Bohlin *et al.*, 2014
BFRs	1.0 ±0.4 3.5 ±0.9	indoor	fully sheltered partially sheltered	Saini et al., 2015
PFASs	1.4 - 4.6	outdoor	fully sheltered	Shoeib *et al.*, 2008
PFASs	3.5	outdoor	fully sheltered	Ahrens *et al.*, 2013
cVMSs	3.3 - 5.7	outdoor	fully sheltered	Ahrens *et al.*, 2014
BFRs	3.8	outdoor	fully sheltered	Pozo *et al*., 2009
PFASs	4	outdoor	fully sheltered	Genualdi *et al*., 2010
OPFRs	3.5 ±1.7	outdoor	fully sheltered	Liu *et al.*, 2016

Figure 1. Housing chamber for passive air sampler.

2 Materials and Methods

2.1 Chemicals

A complete list of target analytes used for indoor air and fingernail samples is presented in Table 6. The list includes three PFASs, three cVMSs, eight BFRs and five OPFRs. In addition, 26 PFASs were analyzed in fingernail samples (Table 7). All solvents used were of the highest purity available. A complete description of chemicals used is given in the appendix (Table A1 and A2) together with information on their purity and manufacturer.

Table 6. *Target FTOHs, cVMSs and FRs analyzed in indoor air and fingernail samples*

Abbreviation	Name	Category
6:2 FTOH	6:2 fluorotelomer alcohol	Fluorotelomer alcohols (FTOHs)
8:2 FTOH	8:2 fluorotelomer alcohol	
10:2 FTOH	10:2 fluorotelomer alcohol	
2,4-DBP	2,4-dibromophenol	Brominated flame retardants (BFRs)
2,6-DBP	2,6-dibromophenol	
2,4,6-TBP	2,4,6-tribromophenol	
BDE-47	2,2',4,4'-tetrabromodiphenyl ether	
BDE-99	2,2',4,4',5-pentabromodiphenyl ether	
BDE-100	2,2',4,4',6-pentabromodiphenyl ether	
BDE-153	2,2',4,4',5,5'-heptabromodiphenyl ether	
BDE-183	2,2',3,4,4',5',6-heptabromodiphenyl ether	
TCEP	tris(2-chloroethyl) phosphate	Organophosphorus flame retardants (OPFRs)
TCIPP	tri(1-chloro-2-propyl) phosphate	
TEHP	tris(2-ethylhexyl) phosphate	
TNBP	tributyl phosphate	
TPeP	tripentyl phosphate	

Abbreviation	Name	Category
D4	Octamethylcyclotera silocane	Cyclic volatile methylsiloxanes (cVMSs)
D5	Decamethylcyclopenta siloxane	
D6	Dodecamethylcyclohexa siloxane	

Table 7. *PFASs analyzed in fingernail samples*

Abbreviation	Name	Category
PFBS	perfluorobutane sulfonate	perfluoroalkane sulfonates (PFSAs)
PFHxS	perfluorohexane sulfonate	
PFOS	perfluorooctane sulfonate	
PFDS	perfluorodecane sulfonate	
PFBA	perfluorobutanoate	perfluoroalkyl carboxylates (PFCAs)
PFPeA	perfluoropentanoate	
PFHxA	perfluorohexanoate	
PFHpA	perfluoroheptanoate	
PFOA	perfluorooctanoate	
PFNA	perfluorononanoate	
PFDA	perfluorodecanoate	
PFUnDA	perfluoroundecanoate	
PFDoDA	perfluorododecanoate	
PFTriDA	perfluorotridecanoate	
PFTeDA	perfluorotetradecanoate	
PFHxDA	perfluorohexadecanoate	
PFOcDA	perfluorooctadecanoate	
FOSA	perfluorooctanesulfonamide	perfluorooctane sulfonamides
N-MeFOSA	N-methylperfluorooctansulfonamide	
N-EtFOSA	N-ethylperfluorooctanesulfonamide	
N-MeFOSE	N-methylperfluorooctanesulfonamido-ethanol	perfluorooctane sulfonamidoeth-anols
N-EtFOSE	N-ethylperfluorooctanesulfonamido-ethanol	
FOSAA	perfluorooctanesulfonamidoacetic acid	perfluorooctane sulfonamidoacetic acids
N-MeFOSAA	N-methylperfluorooctanesulfonamidoacetic acid	
N-EtFOSAA	N-ethylperfluorooctanesulfonamidoacetic acid	
6:2 FTSA	6:2 fluorotelomer sulfonate	x:2 fluorotelomer carboxylates

2.2 Sampling

2.2.1 Preparation of SIPs

The preparation of SIP disks was based on the protocol developed by Shoeib *et al*. (2008). Firstly, PUF disks (14 cm diameter × 1.27 cm thick, surface area 364 cm^2, volume 195 cm^3, Tisch Environmental, Cleves, OH, USA) were washed with clean tap water and then placed on top of an acetone-rinsed aluminium foil in the fume hood to dry for 4-6 hours. The PUF disks were further cleaned by Soxhlet extraction for 24 hours using acetone (400 mL), followed by 6 hours using petroleum ether (400 mL), and then another 18 hours using fresh petroleum ether (400 mL). After the last extraction, the PUF disks were dried in a vacuum desiccator for 48 hours.

Secondly, XAD-4 resin (11.4 g for ten SIPs, Supelco, Bellefonte, PA) was finely ground using Precellys® Evolution (Bertin Technologies, France) and then transferred into a cellulose thimble. The thimble with XAD-4 resin was cleaned by Soxhlet extraction for 6-8 hours using methanol (400 mL), followed by 16-18 hours using dichloromethane (400 mL), and then 6-8 hours using n-hexane (400 mL).

Finally, the clean ground XAD-4 was transferred into an acetone-rinsed glass beaker with n-hexane (1700 mL) to form slurry (6.4 g/L). The slurry was sonicated for 30 minutes and then poured into a crystallizing dish and stirred with a magnetic bar to keep XAD-4 suspended. The PUF disk was dipped in the slurry (30 seconds) and the dipping was repeated 3 times for each PUF disk. With each successive dip, the solvent was allowed to evaporate from each side of the disk to ensure a uniform coating (~0.5 g XAD-4 per disk). The disk was then placed on an acetone-rinsed and pre-heated aluminium foil (30 ℃ - 40 ℃) for 5 minutes and then transferred into a vacuum desiccator for drying.

2.2.2 Sampling

The stainless steel sampler housing chambers (Tisch Environmental, Cleves, OH, USA, Figure 1) were pre-cleaned with water and then rinsed with acetone on both inside and outside. SIP disks were individually housed on the supporting ring in the chamber during the sampling period.The passive samplers were deployed for 14 days between September and November 2016 in three buildings located on the Ultuna campus of the Swedish University of Agricultural Sciences, Uppsala, Sweden (n = 18) and in residences of nine volunteers living in Uppsala, Sweden (n = 9, 5 males and 4 females) (Table 8 and 9). The three buildings on campus were Soil-

Water-Environment center (MVM), Veterinary Medicine and Animal Science Centre (VHC) and Eco-center (EC), which are 5 years, 2 years and approximately 40 years old, respectively. Eco-centrum was renovated in 2008. All three buildings have forced ventilation systems.

Field blanks (n = 2) were collected by exposing the SIPs for 1 minute in the housing chamber at the sampling sites, and they were then treated like real samples. Duplicate samples (n = 3) were collected in the computer room at MVM, the lecture room at VHC and the dining area at EC.

To identify potential factors that can affect the levels of target compounds in indoor air, the nine volunteers filled out questionnaires about the characteristics of the sampling sites. Besides passive air samples, fingernails of the nine participants were collected on the last sampling day and analyzed together with the air samples in order to provide information about human internal levels of target compounds.

Table 8. *Passive air sampling at Ultuna Campus, SLU, Uppsala, Sweden*

Sample Code	Type of room	Building	Start date	End date	Notes
LR1	Lecture room	MVM[a]	2016/9/21	2016/10/5	
CR	Computer room	MVM	2016/9/21	2016/10/5	
CR	Computer room	MVM	2016/9/21	2016/10/5	Duplicate sample
Lab1	Lab	MVM	2016/9/21	2016/10/5	
DA1	Dining Area	MVM	2016/10/31	2016/11/14	Open area
O1	Office	MVM	2016/9/21	2016/10/5	4 persons
O2	Office	MVM	2016/9/22	2016/10/6	1 person
O3	Office	MVM	2016/9/22	2016/10/6	3 person
LR2	Lecture room	VHC[b]	2016/10/10	2016/10/24	
LR2	Lecture room	VHC	2016/10/10	2016/10/24	Duplicate sample
Lab2	Lab	VHC	2016/10/10	2016/10/24	
DA2	Dining Area	VHC	2016/10/10	2016/10/24	Open area
O4	Office	VHC	2016/10/10	2016/10/24	1 person
O5	Office	VHC	2016/10/10	2016/10/24	2 persons
O6	Office	VHC	2016/10/10	2016/10/24	2 persons
DA3	Dining Area	EC[c]	2016/10/26	2016/11/9	Open area
DA3	Dining Area	EC	2016/10/26	2016/11/9	Duplicate sample
Lab3	Lab	EC	2016/10/26	2016/11/9	
LR3	Lecture room	EC	2016/10/27	2016/11/10	
O7	Office	EC	2016/10/26	2016/11/9	2 persons
O8	Office	EC	2016/10/28	2016/11/11	1 person

a. MVM – Soil-Water-Environment center.
b. VHC – Veterinary Medicine and Animal Science Centre.
c. EC – Eco-center.

Table 9. *Volunteers, indoor air samples from their homes and offices, and fingernail samples*

Volunteer	Gender	Building works at	Office sample	Home sample	Fingernail sample	Weight of Fingernail sample (g)
Volunteer 1	male	MVM[a]	O1	H1	FN1	0.09588
Volunteer 2	male	MVM	O2	H2	FN2	0.12327
Volunteer 3	male	MVM	O3	H3	FN3	0.09415
Volunteer 4	male	VHC[b]	O4	H4	FN4	0.08324
Volunteer 5	female	VHC	O5	H5	FN5	0.05092
Volunteer 6	female	VHC	O6	H6	FN6	0.07104
Volunteer 7	female	EC[c]	O7	H7	FN7	0.06670
Volunteer 8	female	EC	O7	H8	FN8	0.08648
Volunteer 9	male	EC	O8	H9	FN9	0.07651

a. MVM – Soil-Water-Environment center.
b. VHC – Veterinary Medicine and Animal Science Centre.
c. EC – Eco-center

2.3 Extraction and clean up

2.3.1 Passive air samplers

Cellulose thimbles were pre-cleaned by Soxhlet extraction for 6 hours using methanol (350 mL), followed by 18 hours using acetone/petroleum ether (1:1, v/v). SIP disk samples were individually placed in the clean cellulose thimbles, spiked with 50 µL internal standards (IS) representative for each substance group analyzed (c = 500, 200 and 5000 pg μL^{-1} for PFASs, FRs and cVMSs, respectively). A complete list of internal standards used together with information of the amount added can be found in the appendix (Table A15). The samples were Soxhlet extracted for 6 hours using petroleum ether/acetone (350 mL, 85:15, v/v). After adding iso-octane (5 mL) as a keeper, the extracts were concentrated to approximately 0.5 mL using an automated evaporation system (TurboVap® II, Biotage, Sweden). Then the extracts were cleaned up on an anhydrous sodium sulfate column to remove any moisture. The analytes were eluted with approximate 10 mL iso-octane and then concentrated under a gentle stream of nitrogen gas to 0.5 mL. Prior to injection, recovery standards (RS) for each substance group analyzed were added (10 µL, c = 1000 pg μL^{-1}) (Table A15).

2.3.2 Fingernials

The fingernail samples (n = 9), were first ground to fine particles and internal standards (IS) representative for each substance group (i.e. PFASs, cVMSs, BFRs

and OPFRs) were added (50 μL, c = 500, 200 and 5000 pg μL^{-1} for PFASs, FRs and cVMSs, respectively, and 100 μL, c = 50 pg μL^{-1} for the additional 26 PFASs). A complete list of internal standards used, together with information of the amount added can be found in the appendix (Table A16). The samples were extracted with dichloromethane (4 mL) under sonication (15 min) and then the extracts were separated from the fingernails by centrifugation (4000 rpm, 5 min). The extraction process was repeated three times and the extract from each time was pooled (in total 12 mL). After reducing the volume to 1 mL by nitrogen evaporation, the extract was split into two portions (0.5 mL each) and the solvent was changed to methanol (for HPLC-MS) and iso-octane (for GC-MS), respectively. Both portions were concentrated to a final volume of 0.5 mL by nitrogen evaporation, and recovery standards (RS) were added before injection. A complete list of recovery standards used together with information of the amount added can be found in the appendix (Table A16). Blanks (n = 2) followed exactly the same procedures, but without fingernails.

2.4 Instrumental analysis

Based on the methods described by Ahrens *et al*. (2013) and Companioni-Damas *et al*. (2012), PFASs and cVMSs were analyzed by gas chromatography coupled with mass spectrometry (Agilent 7890 B Single Quad 5977A MSD; Agilent Technologies, Palo Alto, CA, USA) (GC-MS) using single ion monitoring (SIM) with positive chemical ionization and electron ionization, respectively. FRs were analyzed according Gustavsson *et al.* (2017) using gas chromatography coupled with tandem mass spectrometry (Agilent GC–MS 7890A Triple Quad 7010; Agilent Technologies, Palo Alto, CA, USA) (GC-MS/MS) in multiple reaction monitoring (MRM) mode using electron ionization. The fingernail samples were analyzed for additional 26 PFASs using high-performance liquid chromatography (Agilent 1200; Agilent Technologies, Palo Alto, CA, USA) coupled to tandem mass spectrometry (6460 Triple Quad (-)ESI-MS/MS; Agilent Technologies, Palo Alto, CA, USA) (HPLC-MS/MS) as described by Ahrens et al (2016). More details of the methods can be found in the appendix.

2.5 Quality assurance and statistical analysis

In order to examine the reproducibility, three pairs of duplicate passive air samples were collected at the dining area (DA3), in the computer room (CR) and the lecture room (LR2) during the deployment at EC, MVM and VHC, respectively. The

mean value of each pair of duplicate samples was used as the final results of the three rooms (DA3, CR and LR2).

All analytes were quantified by the isotope dilution method. The criteria of a positive identification for a peak were i) retention time within ± 0.5 minute of that of the reference compound in the calibration solution; ii) signal to noise (S/N) ratio had to be >3; and iii) the quantifier/qualifier ratio had to be within ± 30% of the ratio in the calibration standard.

The method detection limit (MDL) and method quantification limit (MQL) were calculated using the following equations:

$$\mathrm{MDL} = \mathrm{MEAN}_{\mathrm{Blanks}} + 3\times\mathrm{SD}_{\mathrm{Blanks}} \quad (1)$$

$$\mathrm{MQL} = \mathrm{MEAN}_{\mathrm{Blanks}} + 10\times\mathrm{SD}_{\mathrm{Blanks}} \quad (2)$$

For air samples, both blanks (n = 4) and field blanks (n = 2) were used for the calculation, while for fingernail samples the two fingernail blanks were used. Values less than MDL were marked as n.d. (not detected) and were replaced by half of the MDL during statistical analysis.

Kolmogorov-Smirnov test was employed to check if the results of analytes were normally distributed. For the normally distributed data sets, Pearson product-moment correlation coefficient (r_p) was employed to examine the associations between different analytes. Spearman's rank correlation coefficient (r_s) was employed to investigate the relationships between non-normally distributed data sets, between fingernail samples and air samples, and between targeting compounds and potential factors that affect their presence in indoor environment, including age of building, number of electronical equipment, number of people working in the office, number of outdoor wear, volume of the sampling room, renovation, airing frequency and ventilation. In addition, analysis of variation (ANOVA) and cluster analysis were used for identifying potential influencing factors.

2.6 Human exposure assessment

Human daily intake of the targeting compounds in gas phase via inhalation was calculated according to USEPA risk assessment guidance (US EPA, 1989):

$$\mathrm{DED} = \frac{\mathrm{IR}\times\mathrm{C}\times\mathrm{ED}}{\mathrm{BW}} \quad (3)$$

where DED is the daily exposure dose (pg day^{-1} kg BW^{-1}); IR is the inhalation rate, which is assumed to be 13.3 m^3 day^{-1} for an adult (US EPA, 1989); C is the concentration of the compound in air (pg m^{-3}); ED is the exposure duration (hours

per day); BW is bodyweight (kg), which in this case is assumed to be 70 kg for adults.

2.7 Theory on passive air sampling

The uptake of a compound by PAS over time consists of three phases. At the beginning of sampling, there is only a small amount of the compound in the sampler media and the uptake is linear; as the PAS continues absorbing the compound, the uptake enters a curvilinear phase; and finally, when the PAS has been saturated, the uptake reaches equilibrium (Figure 2).

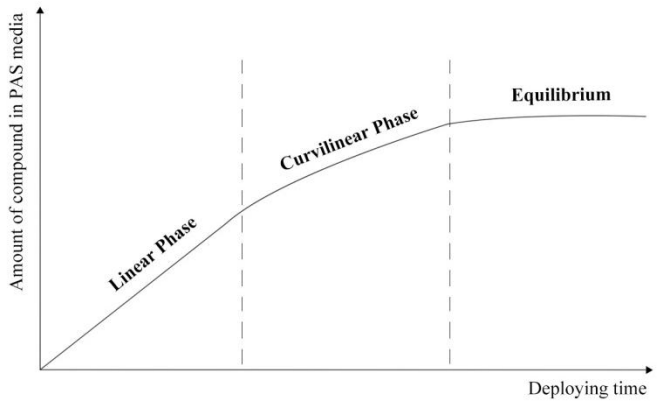

Figure 2. Three phases of uptake of a certain compound by PAS (Bohlin *et al.*, 2007)

The concentration of target compounds in air can be calculated using the following equation:

$$c_A = \frac{m_{SIP}}{V_{AIR}} \quad (4)$$

where c_A is the concentration in air (ng m^{-3}), m_{SIP} is the amount of target compound in SIP (ng SIP^{-1}). V_{AIR} is the air volume sampled during the deployment period, which can be derived using equation 4 according to Ahrens *et al.* (2013, 2014):

$$V_{AIR} = K_{SIP-A} \times V_{SIP} \times \left(1 - exp\left[-\left(\frac{A_{SIP}}{V_{SIP}} \times \frac{k_A}{K_{SIP-A}}\right) \times t\right]\right) \quad (5)$$

where $K_{SIP\text{-}A}$ is the SIP-air partition coefficient, V_{SIP} is the volume of the SIP disk (m^3), A_{SIP} is the planar area of SIP disk (m^2), k_A is the air side mass-transfer coefficient (m d^{-1}) and t is the sampling duration (d).

For samplers where the deployment duration is within the linear phase, equation 4 could be simplified as:

$$V_{Air} = R \times t \quad (6)$$

where R stands for the air sampling rate ($m^3\ d^{-1}$).

A summary of the duration of linear phase for the targeting compounds using SIPs are listed in Table 10. Because the uptake of all analytes was in the linear phase during the sampling period (14 days), equation 6 was used to calculate the air volume sampled. Since uptake rate was not measured in this study, the sampling rate (R-value) of 1.0 $m^3\ d^{-1}$ from Saini *et al*. (2015) was used to derive the air concentration given the similar sampling conditions. However, since SIP disk would collect both gas phase pollutants and a small fraction of particles in air (approximate 10% according to Klánová *et al*. (2008)), the calculated pollutant concentration in air may be slightly higher than the real values.

Table 10. *The duration of linear phase for FTOHs, BFRs, OPFRs and cVMSs in recent studies*

Compound groups	Linear phase	References
FTOHs	> 32 days	Ahrens *et al.*, 2013
BFRs	> 49 days	Saini *et al*., 2015
OPFRs	> 14 days	Liu *et al.*, 2016
cVMs	> 16 days	Ahrens *et al.*, 2014

3 Results and discussion

3.1 Overview

Mean recoveries and standard deviations of mass-labelled internal standards were 52 ± 27%, 70 ± 44% and 58 ± 25% for BFRs, OPFRs and cVMSs in passive air samples, respectively. For FTOHs, the mean recoveries and standard deviation for mass-labelled 8:2 FTOH and mass-labelled 10:2 FTOH were 34 ± 15% and 54 ± 13%, respectively, but for mass-labelled 6:2 FTOH the recovery varied greatly. This may be due to the higher volatility of 6:2 FTOH compared to the other two FTOHs and that other compounds may generate similar fragments during the GC-MS analysis. Therefore, 6:2 FTOH was excluded from the following discussion. MDL of the analytes were 26 and 83 pg m^{-3} for 8:2 FTOH and 10:2 FTOH, respectively, and ranged from 8.0 to 1100 pg m^{-3} for BFRs, 2.7 to 2500 pg m^{-3} for OPFRs and 20 to 42 ng m^3 for cVMSs in indoor air. Relative standard deviation (RSD) of the duplicate samples (n = 3) for FTOHs and cVMSs were between 0.5% and 18%, which indicates a good agreement and reproducibility for these two groups of compounds. But for FRs the RSD varied greatly from 0 to 100% since the concentration of several FRs in the duplicate samples were just around MDL. Detailed QA/QC data for indoor air and fingernail samples can be found in the appendix (Tables A20–A25).

A summary of the results of indoor air samples was presented in Table 11 and Figure 3. The concentrations of the target compounds were found to vary greatly both across the buildings and within each building. The distribution of air sample data sets with detection frequency lager than 50% were evaluated using Kolmogorov-Smirnov test (n = 27, α = 0.05). The results revealed that 2,4,6-TBP, ∑OPFRs, 8:2 FTOH, 10:2 FTOH, ∑FTOHs, D4, D5 and ∑cVMSs were normally distributed, while D6, ∑BFRs and TCEP were not.

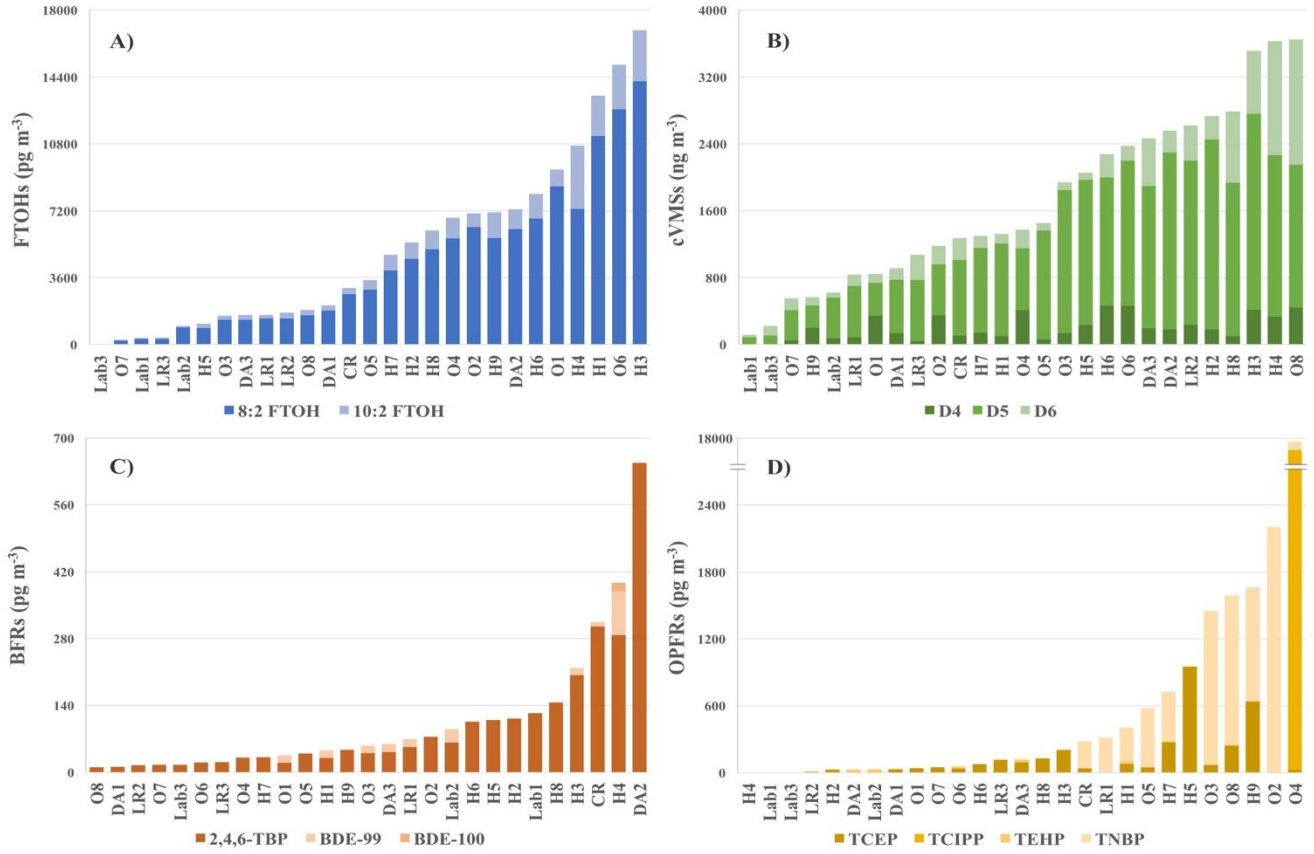

Figure 3. Concentrations of FTOHs (A), cVMSs (B), BFRs (C) and OPFRs (D) in passive air samples (n = 27). CR = computer room, DA = dining area, H = home, Lab = lab room, LR = lecture room, O = office

PFASs. 8:2 FTOH and 10:2 FTOH were found in more than 90% of all air samples (n = 27). The levels of ∑FTOHs in air (sum of 8:2 FTOH and 10:2 FTOH) of all 27 air samples ranged from lower than the MDL (Lab3) to 17000 pg m^{-3} (H3), with an average of 5100 pg m^{-3}. 8:2 FTOH was the most abundant FTOHs and constituted 84% of ∑FTOHs on average.

FRs. Three out of eight targeting BFRs and four out of five targeting OPFRs were detected in the 27 air samples. 2,4,6-TBP was found in all air samples. The average concentration of 2,4,6-TBP was 97 pg m^{-3}, ranged from less than MQL to 650 pg m^{-3}. DA2 contained the highest level of 2,4,6-TBP while Lab3 contained the lowest. BDE-99 and BDE-100 were the only detected PBDEs. BDE-99 was found in one third of the air samples and BDE-100 was only detected in one air sample from homes. The low concentration and detection frequency of PBDEs may be a consequence of their strict restrictions. TCEP was the most frequently detected OPFR (73% of all air samples), but concentration of TNBP (33% of all air samples) was generally higher, with an average of 310 pg m^{-3}. TEHP were found in less than half of the air samples. TCIPP was only found in one sample (O4) with an extraordinarily high concentration of 17300 pg m^{-3}. Since TCIPP was only detected in one sample, it was excluded in the following discussion when the average concentration of ΣOPFRs in different buildings and room types were compared. The range of ∑OPFRs (sum of TCEP, TEHP and TNBP) ranged from less than MDL (Lab1, Lab3 and H4) to 2.21×10^3 pg m^{-3} (O2). On average, the concentrations of OPFRs in air were much higher than BFRs, which is consistent with a previous study in Northern Europe (Cequier *et al*., 2014).

cVMSs. The result of cVMSs in air showed a good agreement with previous studies (Table 4). D4, D5 and D6 were found almost in all 27 air samples, except that D4 was not detected in Lab1 and Lab2. The average concentration of ∑cVMSs (sum of D4, D5 and D6) for all air samples was 1700 ng m^{-3}, ranged from 130 (Lab1) to 3600 ng m^{-3} (O8). All samples were below the health precaution guide value of 400 mg m^{-3} suggested by German Working Group on Indoor Guidelines (2011). D5 was usually the predominant cVMS, with a proportion ranging from 46% to 89% percent (69% on average) of ∑cVMSs.

Fingernails. Two BFRs (246-TBP and BDE-99), three OPFRs (TCEP, TCIPP and TEHP), 8:2 FTOH and 10:2 FTOH, and cVMSs were detected in the nine fingernail samples using GC-MS. 2,4,6-TBP was the most frequently detected analyte (7 out of 9 samples), followed by TEHP (5 out of 9) and TCIPP (4 out of 9). Other compounds were only found in one or two of the nine fingernail samples. Of the 26 targeting PFASs analyzed by LC-MS/MS, PFBS was the only one detected and was only found in two samples. Recovery rate of mass-labelled internal standards ranged from 12% to 169% for GC-MS, and from 1.5% to 68% for LC-MS/MS (Table A24, appendix). The low detection frequency and low recovery

rate of internal standards indicates that the extraction method used for fingernails needs to be further developed before applied to these compounds, or fingernails may not be a good indicator for examining the accumulation of FRs, PFASs and cVMSs in human body as well.

Table 11. *Indoor air levels of FTOHs, BFRs and OPFRs (pg m^{-3}) and cVMSs (ng m^{-3}) (n = 27)*

	Average	Median	SD	MIN	MAX	Detection Frequency (%)
8:2 FTOH	4300	3000	3900	<83	14200	97
10:2 FTOH	850	520	900	<26	3410	97
∑FTOHs	5100	3500	4700	<110	17000	100
2,4,6-TBP	97	42	140	10	650	100
BDE-99	8.3	<15	19	<15	92	33
BDE-100	0.67	<15	3.5	<15	18	3
∑BFRs	110	56	140	10	650	100
TCEP	120	41	210	<9.4	950	73
TCIPP	640	<2500	3300	<2500	17300	3
TEHP	6.4	<3.9	10	<3.9	30	43
TNBP	310	<260	560	<260	2200	33
∑OPFRs[a]	430	120	610	<280	2200	89
D4	200	180	150	<31	470	93
D5	1200	1100	710	86	2300	100
D6	330	170	380	29	1500	100
∑cVMSs	1700	1400	1000	110	3600	100

a. TCIPP was not included in ∑OPFRs.

3.2 Variation among buildings and room types

Indoor air concentrations of FTOHs, BFRs, OPFRs and cVMSs in different buildings and room types are presented in Figure 4.

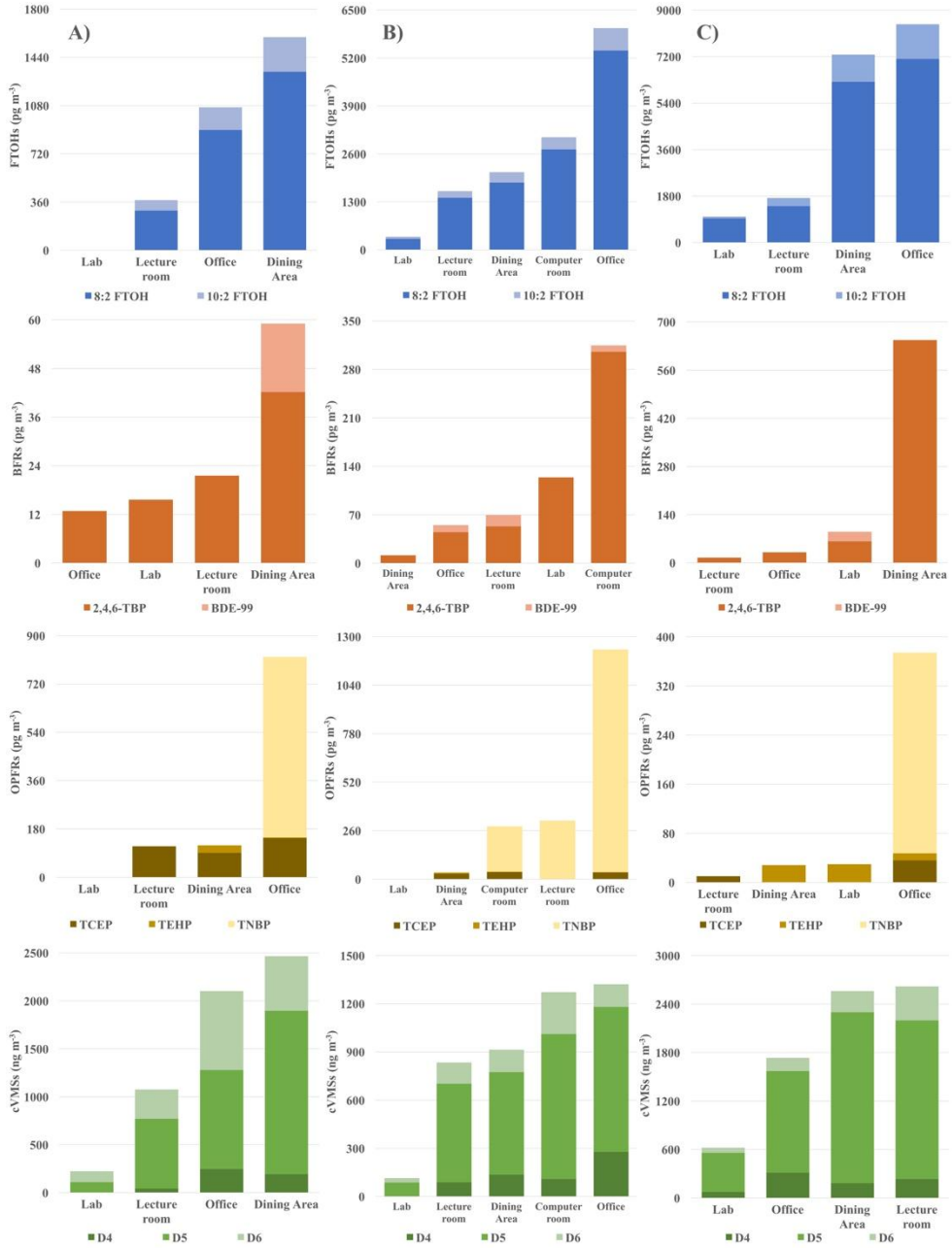

Figure 4. Concentrations of FTOHs, BFRs, OPFRs and cVMSs in different room types at Eco-centrum (column A), MVM (column B) and VHC (column C).

PFASs. Concentrations of ∑FTOHs in air in the three buildings (n = 18) ranged from n.d. (Lab3) to 15000 pg m^{-3} (O6), with an average of 3600 pg m^{-3}. On average, concentration of FTOHs in air was highest in VHC (5900 pg m^{-3}), fol-

lowed by MVM (3600 pg m^{-3}), and EC the lowest (820 pg m^{-3}), which is the reversed order of the ages of the three buildings.

FTOHs in different types of rooms revealed a similar pattern across the three buildings. Generally, FTOH concentrations were higher in dining areas and offices than in labs and lecture rooms. Labs contained the least FTOHs in air in the three buildings, probably because of the improved ventilation (e.g. fume hoods). Among the three labs, Lab2 (VHC) was the largest and had the highest number of analytical instruments (>20 instruments). The size of Lab1 (MVM) is similar to Lab3 (EC) but Lab1 has much more instruments (<10 instruments in Lab1, <3 instruments in Lab3 and >20 instruments in Lab2). The levels of FTOHs in the three labs could be ranked as Lab2 (1000 pg m^{-3}) > Lab1 (350 pg m^{-3}) > Lab3 (n.d.), implicating the number of electronic instruments may be a factor affecting the level of FTOHs in indoor environments.

FRs. BDE-99 and 2,4,6-TBP were the only detected BFRs in the three buildings (n = 18) and 2,4,6-TBP was the predominant one (on average 86 pg m^{-3}). Levels of ∑BFRs (BDE-99 and 2,4,6-TBP) in air in the three buildings ranged from below MQL (Lab3) to 650 pg m^{-3} (DA2), with an average of 92 pg m^{-3}. ∑OPFRs (TCEP, TEHP and TNBP) was higher than ∑BFRs in most cases (average 410 pg m^{-3}) and ranged from n.d. (Lab1 and Lab3) to 2200 pg m^{-3} (O2). When TCIPP was included, O4 had the highest level (18000 pg m^{-3}). The composition of OPFRs in indoor environments varied greatly among different rooms. In most cases, TNBP was the predominant compound, while in other cases TCEP or TEHP was dominant. The variation of BFRs among the buildings followed a same pattern as FTOHs: VHC (on average 140 pg m^{-3}) > MVM (98 pg m^{-3}) > EC (24 pg m^{-3}). For OPFRs, average concentration was, however, the highest in MVM (620 pg m^{-3}), followed by EC (380 pg m^{-3}), and VHC (200 pg m^{-3}) (Figure 4).

Regarding the concentration of FRs in different types of rooms, no clear pattern was observed. Rooms with more electric devices like labs (on average 77 pg m^{-3}), computer room (310 pg m^{-3}), dining areas (on average 240 pg m^{-3}) tended to have higher concentration of BFRs than offices (on average 35 pg m^{-3}) and lecture rooms (on average 35 pg m^{-3}), with some exceptions. Dining areas had the highest concentration of BFRs in EC (DA3, 650 pg m^{-3}) and VHC (DA2, 59 pg m^{-3}), while at MVM the level of BFRs in the dining area (DA1) was below MQL. The distribution of OPFRs seemed opposite to BFRs. The computer room (CR, 310 pg m^{-3}) at MVM had the highest BFRs concentration and it was also the second highest of all air samples from the three buildings. Labs and dining areas had high average concentrations of BFRs (77 and 240 pg m^{-3}, respectively) but quite low average levels of OPFRs (9.9 and 61 pg m^{-3}, respectively). Average concentration of OPFRs in offices (810 pg m^{-3}) was the highest in the three buildings but average concentration of BFRs was the second lowest (35 pg m^{-3}). Such differences suggest

BFRs and OPFRs origin from different sources. Spearman's rank correlation coefficient (r_s) of ∑BFRs and ∑OPFRs showed no correlation between the two groups of FRs in the three buildings (r_s = 0.01, p-value = 0.96).

cVMSs. Concentrations of ∑cVMSs in air in the three buildings (*n* = 18) ranged from 110 (Lab1) to 3600 ng m^{-3} (O8), with an average of 1400 ng m^{-3}. Unlike FTOHs and BFRs, results showed that VHC had the highest average concentration of cVMSs in air (1800 ng m^{-3}), followed by EC (1600 ng m^{-3}) and then MVM (1000 ng m^{-3}). On average, cVMSs in EC and VHC were two times higher than in MVM. D5 was the most abundant cVMSs almost in all samples, but the proportion of D4 and D6 varied. Among the eighteen air samples from the three buildings, twelve samples had a higher concentration of D6 than D4, while the other six contained more D4 than D6 and five out of the six were offices. All the three office samples (O1, O2 and O3) and the lab sample (Lab1) from MVM contained more D4 than D6.

Regarding the levels of cVMSs in different types of rooms, no clear pattern was observed. Like FTOHs, Labs contained the least cVMSs in each building (on average 320 ng m^{-3}), probably because of the better ventilation. Dining area (DA3, 2500 ng m^{-3}), offices (on average 1300 ng m^{-3}) and lecture room (LR2, 2600 ng m^{-3}) contained the highest level of cVMSs at EC, MVM and VHC, respectively. If the air samples were arranged in descending order of concentrations, it could be seen that results of cVMSs had the same order as FTOHs in EC (DA3 > average of offices > LR3 > Lab3) and MVM (average of offices > CR > DA1 > LR1 > Lab1) (Figure 4). However, no such pattern was observed for VHC. The result of Pearson product-moment correlation test (α = 0.05) showed that ∑cVMSs and ∑FTOHs were positive correlated in the three buildings (*n* = 18, r_p = 0.28, p-value = 0.27). Within each building, correlations were observed in samples from EC (*n* = 9, r_p = 0.98, p-value = 0.003) and VHC (*n* = 11, r_p = 0.44, p-value = 0.39), but not in samples from MVM (*n* = 10, r_p = 0.09, p-value = 0.85). For all indoor air samples, ∑cVMSs and ∑FTOHs were significantly correlated (*n* = 27, r_p = 0.51, p-value = 0.007). This may implicate that the concentrations of cVMSs and FTOHs in indoor air may be affected by similar factors or correlated factors in general, but their relationship still need to be further investigated in specific buildings.

3.3 Home and office samples

Home air samples (*n* = 9) and offices air samples (*n* = 8) were collected from nine volunteers (two of them in a same office) working in the three buildings. Homes samples had higher average concentration of ΣFTOHs (8200 pg m^{-3}), ΣBFRs (130 pg m^{-3}) and ΣcVMSs (2200 ng m^{-3}) compared to offices (5700 pg m^{-3}, 35 pg m^{-3}

and 1700 ng m^{-3}, respectively), but lower level of ΣOPFRs in home samples (560 pg m^{-3}) than in office samples (on average 860 pg m^{-3}) (Table 12).

Table 12. *Concentrations of FTOHs (pg m^{-3}), FRs (pg m^{-3}) and cVMSs (ng m^{-3}) in home and office samples*

	Home samples (*n* = 9)			Office samples (*n* = 8)		
	Average	MIN	MAX	Average	MIN	MAX
8:2 FTOH	6700	890	14000	4900	210	13000
10:2 FTOH	1600	250	3400	770	<44	2400
∑FTOHs	8200	1100	1700	5700	260	15000
2,4,6-TBP	120	30	290	31	10	74
BDE-99	14	<15	92	3.8	<15	16
BDE-100	2	<15	18	<15	<15	<15
∑BFRs	130	32	400	35	10	74
TCEP	260	<9.4	950	63	<9.4	240
TCIPP	<2500	<2500	<2500	2200	<2500	17000
TEHP	3.6	<3.9	25.	5.8	<3.9	16
TNBP	200	<260	1000	740	<260	2200
∑OPFRs[a]	460	<280	1700	810	4.0	2200
D4	240	97	460	280	51	470
D5	1600	270	2300	1000	360	1700
D6	440	86	1400	320	900	1500
∑cVMSs	2200	570	3600	1700	550	3600

a. TCIPP was not included in ∑OPFRs.

Results of paired home-office samples were log-transformed (n.d. values were replaced by 0) and were plotted on a X-Y coordinate (Figure 5). If the home sample of a pair of samples contained higher pollutant concentrations, the dot represents that paired samples would be below the line of y = x. Alternatively, if the pollutant concentration in the office sample was higher, the dot would be above line y = x. BDE-99 and BDE-100 were not plotted due to their low detection frequencies. For most compounds, home samples usually had higher concentrations than their corresponding office samples (except D4 and TNBP). Figure 5 showed that though office samples had a higher concentration of ΣOPFRs on average, but in six out of nine pairs of samples, homes contained higher levels of ΣOPFRs than offices. Such discrepancy was due to the particularly high level of TNBP in two office samples (O2 and O3). TEHP and TNBP concentrations were on average higher in office samples, while average concentration of TCEP was higher in home samples. Given the fact that people usually spent more time at home (14 hours on average) than at work (8 hours on average), indoor air at home could be a

critical source of the target compounds when evaluating human exposure via inhalation.

The relationship between 2,4,6-TBP in fingernails and in home and office air samples was investigate using Spearman's rank correlation coefficient. Results showed that, though home samples had higher average concentration of 2,4,6-TBP than office samples, no correlation was found between fingernails and home air samples (n = 9, r_s = 0.07, p-value = 0.86). However, a negative association (n = 9, r_s = -0.48, p-value = 0.23) between fingernails and office air samples was observed. This might be a coincidence and needs to be further investigated.

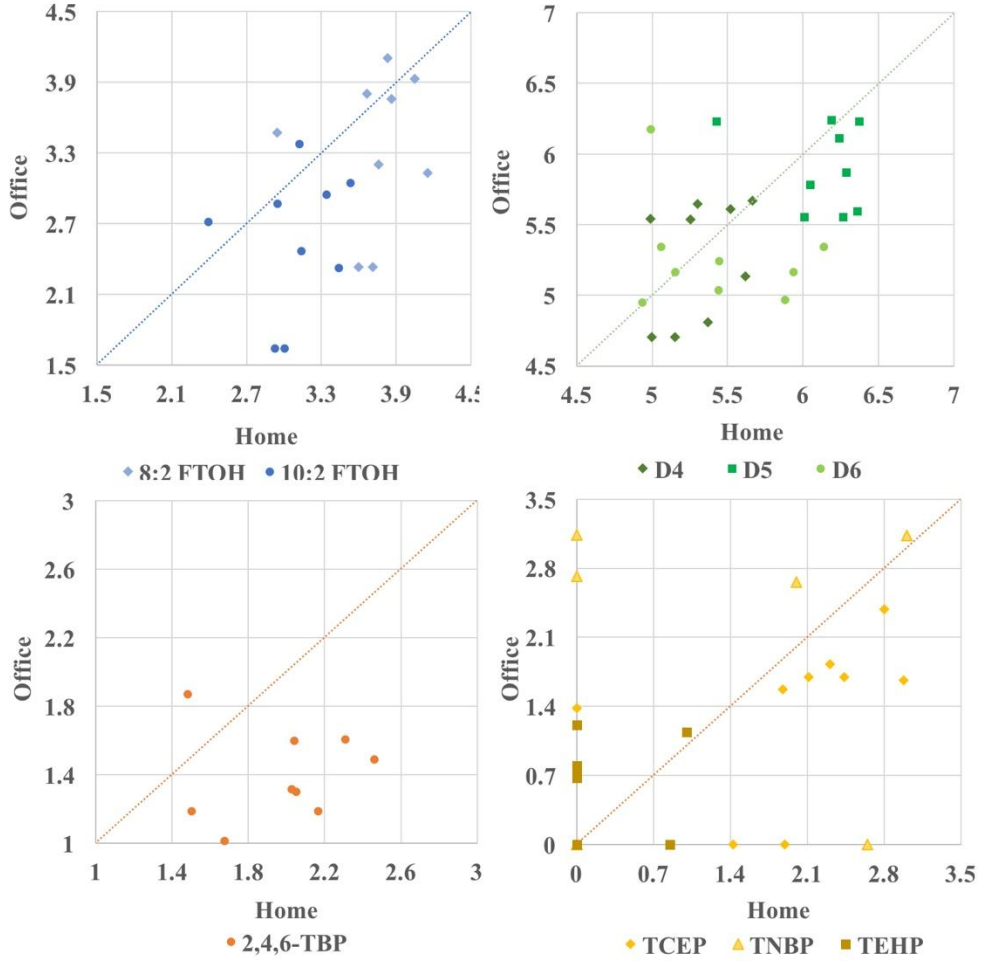

Figure 5. Log-transformed concentration of FTOHs, BFRs, OPFRs (pg m^{-3}) and cVMSs (ng m^{-3}) of paired home and office samples (n = 9 pairs)

3.4 Influencing factors

The home samples (*n* = *9*) and office samples (*n* = 8) were used to analyze the relationship between the analytes and possible influencing factors by employing analysis of variation (ANOVA), cluster analysis and Spearman's rank correlation coefficient (α = 0.05) (Table 13). The factors investigated were age of the building, number of electric equipment, number of outdoor cloth equipment, renovation and ventilation condition. The information was collected by questionnaires filled in by the volunteers (Table A26 in appendix).

Results of Spearman's rank correlation showed that there was a positive, but not significant, correlation (*n* = 17, r_s = 0.36, p-value = 0.15) between FTOHs in air and the number of electric equipment at the sampling sites. For the home samples, it was also found that FTOHs positively associated with the number of outdoor cloth equipment at home (*n* = 9, r_s = 0.52, p-value = 0.055). Thus, electric equipment and outdoor cloth equipment could be potential sources of FTOHs in indoor environments. Additionally, a very weak positive correlation (*n* = 17, r_s = 0.12, p-value = 0.64) between ΣFTOHs and the age of building was observed as well, which means ΣFTOH concentrations increased as the age of the building increasing. This is in contrast with the finding in previous section that the oldest building (EC) had the lowest level of FTOHs while the newest building (VHC) had the highest. The difference was probably because of the different sample size (9 home vs. 3 buildings at SLU). Therefore, based on currently insufficient and vague evidence, we cannot conclude whether the age of the building has affected the levels of FTOHs in its indoor air.

Regarding FRs in the seventeen air samples (*n* = 17) from homes and offices, Spearman's rank correlation coefficient revealed an opposite situation for BFRs and OPFRs. BFRs (mainly 2,4,6-TBP) were found significantly correlated with the number of electronic equipment (r_s = 0.50, p-value = 0.041), while no such correlation was found for OPFRs (ΣOPFRs, r_s = 0.03). Besides, there were also a significant positive correlation between BFRs and the age of building (r_s = 0.60, p-value = 0.011), which may be associated with the restriction on the use of traditional BFRs, while for OPFRs the correlation with building age was negative and not significant (r_s = -0.14, p-value = 0.59). No correlation was found between ΣBFRs and ΣOPFRs, which agreed with the observation of the two compound groups in EC, MVM and VHC in previous section. Such phenomenon indicated the main sources of BFRs and OPFRs were different, and the weak negative correlation between OPFRs and building age might be an implication of the increasingly use of OPFRs as substitutes for traditional BFRs in some products.

As to cVMSs, no correlation was found neither with number of electric devices (*n* = 17, r_s = -0.08, p-value = 0.84) nor with the number of people at the sampling

sites (n = 8, r_s = -0.02, p-value = 0.93), which means though direct human emission could contribute to cVMSs in indoor air, the different usage pattern of personal care products, air circulation within the building and other sources make it challenging to trace the source of cVMSs. However, similar to the three buildings, there was also a positive but not significant correlation between ΣcVMSs and ΣFTOHs in home and office air samples (n = 17, r_p = 0.39, p-value = 0.12), suggesting similar factors affecting the two groups of compounds.

Additionally, ΣcVMSs, ΣBFRs and ΣFTOHs were found positively correlated with each other, while ΣOPFRs was negatively correlated with the other three compound groups in the home and office samples. Since only a few influencing factors were examined in this project, there may be other unidentified factors affecting their presence. The air samples were collected from a wide variety of indoor environments instead of controlled sampling condition, therefore it is challenging to identify which factors were the most influential and whether there are connections among different groups of compounds.

In order to examine the effect of renovation and ventilation, the office and home samples (n = 17) were divided into subgroups according to the last time of renovation of the sampling sites (1-5 years ago or less than 1 year ago) and whether the sampling sites were force-ventilated. However, the results of ANOVA showed no significant difference between the two groups with different renovation time for all the four categories of compounds. ΣBFRs and ΣFTOHs was significantly higher in rooms without forced ventilation system, but for OPFRs and cVMSs, no significant differences observed.

Furthermore, K-means cluster analysis was conducted and the office and home samples were classified into four clusters based on the levels of ΣFTOHs, ΣBFRs, ΣOPFRs and ΣcVMSs (Table A26, appendix). However, no common property was found within each cluster in terms of building age, room volume, rug type, room type, ventilation condition or renovation time, which means the collective effect of influencing factors of the pollutants in indoor environments could be much more complicated than expected and the sample size was too small to identify common properties within each cluster.

Table 13. *Correlation coefficient (r)[a] between groups of analytes in indoor air samples (n = 17) and influencing factors*

	∑OPFRs	∑cVMSs	∑FTOHs	2,4,6-TBP	TCEP	D4	D5	D6	8:2 FTOH	10:2 FTOH	Building Age	Electronic equipment	No. of outdoor cloth	No. of persons
∑BFRs	-0.26	**0.63****	0.43	**0.98*****	-0.03	0.16	**0.54***	0.23	0.34	**0.72****	**0.60***	**0.50***	NA[b]	NA
∑OPFRs[c]		-0.15	-0.37	-0.25	0.37	0.04	-0.23	-0.21	-0.35	-0.41	-0.14	0.03	NA	NA
∑cVMSs			0.39	**0.647****	-0.06	0.42	**0.89*****	**0.65****	0.34	**0.54***	0.39	-0.05	NA	-0.042[d]
∑FTOHs				0.46	-0.27	**0.51***	0.39	0.43	**0.99*****	**0.88*****	0.12	0.36	0.52[e]	NA
2,4,6-TBP					0.02	0.16	**0.59***	0.27	0.38	**0.71****	**0.53***	**0.49***	NA	NA
TCEP						-0.08	0.00	-0.13	-0.28	-0.21	**0.50***	0.06	NA	NA
D4							0.19	0.45	**0.52***	0.43	-0.13	0.11	NA	NA
D5								0.42	0.35	0.48	0.51	0.06	NA	NA
D6									0.08	0.37	0.36	0.07	NA	NA
8:2 FTOH										**0.82*****	0.10	0.35	NA	NA
10:2 FTOH											0.22	0.34	NA	NA

a. Person coefficient (r_p) for normally distributed data and Spearman's rank coefficient (r_s) for non-normally distributed data. Significance level of α = 0.05, *p<0.05, **p<0.01, ***p<0.001.

b. NA – not analyzed.

c. TCIPP was not included in ∑OPFRs.

d. Only office samples (n = 8).

e. Only home samples (n = 9).

3.5 Assessment of human exposure via inhalation

Based on the results of home and office air samples and information from the questionnaires, external daily exposure dose (DED) via gas phase inhalation was calculated for each target compound (Table 14). Average DED of ΣFTOHs, ΣBFRs, ΣOPFRs and ΣcVMSs were 1200, 17, 94 and 340000 pg kg BW^{-1} day^{-1}, respectively. Since the analytes in particle phase were not taken into consideration, the DED values may be conservative. Generally, DED was on average 3 to 7 times higher at homes than in offices for FTOHs, BFRs and cVMs. But for OPFRs, average DED of ΣOPFRs was only slightly higher (1.3 times) at homes than in offices. According to the questionnaires, the volunteers on average spent 14 hours per day at home and 8 hours in offices. The higher average concentrations of ΣFTOHs, ΣBFRs and ΣcVMSs in home samples (Figure 5 and Table 12) and longer exposure duration result in significant higher DEDs at home than in office ($p < 0.05$, ANOVA). For ΣOPFRs, though office samples have a higher average concentration, the longer exposure duration at home led to a higher average DED of ΣOPFRs at home than in office. Among the four groups of analytes, cVMSs had the highest DED (340000 pg kg BW^{-1} day^{-1}).

The relation between DED of 2,4,6-TBP and its concentration in fingernail samples was examined by employing Spearman's rank correlation. The result showed that no correlation was found ($n = 9$, $r_s = 0.07$, p-value = 0.86), indicating exposure through other pathways may have much greater contribution than inhalation. This was consistent with a previous study in which ingestion of drinking water and food were the main exposure pathways of 2,4,6-TBP to the general population (WHO, 2005).

The measured DEDs were compared with reference values and guidelines (Table 14). Since 8:2 FTOHs is a precursor to PFOA and little information was found about guide values of FTOHs in air (Zushi *et al.*, 2012), the provisional tolerable daily intake for PFOA (1.5 μg kg BW^{-1} day^{-1}) suggested by European Food Safety Authority (EFSA 2008) was used as reference DED (RfD) value for 8:2 FTOH. RfDs for BFRs and OPFRs were obtained from Cequier *et al* (2014), with 1.0×10^5, 2.2×10^7, 8.0×10^7 and 2.4×10^7 pg kg BW^{-1} day^{-1} for BDE-99, TCEP, TCIPP and TNBP respectively. RfD of cVMSs was estimated according to the health precaution value (0.4 mg m^{-3}) suggested by German Working Group on Indoor Guidelines (2011). DEDs of FTOHs, BFRs, OPFRs and cVMSs were several orders of magnitude lower than their RfD values, which means the health risk posed by the four groups of compounds via inhalation was rather low. Given that cVMSs are widely used in personal care products and the concentration of cVMSs in indoor

air was several orders of magnitude higher than the other chemicals investigated in this study, more attention should be paid on the health risk posed by this group of compounds.

Although the DEDs of FTOHs, BFRs, OPFRs and cVMSs were far below the RfD values (Table 14), the DEDs are conservative and may be underestimated since only the exposure pathway by gas phase inhalation was investigated and the occurrence of only a selected group of compounds in the four categories was considered in this study. In addition, the toxicity of individual chemicals still need to be further investigated and the toxic effects caused by exposure to chemical mixture are still unclear. Thus, more work is required in terms of health risk assessment and management of PFASs, BFRs, OPFRs and cVMSs.

Table 14. *Estimated daily exposure dose of the targeted compounds (pg kg BW^{-1} day^{-1}) for homes and offices comparing to the reference dose (RfD in pg kg BW^{-1} day^{-1})*[a]

	DED_{home}[c]	DED_{office}[d]	$DED_{home+office}$	RfD
8:2 FTOH	730	290	1000	1.5×10^{6} [e]
10:2 FTOH	170	46	220	NA
∑FTOHs	900	340	1200	NA
246-TBP	13	1.9	15	NA
BDE-99	1.6	0.25	1.8	1.0×10^{5} [f]
BDE-100	0.24	0	0.24	NA
∑BFRs	15	2.1	17	NA
TCEP	30	3.2	33	2.2×10^{7} [f]
TCIPP	0	110	110	8.0×10^{7} [f]
TEHP	0.35	0.31	0.66	NA
TNBP	23	37	60	2.4×10^{7} [f]
∑OPFRs[b]	53	41	94	NA
D4	27000	15000	43000	NA
D5	170000	59000	230000	NA
D6	50000	14000	64000	NA
∑cVMSs	250000	890000	340000	6.3×10^{8} [g]

a. NA – not available
b. TCIPP was not included in ∑OPFRs
c. assuming 14 hours exposure time.
d. assuming 8 hours exposure time.
e. European Food Safety Authority (EFSA), 2008.
f. Cequier *et al.*, 2014.
g. German Working Group on Indoor Guidelines, 2011.

4 Conclusions

Passive air samples (n = 27) were collected from three buildings (n = 18) located at the Ultuna campus of Swedish University of Agricultural Science, Uppsala, Sweden and from residences (n = 9) of nine people from the staff in order to examine the levels of PFASs, BFRs, OPFRs and cVMSs in indoor air. Thirteen out of nineteen target compounds were found in the air samples. FTOHs (*viz*. 8:2 FTOH and 10:2 FTOH), cVMSs (*viz*. D4, D5 and D6) and 2,4,6-TBP were detected in almost all the air samples, while OPFRs and PBDEs had relatively low detection frequency (on average 38% and 18%, respectively). The poor recovery rates of mass-labeled internal standards and low detection frequency of the target compounds in fingernail samples suggested the extraction method of fingernails need to be further developed.

The concentrations of the analytes in the indoor air samples were in line with previous studies (Tables 1–4). D5 was found to be the predominant cVMSs and 8:2 FTOH was the predominant FTOHs in most cases. For OPFRs, TNBP was the predominant compound in offices, while at homes, TCEP was the most abundant. High standard deviation of the results indicated great variation of the concentrations of the four target compound groups both within each building and between buildings, indicating the influence of different sources. Generally, labs in the three sampling buildings contained the lowest FTOH, FR and cVMS concentrations (due to better ventilation), while in dining areas and offices, the average concentration of target compounds were by a factor of approximately 4 higher. Indoor air concentrations of target compounds were on average by a factor of 2 higher in home samples compared to office samples. A significant positive correlation was observed between ΣFTOHs and ΣcVMSs in all air samples ($p < 0.05$), suggesting these two groups of compounds may be influenced by similar factors such as building and room types or by associated factors.

In the seventeen samples from homes and offices, ΣBFRs was found significantly correlated with the age of the building ($p < 0.05$) and the number of electronic equipment at the sampling site ($p < 0.05$). Positive correlations, but not sig-

nificant, were also observed between ΣFTOHs and number of outdoor cloth equipment ($p = 0.055$), and between ΣFTOHs and number of electronic equipment ($p = 0.15$). No correlation was found between number of people and the concentration of ΣcVMSs ($p < 0.05$), indicating that different usage pattern of personal care products, air circulation within the building and other sources make it difficult to trace the source of cVMSs. Concentrations of FTOHs ($p < 0.05$, ANOVA) and BFRs ($p < 0.05$, ANOVA) were found significantly lower in rooms with forced ventilation system than in rooms without.

Daily exposure dose (DED) via inhalation was estimated based on the results of home and office air samples. Generally, DEDs of ΣFTOHs, ΣBFRs and ΣcVMSs were significantly higher at homes than in offices ($p < 0.05$, ANOVA), which is both an effect of the higher level of target compounds and longer time spent at home than in the office. Office air samples contained more ΣOPFRs on average than home samples, but the longer exposure duration at home resulted in a higher average DED at home than in office. Based on available data, estimated DEDs of 8:2 FTOH, BDE-99, TCEP, TCIPP, TNBP and ΣcVMSs were about 3 – 6 orders of magnitude lower than their reference values.

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Appendix

List of chemicals

Table A15. *Chemicals used for the analysis of air samples*

Compounds	Abbreviation	Supplier and purity (%)	
Target analytes			
Octamethylcyclotetrasiloxane	D4	Sigma-Aldrich, St.Louis, MO, USA	98
Decamethylcyclopentasiloxane	D5	Sigma-Aldrich, St.Louis, MO, USA	≥97
Dodecamethylcyclohexasiloxane	D6	Sigma-Aldrich, St.Louis, MO, USA	95
2-Perfluorohexyl ethanol(6:2)	6:2 FTOH	Wellington Laboratories, Guelph, ON, Canada	98
2-Perfluorooctyl ethanol(8:2)	8:2 FTOH	Wellington Laboratories, Guelph, ON, Canada	98
2-Perfluorodecyl ethanol(10:2)	10:2 FTOH	Wellington Laboratories, Guelph, ON, Canada	98
2,4-Dibromophenol	2,4-DBP	Sigma-Aldrich, St.Louis, MO, USA	99.9
2,4,6-Tribromophenol	2,4,6-TBP	Sigma-Aldrich, St.Louis, MO, USA	99.9
2,6-Dibromophenol	2,6-DBP	Sigma-Aldrich, St.Louis, MO, USA	99.9
Tris(2-chloroethyl) phosphate	TCEP	Sigma-Aldrich, St.Louis, MO, USA	97
Tri(1-chloro-2-propyl) phosphate	TCIPP	Sigma-Aldrich, St.Louis, MO, USA	97.5
Tris(2-ethylhexyl) phosphate	TEHP	Sigma-Aldrich, St.Louis, MO, USA	97
Tributyl phosphate	TNBP	Sigma-Aldrich, St.Louis, MO, USA	≥99
Tripentyl phosphate	TPeP	Accustandard, New Haven, CT, USA	97.2
Polybromodiphenyl ethers	PBDEs	Sigma-Aldrich, St.Louis, MO, USA	>98
Internal standards			
Octamethylcyclotetrasiloxane, [methyl-^{13}C4]-	(^{13}C4) D4	MoraVek Biochemicals, CA, USA	>98
Decamethylcyclopentasiloxane, [methyl-^{13}C5]-	(^{13}C5) D5	MoraVek Biochemicals, CA, USA	>99

Compounds	Abbreviation	Supplier and purity (%)	
2-Perfluorohexyl-[1,1-2H2]-[1,2-^{13}C2]-ethanol(6:2)	M6:2 FTOH	Wellington Laboratories, Guelph, ON, Canada	>99
2-Perfluorooctyl-[1,1-^{2}H2]-[1,2-^{13}C2]-ethanol(8:2)	M8:2 FTOH	Wellington Laboratories, Guelph, ON, Canada	>99
2-Perfluorodecyl-[1,1-^{2}H2]-[1,2-^{13}C2]-ethanol(10:2)	M10:2 FTOH	Wellington Laboratories, Guelph, ON, Canada	>99
Mass-labelled 4,4'-dibromodiphenyl ether	MBDE-15	Wellington Laboratories, Guelph, ON, Canada	>98
Mass-labelled 3,3',4,5'-tetrabromodiphenyl ether	MBDE-79	Wellington Laboratories, Guelph, ON, Canada	>98
Mass-labelled 2,2',3,4,4',6-hexabromodiphenyl ether	MBDE-139	Wellington Laboratories, Guelph, ON, Canada	>98
Mass-labelled Tributyl phosphate	M-TNBP	Wellington Laboratories, Guelph, ON, Canada	≥99
Recovery standards			
Mirex	Mirex	Cambridge Isotope Laboratories, Andover, MA, USA	
N,N-dimethylperfluoro-1-octanesulfonamide	Me_2FOSA	Wellington Laboratories, Guelph, ON, Canada	
Mass-labelled 3,3',4,4'-Tetrabromodiphenyl ether	M-BDE77	Wellington Laboratories, Guelph, ON, Canada	
Mass-labelled 2,2′,3,4,4′,5′-Hexabromodiphenyl ether	M-BDE138	Wellington Laboratories, Guelph, ON, Canada	

Table A16. *Chemicals[a] used only for the analysis of fingernail samples by HPLC-(-)ESI-MS/MS*

Compounds	Abbreviation
Target analytes	
perfluorobutane sulfonate	PFBS
perfluorohexane sulfonate	PFHxS
perfluorooctane sulfonate	PFOS
perfluorodecane sulfonate	PFDS
perfluorobutanoate	PFBA
perfluoropentanoate	PFPeA
perfluorohexanoate	PFHxA
perfluoroheptanoate	PFHpA
perfluorooctanoate	PFOA
perfluorononanoate	PFNA
perfluorodecanoate	PFDA
perfluoroundecanoate	PFUnDA
perfluorododecanoate	PFDoDA
perfluorotridecanoate	PFTriDA

Compounds	Abbreviation
perfluorotetradecanoate	PFTeDA
perfluorohexadecanoate	PFHxDA
perfluorooctadecanoate	PFOcDA
perfluorooctanesulfonamide	FOSA
N-methylperfluorooctansulfonamide	N-MeFOSA
N-ethylperfluorooctanesulfonamide	N-EtFOSA
N-methylperfluorooctanesulfonamido-ethanol	N-MeFOSE
N-ethylperfluorooctanesulfonamido-ethanol	N-EtFOSE
perfluorooctanesulfonamidoacetic acid	FOSAA
N-methylperfluorooctanesulfonamidoacetic acid	N-MeFOSAA
N-ethylperfluorooctanesulfonamidoacetic acid	N-EtFOSAA
6:2 fluorotelomer sulfonate	6:2 FTSA
Internal standards	
perfluoro-1-(^{13}C8)octane sulfonamide	$^{13}C_8$-FOSA
N-methyl-d_3-perfluorooctane sulfonamidoacetic acid	d_3-*N*-MeFOSAA
N-ethylper-d_5-fluorooctane sulfonamidoacetic acid	d_5-*N*-EtFOSAA
N-methyl-d_3-perfluorooctane sulfonamide	d_3-*N*-MeFOSA
N-ethyl-d_5-perfluorooctane sulfonamide	d_5-*N*-EtFOSA
N-methyl-d_7-perfluorooctane sulfonamido ethanol	d_7-*N*-MeFOSE
N-ethyl-d_9-perfluorooctane sulfonamido ethanol	d_9-*N*-EtFOSE
perfluoro-($^{13}C_4$)-butanoic acid	$^{13}C_4$-PFBA
perfluoro-($^{13}C_2$)-hexanoic acid	$^{13}C_2$-PFHxA
perfluoro-($^{13}C_8$)-octanoic acid	$^{13}C_4$-PFOA
perfluoro-($^{13}C_5$)-nonanoic acid	$^{13}C_5$-PFNA
perfluoro-($^{13}C_2$)-decanoic acid	$^{13}C_2$-PFDA
perfluoro-($^{13}C_2$)-undecanoic acid	$^{13}C_2$-PFUnDA
perfluoro-($^{13}C_2$)-dodecanoic acid	$^{13}C_2$-PFDoDA
perfluoro-($^{18}O_2$)-hexane sulfonic acid	$^{18}O_2$-PFHxS
perfluoro-($^{13}C_4$)-octane sulfonic acid	$^{13}C_4$-PFOS
perfluoro-($^{13}C_4$)-octanoic acid	$^{13}C_4$-PFOA

a. all chemicals in this table were purchased from Wellington Laboratories, Guelph, ON, Canada. Purity > 98%.

Instrumental settings for analysis of FTOHs

Before extraction, the samples were spiked with 50 μL of internal standard mix (c = 200 pg μL^{-1}). Prior to injection, 10 μL recovery standard (1000 pg μL^{-1}) was added.

- Instrument: Gas chromatography coupled with mass spectrometry (Agilent 7890 B Single Quad 5977A MSD; Agilent Technologies, Palo Alto, CA, USA)
- Injection Volume: 2 µL
- Injection port: Splitless injection
- Injector Temperature: 200 ℃
- Column: DB-WAX (30m, 0.25 mm, 0.25 µm) (J&W Scientific, Agilent Technologies)
- Carrier gas: Helium (flow 2 mL/min)
- Oven Programming:
  - Initial column temp: Hold at 60 ℃ for 2 minutes
  - Increase 2 ℃/min until 70 ℃. Hold at 70 ℃ for 0 minutes
  - Increase 8 ℃/min until 150 ℃. Hold at 150 ℃ for 0 minutes
  - Increase 10 ℃/min until 230 ℃. Hold at 230 ℃ for 0 minutes
- Auxillary Setpoint temperature: 230 ℃
- Source Temperature: 250 ℃
- Quadropole Temperature: 150 ℃
- Source Type: Chemical Ionization
- Detector: Mass selective
- Acquisition Mode: Select Ion Mode (SIM)

Table A17. *Ions used for the GC-MS analysis of FTOHs and cVMSs*

Compounds	Molecular weight	Quantification ion (m/z)	Qualification ion (m/z)
6:2 FTOH	364	365	327
M6:2 FTOH	368	369	331
8:2 FTOH	464	465	427
M8:2 FTOH	468	469	497
10:2 FTOH	564	565	527
M10:2 FTOH	568	569	531
N,N-Me_2FOSA	527	528	444
D4	296	281	265; 249
(^{13}C4) D4	300	285	269; 253
D5	370	355	267; 339
(13C5) D5	375	360	272; 344
D6	444	341	429; 325
MIREX		272	274; 270

Instrumental settings for analysis of cVMSs

Before extraction, the samples were spiked with 50 µL of internal standard mix (c = 5000 pg μL^{-1}). Prior to injection, 10 µL recovery standard (1000 pg μL^{-1}) was added.

- Instrument: Gas chromatography coupled with mass spectrometry (Agilent 7890 B Single Quad 5977A MSD; Agilent Technologies, Palo Alto, CA, USA)
- Injection Volume: 2 µL
- Injection port: Splitless injection
- Injector Temperature: 200 ℃
- Column: DB-5 (60m, 0.25 mm, 0.25 µm) (J&W Scientific, Agilent Technologies)
- Carrier gas: Helium (flow 2 mL/min)
- Oven Programming:
  - Initial column temp: Hold at 35 ℃ for 5 minutes
  - Increase 10 ℃/min until 160 ℃
  - Increase 30 ℃/min until 300 ℃. Hold at 300 ℃ for 10 minutes.
- Auxillary Setpoint temperature: 250 ℃
- Source Temperature: 300 ℃
- Quadropole Temperature: 150 ℃
- Source Type: Electron Ionization
- Detector: Mass selective
- Acquisition Mode: Select Ion Mode (SIM)

Instrumental settings for analysis of FRs

Before extraction, the samples were spiked with 50 µL of internal standard mix (mass labeled PBDEs c = 200 pg μL^{-1}, mass labeled OPFRs c = 250 pg μL^{-1}). Prior to injection, 10 µL recovery standard (1000 pg μL^{-1}) was added.

- Instrument: Gas chromatograph (Agilent Technologies, CA, USA, 7890A) couplet to a tandem mass spectrometer (Agilent Technologies, Triple Quad 7010)
- Injection Volume: 2 µL
- Injection port: Multiple mode inlet (MMI)
- Injector Temperature: 75 ℃ (0.45 min), 600 ℃/min to 325 ℃ (5 min), -20 ℃/min to 150 ℃ (0 min).
- Column: DB-5ms (15 m, 0.25 mm, 0.10 µm) (J&W Scientific, Agilent Technologies)
- Carrier gas: Helium (flow rate 1.5 mL/min)
- Oven Programming:

    - Initial column temp 90 ℃: Hold at 90 ℃ for 2.95 minutes
    - Increase 20 ℃/min until 320 ℃. Hold at 320 ℃ for 5 minutes.
- Auxillary Setpoint temperature: 310 ℃
- Source Temperature: 300 ℃
- Source Type: Electron Ionization
- Quadropole (Q1) Temperature: 150 ℃
- Quadropole (Q2) Temperature: 150 ℃
- Quench flow: 4
- Collison flow: 1.5
- Detector: Mass selective
- Acquisition Mode: Multiple reaction monitoring (MRM)

Table A18. *Ions used for the GC-MS/MS analysis of FRs*

Compound	Parent ion 1	Product ion 1	CE[a]	Parent ion 2	Product ion 2	CE
BDE47	485.7	326	22	483.7	324.1	22
BDE99	565.7	405.8	25	563.7	403.7	25
BDE100	565.7	405.8	22	563.7	403.7	22
BDE153	643.6	488.8	38	483.7	374.9	38
BDE183	723.5	563.4	25	721.5	561.3	25
2,4-DBP	251.8	142.8	10	142.8	117	20
2,6-DBP	251.8	142.8	10	142.8	117	20
2,4,6-TBP	331.7	221.8	30	n.d.[b]	n.d.	n.d.
TNBP	210.9	99.1	10	155.3	99.1	40
TCEP	248.9	124.9	5	204.8	117	5
TCIPP	200.9	99.1	30	276.9	125	10
TEHP	380.7	159	5	302.8	192.9	5
TPeP	168.8	99.1	50	239	99.1	50
M-BDE15	342.8	232	11	230.4	151.1	11
M-BDE77	500.7	390.8	23	496.7	336.8	23
M-BDE79	500.9	338.9	23	338.8	228.9	23
M-BDE138	656.6	496.7	38	496.6	415.8	38
M-BDE139	656.9	496.8	38	498.8	388.9	38
M-TNBP	102.9	83.1	5	167	103.1	40

a. CE = collision energy (V).
b. n.d. = not detected.

Analysis of fingernail samples using HPLC-MS/MS

Before extraction, the samples were spiked with 100 µL of internal standard mix (c = 50 pg μL^{-1}). Prior to injection, 10 µL recovery standard (200 pg μL^{-1}) was added.

The analysis of the 26 PFASs for fingernail samples was conducted, according to Ahrens et al. (2016), by using high performance liquid chromatography (HPLC, Agilent Technologies 1200 Series, Palo Alto, CA, USA) with a triple quadrupole mass spectrometer interfaced with an electrospray ionization source in negative-ion mode ((-)ESI-MS/MS, Agilent 6460 Triple Quadrupole System, Palo Alto, CA, USA).

Aliquots of 10 mL were injected on a Hypersil Gold pre-column (10 2.1 mm, 5 mm particle size, Thermo Scien- tific, Waltham, MA, USA) coupled with a Betasil C18 column (50 2.1 mm, 5 mm particle size, Thermo Scientific, Waltham, MA, USA) using a gradient of 0.350 mL/min Millpore water and methanol (both with 10 mM aqueous ammonium acetate solution (NH4OAc)). The initial gradient was set at 90/50 (v/v) Millipore water/methanol, then decreased for 3 min to 50/50 Millpore water/ methanol and further decreased to 5/95 Millpore water/methanol (hold for 3 min) (total time 20 min). The MS/MS was operated in the multiple-reaction monitoring (MRM) mode at the most sensitive transition from precursor ion to product ion.

Table A19. *Precursor, product ions used for HPLC–MS/MS analysis of PFASs in fingernail samples*

Compound Name	Precursor Ion (m/z)	Product Ion (m/z)	Fragmentor	Collision Energy (V)	Cell Accelerator Voltage (V)
PFBA	213.0	168.9	60	5	4
$^{13}C_4$-PFBA	217.1	172.1	60	5	4
PFPeA	263.0	219.0	60	5	4
PFBS	299.0	99.0	130	40	4
PFBS	299.0	80.0	130	40	4
PFHxA	313.0	313.1	70	5	4
PFHxA	313.0	269.0	70	5	4
$^{13}C_2$-PFHxA	315.0	270.0	60	5	4
PFHpA	363.0	319.1	60	5	4
PFHpA	363.0	168.9	60	10	4
PFHxS	399.0	99.0	140	40	4
PFHxS	399.0	80.0	140	40	4
$^{18}O_2$-PFHxS	403.0	103.0	180	40	4
6:2 FTSA	426.8	406.8	160	20	4
6:2 FTSA	426.8	80.9	160	30	4
PFOA	413.0	369.0	60	5	4
PFOA	413.0	168.9	60	10	4

Compound Name	Precursor Ion (m/z)	Product Ion (m/z)	Fragmentor	Collision Energy (V)	Cell Accelerator Voltage (V)
$^{13}C_8$-PFOA	421.0	376.0	60	5	4
$^{13}C_4$-PFOA	417.0	372.1	60	5	4
PFNA	463.0	419.0	100	5	4
PFNA	463.0	219.0	100	10	4
$^{13}C_5$-PFNA	468.0	423.1	100	5	4
PFOS	499.0	99.0	200	40	4
PFOS	499.0	80.0	200	40	4
$^{13}C_4$-PFOS	503.0	80.0	180	40	4
FOSAA	556.0	556.0	160	5	4
FOSAA	556.0	498.0	160	30	4
FOSAA	556.0	418.9	160	20	4
PFDA	513.0	469.0	100	5	4
PFDA	513.0	169.0	100	5	4
$^{13}C_2$-PFDA	515.0	470.0	100	5	4
N-MeFOSAA	570.0	482.9	130	10	4
N-MeFOSAA	570.0	419.0	130	20	4
d_3-*N*-MeFOSAA	573.0	419.0	140	20	4
FOSA	498.0	498.0	160	5	4
FOSA	498.0	77.9	160	40	4
$^{13}C_8$-FOSA	506.0	77.9	140	40	4
PFUnDA	563.0	518.9	100	10	4
PFUnDA	563.0	169.0	100	20	4
$^{13}C_2$-PFUnDA	565.0	520.0	100	5	4
N-EtFOSAA	584.0	526.0	140	20	4
N-EtFOSAA	584.0	483.0	140	10	4
d_5-*N*-EtFOSAA	589.0	531.0	140	20	4
PFDS	599.0	98.9	200	50	4
PFDS	599.0	80.0	200	50	4
PFDoDA	613.0	568.9	100	5	4
$^{13}C_2$-PFDoDA	615.0	570.0	100	5	4
N-MeFOSA	512.0	512.0	140	5	4
N-MeFOSA	512.0	219.0	140	20	4
N-MeFOSA	512.0	169.0	140	30	4
d_3-*N*-MeFOSA	515.0	169.0	140	30	4
N-MeFOSE	616.0	616.0	100	5	4
N-MeFOSE	616.0	59.1	100	10	4
d_7-*N*-MeFOSE	623.1	59.0	100	10	4
PFTriDA	663.0	618.9	100	5	4
N-EtFOSA	526.0	526	140	5	4

Compound Name	Precursor Ion (m/z)	Product Ion (m/z)	Fragmentor	Collision Energy (V)	Cell Accelerator Voltage (V)
N-EtFOSA	526.0	219	140	20	4
d_5-*N*-EtFOSA	531.0	169	140	30	4
N-EtFOSE	630.0	630	110	5	4
N-EtFOSE	630.0	59.0	110	10	4
d_9-*N*-EtFOSE	639.1	59.0	100	10	4
PFTeDA	713.0	669.0	120	5	4
PFHxDA	813.0	769.0	120	5	4
PFOcDA	913.0	868.9	120	5	4

MDLs, MQLs, recoveries and duplicate samples

Table A20. *MDLs, MQLs and detection frequencies of the target compounds in indoor air samples*

Compounds	MDL (pg μL^{-1})	MQL (pg μL^{-1})	MDL in air (pg m^{-3})	MQL in air (pg m^{-3})	Detection Frequency (%)
8:2 FTOH	2.3	4.2	83	150	97
10:2 FTOH	0.73	1.2	26	44	97
2,4-DBP	150	400	1000	2800	0
2,6-DBP	160	420	1100	3000	0
2,4,6-TBP	1.1	2.9	8.0	20	100
BDE-47	1.7	3.2	12	23	0
BDE-99	2.1	4.5	15	32	33
BDE-100	2.1	4.8	15	34	3
BDE-153	1.1	2.9	8.0	21	0
BDE-183	0.16	0.36	1.2	2.6	0
TCEP	1.3	3.3	9.4	24	73
TCIPP	350	970	2500	6900	3
TEHP	0.54	1.6	3.9	11	43
TNBP	37	100	260	720	33
TPeP	0.37	1.0	2.7	7.4	0
D4	860	2000	31 ng m^{-3}	72 ng m^{-3}	93
D5	1200	2200	42 ng m^{-3}	80 ng m^{-3}	100
D6	560	1100	20 ng m^{-3}	40 ng m^{-3}	100

Table A21. *MDLs, MQLs and detection frequencies of the target compounds in fingernail samples (GC-MS and GC-MS/MS)*

Compounds	MDL (ng mL^{-1})	MQL (ng mL^{-1})	MDL (ng g^{-1} dw)	MDL (ng g^{-1} dw)	Detection Frequency (%)
6:2 FTOH	2.5	3.8	31	46	0
8:2 FTOH	0.88	1.1	11	14	11
10:2 FTOH	0.60	1.0	7.3	12	11
24-DBP	33	58	80	141	0
26-DBP	1300	1300	3100	3300	0
246-TBP	0.66	0.76	1.6	1.8	78
BDE-47	0.79	1.7	1.9	4.3	11
BDE-99	3.3	5.9	8.1	14	22
BDE-100	3.0	6.6	7.4	16	0
BDE-153	1.7	2.5	4.2	6.2	0
BDE-183	0.90	2.1	2.2	5.2	0
TCEP	0.22	0.36	0.53	0.88	11
TCIPP	3.1	5.3	7.6	13	44
TEHP	0.050	0.12	0.12	0.29	56
TNBP	3.5	6.1	8.5	15	0
TPeP	0.046	0.13	0.11	0.32	0
D4	300	510	3600	6300	22
D5	570	970	7000	12000	22
D6	450	730	5500	8900	22

Table A22. *MDLs, MQLs and detection frequencies of 26 PFASs in fingernail samples (HPLC-MS/MS)*

Compounds	MDL (ng mL^{-1})	MQL (ng mL^{-1})	MDL (ng g^{-1} dw)	MDL (ng g^{-1} dw)	Detection Frequency (%)
6:2 FTS	0.78	2.2	9.5	28	0
PFBA	410	1100	5000	13000	0
PFPeA	450	1200	5500	15000	0
PFHxA	120	330	1500	4100	0
PFHpA	7.1	17	88	210	0
PFOA	5.9	12	72	150	0
PFNA	0.21	0.35	2.5	4.3	0
PFDA	0.29	0.39	3.5	4.8	0
PFUnDA	0.25	0.30	3.1	3.7	0
PFDoDA	3.9	10	48	120	0
PFTriDA	1.3	3.2	16	39	0
PFTeDA	3.6	7.9	44	97	0
PFHxDA	3.6	8.2	44	100	0

Compounds	MDL (ng mL^{-1})	MQL (ng mL^{-1})	MDL (ng g^{-1} dw)	MDL (ng g^{-1} dw)	Detection Frequency (%)
PFOcDA	5.4	13	67	160	0
EtFOSA	0.27	0.33	3.3	4.0	0
EtFOSAA	0.063	0.11	0.77	1.4	0
EtFOSE	0.25	0.32	3.1	3.9	0
FOSA	0.28	0.35	3.4	4.3	0
FOSAA	1.6	4.7	20	58	0
MeFOSA	0.26	0.31	3.2	3.7	0
MeFOSAA	0.27	0.41	3.4	5.0	0
MeFOSE	0.30	0.64	3.7	7.8	0
PFBS	0.090	0.14	1.1	1.8	22
PFDS	0.36	0.61	4.4	7.5	0
PFHxS	0.33	0.94	4.0	12	0
PFOS	2.5	7.0	30	86	0

Table A23. *Recovery rate of the internal standards for passive air samples*

Compounds	Average (%)	Standard Deviation
^{13}C-D4	51	15
^{13}C-D5	64	31
M-BDE15	27	17
M-BDE79	56	12
M-BDE139	70	18
M-TNBP	70	46
M6:2FTOH	16	56
M8:2FTOH	34	15
M10:2FTOH	55	13

Table A24. *Recovery rate of the internal standards for fingernail samples*

Compounds	Average (%)	Standard Deviation
^{13}C-D4	169	36
^{13}C-D5	169	38
M-BDE15	12	3.8
M-BDE79	42	16
M-BDE139	35	14
M-TNBP	146	68
M6:2 FTOH	0.3	0.1
M8:2 FTOH	17	4.7
M10:2 FTOH	40	9.2
$^{18}O_2$-PFHxS	26	15

Compounds	Average (%)	Standard Deviation
$^{13}C_4$-PFBA	1.4	1.5
$^{13}C_2$-PFHxA	4.9	4.5
$^{13}C_4$-PFOA	16	8.4
$^{13}C_5$-PFNA	13	10
$^{13}C_2$-PFDA	17	12
$^{13}C_2$-PFUnDA	20	15
$^{13}C_2$-PFDoDA	22	15
d_5-*N*-EtFOSA	46	4.0
d_5-*N*-EtFOSAA	68	35
d_9-*N*-EtFOSE	50	6.9
$^{13}C_8$-FOSA	50	4.0
d_3-*N*-MeFOSAA	35	24
d_3-*N*-MeFOSA	42	4.3
d_7-*N*-MeFOSE	40	3.7
$^{13}C_4$-PFOS	36	13

Table A25. *Results of duplicate passive air samples (FTOHs, BFRs and OPFRs in pg m^{-3},cVMSs in ng m^{-3})*

	EC			MVM			VHC		
	DA3	DA3	RSD (%)	CR	CR	RSD (%)	LR2	LR2	RSD (%)
246-TBP	57	27	35	590	25	92	15	15	0
BDE-99	16	18	3.7	0	18	100	0	0	0
BDE-100	0	0	0	0	0	0	0	0	0
TCEP	0	180	100	51	24	37	9.9	10	0.5
TCIPP	0	0	0	0	0	0	0	0	0
TEHP	18	38	34	4.5	0	100	0	0	0
TNBP	0	0	0	490	0	100	0	0	0
8:2 FTOH	1400	1200	8.6	3100	2300	15	1400	1400	0
10:2 FTOH	270	240	5.6	390	270	18	320	310	1.6
D4	200	180	5.4	130	90	17	240	220	4.3
D6	590	540	4.5	290	230	12	410	430	2.4

RSD – relative standard deviation.

Table A26. *Overview of sampling in homes (H) and offices (O) of nine volunteers and results of cluster analysis*

	Building Age (years)	Last renova-tion[b]	Area (m^2)	Volume (m^3)	Number of Electronic equipment	Forced ventila-tion	Number of Gore-tex equipment	Airing Frequency[c]	Rug type	Cluster
H1[a]	60	3	50	150	3	No	5	2	Synthetic	1
H2	50	3	20	48	9	No	2	1	Synthetic	2
H3	88	3	25	69	18	No	8	3	None	1
H4	40	2	16	40	10	No	2	1	Wool	1
H5	50	3	30	75	5	Yes	1	3	Synthetic	3
H6	40	3	7	16	10	No	2	0	None	4
H7	35	3	35	84	15	Yes	6	3	None	2
H8	60	2	18	45	7	Yes	NA[d]	NA	Synthetic	2
H9	40	3	6	14	10	Yes	3	0	Synthetic	4
O1	5	2	7	21	6	Yes	NA	NA	Synthetic	4
O2	5	2	20	50	11	Yes	NA	NA	Synthetic	4
O3	5	2	24	72	9	Yes	NA	NA	Synthetic	3
O4	2	2	9	30	2	Yes	NA	NA	Synthetic	2
O5	2	2	7	21	4	Yes	NA	NA	Synthetic	3
O6	2	2	7	21	6	Yes	NA	NA	Synthetic	1
O7	40	3	32	77	3	Yes	NA	NA	Synthetic	3
O8	40	3	8	40	2	Yes	NA	NA	Synthetic	3

a. The numbers indicate the volunteers, e.g. H1 and O1 belong to the same person.
b. Last renovation: 1 = less than 1 year; 2 = 1~5 years ago; 3 = more than 5 years ago.
c. Airing frequency: 0 = seldom; 1 = 1~2 times per week; 2 = 3~5 times per week; 3 = daily.
d. NA – not available.