



Human dietary exposure to perfluoroalkyl substances in Catalonia, Spain. Temporal trend

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ABSTRACT

In this study, we assessed the levels of 18 perfluoroalkyl substances (PFASs) in the most widely consumed foodstuffs in Catalonia, Spain, as well as the total dietary intake of these compounds. Forty food items were analysed. Only perfluoropentanoic acid (PFPeA), perfluorohexadecanoic acid (PFHxDA) and perfluorooctanoicdecanoic acid (PFOcDA) were not detected in any sample. Perfluorooctane sulfonate (PFOS) was the compound found in the highest number of samples (33 out of 80), followed by perfluorooctanoic acid (PFOA), perfluoroheptanoic acid (PFHpA), perfluorohexane sulfonic acid (PFHxS), perfluorodecanoic acid (PFDA) and perfluorodecane sulfonic acid (PFDS). Fish and shellfish was the food group in which more PFASs were detected and where the highest PFAS concentrations were found. The highest dietary intakes corresponded to children, followed by male seniors, with values of 1787 and 1466 ng/day, respectively. For any of the age/gender groups of the population, the Tolerable Daily Intakes (TDIs) recommended by the EFSA were not exceeded. In general terms, PFAS levels found in the current study are lower than the concentrations recently reported in other countries.

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1. Introduction

Perfluoroalkyl acids (PFAAs) and their salts, PFASs (perfluoroalkyl sulfonic acids) and PFCAs (perfluoroalkyl carboxylic acids), are stable chemicals comprising a carbon chain surrounded by fluorine atoms with a functional group located at the end of the carbon chain. Because these substances repel oil, grease and water, they have wide consumer and industrial applications, including protective coating for fabrics and carpets, paper coatings, insecticides, paints, cosmetics and fire-fighting foams (Paul, Jones, & Sweetman, 2009). These substances are known globally as perfluoroalkyl substances (PFASs). Perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA) are the two PFASs made in the largest quantities. They are the most investigated and the most commonly detected. PFOS is classified as a persistent and bio-accumulative substance (OECD, 2002). The industrial production of PFOS and some of its derivatives was phased out by the major producer, 3M, in 2002, while the European Union (EU) banned most uses of this compound from 2008 (EC, 2006). PFOS has been very recently included in the EU list of priority substances (PS) in the field of water policy, i.e., the chemicals identified among those present-

ing a significant risk to or via the aquatic environment, according to the Water Framework Directive (EC, 2012). In 2009, PFOS was included in Annex B of the Stockholm Convention list of persistent organic pollutants (Buck et al., 2011). In turn, US EPA launched a voluntarily stewardship programme to reduce PFOA emitted and contained in products by 95% by 2010, as well as to work towards elimination by 2015 (US EPA, 2012). However, hundreds of related chemicals, such as homologues with shorter or longer alkyl chains, which potentially may degrade to PFASs, have not yet been regulated.

In general, PFASs are extremely persistent, bio-accumulative and of toxicological concern (Dewitt, Peden-Adams, Keller, & Germolec, 2012; Houde, De Silva, Muir, & Letcher, 2011; Suja, Pramanik, & Zain, 2009; Zhang et al., 2011). In recent years, a number of studies involving PFASs have focused on increasing general knowledge on their toxicity (Florentin, Deblonde, Diguio, Hautemaniere, & Hartemann, 2011; Fuentes, Colomina, Vicens, Franco-Pons, & Domingo, 2007; Fuentes, Vicens, Colomina, & Domingo, 2007; Johansson, Eriksson, & Viberg, 2009; Ribes, Fuentes, Torrente, Colomina, & Domingo, 2010), environmental distribution and fate (Houde et al., 2011; Martin, Whittle, Muir, & Mabury, 2004; Nakata et al., 2006; Yamashita et al., 2005), as well as potential human health risks of exposure to these pollutants, especially to PFOS and PFOA (Cornelis et al., 2012; Domingo, 2012; Egeghy & Lorber, 2011; Haug, Huber, Becher, & Thomsen, 2011; Liu et al., 2010; Zhao

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et al., 2011). However, significant gaps still exist on that knowledge.

Human exposure to PFASs, mainly PFOS and PFOA, is due to a variety of environmental and product-related sources. However, this exposure has been suggested to be mainly through the diet, including drinking water (D'Hollander, de Voogt, de Coen, & Bervoets, 2010; Domingo, 2012; Ericson et al., 2008; Fromme, Tittlemier, Völkel, Wilhelm, & Twardella, 2009; Kärrman et al., 2007, 2009; Picó, Farré, Llorca, & Barceló, 2011; van Asselt, Rietra, Römkens, & van Der Fels-Klerx, 2011). On the other hand, recent investigations have shown that PFASs are also present in house dust at levels that may represent an important pathway for human exposure (Cornelis et al., 2012; D'Hollander et al., 2010; Ericson Jogsten, Nadal, van Bavel, Lindström, & Domingo, 2012; Strynar & Lindstrom, 2008). In order to increase the general knowledge on PFASs, in 2006 we initiated in our laboratory a wide programme aimed at increasing the information on human health risks of these compounds. We assessed whether diet, including drinking water, could make a significant contribution to human exposure to PFASs, as well as the role that food processing and packaging could play as a source of PFASs through dietary intake (Ericson et al., 2008, 2009; Ericson, Nadal, van Bavel, Lindström, & Domingo, 2008; Jogsten et al., 2009). In addition, we measured the levels of PFASs in human blood, milk and the liver of subjects belonging to the population for which dietary exposure to these pollutants was assessed (Ericson et al., 2007; Kärrman et al., 2010). Recently, we also determined the concentrations of a number of PFASs in house dust and indoor air samples from selected homes in Catalonia, Spain (Ericson Jogsten et al., 2012). Based on previous studies on dietary intake and drinking water consumption, we noted that house dust and indoor air seem to contribute significantly less to PFAS exposure within the Catalan population.

The purpose of the present study was to establish the temporal trend in the levels of PFASs found in the most widely consumed foodstuffs in Catalonia, as well as the total dietary intake of these compounds. Food items belonging to the same food groups assessed in our previous survey (Ericson et al., 2008) and some additional foodstuffs were collected and analysed for various PFASs. Here, we present the concentrations of PFASs in a number of food items corresponding to this last survey, as well as the dietary intake of these pollutants by the population of Catalonia. Finally, current dietary intake is also compared with human dietary intakes of PFASs recently reported for various countries.

2. Materials and methods

2.1. Sampling

In September 2011, foods were purchased in 12 representative cities of Catalonia, all with more than 20,000 inhabitants: Barcelona, l'Hospitalet de Llobregat, Vilanova i la Geltrú, Mataró, Sabadell, Terrassa, Girona, Tarragona, Reus, Tortosa, Lleida and Manresa. Globally, these cities represent approximately 72% of the population of Catalonia. Food samples were obtained at each locality in 4 shops/stores of different size (local markets, small stores, supermarkets and big grocery stores). Foods selected for PFAS analysis were among the most consumed in Catalonia (Serra-Majem et al., 2003). Analysed food samples included a total of 40 items: meat (veal steak, loin of pork, chicken breast, and steak of lamb) and meat products (boiled ham, "Frankfurt"-type sausage, and cured ham); fish and shellfish (sardine, tuna, anchovy, swordfish, salmon, hake, red mullet, sole, cuttlefish, clam, mussel, and shrimp); vegetables and tubers (lettuce, tomato, potato, and carrot); fresh fruits (apple, orange, and banana); milk and dairy products (whole and semi-skimmed milk, yogurt, cheese I – low fat, cheese II – medium fat, and cheese III – extra fat); cereals (French bread, and pasta); pulses (lentils); industrial bakery (cookies); eggs (hen eggs); oils and fats (olive oil), and canned products (sardine and tuna). For each food item, two composite samples were prepared for analysis. Each composite sample consisted of 24 individual units. Only edible parts of each food item were included in the composites. Samples were freeze-dried at -80°C with a Cryodos Telstar lyophilizer for 24 h and then stored at -20°C until analysis of PFASs. The list of PFASs analysed is shown in Table 1.

2.2. Sample preparation and instrumental analysis

2.2.1. Chemicals

Thirteen PFCAs (C4–C14, C16, C18, $^{13}\text{C}_4$ -labeled C4, C6, C8–C12, $^{13}\text{C}_8$ -PFOA) and four PFASs (C4, C6, C8, C10, $^{13}\text{C}_4$ -labelled C6, C8, $^{13}\text{C}_8$ -PFOS) were obtained from Wellington Laboratories (Guelph, Ontario, Canada). Performance standard 7H-PFHpA (98% in methanol) was purchased from ABCR (Karlsruhe, Germany). Methanol and water were of HPLC grade and purchased from Fluka (Steinheim, Germany), Supelclean ENVI-carb (120/400 mesh) was purchased from Supelco (Bellafonte, PA, USA) and sodium acetate was purchased from E. Merck (Darmstadt, Germany). Laboratory

Table 1
Perfluoroalkyl substances, abbreviations, molecular formulae and traces monitored during mass spectrometric analysis.

Compound	Abbreviation	Molecular formula	Primary trace	Secondary trace
<i>Perfluoroalkyl carboxylic acids</i>	<i>PFCAs</i>			
Perfluorobutanoic acid	PFBA	$\text{C}_4\text{F}_7\text{O}_2\text{H}$	213 > 168.90	
Perfluoropentanoic acid	PFPeA	$\text{C}_5\text{F}_9\text{O}_2\text{H}$	263.2 > 218.9	
Perfluorohexanoic acid	PFHxA	$\text{C}_6\text{F}_{11}\text{CO}_2\text{H}$	313.25 > 269.25	
Perfluoroheptanoic acid	PFHpA	$\text{C}_7\text{F}_{13}\text{CO}_2\text{H}$	363.18 > 319	363.18 > 168.8
Perfluorooctanoic acid	PFOA	$\text{C}_8\text{F}_{17}\text{CO}_2\text{H}$	413.1 > 369.2	413.1 > 169.1
Perfluorononanoic acid	PFNA	$\text{C}_9\text{F}_{19}\text{CO}_2\text{H}$	462.99 > 419	462.99 > 219
Perfluorodecanoic acid	PFDA	$\text{C}_{10}\text{F}_{21}\text{CO}_2\text{H}$	513.15 > 469.15	513.05 > 168.8
Perfluoroundecanoic acid	PFUnDA	$\text{C}_{11}\text{F}_{23}\text{CO}_2\text{H}$	563 > 519.1	563 > 269.1
Perfluorododecanoic acid	PFDoDA	$\text{C}_{12}\text{F}_{25}\text{CO}_2\text{H}$	613.01 > 569	613.01 > 168.9
Perfluorotridecanoic acid	PFTriDA	$\text{C}_{13}\text{F}_{27}\text{CO}_2\text{H}$	663.09 > 619.10	663.09 > 169
Perfluorotetradecanoic acid	PFTeDA	$\text{C}_{14}\text{F}_{29}\text{CO}_2\text{H}$	713.24 > 669.1	713.24 > 169
Perfluorohexadecanoic acid	PFHxDA	$\text{C}_{16}\text{F}_{33}\text{CO}_2\text{H}$	813.13 > 769.2	813.3 > 169
Perfluorooctadecanoic acid	PFODaDA	$\text{C}_{18}\text{F}_{37}\text{CO}_2\text{H}$	913.2 > 869.2	913.2 > 169
<i>Perfluoroalkyl sulfonic acids</i>	<i>PFASs</i>			
Perfluorobutane sulfonic acid	PFBS	$\text{C}_4\text{F}_9\text{SO}_3^-$	299.2 > 98.85	299.2 > 79.8
Perfluorohexane sulfonic acid	PFHxS	$\text{C}_6\text{F}_{13}\text{SO}_3^-$	399.1 > 98.9	399.1 > 79.85
Perfluorooctane sulfonate	PFOS	$\text{C}_8\text{F}_{17}\text{SO}_3^-$	498.9 > 98.85	498.9 > 79.85
Perfluorodecane sulfonic acid	PFDS	$\text{C}_{10}\text{F}_{21}\text{SO}_3^-$	599 > 79.70	
Tetrahydroperfluorooctane sulfonic acid	THPFOS	$\text{C}_6\text{F}_{13}\text{C}_2\text{H}_4\text{SO}_3^-$	427.0 > 407	427.0 > 80.7

Table 2
Mean concentrations (in pg/g fw) of the 18 PFASs studied according to the groups of foodstuffs analysed.

Food groups	PFBA	PFPeA	PFBS	PFHxA	PFHpA	PFHxS	PFOA	THPFOS	PFNA	PFOS	PFDA	PFUnDA	PFDS	PFDoDA	PFTfDA	PFTeDA	PFHxDA	PFoCDA
Meat and meat products	<90	<12	<2.4	<30	280	3.2	<300	43	<79	34	<7.9	<29	<0.46	<8.7	<13	<13	<99	<86
Fish and seafood	<110	<14	54	67	270	45	2600	410	500	2700	190	430	8.7	60	210	67	<120	<100
Vegetables	<140	<19	13	<47	360	4.5	370	1100	<370	100	<37	140	<2.2	<41	<60	<60	<460	<400
Tubers	<110	<15	<3.0	<36	<340	<1.9	<360	<43	<95	<5.0	16	<35	<0.56	<10	<15	<15	<120	<100
Fruits	<110	<15	<3.0	<36	<350	<1.9	<360	<44	<96	<5.0	11	34	<0.56	<10	16	19	<120	<100
Eggs	<120	<16	<3.2	<39	200	<2.0	<390	<47	<100	<5.3	<10	<38	<0.60	<11	<17	<17	<130	<110
Milk	<150	<20	<4.1	<50	240	<2.6	390	<61	<130	<6.9	<13	46	<0.78	<15	<21	<22	<170	<140
Dairy products	68	<9.0	<1.8	31	170	<1.1	190	<27	40	<3.0	<3.3	12	<0.34	<6.4	<9.4	<9.5	<73	<63
Cereals	<38	<5.1	<1.0	<12	<120	<0.6	<120	<15	<33	<1.7	<3.3	<12	2.9	<3.6	<5.4	<5.4	<42	<36
Pulses	<78	<11	<2.1	<26	<250	<1.3	<260	<31	<68	6.1	<6.8	<25	0.40	<7.5	<11	<11	<86	<74
Oils	<43	<5.9	<1.2	<14	<140	<0.7	<140	<17	<38	1.1	<3.8	<14	<0.22	<4.2	<6.1	<6.2	<48	<41
Industrial bakery	<33	<4.4	<0.89	<11	<100	<0.56	<110	<13	<29	0.70	<2.9	<11	<0.17	<3.1	<4.6	<4.7	<36	<31

ware including filters was carefully rinsed with methanol before use.

Labelled extraction standards (1 ng PFCAs/PFSAs) were added to 10 g of freeze-dried food sample, and 10 ml 0.2 M sodium hydroxide in methanol was then added. After 30 min, 40 ml methanol was added and samples shaken vigorously for 20 min before addition of 1 ml 1 M hydrochloric acid. Samples were centrifuged at 5000g for 20 min and the supernatant was removed. The extraction was repeated with an additional 40 ml of methanol, the two supernatants were combined and evaporated using a rotary evaporator to 10 ml. Water was added to reach a methanol content of 40% prior to extraction using Waters Oasis weak anion exchange sorbent (6 cc, 150 mg) previously cleaned with 4 ml methanol and 4 ml of water. Samples were loaded onto the WAX cartridges at a speed of one drop per second. Prior to elution with 4 ml 2% ammonium hydroxide in methanol, WAX cartridges were eluted with 4 ml 25 mM acetate buffer solution, pH 4, and 4 ml 40% methanol. For lipid removal, samples were evaporated to a volume of 2 ml, 1 ml of hexane was added, and samples were shaken for 30 s. The supernatant was removed and discarded. This procedure was then repeated twice more. Additional clean-up was performed using 50 mg of Supelclean EnviCarb (120/400 mesh from Supelco, Bellefonte, PA, USA) added to the extracts. Extracts were filtered using a 2 µm nylon filter before setting the final volume to 500 µl, including labelled recovery standards to monitor the performance of the extraction standards and 300 µl 2 mM ammonium acetate.

2.2.2. Instrumental analysis and quality assurance

Levels of ionic PFASs were analysed using an ultra performance liquid chromatograph (UPLC) tandem mass spectrometer (MS/MS). The Acquity UPLC system was coupled to a Quattro Premier XE (Waters Corporation, Midford, USA) with an atmospheric electrospray interface operating in negative ion mode. Separation was performed on an Acquity BEH C18 column (2.1 × 100 mm, 1.7 µm) kept at 50 °C. A PFAS isolator column (Waters Corporation, Midford, USA) was inserted between the pump and the injector to retain any fluorochemicals originating from the UPLC system. Injection volume was set to 10 µl, and the flow rate was set to 300 µl/min. A gradient program was employed delivering mobile phases of 2 mM ammonium acetate in methanol and 2 mM ammonium acetate in water. Multiple reaction monitoring (MRM) of molecular anion [M-H]⁻ for PFCAs, [M]⁻ for PFSAs, and product ions [M-COOH]⁻ and [FSO₃]⁻ for carboxylates and sulfonates, respectively, was used. The concentration of the analytes in the samples was calculated using internal standard quantification. The internal standard closest in retention time was used for those compounds for which a corresponding labeled internal standard was not available. Authors participated in the 2009 worldwide inter-laboratory study on PFASs in fish muscle and water, as confirmation of the quality control in the analytical work. All analytes, their abbreviations and transitions used during instrumental analysis are presented in Table 1.

2.3. Intake of PFASs

Total dietary PFAS intake of each food group was calculated by summing the results of multiplying the respective PFAS concentration in each specific food item by the amount consumed of that item. Finally, total dietary intake of PFASs through the 40 foodstuffs included in this study was obtained by summing the respective intakes from each food group. When a concentration was under the limit of detection (LOD), daily intakes were calculated assuming that respective values would be equal to zero and to one-half of that LOD (ND = 0 and ND = 1/2 LOD, respectively). Dietary PFAS intake was estimated for the following age/gender

groups: children (6–9 years; assumed average body weight 24 kg), male adolescents (10–19 years; assumed average body weight 56 kg), female adolescents (10–19 years; assumed average body weight 53 kg), male adults (20–65 years; assumed average body weight 70 kg), female adults (20–65 years; assumed average body weight 55 kg), male seniors (>65 years; assumed average body weight 65 kg), and female seniors (>65 years; assumed average body weight 60 kg).

3. Results and discussion

Data corresponding to the 80 individually analysed composite food samples show that only PFPeA, PFHxDA and PFOcDA could not be detected in any sample. The remaining PFASs were detected in at least one sample (i.e., PFBA in semi-cured cheese). PFOS was the compound found in the highest number of samples (33 out of 80). PFOA, PFHpA, PFHxS, PFDA and PFDS were the compounds that, concurrently with PFOS, were detected in the greatest number of food samples. PFOA was found in anchovy, red mullet, sole, clams and mussels, as well as in lettuce, carrot, semi-skimmed milk and yogurt, while PFHpA could be detected in samples of swordfish, salmon, red mullet, sole and clams, as well as in lettuce, eggs, whole and semi-skimmed milk and yogurt. In turn, PFHxS was found in six species of fish and shellfish, as well as in boiled ham, “Frankfurt”-type sausages, canned sardine and lettuce. PFDA was detected in seven seafood species and in potatoes, apple, or-

ange and canned products (sardine and tuna), while PFDS could be found in six fish and shellfish species and also in canned tuna and pasta. The highest values of PFASs were found in fish and shellfish samples. These highest levels (fresh weight) were: 46,000 pg PFOA/g in a composite sample of mussels, 17,000 pg PFOA/g in a sample of clams, 13,000 pg PFOS/g in a sample of red mullet, 11,000 pg PFOS/g in a sample of sole, and 9300 pg PFOS/g in an anchovy sample. The remaining PFAS levels were all below 5000 pg/g, with the highest values always observed in fish and shellfish samples. The levels of detected PFASs in other groups of foodstuffs were considerably lower, ranging from 840 to 1100 pg THPFOS/g in a composite sample of lettuce and 0.68 to 0.71 pg PFOS/g in the composite samples of cookies.

The mean concentrations of the 18 PFASs in 12 groups of analysed foodstuffs are summarized in Table 2. It can be seen that fish and shellfish was the group in which more PFASs were detected and where the highest PFAS concentrations were found. PFOS and PFOA showed the highest levels, 2700 and 2600 pg/g fw, respectively, while in contrast, PFBA, PFPeA, PFHxDA and PFOcDA were the only PFASs in that group whose mean concentrations were under their respective LODs. The remaining food groups all showed PFAS levels notably lower than those of PFOS. Among these, 7 and 6 PFASs could be detected in vegetables and dairy products, respectively. In contrast, there were five food groups in which only one PFAS per group, mainly PFOS, was detected. Recently, we determined the levels of 13 PFASs in samples of seven different species of fish and shellfish (sardine, tuna, hake, red mul-

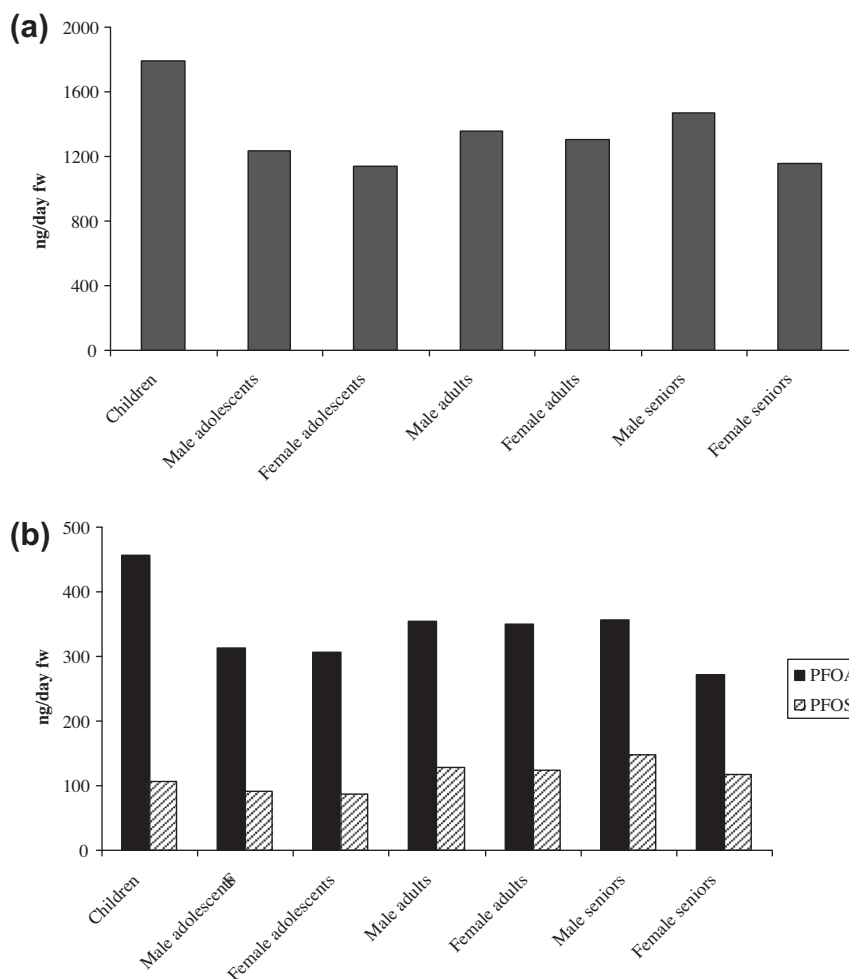


Fig. 1. (a) Dietary intake of total PFASs by the population of Catalonia. (b) Intake of PFOA and PFOS by the population of Catalonia.

Table 3
Dietary intake of total PFASs by the population of Catalonia (in ng/kg body weight/day).

		PFBA	PFBS	PFHxA	PFHpA	PFHxS	PFOA	THPFOS	PFNA	PFOS	PFDA	PFUnDA	PFDS	PFDoDA	PFTTrDA	PFTeDA
Children	ND = 0	0.03	0.10	0.10	0.34	0.08	4.73	3.03	0.65	4.24	0.35	0.79	0.03	0.07	0.31	0.12
	ND = 1/ 2 LOD	5.42	0.24	1.87	12.8	0.17	19.0	5.28	5.83	4.48	0.82	2.50	0.06	0.66	1.15	0.97
Boys adolescents	ND = 0	0.00	0.01	0.01	0.07	0.02	0.83	0.34	0.22	1.58	0.14	0.55	0.02	0.08	0.38	0.12
	ND = 1/ 2 LOD	1.63	0.05	0.54	4.01	0.04	5.59	0.97	1.52	1.65	0.27	0.97	0.02	0.23	0.61	0.34
Girls adolescents	ND = 0	0.00	0.01	0.01	0.08	0.02	1.64	0.51	0.21	1.57	0.12	0.68	0.02	0.10	0.50	0.16
	ND = 1/ 2 LOD	1.49	0.05	0.50	3.63	0.04	5.77	1.07	1.38	1.63	0.24	1.08	0.02	0.24	0.71	0.37
Male adults	ND = 0	0.00	0.02	0.01	0.06	0.02	1.55	0.60	0.22	1.80	0.14	0.67	0.01	0.10	0.54	0.18
	ND = 1/ 2 LOD	1.23	0.05	0.41	3.21	0.04	5.05	1.06	1.21	1.84	0.23	1.02	0.02	0.22	0.70	0.34
Female adults	ND = 0	0.00	0.04	0.02	0.08	0.03	2.33	0.80	0.28	2.20	0.17	0.80	0.01	0.12	0.64	0.21
	ND = 1/ 2 LOD	1.51	0.08	0.51	3.76	0.06	6.37	1.36	1.47	2.26	0.28	1.22	0.02	0.27	0.84	0.41
Male seniors	ND = 0	0.00	0.03	0.02	0.06	0.03	1.53	0.65	0.25	2.23	0.15	0.95	0.01	0.15	0.82	0.28
	ND = 1/ 2 LOD	1.41	0.07	0.48	3.73	0.06	5.50	1.18	1.39	2.29	0.24	1.36	0.02	0.28	1.01	0.47
Female seniors	ND = 0	0.00	0.02	0.01	0.07	0.02	1.10	0.47	0.26	1.92	0.15	0.69	0.01	0.11	0.57	0.19
	ND = 1/ 2 LOD	1.29	0.05	0.43	3.24	0.04	4.55	0.96	1.27	1.97	0.24	1.06	0.02	0.23	0.75	0.37

Calculations were not performed for those PFASs that were undetected in all food samples (PFPeA, PFHxDA and PFOcDA).

let, cuttlefish, mussel and prawn), all caught in coastal areas of Catalonia (unpublished data). Among the PFASs analysed, only seven compounds could be detected in at least one composite sample. PFBS, PFHxA, PFHpA, PFDS, PFDA and PFTDA were undetected in all samples. PFOS was, by far, the PFAS showing the highest mean concentration in fish and shellfish (2.70 ng/g fw), being detected in all species with the exception of mussels. High PFOS levels were found in sardine and red mullet (8.32 and 7.24 ng/g fw, respectively). With respect to PFOA (mean level, 0.074 ng/g fw), the highest concentrations were detected in prawn and hake (0.098 and 0.091 ng/g fw, respectively). The current mean concentration of PFOS in fish and shellfish (2.7 ng/g fw) is quite similar to that of our previous survey (2.70 ng/g fw). In contrast, the mean PFOA concentration (2.6 ng/g) was notably higher than that found recently (0.074 ng/g). The origin of the samples could explain this difference, as might the fact that the analysed fish and shellfish species were not the same in both investigations.

With respect to dietary exposure, Fig. 1 (top) depicts the total intakes of PFASs (Σ PFASs) corresponding to the seven groups in which the Catalan population was classified in the current study. It can be seen that the highest intakes corresponded to the group of children, followed by male seniors (aged >65), with values of 1787 and 1466 ng/day, respectively. However, it must be noted that the PFAS intake for children was calculated according to the daily consumption of the different food groups, not considering the individual foodstuffs as was carried out for the remaining age/gender groups. This might mean that the intake of PFASs could be slightly overestimated for children. On the other hand, female teenagers and female seniors, both with a similar value, were the age/gender groups showing the lowest intake of PFASs. If we consider only the intake of the two most well studied PFASs, PFOS and PFOA, which generally are also those mainly found in food samples, the highest and lowest intakes of PFOS corresponded to male seniors (149 ng/day) and female adolescents (86.3 ng/day), respectively (Fig. 1, bottom). For PFOA, children were the group showing the highest intake (456 ng/day), while female seniors showed the lowest intake (273 ng/day). The intakes of the 18 PFASs here analysed, expressed in ng/kg body weight/day, are given in Table 3. Intakes were estimated assuming both: ND = 0 and ND = 1/2 LOD. When we assumed that ND = 0, the highest and lowest PFOS intakes corresponded to children (4.24 ng/kg/day) and adolescents (1.58 and 1.57 ng/kg/day for males and females, respectively), while those concerning PFOA corresponded to chil-

dren (4.73 ng/kg/day) and female seniors (1.10 ng/kg/day), respectively. On the other hand, when the results were calculated assuming ND = 1/2 LOD, the highest and lowest PFOS intakes corresponded to children (4.48 ng/kg/day) and adolescents (1.63 and 1.65 ng/kg/day for males and females, respectively), while those concerning PFOA corresponded to children (19.0 ng/kg/day) and female seniors (4.55 ng/kg/day), respectively.

Recently, Domingo (2012) reviewed the state of the science regarding the concentrations of PFASs in foodstuffs, as well as human dietary exposure to these compounds and their health risks. It was found that information about exposure to PFASs through the diet was rather limited. In Europe, information was only available for Denmark, Spain, Norway, Germany, United Kingdom and Sweden, while in the rest of the world, only reports for Canada and USA in North and South America, and China and Japan in Asia, were found. No scientific reports from countries such as France, Italy, Austria, Finland or Switzerland in Europe, India in Asia, and Brazil, Mexico, Chile and Argentina in South America, or Australia, among many other countries, are currently available.

With respect to Spain, Ericson et al. (2008) determined the dietary intake of 11 PFASs by the population of Tarragona County (Catalonia). PFAS levels were measured in 36 composite samples of foodstuffs, which were purchased randomly in various locations of that County in July 2006. Exposure to PFASs through the diet was estimated for various age/gender groups. Only PFOS (in vegetables, fish and seafood, meats, eggs and dairy products), PFOA (in whole milk) and PFHpA (in whole milk) were detected in at least one food sample. For a standard adult man (70 kg body weight), the daily dietary intake of PFOS was estimated to be 62.5 or 74.2 ng (assuming ND = 0 or ND = 1/2 LOD, respectively). Fish, followed by dairy products and meats were the main contributors to PFOS intake. For an adult man, the intake of PFOS, and those of PFOA and PFHpA, was lower than those previously reported for Canada (Tittlemier et al., 2007), and considerably lower than that found in the United Kingdom (FSA, 2006), the only two countries where until the date in which our previous study (Ericson et al., 2008) was carried out, results concerning human dietary intake of PFASs had been estimated. In the present study, PFOS were detected in meat and meat products, fish and shellfish, vegetables and tubers, pulses, oils and bakery products. This leads to a dietary intake for an adult man of 126 or 129 ng/day (assuming ND = 0 or ND = 1/2 LOD, respectively), which is equivalent to 1.80 or 1.84 ng/kg/day (for a body weight of 70 kg). Regarding PFOA, the human daily intake through

the diet was 108 (ND = 0) or 354 ng (ND = 1/2 LOD), which is equivalent to 1.55 or 5.05 ng/kg/day, respectively. These intakes are considerably lower than the recommendations made by various international regulatory bodies. Thus, an evaluation of the TDIs for PFOS and PFOA was performed by the UK Committee on Toxicity of Chemicals in Food, Consumer Products and the Environment (COT, 2006a, 2006b). The COT recommended a TDI of 300 ng/kg for PFOS, while for PFOA a TDI of 3000 ng/kg was suggested. In turn, the CONTAM Panel of the EFSA recommended TDIs of 150 and 1500 ng/kg/day, for PFOS and PFOA, respectively (EFSA, 2008; EFSA, 2011). These latter values are lower than those previously proposed by the COT, but they are still notably higher than the daily intakes for PFOS and PFOA estimated in the present study. Although in this survey the number of analysed foodstuffs has been limited to 40, among these are those food groups and food items likely to contain the highest PFOS and PFOA concentrations, and, therefore, they would make the greatest contribution to the daily dietary intake to these compounds.

The potential role that some food processing and packaging might play as a source of PFASs through the diet was also investigated in our laboratory (Jogsten et al., 2009). The levels of 11 PFASs were determined in composite samples of veal steak (raw, grilled and fried), pork loin (raw, grilled and fried), chicken breast (raw, grilled and fried), black pudding (uncooked), liver lamb (raw), marinated salmon (home-made and packaged), lettuce (fresh and packaged), pate of pork liver, foie grass of duck, “Frankfurt”-type sausages, chicken nuggets (fried), and common salt. Only four PFASs (PFHxS, PFOS, PFHxA and PFOA) were detected in at least one composite sample. Once more, PFOS was the most frequently detected compound (8/20 of the food items analysed), while PFHxA was detected in samples of raw veal, chicken nuggets, “Frankfurt”-type sausages, and packaged lettuce. It was concluded, based on those results, that it was not sufficiently clear whether cooking with non-stick cookware, or the specific packaging of some foods, would contribute to significantly modify human exposure to PFASs.

With respect to recent studies performed in other countries, in Canada, Ostertag, Chan, Moisey, Dabeka, and Tittlemier (2009) measured the concentration of various PFASs in composite samples collected for the 1998 Health Canada Total Diet Study, and estimated dietary exposure for the Canadian population (older than 12 years of age) using previously collected dietary data ($n = 1721$). The exposure levels were below the provisional TDI provided by the German Drinking Water Commission (100 ng/kg for PFOS) (Fromme et al., 2009). The authors noted that dietary exposure to total PFASs had not changed over time. However, the contribution of PFOS to total PFAS exposure might have increased between 1998 and 2004. In Norway, Haug et al. (2010b) determined the levels of 19 PFASs in serum from 175 participants in the Norwegian Fish and Game Study, evaluating also the relationship with respect to food consumption by means of multiple linear regression analysis. It was found that PFAS concentrations in serum were significantly associated with consumption of lean fish, fish liver, shrimps and meat. The estimated dietary intakes of PFOA, PFUnDA, and PFOS were 0.60, 0.34 and 1.5 ng/kg body weight/day, respectively. As in most of the available studies, fish and shellfish was the group showing the major dietary source of the estimated intakes of PFOA (38%), PFUnDA (93%), and PFOS (81%). The authors observed that estimated dietary intakes of these three selected PFASs were significantly associated with corresponding serum levels. In one of our previous studies (Ericson et al., 2008), we also suggested a positive correlation between dietary intake and blood levels of PFOS, being corroborated in further analyses (Ericson Jogsten et al., 2012) in which drinking water was estimated as a relevant route only in extreme circumstances. However, according to those results we could not corroborate that dietary intake

was the main route of exposure governing blood concentrations of other PFASs. In another Norwegian study by the same researchers (Haug et al., 2010a), the concentrations of PFASs were determined in 21 samples of meat, fish, bread, vegetables, milk, drinking water and tea from the Norwegian market. Based on these 21 measurements together with consumption data for the general Norwegian population, a rough estimation of the total dietary intake of PFASs was found to be 100 ng/day. PFOA and PFOS contributed approximately 50% to the total intake. When dividing the population into gender and age groups, estimated intakes decreased with increasing age and was higher in males than females. The results of our current study do not support this last finding.

In a recent study performed in the Netherlands by Noorlander, van Leeuwen, Te Biesebeek, Mengelers, and Zeilmaker (2011), 6 out of 14 analysed PFASs could be quantified in the majority of the food categories evaluated (PFHpA, PFOA, PFNA, PFDA, PFHxS and PFOS). The highest concentration of the sum of these six compounds was also found in fish and shellfish: crustaceans (825 pg/g, PFOS: 582 pg/g) and lean fish (481 pg/g, PFOS: 308 pg/g). The median long-term intake for PFOS was 0.3 ng/kg bw/day and for PFOA, 0.2 ng/kg bw/day, values which are clearly lower than those estimated in the present study. However, the comparison among these dietary intakes is rather hard to make, as there are notable differences between both surveys in the food items analysed. In a Chinese study (Zhang et al., 2011), the estimated daily intake of PFOS and PFOA via fish and seafood consumption ranged from 0.10 to 2.51 ng/kg/day and from 0.13 to 0.38 ng/kg/day, respectively, for different age groups (i.e., toddlers, adolescents and children, and adults) from selected Chinese locations (i.e., Tianjin, Nanchang, Wuhan and Shenyang). On the other hand, in Flanders (Belgium) Cornelis et al. (2012) recently measured the concentrations of PFOS and PFOA in settled dust in homes and offices, in a selection of food items from local origin, in drinking-water, as well as in human serum. It was found that exposure to these compounds was dominated by food intake, while other exposure pathways contributed only marginally. Dietary exposure of children to PFOS was dominated by intake from potatoes (48%), followed by fish and seafood, dairy products, eggs and fruit (all contributing for about 10%). In adults, intake was dominated by fish and seafood (57%), followed by potatoes (28%). In turn, the intake of PFOA in children resulted mainly from fruits (30%) and vegetables (20%), with fish and seafood constituting only a small fraction, whereas the exposure of adults resulted from fish and seafood, potatoes, fruit and vegetables with almost equal contributions of about 20% from each item.

In summary, the results of the present survey corroborate that fish and shellfish is the food group showing the highest PFAS levels, in general, and those of PFOS and PFOA, in particular. PFOS was the most detected compound in the 80 analysed samples, while PFOA was detected in a lower number of samples (10 out of 80), but it was the compound in which the highest concentration of all PFASs was detected (a composite sample of mussels). In general terms, the PFAS levels found in the current study are lower than concentrations recently reported in other countries, being similar to those previously found in our laboratory. The differences among the present data and those from other countries may be due to the differences in the experimental designs (food items analysed, number of samples, PFASs analysed, food processing, etc.). Finally, and concerning human health risks from dietary exposure to PFASs in Catalonia, it is important to note that for any of the age/gender groups of population estimated, the TDIs recommended by the EFSA were not exceeded. As food consumption may be uncertain and/or variable, even for individuals within the same population group, probabilistic tools will be applied in future investigations in order to estimate the variability/uncertainty in the dietary exposure to PFASs.

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