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Per- and Polyfluoroalkyl Substances (PFASs) in Food and Human Dietary Intake: A Review of the Recent Scientific Literature

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ABSTRACT: Because of the important environmental presence and the potential human toxicity of per- and polyfluorinated alkyl substances (PFASs), in recent years the social and scientific interest in these compounds has notably increased. Special attention has been paid to perfluorooctanesulfonate (PFOS) and perfluorooctanoic acid (PFOA), the most extensively investigated PFASs. Although human exposure to PFASs may occur through different pathways, dietary intake seems to be the main route of exposure to these compounds. In 2012, we published a wide revision on the state of the science regarding the concentrations of PFASs in foodstuffs, the human dietary exposure to these compounds, and their health risks. In the present review, we have updated the information recently (2011−2016) published in the scientific literature. As in our previous review, we have also observed considerable differences in the PFASs detected—and their concentrations—in the food items analyzed in samples from a number of regions and countries. However, fish and other seafood seem to be the food group in which more PFASs are detected and where the concentrations of these compounds are higher. On the basis of the recommendations of the EFSA on the maximum dietary intakes of PFOS and PFOA, human health risks would not be of concern for nonoccupationally exposed populations, at least in the very limited countries for which recent data are available.

KEYWORDS: *per- and polyfluorinated alkyl substances (PFASs), PFOS, PFOA, dietary intake, health risks*

INTRODUCTION

PFASs and their derivatives are a group of chemicals with a great number of applications. They can offer resistance to heat, to other chemicals, or to abrasion, being also used as dispersion, wetting, or surface-treatment agents. Due to this, for over 60 years, the properties of these substances have been utilized in a very wide range of products and applications (food packaging, nonstick cookware, cleaning agents, surfactant, coating materials, etc.).[1]

However, the social and scientific interest/concern for their environmental impacts and the potential health risks in humans is much more recent. Thus, using Scopus (https://www.scopus.com/) as database and "perfluorinated compounds"—an old but more extended term to refer to PFASs—and "human health risks" as search terms, the first citation does not appear until 1989.[2] The second indexed paper was published 12 years after,[3] although it is not an experimental study. During that time, a review paper on extraperoxisomal targets of peroxisome proliferators, in which perfluorinated straight-chain monocarboxylic fatty acids are revised, can be also found.[4] The scientific information during subsequent years is also very scarce, ranging between two and seven citations in 2004 and 2009, respectively. In contrast, during the current decade, the number of citations has notably increased, with the maximum number (20) corresponding to 2013 (15 citations in 2015). Moreover, using "perfluorinated compounds" and "environmental impact" as terms of search, the number of citations has maintained a very similar trend, with 26 and 20 references in 2014 and 2015, respectively.

As for many other organic substances of environmental concern, food consumption is one of the main routes of human exposure to PFASs.[5−11] In recent years, a number of investigators have determined the concentrations of PFASs in foodstuffs, whereas some studies have also assessed the dietary intake of some of the most well-known PFASs, mainly PFOS and PFOA. In 2011, we assessed the state-of-the-art regarding human health risks of the dietary exposure to PFASs.[7] Because of the interest that this topic still generates in both public opinion and the scientific community, we have now updated the existing information related to this topic.

PFAS CONCENTRATIONS IN FOOD AND DIETARY INTAKE IN A NUMBER OF COUNTRIES

PubMed (https://www.ncbi.nlm.nih.gov/pubmed) and Scopus were used as databases, with the search terms "PFASs and food" and "PFASs and dietary intake". Notwithstanding, a complementary search was carried out by using the old term "perfluorinated compounds". Publications from regulatory organizations and other authorities, as well as gray literature available through the Internet, were intentionally excluded. Therefore, this review was focused only on scientific publications. The period of this new review covered between May 2011 and October 2016. Although only scientific publications within this period were gathered, some of the reported data may be referred to samples collected or analyzed in previous years.[12] In general terms, most of the studies here reviewed have been carried out in European countries, only some of them being conducted in North America and Asia (Table 1). Besides checking papers reporting the levels of

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Table 1. Summary of Recent (2011−2016) Publications on the Occurrence of PFASs in Food and Their Dietary Intake by Populations in Several Countries

country	characteristics of the study	PFASs assessed	remarks	dietary intake (in ng/kg bw/day)[a]	ref
Germany	analysis of PFASs in samples of fish fillet and correlation with plasma levels of anglers at Lake Möhne, Sauerland	10 PFASs, including PFOS and PFOA	dose-dependent relationship observed between fish consumption and internal exposure to PFOS	PFOS: max, 17	Hölzer et al.[13]
The Netherlands	determination of PFASs in food from Dutch reatil sotre chains and assessment of the dietary intake	14 PFASs, including PFOS and PFOA	crustaceans and lean fish showed the highest concentrations of the quantified PFASs	PFOS: median, 0.3; high-level intake, 0.6	Noorlander et al.[14]
				PFOA: median, 0.2; high-level intake, 0.5	
Spain	human exposure to PFASs through foodstuffs of wide consumption in Catalonia	18 PFASs, including PFOS and PFOA	fish and shellfish comprised the food group showing the highest levels of PFASs, in general, and those of PFOS and PFOA, in particular	adults: PFOS, 1.84; PFOA, 5.05	Domingo et al.[15]
	analysis of PFASs in samples of fish and shellfish collected from coastal areas of Catalonia, as well as in drinking water	13 PFASs, including PFOS and PFOA	PFOS was the individual compound with the greatest concentrations in fish and shellfish	PFOS: mean, 1.02 through food; 0.18 through water	Domingo et al.[16]
France	PFAS determination in mollusks collected along the coasts on mainland France	17 PFASs, including PFOS and PFOA	highest levels of PFOS were found in samples from the English Channel, contrasting with those from the Mediterranean coast	na	Munschy et al.[17]
	health risk assessment related to the presence of PFASs in food collected for the 2n French TDS	16 PFASs, including PFOS and PFOA	max levels of PFOS and PFOA in mollusks and crustaceans	means: PFOS, 0.66; PFOA, 0.74	Rivière et al.[18]
				95th percentile: PFOS, 1.15; PFOA, 1.50	
	dietary intake of PFASs for three different groups of French adult populations	15 PFASs, including PFOS and PFOA	PFAS levels in freshwater fish were higher than those in marine fish and seafood; PFOS was the largest contributor in freshwater fish	highest PFOS intake: freshwater fish consumers (7.5)	Yamada et al.[20]
				highest PFOA intake: high seafood consumers (1.2)	
	risk/benefit analysis of fish consumption by comparing PFAS levels and fatty acid composition	15 PFASs, including PFOS and PFOA	carp, perch, and mullet presented the most unfavorable balance profile due to their high level of PFASs and low level of n-3 long-chain polyunsaturated fatty acids (LC-PUFAs)	na	Yamada et al.[21]
Sweden	estimation of the dietary intake of PFASs by analyzing a set of archived food market basket samples	11 PFASs, including PFOS and PFOA	dietary intake of PFOS and PFOA was higher than estimated exposure through dust ingestion and drinking water; dietary intake has been fairly constant over the past decade	PFOS: 1.44, 0.86, and 1.00 in 1999, 2005 and 2010, respectively	Vestergren et al.[22]
				PFOA: 0.35, 0.50, and 0.69 in 1999, 2005, and 2010, respectively	
	analysis of PFASs in archive samples of eggs, milk, and farmed rainbow trout	11 PFASs, including PFOS and PFOA	PFOS levels in fish and eggs, as well as PFOA in eggs, decreased significantly with time	na	Johansson et al.[23]
	determination of PFASs and precursor compounds in archived food market basket samples	15 PFASs, including PFOS and PFOA, and a number of precursor compounds, including FOSA, FOSAA and diPAPs	all precursors were predominantly found in meat, fish, and/or eggs; PFOS precursors contributed 1−4% as indirect source of PFOS intake	PFOS and precursors: 1.64, 0.88, and 0.73 in 1999, 2005, and 2010, respectively	Gebbink et al.[12]
4 European countries (Belgium, Czech Republic, Italy and Norway)	design of a harmonized sampling survey to analyze PFASs in 20 vegetable species	14 PFASs, including PFOS and PFOA	perfluorinated carboxylic acids (PFCAs) were the main group of detected PFASs; intake of potatoes governed exposure to PFOA	PFOA (only vegetables): 0.040	Herzke et al.[25]
	PFAS determination in 50 selected pooled samples representing 15 food commodities	21 PFASs, including PFOS and PFOA	PFOS was the most frequently detected analyte; country profile: Belgium ≫ Norway, Italy > Czech Republic	mean intake of PFOS: fish, 0.03−0.11; seafood, 0.03−0.05; liver, 0.01−0.03	Hlouskova et al.[26]
	PFAS determination in fruits, cereals, sweets, and salt	12 PFASs, including PFOS and PFOA	PFOA was the most abundant compound, whereas PFOS showed the greatest concentrations	mean for high consumers (95th percentile): PFOS, 1.00; PFOA, 0.35	D'Hollander et al.[28]

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Table 1. continued

country	characteristics of the study	PFASs assessed	remarks	dietary intake (in ng/kg bw/day)[a]	ref
	dietary exposure to PFASs according to data from the PERFOOD EU project	7 selected PFASs	foods of plant origin are most important for dietary exposure to PFOA, whereas foods of animal and plant origins contribute equally to PFOS intake	estimated average dietary intake for adults and children is generally below or close to 1	Klenow et al.[27]
Italy	study of the presence of PFASs in meals served weekly for lunch in Italian school canteens	7 PFASs, including PFOS and PFOA	negligible contribution from food processing and serving to meal contamination	PFOS: 0.5−1.4 PFOA: 0.3−1.1	Dellatte et al.[29]
	analysis of PFASs in human milk and food samples from the city of Sienna and its province	PFOS and PFOA	only PFOS was found in food samples; fish showed the greatest levels	na	Guerranti et al.[30]
	PFAS determination in samples of European whitefish and European perch from Lake Maggiore	PFOS and PFOA	PFOS was more frequently detected in freshwater fish samples than PFOA	PFOS: median, 11.9; 95th percentile, 54.39	Squadrone et al.[31]
	PFAS determination in samples of European perch from Lake Varese	PFOS and PFOA	results did not show a particularly alarming level of PFAS pollution	PFOS: median, 5.15; 95th percentile, 23.55	Squadrone et al.[32]
	monitoring the presence of PFASs in Italian cow's milk	PFOS and PFOA	contamination by PFOS was frequent, but at low concentrations	na	Barbarossa et al.[33]
Greece	analysis of PFASs in raw and cooked samples of several species of Mediterranean finfish and shellfish	12 PFASs, including PFOS and PFOA	PFOS was the predominant compound; PFAS levels usually higher after cooking	PFOS: max in picarel, 10.48 PFOA: max in shrimp, 0.20	Vassiliadou et al.[34]
Greece and The Netherlands	PFAS analysis in home-produced and commercially produced chicken eggs	11 PFASs, including PFOS and PFOA	median levels of PFASs in eggs from The Netherlands were almost 3 times higher than those from Greece	PFOS (only eggs) children, median 3.5, max 24.8 adults: median 1.1, max 7.6	Zafeiraki et al.[35]
Finland	determination of PFASs in various edible fish species	13 PFASs, including PFOS and PFOA	PFASs were present in all Baltic Sea and freshwater species, but not in farmed fish; PFOS was the most abundant compound	na	Koponen et al.[36]
Faroe Islands	analysis of PFASs in food samples and purified drinking water	15 PFASs, including PFOS and PFOA	PFOS was most frequently detected compound	na	Eriksson et al.[37]
Greenland	contamination levels of PFASs and other POPs in traditional seafood items and assessment of the dietary intake	14 PFASs, including PFOS and PFOA	PFASs were below detection limits in most fish fillet samples, varying from 2.9 ng/g ww in whale beef to 13.5 ng/g ww in seal beef	PFOS: 6.89	Carlsson et al.[38]
China	risk assessment of PFAS exposure through consumption of freshwater and marine fish from Hong Kong and Wiamen	10 PFASs, including PFOS and PFOA	risks and potential effects of PFASs for the health of the coastal population in the Pearl River Delta are of concern	means: PFOS (Hong Kong) 2.4; PFOS (Xiamen) 5.1; PFOA (Hong Kong) 3.3; PFOA (Xiamen) 3.0	Zhao et al.[40]
	contribution of food to the exposure to PFASs and comparison with ingestion through drinking water and indoor dust	10 PFASs, including PFOS and PFOA	drinking water was a minor source of PFOS exposure	PFOS: 0.10−2.51 PFOA: 0.13−0.38	Zhang et al.[41]
	determination of PFAS levels in samples of fatty fish and shellfish from Chinese coastal areas	13 PFASs, including PFOS and PFOA	PFOS was the dominant compound in fatty fish, whereas PFAS was the most abundant in shellfish	PFOS: 0.037−0.694 PFOA: 0.008−0.914	Wu et al.[42]
	PFAS determination in seafood samples collected in Bohai Bay	14 PFASs, including PFOS and PFOA	PFOS and PFOA were the most frequent compounds; species at higher trophic levels accumulate more PFASs than benthic invertebrates	PFOS: 2.44 PFOA: 0.50	Yang et al.[43]
	PFAS determination in a number of tissues of popular farmed freshwater fish from Beijing	14 PFASs, including PFOS and PFOA	overall tissue distribution of PFOS: fish blood > liver > brain > muscle	PFOS: 0.24 total PFASs: 0.44	Shi et al.[44]

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Table 1. continued

country	characteristics of the study	PFASs assessed	remarks	dietary intake (in ng/kg bw/day)[a]	ref
	residues of PFASs investigated in fish samples from Danjiangkou reservoir and Hamjiang River	8 PFASs, including PFOS and PFOA	frequent consumption of yellow croaker fish may pose an unacceptable risk to human health	PFOS: urban, 0.42–0.84; rural, 0.37 PFOA: urban, 0.02–0.18; rural, 0.06	He et al.[45]
South Korea	analysis of PFASs in 397 food items, of 66 types, and evaluation of the dietary intake	16 PFASs, including PFOS and PFOA	fish and dairy products were the major contributors of PFOS and PFOA, respectively; tap water intake might have also a key role in PFOA intake	PFOS: 0.47–3.03 PFOA: 0.17–1.68	Heo et al.[47]
	correlation study of PFASs in blood serum and composite diet samples	16 PFASs, including PFOS and PFOA	because of the low number of samples, no correlations between PFAS levels in diet and serum were found	na	Kim et al.[48]
Canada	PFAS contamination in sport fish species collected downstream of the Hamilton International Airport, Ontario	10 PFASs, including PFOS and PFOA	PFOS dominated the PFAS profile in the different fish species and sampling locations	na	Gewurtz et al.[49]
	role of cooking for reducing exposure to PFASs studied by analyzing raw, baked, broiled, and fried fish fillets	12 PFASs, including PFOS and PFOA, as well as diPAPs	relatively minor differences in changes in the fish PFAS amounts after cooking depended on fish species and cooking method used	na	Bhavsar et al.[50]
United States	greenhouse study to investigate the potential uptake of PFASs by lettuce and strawberry	9 PFASs, including PFOS and PFOA	PFASs can enter and bioaccumulate in food crops irrigated with reclaimed water	na	Blaine et al.[53]
	characterization of PFASs in freshwater fish on a national scale	13 PFASs, including PFOS and PFOA	PFOS was the most frequently detected compound	na	Stahl et al.[54]
4 countries: Brazil, Saudi Arabia, Spain, and Serbia	analysis of PFASs in 283 food items from representative countries of the diet in South America, western Asia, Mediterranean area, and southeastern Europe	21 PFASs, including PFOS and PFOA	fish and seafood were identified as the major PFASs contributors to the diet in all countries	total PFASs: 33–54	Pérez et al.[55]

[a]When not provided in these units, the dietary intake for the adults was recalculated by assuming 70 kg as body weight. na, not available.

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PFASs in foodstuffs, this review was particularly aimed at evaluating the state of the art on the dietary intake of PFASs, which is estimated by multiplying PFAS concentrations and food consumption data.

European Countries. *Germany.* Our search terms produced one published paper during the assessed period. It belongs to Hölzer et al.,[13] who measured the concentrations of various PFASs in samples of several fish species collected at Lake Möhne (Sauerland area). The levels of PFOS ranged between 4.5 and 150 ng/g, the highest median PFOS concentrations being found in perches (96 ng/g) and eels (77 ng/g), followed by pikes (37 ng/g), whitefish (34 ng/g), and roaches (6.1 ng/g). Interestingly, in a food surveillance program only 11% of fishes at retail sale were found to contain PFOS at detectable levels. In this same study, plasma samples of anglers of the zone were also collected and analyzed for the same PFASs. For PFOS, the concentrations in plasma of anglers consuming fish two to three times per month were 7 times higher, compared to those without any fish consumption from Lake Möhne.

The Netherlands. Noorlander et al.[14] determined the levels of 14 PFASs in foodstuffs (pooled samples) purchased in several Dutch retail store chains with nationwide coverage. Six PFASs (PFHpA, PFOA, PFNA, PFDA, PFHxS, and PFOS) could be quantified in most food categories. The highest concentration of the sum of these six compounds was found in crustaceans (825 pg/g product; PFOS, 582 pg/g product) and in lean fish (481 pg/g product; PFOS, 308 pg/g product). In contrast, beef, fatty fish, flour, butter, eggs, and cheese showed lower concentrations (20 and 100 pg/g product; PFOS, 29−82 pg/g product). Milk, pork, bakery products, chicken, vegetable, and industrial oils showed concentrations <10 pg/g product (PFOS not detected). The median long-term intake for PFOS was 0.3 ng/kg body weight (bw)/day and for PFOA, 0.2 ng/kg bw/day. These intakes were well below the TDI values of both compounds (PFOS, 150 ng/kg bw/day; PFOA, 1500 ng/kg bw/day). The results of that survey showed that vegetables/fruits and flour were important contributors of PFOA intake, whereas milk, beef, and lean fish were important contributors of PFOS intake.

Spain. Human exposure to 18 PFASs through the most widely consumed foodstuffs in Catalonia (Spain) was assessed by Domingo et al.[15] Two samples of 40 different food items were analyzed. PFOS was the compound found in the highest number of samples (33 of 80), followed by PFOA, PFHpA, PFHxS, PFDA, and PFDS. Fish and shellfish comprised the food group in which more PFASs were detected and in which 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. However, the TDIs recommended by the EFSA (150 and 1500 ng/kg bw/day for PFOS and PFOA, respectively),[10] were not exceeded for any of the age/gender groups of the population of Catalonia. When the PFAS levels found in that survey were compared with those previously reported for other countries, the concentrations found in foodstuffs purchased from Catalonia were generally lower.[7] Additionally, the concentrations of 13 PFASs were also determined in samples of fish and shellfish collected from coastal areas of Catalonia.[16] Only seven PFASs were detected in at least one composite sample, with PFOS showing the highest mean concentration (2.70 ng/g fresh weight (fw)) and being detected in all species with the exception of mussels. In turn, the highest levels of PFOA (mean = 0.074 ng/g fw) corresponded to prawn and hake (0.098 and 0.091 ng/g fw, respectively). The intake of PFASs through the consumption of the analyzed foodstuffs was below the recommended TDI for those compounds for which information is currently available.

France. Munschy et al.[17] determined the levels of PFASs in mollusks collected along the coasts on mainland France (Atlantic coast, Mediterranean Sea, and English Channel). The median concentrations of PFOS were 0.18, 0.09, and 0.04 ng/g wet weight (ww) in samples from the English Channel, the Atlantic coast, and the Mediterranean coast, respectively. No estimation on human exposure to PFASs through mollusk consumption was performed in that study. However, Rivière and co-workers[18] assessed the human health risks related to the presence of 16 PFASs measured in food samples collected for the second Total Diet Study (TDS) performed in France.[19] PFOA was quantified in meats, poultry, and game, delicatessen meats, seafood products, vegetables (excluding potatoes), water, and mixed dishes. The highest mean concentrations were found in mollusks and crustaceans (0.044 μg/kg fw), whereas PFOS was quantified in meats, delicatessen meats, seafood products, vegetables, water, and mixed dishes. As for PFOA, the highest mean concentrations were also noted in mollusks and crustaceans (0.19 μg/kg fw). In adults, mean exposures to PFOA and PFOS were estimated at 0.74 and 0.66 ng/kg bw/day, respectively. At the 95th percentile, exposures were estimated at 1.50 and 1.15 ng/kg bw/day for PFOA and PFOS, respectively. These values should not mean risks for human health. However, due to the lack of information for the other PFASs, for which toxicological properties are not well-known, health risks could not be properly evaluated for compounds other than PFOS and PFOA. In another study, Yamada et al.[20] assessed the dietary exposure to 15 PFASs for three different groups of French adult populations: high seafood consumers, high freshwater fish consumers, and pregnant women. The analysis of fish samples showed that freshwater fish was the most contaminated group, with a level up to 168.4 ng/g ww (sum of 15 analyzed PFASs: PFBA, PFPA, PFHxA, PFHpA, PFOA, PFNA, PFDA, PFUnA, PFTrDA, PFTeDA, PFBS, PFHxS, PFHpS, PFOS, and PFDS), followed by marine fish and seafood (6.8 and 7 ng/g ww, respectively). Freshwater fish contamination was mainly due to PFOS (75%), whereas that of marine fish was due to PFOA (24%), PFOS (20%), PFHxA (15%), PFHpA (11%), and PFBA (11%).[21] It was found that high freshwater fish consumers were the most exposed to PFOS (7.5 ng/kg bw/day), PFUnA (1.3 ng/kg bw/day), PFDA (0.4 ng/kg bw/day), and PFHpS (0.03 ng/kg bw/day). In turn, high seafood consumers were the most exposed to PFOA (1.2 ng/kg bw/day), PFNA (0.2 ng/kg bw/day), and PFHxS (0.06 ng/kg bw/day).

Sweden. Vestergren and co-workers[22] estimated the dietary intake of PFASs using a highly sensitive method to analyze a set of archived (1999, 2005, and 2010) food market basket samples. Fish and meat were the main contributors to the dietary exposure to PFOS (860−1440 pg/kg bw/day), PFUnDA (90−210 pg/kg bw/day), PFDA (50−110 pg kg bw/day), and PFNA (70−80 pg/kg bw/day), but not for PFOA (350−690 pg/kg bw/day). An interesting conclusion of that survey was that the dietary intake of PFASs had been rather constant over the past decade, in which important manufacturing changes occurred. However, in a subsequent study conducted by the same researchers,[23] in which archived

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samples (1999–2010) of eggs, milk, and farmed rainbow trout were analyzed for determining the levels of PFASs, significant decreasing trends were observed for the levels of PFOS and PFHxS in fish and eggs. The levels of PFOS in fish and eggs decreased by factors of 10 and 40, respectively, whereas in eggs there was also a significant decreasing trend in the levels of PFOA. In a recent study by the same research group,[12] in which archived food samples were again analyzed, PFOS precursors were detected in all food year (1999, 2005, and 2010) pools, with the highest concentrations corresponding to 1999. Six diPAPs were detected in the year pools with the highest ΣdiPAP concentrations found in 1999 and 2005. All precursors were predominantly found in meat, fish, and eggs, based on analysis of individual food groups from 1999, whereas the lowest precursor contributions were generally found in food samples from 2010. Very recently, Sjogren and co-workers[24] reported the results of a study having as its main objective the examination of the cross-sectional relationship between blood levels in an elderly Swedish population of selected PFASs and adherence to three predefined dietary patterns: (a) a WHO-recommended diet, (b) a Mediterranean-like diet, and (c) an LCHP diet. In that study, which included 844 elderly Swedish men and women, all dietary patterns were positively associated with blood levels of PFASs, the highest body burden of these compounds being found in individuals with high adherence to a Mediterranean-like diet, which is characterized by high fish consumption. In contrast, subjects who more closely followed the officially recommended diet showed a lower body burden of PFASs.

Joint Study in Four European Countries (Belgium, Czech Republic, Italy, and Norway). Herzke et al.[25] designed a harmonized sampling survey collecting similar food items in a uniform procedure, enabling direct comparison between four European countries: Belgium, Czech Republic, Italy, and Norway. In a first study of that survey, only vegetables (20 species) were analyzed. In addition to fresh vegetables, processed vegetable products such as ready-to-cook pommes frites, frozen minced spinach, and mixed lettuce were also sampled and analyzed for PFAS concentrations. Only low levels of PFASs were detected in all analyzed samples. PFCAs were the main group of detected PFASs, PFOA being the most abundant PFCA (with the exception of samples from Czech Republic). However, dietary intake estimates for PFOA showed low human exposure due to vegetable consumption for adults and children. In a parallel study, Hlouskova et al.[26] measured the concentrations of 21 PFASs in samples of meat, fish, and seafood, milk and dairy products, and eggs. Five PFASs were not detected in any of the analyzed samples. PFOS was the most frequently detected analyte (range = 0.98–2600 ng/kg). The concentration ranges of individual compounds were the following: 2.33–76.3 ng/kg for PFSAs (without PFOS), 4.99–961 ng/kg for PFCAs, 10.6–95.4 ng/kg for PFPAs, and 1.61–519 ng/kg for FOSA. The levels of PFASs in the analyzed foodstuffs decreased in the order seafood > pig/bovine liver ≫ freshwater/marine fish > hen egg > meat ≫ butter. Subsequently, dietary exposure to seven selected PFASs (including PFOS and PFOA) was estimated for the population of the Czech Republic, Italy, Belgium, and Norway, the countries where the food samples were collected.[27] For both adults and children, the intake was generally below or close to 1 ng/kg bw/day for all of the selected PFASs. According to the TDIs proposed by the EFSA[10] for PFOS (150 ng/kg bw/day) and PFOA (1500 ng/kg bw/day), no concerns were identified. As expected, the different dietary exposure patterns among countries, which is a consequence of different food consumption and contamination patterns, showed different results among countries. Fruits and vegetables were the most important contributors to the dietary exposure to PFOA, whereas for PFOS, food of animal and plant origin showed a rather similar contribution. In a recent study also conducted in the context of the same European project, D'Hollander et al.[28] determined the concentrations of 12 PFASs in samples of 14 food items (fruits, cereals, sweets, and salt) collected in the same four European countries. Ten PFASs could be detected in 67% of the analyzed samples. PFOA was the most abundant compound (detection frequency of 49%) followed by PFOS (29%), whereas PFHxA and PFNA were found in 20% of the samples. The highest concentration corresponded to PFOS (539 pg/g), followed by PFNA, PFHxS, PFHpA, PFHxA, and PFOA, all showing similar maximum levels (approximately 200 pg/g). The dietary intake of PFOS and PFOA was clearly below the tolerable levels,[10] although they contributed to the total daily intake.

Italy. The dietary intake by children of selected PFASs (PFHxA, PFOA, PFNA, PFDA, PFUnDA, PFHxS, and PFOS) through meals served at school canteens was estimated by Dellatte et al.[29] For PFOA and PFOS, Italian school-age children showed intakes ranging from 0.3 to 1.1 ng/kg bw/day and from 0.5 to 1.4 ng/kg bw/day, respectively,[29] well below the corresponding TDIs (1500 and 150 ng/kg bw/day, respectively).[10] In another Italian study,[30] the concentrations of PFOS and PFOA were determined in samples of cereal-based products, fish and seafood, meat, eggs, milk and dairy products, fruits and vegetables, oils and fats, fruit juices, and honey purchased from the city of Siena. For PFOS, fish samples were the most contaminated (7.65 ng/g), whereas mean levels in meat and milk and dairy products were similar (1.43 and 1.35 ng/g, respectively). However, in samples of the remaining foodstuffs (cereal-based foods, eggs, vegetables, honey, and beverages), PFOS could not be detected. The estimation of the dietary intake of PFOS was not carried out in that study. Interestingly, PFOA could not be detected in any food sample. On the other hand, and with respect to freshwater fish, Squadrone et al.[31] measured PFOS and PFOA in samples of European whitefish and European perch collected from Lake Maggiore. PFOA was not detected in any sample, but PFOS was found in all samples, with levels of up to 46.0 ng/g fw. The authors concluded that PFOS intake through fish consumption from Lake Maggiore could be a significant source of dietary PFOS exposure. A similar study was also done with samples of the same species collected from Lake Varese. PFOS was detected in all samples with concentrations up to 17.2 ng/g. The conclusion was completely similar: fish from Lake Varese could be also a significant source for dietary PFOS exposure.[32] On the other hand, Barbarossa et al.[33] analyzed the concentrations of PFOS and PFOA in 67 samples of different types of cow's milk from Italy. PFOS was often present, but at relatively low concentrations (up to 97 ng/L). In contrast, PFOA was rarely found. According to the authors, their results, as well as previous data from the scientific literature, would indicate that cow's milk should not be a major source of PFASs, compared with other foods such as fish and seafood.

Greece. Vassiliadou and co-workers[34] determined the concentrations of 12 PFASs in 7 species of finfish (anchovy, bogue, hake, picarel, sardine, sand smelt, and striped mullet) and 3 species of other seafood (mussel, shrimp, and squid).

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Analyses of PFASs were conducted not only in raw samples but also in samples of these species fried in olive oil or grilled. PFASs above the detection limit were found in all raw samples with the exceptions of sardine, mussel, and squid. PFOS was the most abundant compound, with its highest level detected in picarel (20.4 ng/g fw). With respect to the effects of cooking, the PFAS levels were usually higher after frying or grilling than in raw samples. The estimated daily intake for PFOS and PFOA through consumption of the fish and seafood species analyzed in that survey was found to be well below the values proposed by the EFSA.[10] More recently, Zafeiraki et al.[35] analyzed the content of PFASs in home-produced and commercially produced eggs of chickend from Greece and The Netherlands. The egg yolks were analyzed for 11 PFASs by liquid chromatography−tandem mass spectrometry using isotope dilution. PFAS levels in yolk were higher in home-produced eggs from The Netherlands (median = 3.1; range, < LOQ−31.2 ng/g) and Greece (median = 1.1; range, <LOQ−15.0 ng/g) compared to the eggs collected from supermarkets.

Finland. Koponen et al.[36] determined the concentrations of PFASs in fish species—collected from the Baltic Sea—commonly consumed in Finland. The main goal of the study was to collect data to be used for future dietary intake of PFASs and human exposure assessments. The species analyzed were Baltic herring, pike-perch, perch, burbot, whitefish, salmon, and vendace. In addition, perch and pike-perch were collected from Helsinki Vanhankaupunginlahti bay, whereas perch was also collected from Lake Päijänne. Farmed fish species included whitefish and rainbow trout. As found in most recent studies on the same topic, PFOS was again the most abundant compound in each fish species. Another six PFASs (PFOA, PFNA, PFDA, PFUnA, PFDoA, and PFTrA) were also detected in fish samples. It was noted that farmed fish in Finland was not a significant source of PFAS for humans, although the importance of fish on the total PFAS intake and human exposure in Finnish population was unclear.

Faroe Islands and Greenland. In locally produced food items from the Faroe Islands, Eriksson et al.[37] analyzed the levels of 15 PFASs collected in 2011/2012. Food samples included dairy products (milk, yogurt, and crème fraiche), fish, and potatoes. Of the 15 PFASs measured, 13 could be detected in at least one sample. The frequency detection of PFASs in food items was PFOS > PFUnDA > PFNA > PFOA, but PFUnDA predominated in milk and wild fish (mean concentrations = 170 pg/g). Although no exposure assessment was conducted in that survey, the authors concluded that consumption of the analyzed food items was not expected to exceed the TDI proposed by the EFSA[10] for PFOS and PFOA. On the other hand, Carlsson et al.[38] measured the concentrations of PCBs, PBDEs, and PFASs in traditional Greenlandic seafood items: fresh salmon fillet, smoked salmon fillet, smoked halibut fillet, commercial whale beef narwhal mattak (Greenlandic traditional food consisting of blubber and parts of the skin), and commercial seal meat. With respect to PFASs, 13 compounds were analyzed. Again, PFOS was the most common PFASs, whereas the sum of PFASs was below the limit of detection in most fish fillet samples, varying from 2.9 ng/g ww in whale beef to 13.5 ng/g ww in seal beef.

Because of their high consumption of marine mammals, the Greenlandic population has been a target population in some biological monitoring studies. A recent example is the investigation of Long et al.,[39] who collected blood samples and questionnaire data from 207 pregnant women, PFASs, legacy POPs, and metals being determined in all of the samples. Although lower levels of PFOS and PFOA were found compared to data from other Arctic regions, PFHxS and PFNA sustained the same level, indicating that the Greenlandic Inuit are still exposed to PFASs.

Asia. *China.* Zhao and co-workers[40] determined the concentrations of various PFASs in freshwater (10 species) and marine fish (10 species) collected from Hong Kong and Xiamen, China. Risk assessment of exposure of local people to PFASs through fish consumption was subsequently assessed. PFOS, PFOA, PFNA, PFDA, PFUnDA, and PFTrDA were detected. Total concentrations of PFASs ranged between 0.27 and 8.4 ng/g and between 0.37 and 8.7 ng/g for Hong Kong and Xiamen, respectively. PFOS was the predominant PFAS in fish, ranging between 0.27 and 4.5 ng/g, wet weight (ww). The authors also calculated the HR by dividing the average daily intake by the RfD. The HR of PFOS for all fish was <1.0, whereas that of PFOA was <0.01 for all analyzed species of fish. In another study also conducted in China, the levels of 10 PFASs were analyzed in samples of edible freshwater fish and seafood.[41] The estimated daily intakes of PFOS and PFOA via fish and seafood consumption ranged from 0.10 to 2.51 ng/kg bw/day and from 0.13 to 0.38 ng/kg bw/day, respectively, for different age groups (toddlers, adolescents and children, and adults) from four selected locations. Fish and seafood consumption of the analyzed species accounted for 7−84% of PFOS intake, indicating important regional differences in human dietary exposure to PFOS. Additional data on the concentrations of PFASs in seafood from China were reported by Wu et al.,[42] who analyzed the levels of 13 PFASs in 47 fatty fish and 45 shellfish samples, collected from six coastal provinces. The daily intakes of PFASs via seafood consumption by residents, as well as the healthy effect of local inhabitants in these six regions, were subsequently estimated. PFASs were detected in 92% of the 92 seafood samples, PFOS, PFOA, PFNA, and PFUnDA being the most predominant compounds, whereas the remaining nine PFASs were found in only a few samples (<30%) at trace concentrations. The levels of PFOS ranged from <1.4 to 1627 pg/g ww in fatty fish, whereas those of PFOA in shellfish ranged from <5.4 to 7543 pg/g ww. With regard to the estimated daily intakes of PFOS and PFOA, the authors stated that values were much lower than the TDIs recommended by the EFSA.[10] In turn, Yang et al.[43] measured the levels of 10 PFASs in seafood products from Bohai Bay. The sum of the levels of the PFASs was in the range between ND (not detected) and 194 ng/g dry weight and between 4.0 and 304 ng/g dry weight for invertebrates and fish, respectively. The concentrations of PFOS and PFOA in the seafood were lower than those reported in other countries; therefore, they did not constitute a source of immediate risk to public health. A similar conclusion with respect to health risks was also reached by Shi et al.,[44] who measured 14 PFASs in samples of various tissues of farmed freshwater fish from Beijing. A low health risk for the residents of this city, due to exposure to PFASs through fish consumption, was found. Recently, He and co-workers[45] determined the levels of 8 PFASs in 15 fish samples from the Danjiangkou reservoir and the Hanjiang River in China. The human health risks resulting from the consumption of potentially contaminated fish were also determined. Seven of the 8 PFASs could be quantified in all samples, PFOA being the least frequently detected (47%). In fish muscles, the total PFAS concentrations ranged between 2.01 and 43.8 ng/g dry weight. After risk assessment, the authors concluded that most of the

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fish species analyzed could be considered as safe for consumption. However, they also suggested the residents of Danjiangkou and Xiangyang should be cautious when consuming yellow croaker, a fish with a HR of 0.2.

South Korea. Ji et al.[46] determined the serum levels of 13 PFASs in the general adult population of Daegu (Soth Korea) and investigated the dietary contribution (including drinking water) of these PFASs. An extensive questionnaire was designed to obtain the consumption patterns of various foods that might contain PFASs. Potato consumption was identified to be a significant contributor to the concentrations of PFOA in serum. The PFUnDA and PFTrDA levels were positively associated with the intake of fish/shellfish. In turn, Heo et al.[47] measured the concentrations of 16 PFASs in foods (397 of 66 types) and determined the main contributors to human exposure to these compounds. The foodstuffs were classified into seven categories (fish and shellfish, meat and meat products, vegetables and fruits, processed products, dairy products, beverages, and others). Fish and shellfish was the group containing the highest mean ΣPFAS concentration (2.34 ng/g), followed by meat and meat products (1.61 ng/g), processed products (0.85 ng/g), and dairy products (0.57 ng/g). Fish was the major contributor for PFOS and dairy products for PFOA. The estimated dietary intakes of PFOS and PFOA by Korean adults were 0.47−3.03 and 0.17−1.68 ng/kg bw/day, respectively, which were lower than the acceptable TDI limits suggested by the EFSA[10] for these compounds. On the other hand, Kim et al.[48] investigated the relationship between the concentrations of 16 PFASs in blood serum of the general population of Busan (South Korea) and the dietary exposure to these PFASs. Food samples consisted of one-day composite diet samples (breakfast, lunch, and dinner) from 20 of the serum donors. The total PFAS concentrations in the composite food samples ranged from 0.016 to 1.58 ng/g. Only PFOS, PFPeA, PFHxA, PFHpA, PFOA, PFDA, PFUnDA, PFDoDA, PFTrDA, and PFTeDA could be detected (between 5 and 95%). Although food is one of the main routes of human exposure to PFASs, the authors did not find correlations between the PFAS concentrations in the one-day composite diet samples and the serum samples, which was attributed to the lack of representativeness of a one-day composite diet sample.

North America. *Canada.* Gewurtz et al.[49] determined the extent of PFAS contamination in sport fish species collected downstream of a firefighting training facility at the Hamilton International Airport, ON (Canada), to explore if the airport could be a potential source of PFASs. The PFOS concentrations in fish (15 analyzed species) were also compared to consumption advisory benchmarks. Interestingly, PFOS concentrations in seven fish species collected from the three blocks closest to the airport exceeded the 95th percentile concentration of values reported in the scientific literature. PFOS concentrations in the sampling blocks farthest from the airport were significantly lower, being in line with fish concentrations previously reported in the literature and generally below consumption restriction benchmarks. For PFCAs, sport fish concentrations were comparable to, or below, the average concentrations reported in the literature. In a quite different study, the effectiveness of three cooking methods (baking, broiling, and frying) to reduce PFAS levels was examined in four fish species (Chinook salmon, common carp, lake trout, and walleye) sampled from rivers in Ontario (Canada).[50] Five groups of PFASs (PFCAs, PFSAs, PFPAs, diPAPs, and PFPIAs) were analyzed in raw, baked, broiled, and fried fish fillets. In general terms, the scarce changes in the fish PFAS levels found after cooking depended on fish species and cooking method used. The authors concluded that cooking sport fish is generally not an effective approach to reduce dietary exposure to PFASs in general, and PFOS in particular, which is in good agreement with the results of previous studies.[51,52]

United States. Surprisingly, there is no information in the scientific literature on the dietary exposure to PFASs for the general population of the United States. In the period between our previous review[7] and the current one, only a couple of studies have reported data on foods contaminated by PFASs, both being rather tangential to the objective of the present paper. Thus, Blaine et al.[53] determined the PFAS uptake in samples of lettuce and strawberry irrigated with reclaimed water, showing that these compounds could enter and bioaccumulate in food crops. In turn, Stahl and co-workers[54] determined the concentrations of PFASs in freshwater fish samples from U.S. urban rivers and the Great Lakes. The maximum PFOS levels were 127 and 80 ng/g in urban river and Great Lakes samples, respectively.

Other Countries. According to the databases used to prepare this review, there is no recent available information on human dietary exposure for countries such as Australia, Japan, or India, as well as for many other countries of the five continents. With respect to this, recently Pérez et al.[55] assessed the concentrations of 21 PFASs in 283 food items from Brazil (38), Saudi Arabia (35), Spain (174), and Serbia (36), countries that were selected as representatives of South America, western Asia, Mediterranean countries, and south-eastern Europe. In all four countries, the major dietary contributor to PFASs was the group of fish and seafood, the total PFAS food intake being between 2300 and 3800 ng/person/day for the different countries. The tolerable daily intakes for those PFASs for which information is available were not exceeded in any of the four countries.

■ SUMMARY AND CONCLUSIONS

One of the most relevant conclusions drawn in our previous review[6] was that in most countries, for which information on the dietary intake of PFASs was available in the scientific literature, food intake was the most important source of exposure to these compounds, and particularly to PFOS and PFOA, the two most investigated PFASs. However, on the basis of the recommendation of the EFSA[10] on the maximum dietary intakes of PFOS and PFOA, in those countries, human health risks would not be of concern for the nonoccupationally exposed populations. Considering this, the potential role of nondietary exposure sources, such as drinking water consumption and dust inhalation, cannot be disregarded when human exposure to PFASs is evaluated.

In the present revision (2011−2016), we have reached the same conclusion for those rather limited number of countries for which scientific data are available. In relation to this, we note the lack of information on the dietary intake of PFASs in countries of great importance in the international context and with well-reputed food agencies. Australia, Japan, or United States are excellent examples of this lack of data, but also India and other Asian countries, New Zealand, or Middle East countries. Interestingly, there is no data published in the scientific literature on the levels of PFASs in foodstuffs from Africa or Central and South America, with only some

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exceptions.[54] In our last review,[7] we already pointed out Europe as the area with the highest number of scientific publications regarding the dietary intake of PFASs. European countries still hold the first position, with France and Italy as countries providing a good number of studies. Similarly, a rapid increase in the number of publications has been reported in Asia, although these are restricted to only China and South Korea. In turn, the limited amount of data from North America needs to be highlighted, whereas even fewer data are available in other areas worldwide, such as South America or Africa. It is important to note that taking into the account the tremendous differences in the dietary habits among countries, as well as the origin of the foodstuffs, the results obtained in the countries for which information is available cannot be obviously extrapolated to other regions and countries. This lack of data makes it impossible to know whether there are health risks of the dietary intake of PFASs for the populations of numerous countries where no food monitoring studies have been conducted.

As we also noted in our previous review,[7] in this revision we have also observed considerable differences in the PFASs detected—and their concentrations—in the food items analyzed in samples of different regions and countries. However, fish and other seafood seem to be the food group in which more PFASs are detected and for which the concentrations of these compounds are higher. This means that in certain countries, individuals consuming great amounts of fish and shellfish are assuming certain risks, which are not currently quantified. Furthermore, although PFOA and PFOS still continue to be the most remarkable PFASs, increasing attention is being paid to short-chain-length compounds, especially after these new compounds are used as replacements for PFOS and PFOA,[56] the production of which has been banned in certain countries.

As for any potentially toxic environmental pollutant, to prevent health risks from exposure to PFASs in food, it is essential to know what the current dietary intakes are. Because information is still very limited for most countries, food agencies should pay attention to this issue.

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Notes

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ABBREVIATIONS USED

PFASs, per- and polyfluorinated alkyl substances; PFBA, perfluorobutanoic acid; PFPA, perfluoropentanoic acid; PFHxA, perfluorohexanoic acid; PFHpA, perfluoroheptanoic acid; PFOA, perfluorooctanoic acid; PFNA, perfluorononanoic acid; PFDA, Perfluorodecanoic acid; PfUnDA, perfluoroundecanoic acid; PFDoDA, perfluorododecanoic acid; PFTrDA, perfluorotridecanoic acid; PFTeDA, perfluorotetradecanoic acid; PFBS, perfluorobutanesulfonate; PFHxS, perfluorohexanesulfonate; PFHpS, perfluoroheptanesulfonate; PFOS, perfluorooctanesulfonate; PFDS, perfluorodecanesulfonate; FOSA, perfluorooctanesulfonate; diPAPs, polyfluoroalkyl phosphoric acid diesters; PFSAs, perfluoroalkyl sulfonates; PFCAs, perfluoroalkyl carboxylates; PFPAs, perfluoroalkyl phosphonic acids; PFPIAs, perfluoroalkyl phosphinic acids; LCHP, low-carbohydrate high-protein; EFSA, European Food Safety Authority; WHO, World Health Organization; TDI, tolerable daily intake; PCBs, polychlorinated biphenyls; PBDEs, polybrominated diphenyl ethers; HR, hazard ratio; RfD, reference dose

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