

1. Introduction

Perfluoroalkylated substances (PFASs; $C_nF_{2n+1}R$) are used as intermediates or ingredients of surface protectors and surfactants in a wide variety of industrial and consumer applications, due to their properties such as resistance to chemicals agents and heat, dielectric properties, low friction properties and surface energy (OECD, 2013). There are many types of PFAS substances, including polymeric and non-polymeric structures, however, among them perfluoroalkyl acids (PFAAs) stand out. PFAAs structure consists of a fully fluorinated hydrophobic alkyl chain attached to a hydrophilic end group, being hydrophobic property increased with perfluorocarbon chain length (Navarro et al., 2018a). PFASs have been extensively used since the 1950s; nonetheless, evidence that they cause harm to people and ecosystems health has forced the implementation of regulations regarding their use worldwide. Consequently, based on assuming lower bioaccumulation rates and more rapid elimination in multiple organisms tested for short-chain PFASs (Olsen et al., 2009), since the turn of the millennium there has been a world-wide trend towards restricting the use of long-chain PFAAs ($C_nF_{2n+1}SO_3H$ with $n \geq 6$ for perfluoroalkane sulfonic acids (PFSAs) and $C_nF_{2n+1}CO_2H$ with $n \geq 7$ for perfluoroalkyl carboxylic acids (PFCAs); as proposed by Buck et al., 2011) by shorter ones; among it is worth mentioning the inclusion of perfluorooctanesulfonate (PFOS) and perfluorooctanoic sulfonic acid (PFOA) under Stockholm Convention (UNEP, 2009) and the currently proposal for listing of perfluorohexane sulfonic acid (PFHxS). Apparently, these restrictions come late, since PFOS major global manufacturer (3M Company produced ~80% of the global market; Martin et al., 2010) phased out of its products containing C6, C8, and C10 perfluorochemicals and replaced them with shorter C4 products (3M Company, 2000). Nevertheless, while manufacturers in most Organisation for Economic Cooperation and Development (OECD) countries planned to discontinue the production and use of long-chain PFAAs by 2015, new producers in developing and transition countries have started to manufacture them (OECD, 2015; UNEP, 2017).

Although PFASs have been used in a wide range of consumer and industrial applications, some major historical and current uses could be easily associated to their presence in home environments: electronics (flame retardants), construction products (additives blended in paints and coatings), household products (wetting agents or surfactant in floor polishes and cleaning agents), fire-fighting (film formers), metal plating (wetting agent), biocides (insect baits for control of leaf-cutting ants) or even in the polymer manufacture (processing aids). These chemicals will slough off from applied materials by use, abrasion and/or direct volatilization, and accumulate in the dust, being house dust monitoring a reliable tool to evaluate substitution tendencies of regulated chemicals by unregulated ones. However, two types of dust (suspended and settle dust) could be sampled. According to Cequier et al., 2014, monitoring of either settle dust or floor dust can be considered equally representative of the indoor environment for some POPs. Nevertheless, while data derived from elevated surfaces (suspended dust) may reflect adult exposure better, concentrations obtained in dust collected from the floor, seems to be more adequate to perform exposure assessments for children and toddlers. During these first years of growth and development, these population groups could be exposed to higher PFASs pollution, especially in developed countries, because they spend lots of time crawling on indoor floors, and present higher dust ingestion rates following hand-to-mouth behaviour (Trudel et al., 2008; U.S. EPA, 2017; Winkens et al., 2017).

Considering all mentioned above, it is of great interest to: i) evaluate tendencies of pollutant substitution degree, ii) investigate potential sources and iii) update human exposure assessments to these chemicals. To address these objectives, house dust samples collected from three European countries (Belgium, Italy and Spain) were analysed for short-chain and long-chain PFASs and compared to data previously reported. Moreover, potential sources related to building

and outdoor surrounding characteristics and occupant habits were investigated. Finally, estimated daily intakes via dust ingestion were calculated for toddlers and adults at median and worst case scenario and compared to oral Reference Dose (RfD) and dietary exposure estimated to provide an indication of the significance of dust ingestion to overall human exposure.

2. Materials and methods

2.1. Chemical and reagents

Samples were analysed for 20 PFASs including PFBS, PFHxS, PFOS, perfluorodecanesulfonate (PFDS), perfluorobutanoic acid (PFBA), perfluoropentanoic acid (PFPeA), perfluorohexanoic acid (PFHxA), perfluoroheptanoic acid (PFHpA), perfluorooctanoic acid (PFOA), perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA), perfluoroundecanoic acid (PFUDA), perfluorododecanoic acid (PFDoA), perfluorotridecanoic acid (PFTTrDA), perfluorotetradecanoic acid (PFTeDA), perfluorohexadecanoic acid (PFHxDA), perfluorooctadecanoic acid (PFODA), perfluorooctanesulfonamide (FOSA), *N*-methyl perfluorooctanesulfonamide (N-MeFOSA) and *N*-ethyl perfluorooctanesulfonamide (N-EtFOSA). Surrogate standard (LCS including MPFAC-MXA, N - d_3 -MeFOSA and N - d_5 -EtFOSA), internal standard spiking (ISS; $^{13}C_9$ -PFNA) and calibration (LCS, ISS and natives PFAC-MXB, FOSA, N-MeFOSA and N-EtFOSA) solutions were purchased from Wellington Labs (Canada). Acetonitrile and methanol were obtained from Scharlau (Barcelona, Spain). EnviCarb cartridges (500 mg, 6 mL) were provided from Sigma-Aldrich (St. Louis, MO, USA).

2.2. Sample collection

A total of 65 household dust samples were collected throughout Belgium ($n = 22$), Italy ($n = 22$) and Spain ($n = 21$). Samples were obtained from the houses of the partners of Test-Achats (Belgium), Altroconsumo (Italy) and OCU Ediciones SA (Spain). Home occupants were asked to vacuum the entire floor home from September 2016 to January 2017 with their domestic vacuum cleaners. This sampling strategy collects settle dust from the entire home, in which indoor sources were averaged together and cross-contamination from outdoor sources cannot be disregarded. Nevertheless, concentrations obtained in dust collected from the floor in which infants and toddlers spend lots of time could be more adequate to calculate exposure assessment for this population group. After sampling was finished, vacuum cleaner bags were covered with aluminium film, introduced in polyethylene sealable bags and sent together with a filled questionnaire regarding building and outdoor surrounding characteristics and occupant habits (see Table S1 for details). Once arrived at the laboratory, bulk dust samples were sieved (500 μm), homogenized and stored at $-20^\circ C$ until analysis.

2.3. Sample analysis

The analytical method used is a variation of a previously published (Navarro et al., 2017). An amount of 0.5 g of house dust spiked with surrogate standards (MPFAC-MXA, N - d_3 -MeFOSA and N - d_5 -EtFOSA) were extracted twice with 10 mL of acetonitrile in a polypropylene tube, vortexed 0.5 min, agitated 10 min, ultrasonicated at $40^\circ C$ for 30 min and centrifuged for 15 min at 2000g. Extracts were then combined and evaporated to 2 mL and passed through EnviCarb cartridges. The purified extract was reduced to 140 μL . 240 μL of methanol and 240 μL of 2 mM ammonium acetate in Milli-Q water were added to the final extract spiked with internal standard spiking solution ($^{13}C_9$ -PFNA) prior to instrumental analysis. PFASs were determined by HPLC-MS/MS (Varian 212 Liquid Chromatograph coupled to a Varian 320 triple quadrupole MS) as described elsewhere (Navarro et al., 2018b).

2.4. Quality assurance/quality control

House dust standard reference material (SRM2585; National Institute of Standards and Technology (NIST)) was analysed ($n = 3$) and compared to certified concentrations and data reported in the literature to check method accuracy and precision (Table S2). The average recovery of surrogates in house dust samples from the three European countries ranged from 87 to 114%. The limits of quantification (LOQs) of the method were calculated as the concentration corresponding to a signal-to-noise ratio ≥ 10 (Table S2). Injections of methanol (HPLC), as instrumental blanks, were used to assess instrumental contamination. Two field blanks, vacuuming diatomaceous earth, were also collected at each country. Field and procedural blanks were treated as samples through the entire process. Concentrations of all analytes in procedural blanks were below LOQs. Mean country-specific field blank levels, $< 5\%$ of sample concentrations in all cases, were subtracted from samples.

2.5. Statistical analyses

Statistical analyses were performed with SPSS 14.0 for Windows software. Analyte concentrations were not normally distributed (Shapiro-Wilk and Anderson-Darling test), thus Spearman rank correlation coefficient was derived to evaluate bivariate relationship and H Kruskal-Wallis or U-Mann Whitney tests were used to investigate differences between groups. In order to include all samples, those with concentrations below LOQ were replaced by the LOQ divided by the square root of two. For exploring bivariate correlations (Spearman test) values below LOQ were removed.

3. Results and discussion

Polyfluoroalkyl substances were quantified in all samples, ranging from 3.13 to 155 ng/g (12.9 ng/g; median, Table 1). The three European countries presented similar Σ PFASs (sum of PFSAs, PFCAs and perfluoroalkane sulfonamides (PFOSAs)) levels, but in all cases, even considering samples collected in each country separately Σ PFCAs concentrations were statistically higher ($p < 0.01$) than those obtained for Σ PFSAs. Quantification frequency of PFOSAs decreased significantly

compared to PFAAs, being FOSA only detected in eight samples, while MeFOSA and EtFOSA were below LOQs in all cases. In general, PFAAs levels in house dust are in the range of ng/g, but nonetheless concentrations can vary widely from country to country (Eriksson and Kärman, 2015; Jian et al., 2017; Lucattini et al., 2018; Moschet et al., 2018), which may be attributed to diversified lifestyles in different cultures, building and decorating characteristics, residents' habits or consumer products, but also to the economic status of a region (Jian et al., 2017). Shoeib et al., 2016 demonstrated that there is a significant positive correlation between PFOS and PFOA house dust levels and Human Development Indexed (HDI) which authors explained by the fact that inclusion of these chemicals in commercial merchandise results in an additional cost that is probably more difficult to assume in less developed countries. Belgium, Italy and Spain presented similar HDIs (0.896, 0.887 and 0.884; UNDP, 2016).

3.1. Temporal trends of polyfluoroalkyl substances

To our knowledge, there are two previous studies evaluating PFASs in Belgium (Cornelis et al., 2012; D'Hollander et al., 2010; Table S3) household dust samples. In both studies, sampling of settle dust in 2008 was performed following a standardized protocol proposed by Harrad et al., 2008 that differs from the present study. However, sum of some PFAAs (PFBS, PFHxS, PFOS, PFBA, PFHxA, PFOA, PFNA and PFDA) reported by D'Hollander et al., 2010 (2.9, 0.1–406 ng/g; median, min-max) are within the range obtained here (7.65, 1.71–95.9 ng/g) for these compounds, which at first may suggest no temporal trend variation. Nevertheless, it is important to notice that while median PFOS (0.50–0.73 ng/g; D'Hollander et al., 2010 and Cornelis et al., 2012) and PFHxS (0.10 ng/g; D'Hollander et al., 2010) concentrations obtained in 2008 closely resembles the ones quantified in the present study (0.77 and 0.13 ng/g; PFOS and PFHxS), levels of PFBS seem to have increased in the last ten years (0 to 0.40 ng/g; D'Hollander et al., 2010 – present study). It is worth to mention that, such a finding was not observed for some carboxylate PFAAs, which reflect similar concentrations in both studies (0.20–0.11, 0.30–0.41, 0.70–1.54 and 0.20–0.45 ng/g; 2008–2017 median values for PFBA, PFHxA, PFOA and PFDA). In Spanish case, previous studies exhibited PFOS median concentrations (2.45 and

Table 1
Quantifications frequencies (Qf) and median (min.-max.) concentrations (ng/g) obtained in household dust from Belgium, Italy and Spain.

	Total (n = 65)		Belgium (n = 22)		Italy (n = 21)		Spain (n = 21)	
PFBS	32%	0.40 (n.d.–56.7)	27%	0.40 (n.d.–56.7)	18%	0.40 (n.d.–11.6)	52%	0.70 (n.d.–12.0)
PFHxS	45%	0.13 (n.d.–11.3)	23%	0.13 (n.d.–11.3)	36%	0.13 (n.d.–3.62)	76%	0.95 (n.d.–7.16)
PFOS	63%	0.28 (n.d.–11.9)	73%	0.77 (n.d.–6.81)	73%	0.33 (n.d.–11.9)	43%	0.03 (n.d.–2.45)
Σ PFSAs ^a	94%	2.19 (n.d.–59.6)	86%	2.30 (n.d.–59.6)	100%	1.76 (0.64–15.2)	100%	2.68 (0.86–15.0)
PFBA	18%	0.11 (n.d.–20.9)	32%	0.11 (n.d.–20.9)	9%	0.11 (n.d.–6.99)	14%	0.11 (n.d.–2.51)
PFPeA	45%	0.02 (n.d.–21.3)	27%	0.02 (n.d.–2.44)	64%	0.21 (n.d.–2.48)	43%	0.02 (n.d.–21.3)
PFHxA	57%	0.31 (n.d.–28.3)	59%	0.41 (n.d.–11.0)	55%	0.34 (n.d.–28.3)	57%	0.31 (n.d.–5.10)
PFHpA	66%	1.00 (n.d.–105)	55%	0.32 (n.d.–105)	77%	2.79 (n.d.–29.9)	67%	0.49 (n.d.–55.4)
Σ PFCAs(<7) ^b	91%	3.14 (n.d.–108)	82%	2.14 (n.d.–108)	95%	5.40 (n.d.–60.7)	95%	2.02 (n.d.–55.6)
PFOA	100%	1.41 (0.21–53.0)	100%	1.54 (0.31–24.2)	100%	1.56 (0.21–53.0)	100%	1.00 (0.42–12.5)
PFNA	46%	0.04 (n.d.–9.04)	36%	0.04 (n.d.–9.04)	55%	0.10 (n.d.–6.54)	48%	0.04 (n.d.–5.70)
PFDA	74%	0.49 (n.d.–25.8)	86%	0.45 (n.d.–25.8)	64%	0.52 (n.d.–15.8)	71%	0.36 (n.d.–17.4)
PFUdA	75%	0.17 (n.d.–7.68)	82%	0.15 (n.d.–7.68)	91%	0.29 (n.d.–2.55)	52%	0.07 (n.d.–3.09)
PFDoA	74%	0.28 (n.d.–19.7)	82%	0.30 (n.d.–19.7)	82%	0.38 (n.d.–5.52)	57%	0.12 (n.d.–14.5)
PFTTrDA	60%	0.23 (n.d.–11.1)	77%	0.27 (n.d.–11.0)	73%	0.79 (n.d.–6.83)	29%	0.03 (n.d.–11.1)
PFTeDA	89%	1.11 (n.d.–38.4)	95%	0.70 (n.d.–27.7)	91%	1.61 (n.d.–16.1)	81%	0.73 (n.d.–38.4)
PFHxDA	80%	0.75 (n.d.–14.9)	68%	0.28 (n.d.–8.92)	100%	1.10 (0.43–4.83)	71%	0.44 (n.d.–14.9)
PFODA	66%	0.46 (n.d.–4.96)	59%	0.24 (n.d.–4.81)	73%	0.84 (n.d.–4.62)	67%	0.47 (n.d.–4.96)
Σ PFCAs(≥ 7) ^c	100%	5.92 (1.13–129)	100%	4.65 (1.70–129)	100%	7.17 (2.40–66.2)	100%	3.78 (1.13–120)
Σ PFCAs ^d	100%	8.68 (2.05–149)	100%	6.92 (2.11–149)	100%	15.2 (2.80–125)	100%	8.68 (2.05–125)
FOSA	12%	0.01 (n.d.–1.05)	18%	0.01 (n.d.–0.43)	9%	0.01 (n.d.–1.05)	10%	0.01 (n.d.–0.03)
Σ PFAAs ^e	100%	12.9 (3.13–155)	100%	10.2 (3.13–155)	100%	19.7 (3.95–137)	100%	11.3 (3.98–137)

n.d. = not detected.

^a Sum of perfluoroalkane sulfonic acids: PFBS, PFHxS and PFOS;

^b Sum of perfluoroalkyl carboxylic acids: PFBA, PFPeA, PFHxA, PFHpA;

^c Sum of perfluoroalkyl carboxylic acids PFOA, PFNA, PFDA, PFUdA, PFDoA, PFTTrDA, PFTeDA, PFHxDA and PFODA;

^d Sum of perfluoroalkyl carboxylic acids: PFBA, PFPeA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUdA, PFDoA, PFTTrDA, PFTeDA, PFHxDA and PFODA;

^e Sum of Σ PFSAs, Σ PFCAs and FOSA.

5.29 ng/g; Ericson Jogsten et al., 2012 and Eriksson and Kärrman, 2015) up to three orders of magnitude higher than the one obtained here (0.03 ng/g). These studies collected samples from vacuum cleaner bags in 2009, so the comparison of results is more plausible and could evidence a decrease in the use of PFOS in Spain. To the best of the author's knowledge, this is the first study reporting PFAS concentration in house dust from Italian homes.

3.2. Perfluoroalkyl acids (PFSA and PFCAs)

Interesting differences in terms of PFSA composition profile were found between countries (Fig. 1). While similar PFOS and PFBS contributions to Σ PFASs were obtained from Belgian and Italian house dust, PFBS clearly dominated in Spanish case, indicating a higher PFOS substitution degree in the latter country. PFOS was the first PFASs to which global extent environmental contamination was demonstrated. It was originally included in the OSPAR List of Chemicals for Priority Action (2003), then subjected to the European Chemical Regulation, REACH Annex XVII restricted substances list (entry 53; EC, 2009) and finally listed as a persistent organic pollutant (POP) with its salts and the perfluorooctane sulfonyl fluoride (PFOSF) under Stockholm Convention (UNEP, 2009) and therefore removed from REACH and subjected to EU POPs regulation (Re EU No 757/2010; EC, 2010). Nevertheless, it could be expected relative high presence in worldwide house dust since PFOS was included in the Annex B of Stockholm Convention. This inclusion only restricts the use of the chemical and allows the PFOS production by listing specific exemptions that currently coincide with all major historical uses (leather and apparel, rubber and plastics, fire-fighting foam, carpets, textiles and upholstery, photo-imaging and insecticide). Information gathered using questionnaires was used to evaluate possible relationships with building characteristics, occupant habits and/or outdoor surroundings of potential emission sources. Nevertheless, PFOS concentrations only correlated positively with the building edification age ($r_s = 0.406$; $p < 0.05$; Table S4). This result implies a lower PFOS content in newer buildings, which could again reinforce a decrease in the use of this chemical in Europe. PFOS and PFHxS share several application areas for consumer goods such as papermaking, sealants, carpets, leather, apparel, textiles among others. Therefore it is not strange to find a positive association between PFOS and PFHxS ($r_s = 0.616$ $p < 0.01$) in European house dust. Interestingly, a moderate correlation was obtained between PFHxS and the percentage of the floor covered by textiles ($r_s = 0.456$ $p < 0.05$), that could be easily associated with its previously mentioned uses. Finally, no correlations were detected

between PFOS and PFHxS with PFBS ($p > 0.05$), suggesting different application areas. The potassium salt of PFBS is marketed as flame retardants for polycarbonate resins used in electronics (OECD, 2013). However, no correlations were found between PFBS dust content and data obtained in the questionnaires related to electric and electronic devices (EED) presence and/or use at homes, which allowed the drawing of conclusions.

Quantification of PFOA above LOQ in all samples obtained from the three countries (0.21 to 53, 1.41 ng/g; min-max, median; Table 1) clearly reflects the great use of this chemical in Europe. PFOA has been mainly used as a polymerization aid in the manufacturing of fluoropolymers and in aqueous fluoropolymer dispersions, which are used for paints, photographic film additives and in the textile finishing industry (Vierke et al., 2012). PFOA can be detected in numerous consumer products with water, grease and dirt repellent properties like carpets, outdoor jackets, and impregnating agents in paper, textiles, paints and lacquers. Therefore it is not strange that it dominated PFCA pattern (Fig. 1). PFOA, its salts and PFOA-related substances have similar hazard profile to PFOS and were added to REACH annex XII (entry 68) on 14 June 2017 (Reg EU 2017/1000; EC, 2017) and recently listed with its salts and PFOA-related compounds in Annex A (elimination) of the Stockholm Convention. Nevertheless, as happened for PFOS and PFHxS, PFOA production was also phased out in 2002 by 3 M Company, measure that joined other major PFAS manufactures in 2006 signing on to the U.S. EPA "2010/2015 PFOA Stewardship Program" with commitments to work towards the reduction (95% by 2010) and elimination (2015) of emissions and product content of PFOA and higher homologues and precursors. In addition, very long-chain PFCAs (PFUdA, PFDoA, PFTrDA, and PFTeDA; C11-C14 PFCAs) are recognized as very persistent and very bioaccumulative (vPvB) substances and included in the Candidate List of Substances of Very High Concern (SVHCs). Therefore it could be expected that as happened for PFASs, driven by these regulatory policies, levels of short-chain PFCA will increase in environmental media. However, on contrary to what was found for sulfonates, long-chain PFCAs presented higher contribution to Σ PFCAs (Table 1 and Fig. 1) and still aroused higher concentrations than short-chain carboxylic acids in house dust from Belgium ($p < 0.01$), Italy and Spain ($p < 0.05$). Alternative short-chain products are technically much less performing. Therefore new producers (largely in continental Asia) that are not part of the 2010/2015 PFOA Stewardship Program have begun to produce long-chain PFCAs and their precursors (Wang et al., 2014) to meet continuing international market demands and hence, long-chain PFCAs cannot be ruled out. Interestingly, long-

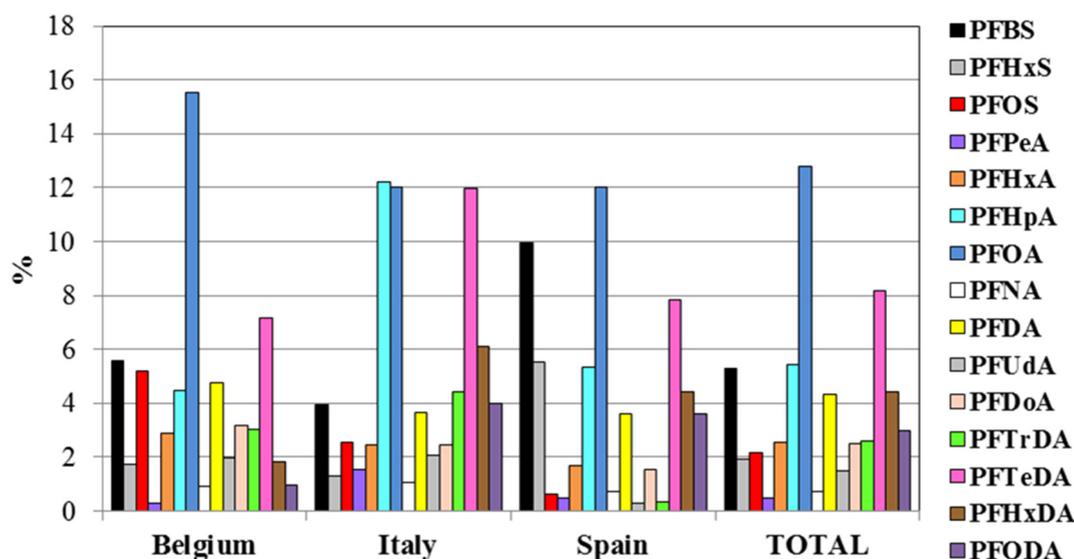


Fig. 1. Median percentage of total PFASs in dust samples from Belgium, Italy and Spain for analytes quantified in >30% of samples.

chain PFCAs (PFOA, PFNA, PFDA, PFUdA, PFDaA, PFTTrDA, PFTTeDA, PFHxDA and PFODA) were strongly correlated with one another ($r_s > 0.487$, $p < 0.01$; Table S4), decreased statistical significance with PFHpA (C6) and lost with most of short-chain PFCAs. The significant associations between these PFCAs seem to indicate that these compounds may be found together in commercial products or may be used in multiple products often used together.

Analysis of home surroundings and PFAS content revealed statistically higher Σ PFCAs, Σ PFCAs $n \geq 7$, PFUdA, PFOA and PFHpA concentration in industrial sites compared to urban or agricultural locations (Table S5), suggesting industrial processes as a potential source of these chemicals in Europe. Concentrations found in dust from Italy presented higher levels ($p < 0.05$) of Σ PFCAs and Σ PFCAs $n \geq 7$ compared to Belgium and Spain. Italy has not ratified the Stockholm Convention, but it is subjected to European policies. In this sense, the result of the public consultation conducted by the ECHA (ECHA, 2017) and the opinion of the Committee for Risk Assessment (RAC) and the Committee for Socio-economic Analysis (SEAC) (ECHA, 2018) for proposing a restriction on the manufacturing, use, placing on the market and import of C₉–C₁₄ PFCAs, their salts and precursors to prevent a switch by industry using PFOA-based substances, that to longer chain PFCAs, could play an important role in the future. In the European Union, PFOA shall not be manufactured or placed on the market as substances on their own from 4 July 2020 in the European Union (EC, 2017).

Strong positive correlations were obtained between PFOS and PFOA ($r_s = 0.705$, $p < 0.01$; Table S4) and these with PFHxS ($r_s > 0.438$, $p < 0.05$). First paper reporting data for PFOA and PFOS in house dust (Moriwaki et al., 2003) also found a positive correlation between them, result that was also observed for dust from elevate surfaces (Björklund et al., 2009; Haug et al., 2011) and settle dust collected with specific protocols (D'Hollander et al., 2010; Fraser et al., 2013; Goosey and Harrad, 2011) or vacuum cleaner bags (Knobeloch et al., 2012; Kubwabo et al., 2005; Shoeib et al., 2011, 2016; Strynar and Lindstrom, 2008; Xu et al., 2013). Positive correlation found between PFOA, PFOS and PFHxS levels and the amount of carpeting in homes have been associated to the use of carpet surface treatment products (Kubwabo et al., 2005; Shoeib et al., 2011). However, as mentioned before this was only observed in the present study for PFHxS. Remarkably, some PFCAs, such as PFNA, PFDA, PFTTeDA and PFHxDA correlated well with PFHxS ($r_s > 0.544$, $p < 0.01$) but lost statistical significance with PFOS, possibly suggesting two different sources of these materials. The type of building was revealed as a factor to be taken into account in terms of PFAS content. Dust collected from flats exhibited higher PFHxS, PFTTeDA, PFHxDA and PFODA concentrations than detached houses. A similar result was reported by Björklund et al., 2009 for PFOS and PFOA in dust collected during 2006/2007 in Stockholm city (Sweden), but in this case, sampling was done in surfaces at least one meter above the floor.

3.3. Perfluoroalkyl sulfonamides

Interestingly, from the three perfluoroalkyl sulfonamides (PFOSAs; FOSA, MeFOSA and EtFOSA) only 8 samples aroused FOSA level (from 0.01 to 1.05 ng/g) above LOQ. Presence of these PFOSAs was not previously reported in household dust from Belgium and Italy. Besides, Ericson Jogsten et al., 2012 measured MeFOSA in one dust sample from Spain at a concentration of 0.065 ng/g, but it could not be confirmed in the secondary transition when analysed by GC–MS/MS. Generally, low levels of PFOSAs were found in dust from Canada (Karásková et al., 2016; Shoeib et al., 2005), Norway (Haug et al., 2011), Czech Republic (Karásková et al., 2016), Germany, United Kingdom, Australia, (Kato et al., 2009) and United States (Karásková et al., 2016; Kato et al., 2009). Although EtFOSA was under LOQ in our samples, higher levels in house dust have been related to its use as an insecticide (Goosey and Harrad, 2011). EtFOSA is the active ingredient of Sulfluramid or Sulfluramid, a pesticide introduced as an alternative

to Mirex that has been applied in ant baits to control leaf-cutting ants, red imported fire ants and termites (OECD, 2013). Sulfluramid holds particular importance in Latin America, concretely, Brazil has an exemption from the Stockholm Convention to produce and use Sulfluramid to manage leaf-cutting ants, which are agricultural pests throughout the entire neotropical region. However, in the EU, PFOS-related substances are not used in the manufacture of pesticides (UNEP, 2007).

3.4. Human exposure assessment

Exposure from indoor dust occurs through ingestion of settle dust, inhalation of suspended dust and dermal absorption of both matrices. However, assuming that i) present study has evaluated settle dust and ii) studies conducted by Winkens et al., 2017 concluded that reliable methods to calculate dermal uptakes are not available, human exposure assessment was performed considering only dust intake via oral ingestion. Thus, Estimated Daily Intakes ($EDI_{dust\ ingestion}$; ng/day) were calculated multiplying PFAS concentrations C_{dust} (ng/g), dust ingestion rates (IR_{dust} ; mg/day), and the gastrointestinal absorption fraction (AF , 0.94; mean value obtained from Björklund et al., 2009; Egeghy and Lorber, 2011; Ericson Jogsten et al., 2012; Goosey and Harrad, 2011; Liu et al., 2011; Shoeib et al., 2005, 2011, 2016; Tian et al., 2016; Xu et al., 2013), as it is shown in Eq. (1). EDI values were calculated for toddlers (7 months to 4 years; U.S. EPA, 2017) and adults (20 to 59 years) at median and worst-case scenarios. To do that, 50th (P50) and 95th (P95) percentile for total concentrations were used, since similar ($p > 0.05$) PFAS concentrations were obtained in the three countries. Dust ingestion rates of 50–100 mg/day (toddlers) and 20–60 mg/day (adults) for the central and upper percentiles were obtained from U.S. EPA, 2017.

$$EDI_{dust\ ingestion} = C_{dust} \times IR_{dust} \times AF \quad (1)$$

The $EDI_{dust\ ingestion}$ values at the median and worst case scenario for PFASs ranged from 0.28 to 7.74 (adults) and from 0.69 to 12.9 ng/d (toddlers), respectively (Table 2). Calculated exposures for PFOS (0.31–0.52 ng/d; adults and toddlers) and PFOA (0.85–1.41 ng/d) at worst case scenario were in the range of those previously reported for Belgian (0.4–3.8 and 0.2–2.5 ng/d for PFOS and PFOA; D'Hollander et al., 2010) and Spanish (1.3–2.6 and 4.0–8.1; Ericson Jogsten et al., 2012) population. Normalized daily intakes (ng/kg b.w./day) calculated assuming 13.8 and 80 kg body weight for toddlers and adults (U.S. EPA, 2011), even at worst case scenarios, were well (>150 times) below oral Reference Dose (RfD) or tolerable daily intakes (TDI). In 2012 European Food Safety Authority (EFSA) published a scientific report of perfluoroalkylated substances in food, in which PFOS and PFOA dietary exposures were estimated for 13 European countries including Belgium, Italy and Spain (EFSA, 2012). Adult estimated daily intakes via house dust ingestion for PFOS and PFOA calculated in the present study, even at worst case scenario (0.004 and 0.01 ng/kg b.w./day; PFOS-PFOA Table 2), represented 1 and 13% of the European dietary exposure (0.27 and 0.08 ng/kg b.w./day) calculated using lower bound mean occurrence, respectively. However, in toddlers case, percentages for the same parameters increase to 6 and 51% of dietary exposure (0.58 and 0.20 ng/kg b.w./day; PFOS and PFOA). These results evidenced that food ingestion is the primary route of exposure for adults, even though the contribution from dust ingestion turns up to the half of food ingestion for toddlers at worst case scenarios.

4. Conclusions

Concentrations of PFASs were quantified in 65 house dust samples obtained from Belgium, Italy and Spain. Three European countries presented similar Σ PFASs levels, but in all cases, even considering samples collected in each country separately, Σ PFCAs concentrations were higher than those obtained for Σ PFASs. Comparison with previously published data revealed a PFBS in samples collected from

Table 2

Estimated daily intakes (EDI) via house dust ingestion (ng/kg b.w./day) calculated for toddlers and adults at central (P50) and upper (P95) scenarios. Reference dose (RfD) and total daily intakes (TDI) in ng/kg b.w./day.

	EDI (ng/day)						EDI (ng/Kg b.w./day)						RfD	TDI
	Toddler			Adults			Toddler			Adults				
	P50	–	P95	P50	–	P95	P50	–	P95	P50	–	P95		
PFBS	0.02	–	1.07	0.01	–	0.64	0.001	–	0.08	9.4E-05	–	0.01	430 ^{a,b} –20000 ^c	
PFHxS	0.01	–	0.33	0.002	–	0.20	4.4E-04	–	0.02	3.1E-05	–	0.002	20 ^b	20 ^g
PFOS	0.01	–	0.52	0.01	–	0.31	0.001	–	0.04	6.6E-05	–	0.004	20 ^d	20 ^g –150 ^h
∑PFASs	0.10	–	1.43	0.04	–	0.86	0.01	–	0.10	5.1E-04	–	0.01		
PFBA	0.01	–	0.20	0.002	–	0.12	3.6E-04	–	0.01	2.5E-05	–	0.002	2900 ^e	
PFPeA	0.001	–	0.23	3.5E-04	–	0.14	6.4E-05	–	0.02	4.4E-06	–	0.002		
PFHxA	0.01	–	0.82	0.01	–	0.49	0.001	–	0.06	7.3E-05	–	0.01		
PFHpA	0.05	–	1.34	0.02	–	0.80	0.003	–	0.10	2.4E-04	–	0.01	20 ^b	
PFOA	0.07	–	1.41	0.03	–	0.85	0.005	–	0.10	3.3E-04	–	0.01	20 ^f	160 ^g –1500 ^h
PFNA	0.002	–	0.48	7.5E-04	–	0.29	1.4E-04	–	0.03	9.4E-06	–	0.004	20 ^b	
PFDA	0.02	–	1.05	0.01	–	0.63	0.002	–	0.08	1.1E-04	–	0.008		
PFUdA	0.01	–	0.22	0.003	–	0.13	0.001	–	0.02	4.1E-05	–	0.002		
PFDoA	0.01	–	0.50	0.01	–	0.30	0.001	–	0.04	6.6E-05	–	0.004		
PFTrDA	0.01	–	0.62	0.004	–	0.37	0.001	–	0.05	5.4E-05	–	0.005		
PFTeDA	0.05	–	1.56	0.02	–	0.93	0.004	–	0.11	2.6E-04	–	0.01		
PFHxDA	0.04	–	0.42	0.01	–	0.25	0.003	–	0.03	1.8E-04	–	0.003		
PFODA	0.02	–	0.31	0.01	–	0.18	0.002	–	0.02	1.1E-04	–	0.002		
∑PFCA(<7)	0.15	–	2.69	0.06	–	1.62	0.01	–	0.20	7.4E-04	–	0.02		
∑PFCA(≥7)	0.36	–	6.19	0.14	–	3.71	0.03	–	0.45	0.002	–	0.05		
∑PFCS	0.52	–	11.8	0.21	–	7.05	0.04	–	0.85	0.003	–	0.09		
∑PFASs	0.69	–	12.9	0.28	–	7.74	0.05	–	0.94	0.003	–	0.10		

^a MDH, 2017

^b MassDEP, 2018.

^c U.S. EPA, 2014.

^d U.S. EPA, 2016a.

^e MDH, 2018.

^f U.S. EPA, 2016b.

^g FSANZ, 2016.

^h EFSA, 2008.

Belgium while no variations were found for the rest of analytes. Conversely, samples obtained from Spain indicated a decrease in PFOS use in this country. These tendencies could indicate a possible PFOS alternative use. In PFCAs cases, long-chain PFCAs still aroused higher concentrations than short-chain carboxylic acids in house dust from the three countries. To the best of the author's knowledge, this is the first study reporting PFAS concentration in house dust from Italian homes.

Positive correlations were found between PFOS and PFHxS. This result could suggest a potential common origin. However, it could not be identified related to building characteristics, occupant habits and/or outdoor surroundings. PFOS concentrations showed a positive association with the building edification age, which could highlight a decrease in the use of this chemical in Europe. On the other hand, PFHxS levels were associated with the percentage of the floor covered by textiles. ∑PFCAs, long-chain PFCAs, PFUdA, PFOA, and PFHpA concentrations obtained in homes located in industrial sites showed higher levels compared to urban or agricultural locations, revealing the industrial processes as a potential source of these chemicals in Europe. The type of building was revealed also as a factor to be taken into account in terms of PFHxS, PFTeDA, PFHxDA and PFODA house dust content. Dust collected from flats presented higher concentrations for these chemicals than detached houses. Finally, EDI_{dust ingestion} values calculated for toddlers and adults even in the worst case scenario were well below RfDs and TDIs. Nonetheless, for toddlers, the age class having higher exposure, the significance of dust ingestion to overall PFAS exposure could reach values of 50%, pointing out the house dust as a potential pollution source in indoor places.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2019.05.463>.

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