Perfluorinated Compounds 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 poly-fluorinated alkyl substances (PFASs), also known as perfluorinated compounds (PFCs), in recent years, the social and scientific interest on these compounds has notably increased. Special attention has been paid to perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA), the most extensively investigated PFASs. Although human exposure to PFCs 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 PFCs 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 PFCs 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 PFCs are detected, and where the concentrations of these compounds are higher. Based on the recommendations of the EFSA on the maximum dietary intakes of PFOS and PFOA, human health risks would not be of concern for non-occupationally exposed populations, at least in the very limited countries for which recent data are available.

KEYWORDS: *perfluorinated compounds (PFCs), PFOS, PFOA, dietary intake, health risks.*

■ INTRODUCTION

PFASs, also known as PFCs (per- and poly-fluorinated chemicals), 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).

However, the social and scientific interest/concern on 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" and "human health risks" as search terms, the first citation does not appear until 1989.[1] The second indexed paper was published 12 years after,[2] although it is not an experimental study. During that time, a review-article on extraperoxisomal targets of peroxisome proliferators, in which perfluorinated straight-chain monocarboxylic fatty acids is revised, can be also found.[3] The scientific information during subsequent years is also very scarce, ranging between 2 and 7 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 PFCs.[4-10] In recent years, a number of investigators have determined the concentrations of PFCs in foodstuffs, while some studies have also assessed the dietary intake of some of the most well known PFCs, mainly PFOS and PFOA. In 2011, we assessed the state-of-the-art regarding human health risks of the dietary exposure to PFCs.[6] Because of the interest that this topic still generates in both, the public opinion and the scientific community, we have now updated the existing information related with this topic. PubMed (https://www.ncbi.nlm.nih.gov/pubmed) and Scopus have been used as databases. The period of this new review has covered between May 2011 and October 2016. Most of the studies here reviewed have been carried out in European countries, being only some of them conducted in North America and Asia (Table 1).

■ PFC CONCENTRATIONS IN FOOD AND DIETARY INTAKE IN A NUMBER OF COUNTRIES

European countries

Germany. Only a study has been published during the assessed period. It belongs to Hölzer et al.,[11] who measured the concentrations of various PFCs in samples of several fish species collected at the Lake Möhne (Sauerland area). The levels of PFOS ranged between 4.5 and 150 ng/g, being the highest median PFOS concentrations 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 PFCs. For PFOS, the concentrations in plasma of anglers consuming fish 2-3 times per month were 7 times higher, compared to those without any fish consumption from Lake Möhne.

The Netherlands. Noorlander et al.[12] determined the levels of 14 PFCs in foodstuffs (pooled samples) purchased in several Dutch retail store chains with nation-wide coverage. Six PFCs (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 lower than 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, while 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.[13] Two samples of 40 different food items were analyzed. PFOS was the compound found in the highest number of samples (33 out of 80), followed by PFOA, PFHpA, PFHxS, PFDA and PFDS. Fish and shellfish was the food group where 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. However, the TDIs recommended by the EFSA[34] 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.[6] Additionally, the concentrations of 13 PFCs were also determined in samples of fish and shellfish collected from coastal areas of Catalonia.[14] Only seven PFCs 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 amounts of PFCs ingested were below the recommended TDIs for those compounds for which information is currently available.

France. Munschy et al.[15] determined the levels of PFCs in mollusks collected along the coasts on mainland France (Atlantic coast, Mediterranean Sea and English Channel). The median concentrations of PFOS were 0.18 ng/g wet weight (ww), 0.09 ng/g ww and 0.04 ng/g ww in samples from the English Channel, the Atlantic coast and the Mediterranean coast, respectively. No estimation on human exposure to PFCs through mollusks consumption was performed in that study. However, Rivière and co-workers[16] 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.[17] 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), while 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 ng/kg bw/day and 1.15 ng/kg bw/day for PFOA and PFOS, respectively. These values should not mean risks for the general population. However, due to the lack of information for the remaining analyzed compounds, health risks could not be evaluated in that survey. In another study, Yamada et al.[18] assessed the dietary exposure to 15 PFASs for 3 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%), while that of marine fish was due to PFOA (24%), PFOS (20%), PFHxA (15%), PFHpA (11%) and PFBA (11%).[19] 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[20] 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 bw/kg/day) and PFNA (70-80 pg/kg bw/day), but not for PFOA (350-690 pg/kg/day). An interesting conclusion of that survey was that the dietary intake of PFASs had been rather constant over the last decade, in which important manufacturing changes occurred. However, in a subsequent study conducted by the same researchers,[21] in which archived 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, while in eggs there was also a significant decreasing trend in the levels of PFOA. In a recent report by the same research group,[22] where 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 ∑diPAP 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, while the lowest precursor contributions were generally found in food samples from 2010. Very recently, Sjogren and co-workers[23] reported the results of a study whose main objective was to examine the cross-sectional relation 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, being the highest body burden of these compounds found in individuals with high adherence to a

Mediterranean-like diet. 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.[24] 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 as ready-to-cook pommes frites, frozen minced spinach, and mixed lettuce were also sampled and analyzed for PFASs concentrations. Only low levels of PFASs were detected in all analyzed samples. PFCAs were the main group of detected PFASs, being PFOA 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.[25] measured the concentrations of 21 PFASs in samples of meat, fish and seafood, milk and dairy products, and eggs. Sixteen PFASs could be detected in at least one sample. 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, the dietary exposure to 7 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.[26] For both, adults and children, the intake was generally below or close to 1 ng/kg bw/day for all the selected PFASs. According to the TDIs proposed by the EFSA[34] 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, while 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.[27] 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%), while 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,[34] 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.[28] For PFOA and PFOS, Italian school-age children showed intakes ranging 0.3-1.1 and 0.5-1.4 ng/kg bw/day, respectively, well below the corresponding TDIs.[34] In another Italian study,[29] the concentrations of PFOS and PFOA were determined in samples of cereal-based products, fish and seafood, meat, eggs, milk and dairy products, fruit 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), while 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 food, 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.[30] 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 the Lake Varese. PFOS was detected in all samples with concentrations of 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.[31] On the other hand, Barbarossa et al.[32] analyzed the concentration of PFOS and PFOA in 67 samples of different types of cow 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 milk should not be a major source of PFASs, compared with other foods such as fish and seafood.

Greece. Vassiliadou and co-workers[33] determined the concentrations of 12 PFCs 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). Analyses of PFCs were conducted not only in raw samples, but also in samples of these species fried in olive oil or grilled. PFCs 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 PFC 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.[34] More recently, Zafeiraki et al.[35] analyzed the content of PFCs in home produced and commercially produced chicken eggs of chicken 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)

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, while perch was also collected from Lake Päijänne. Farmed fish species included whitefish and rainbow trout. As it has been found in most recent studies on the same topic, PFOS was again the most abundant compound in each fish species. Other 6 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, while 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 Faroe Islands, Eriksson et al.[37] analyzed the levels of 15 PFASs collected in 2011/2012. Food samples included dairy products (milk, yoghurt 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 the following: 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[34] 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, while the sum of PFASs was below the limits of detection in most fish fillet samples, varying from 2.9 ng/g ww in whale beef, and 13.5 ng/g ww in seal beef.

Asia

China. Zhao and co-workers[39] determined the concentrations of various PFCs in freshwater (10 species) and marine fish (10 species) collected from Hong Kong and Xiamen, China. Risk assessment of local people exposure to PFCs through fish consumption was subsequently assessed. PFOS, PFOA, PFNA, PFDA, PFUnDA and PFTrDA were detected. Total concentrations of PFCs ranged between 0.27-8.4 ng/g and 0.37-8.7 ng/g for Hong Kong and Xiamen, respectively. PFOS was the predominant PFAS in fish, ranging between 0.27 and 4.5 ng g^{-1}, wet weight (ww). The Hazard Ratio (HR) of PFOS for all fish was < 1.0, while that of PFOA was < 0.01 for all analyzed species of fish. In another study also conducted in China, the levels of 10 PFCs were analyzed in samples of edible freshwater fish and seafood.[40] The estimated daily intake of PFOS and PFOA via fish and seafood consumption ranged from 0.10 to 2.51 ng/kg bw/day and 0.13 to 0.38 ng/kg bw/day, respectively, for different age groups (toddlers, adolescents and children, and adults) from 4 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 PFCs in seafood from China were reported by Wu et al.[41], who analyzed the levels of 13 PFCs in 47 fatty fish and 45 shellfish samples, collected from 6 coastal provinces. The daily intakes of PFCs via seafood consumption by residents, as well as the healthy effect of local inhabitants in these 6 regions, were subsequently estimated. PFCs were detected in 92% of the 92 seafood samples, being PFOS, PFOA, PFNA and PFUnDA the most predominant compounds, while the remaining 9 PFCs were only found in a few samples (< 30%) at trace concentrations. The levels of PFOS ranged from <1.4 to 1627 pg/g ww in fatty fish, while those of PFOA in shellfish ranged from < 5.4 to 7543 pg/g ww. Regarding the estimated daily intakes of PFOS and PFOA, these were much lower than the TDIs recommended by the EFSA.[34] In turn, Yang et al.[42] measured the levels of 10 PFASs in seafood products from the Bohai Bay. The sum of the levels of the PFASs were in the range between ND (not detected) to 194 ng/g dry weight, and between 4.0 to 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 mean a source of immediate risk to public health. A similar conclusion with respect to health risks was also reached by Shi et al.[43], who measured 14 PFCs in samples of various tissues of farmed freshwater fish from Beijing. A low health risks for the residents of this city, due to exposure to PFCs through fish consumption, was found. Recently, He and co-workers[44] determined the levels of 8 PFCs in 15 fish samples from the Danjiangkou reservoir and the Hanjiang River in China. The human health risks resulting from the consumption of potential contaminated fish were also determined. Seven of the 8 PFCs could be quantified in all samples, being PFOA the least frequently detected (47%). In fish muscles, the total PFC concentrations ranged between 2.01 and 43.8 ng/g dry weight. After risk assessment, the authors concluded that most of the fish species analyzed could be considered as safe for consumption. However, they also suggested to the residents of Danjiangkou and Xiangyang to be cautious when consuming yellow croaker, a fish with a HR of 0.2.

South Korea. Ji et al.[45] 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.[46] 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 7 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 low than the acceptable TDI limits suggested by the EFSA[34] for these compounds. On the other hand, Kim et al.[47] investigated the relationship between the concentrations of 16 PFCs in blood serum of the general population of Busan (South Korea) and the dietary exposure to these PFCs. Food samples consisted of one day composite diet samples (breakfast, lunch and dinner) from 20 of the serum donors. The total PFC 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 PFCs, the authors did not find correlations between the PFC concentrations in the one day composite diet samples and the serum samples. It was attributed to the lack of representativeness of a 1-day composite diet sample.

North America

Canada. Gewurtz et al.[48] determined the extent of PFAS contamination in sport fish species collected downstream of a firefighting training facility at the Hamilton International Airport, Ontario, 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 7 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 furthest 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 PFC levels were examined in four fish species (Chinook salmon, common carp, lake trout and walleye) sampled from rivers in Ontario, Canada.[49] 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.[50,51]

USA. Surprisingly, there is not information in the scientific literature on the dietary exposure to PFASs for the general population of the USA. In the period between our previous review[6] and the current one, only a couple of studies have reported data on foods contaminated by PFASs, being both rather tangential to the objective of the present paper. Thus, Blaine et al.[52] 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[53] determined the concentrations of PFCs in freshwater fish samples from US 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-article, there is not recent available information on the 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.[54] 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-Esatern Europe. In all 4 countries, the major dietary contributor to PFASs was the group of fish and seafood, being the total PFASs food intake 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 4 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 PFCs was available in the scientific literature, food intake was being the most important source of exposure to these compounds, and particularly to PFOS and PFOA, the two most investigated PFCs. However, based on the recommendation of the EFSA[34] on the maximum dietary intakes of PFOS and PFOA, in those countries, human health risks would not be of concern for the non-occupationally exposed populations.

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 it, we would like to remark the lack of information on the dietary intake of PFCs in countries of great importance in the international context and with well reputed food agencies. Australia, Japan, or USA could be excellent examples of this lack of data, but also India and other Asian countries, New Zealand, or Middle East countries. Interestingly, there is not any data published in the scientific literature on the levels of PFCs in foodstuffs from

Africa or Central and South America, with only some exception.[54] 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. Consequently, it is impossible to know whether there are health risks of the dietary intake of PFCs for the populations of numerous countries.

As we also noted in our previous review,[6] in this revision we have also observed considerable differences in the PFCs 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 PFCs are detected, and where the concentrations of these compounds are higher. It means that in certain countries, individuals consuming great amounts of fish and shellfish could are assuming certain risks, which are not currently quantified.

In recent years, the toxicological information on PFCs has notably increased and a number of toxic effects of these compounds are now well established.[7,55-57] With respect to the carcinogenicity of PFCs, there are still some doubts on the carcinogenic potential of some of these compounds such as PFOA,[58] although the epidemiologic evidence –as a whole- does not seem to support the hypothesis of a causal association between PFOA or PFOS exposure and cancer in humans.[59]

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

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The authors declare no competing financial interest.

■ ABBREVIATIONS USED

PFASs, per- and poly-fluorinated alkyl substances; PFCs, perfluorinated compounds; 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, perfluorobutane sulfonate; PFHxS, perfluorohexane sulfonate; PFHpS, perfluoroheptane sulfonate; PFOS, perfluorooctane sulfonate; PFDS, Perfluorodecane sulfonate ; FOSA, perfluorooctane sulfonate; 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.

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

Country	Characteristics of the study	PFCs assessed	Remarks	Dietary intake (in ng/kg body weight/day)[a]	Reference
Germany	Analysis of PFCs in samples of fish fillet and correlation with plasma levels of anglers at Lake Möhne, Sauerland	10 PFCs, including PFOS and PFOA	A dose-dependent relationship between fish consumption and internal exposure to PFOS was observed	PFOS: Max: 17	Hölzer et al.[11]
The Netherlands	Determination of PFCs in food from Dutch reatil sotre chains, and assessment of the dietary intake	14 PFCs, including PFOS and PFOA	Crustaceans and lean fish showed the highest concentrations of the quantified PFCs	PFOS: Median: 0.3 High-level intake: 0.6 PFOA: Median: 0.2 High-level intake: 0.5	Noorlander et al.[12]
Spain	Human exposure to PFCs through foodstuffs of wide consumption in Catalonia.	18 PFCs, including PFOS and PFOA	Fish and shellfish was the food group showing the highest levels of PFCs, in general, and those of PFOS and PFOA, in particular	In adults: PFOS: 1.84 PFOA: 5.05	Domingo et al.[13]
	Analysis of PFCs in samples of fish and shellfish collected from coastal areas of Catalonia, as well as in drinking water	13 PFCs, 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.[14]
France	PFC determination in mollusks collected along the coasts on mainland France	17 PFCs, including PFOS and PFOA	Highest levels of PFOS were found in samples from the English Channel, contrasting with those from the Mediterranean coast	n.a.	Munschy et al.[15]
	Health risk assessment related to the presence of PFCs in food collected for the 2n French TDS	16 PFCs, including PFOS and PFOA	Maximum levels of PFOS and PFOA in mollusks and crustaceans.	Mean: PFOS: 0.66 PFOA: 0.74 95th percentile: PFOS: 1.15 PFOA: 1.50	Rivière et al.[16]
	Dietary intake of PFCs for 3 different groups of French adult populations.	15 PFCs, including PFOS and PFOA	PFCs 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) Highest PFOA intake: high seafood consumers (1.2)	Yamada et al.[18]

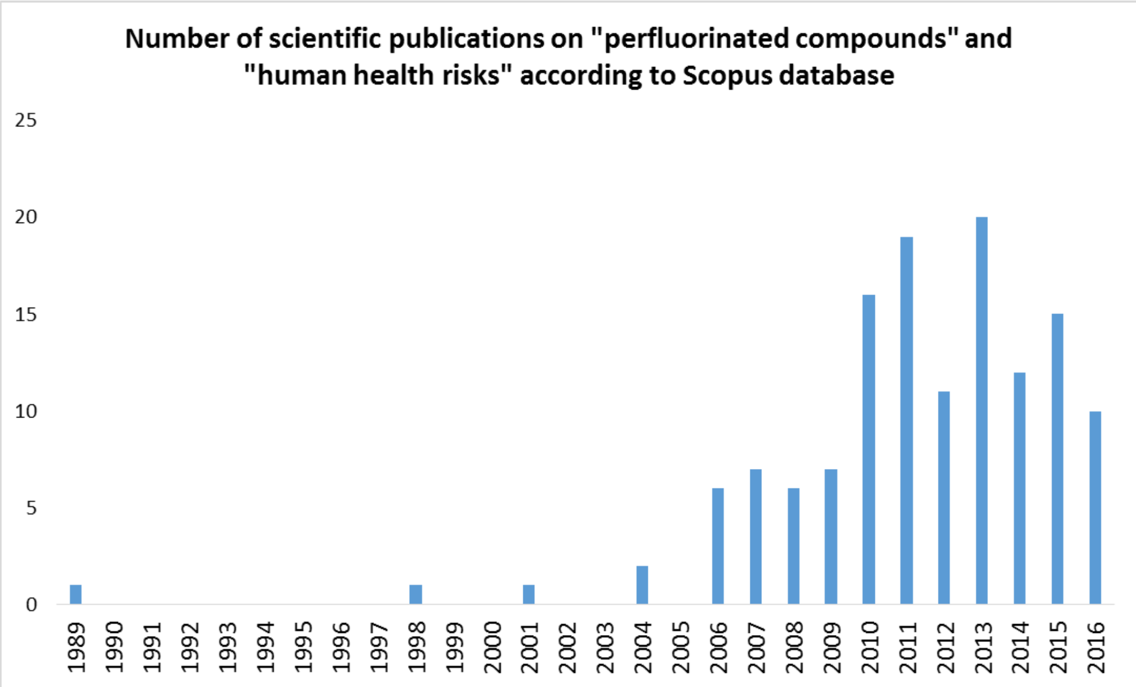
Sweden	Estimation of the dietary intake of PFCs by analyzing a set of archived food market basket samples.	11 PFCs, including PFOS and PFOA	Dietary intake of PFOS and PFOA was higher than estimated exposure through dust ingestion and drinking water. Dietary intake has been fairly constant over the past decade.	PFOS: 1.44, 0.86 and 1.00 in 1999, 2005 and 2010, respectively. PFOA: 0.35, 0.50 and 0.69 in 1999, 2005 and 2010, respectively.	Vestergren et al.[20]
	Analysis of PFCs in archives samples of eggs, milk and farmed rainbow	11 PFCs, including PFOS and PFOA	PFOS levels in fish and eggs, as well as PFOA in eggs, decreased significantly with time.	n.a.	Johansson et al.[21]
	Determination of PFCs and precursor compounds in archived food market basket samples	15 PFCs, 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.[22]
Four European countries (Belgium, Czech Republic, Italy and Norway)	Design of a harmonized sampling survey to analyze PFCs in 20 vegetable species	14 PFCs, including PFOS and PFOA	Perfluorinated carboxylic acids (PFCAs) were the main group of detected PFCs. The intake of potatoes governed the exposure to PFOA	PFOA (only vegetables): 0.040	Herzke et al.[24]
	PFC determination in 50 selected pooled samples representing 15 food commodities	21 PFCs, 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.[25]
	PFC determination in fruits, cereals, sweets and salt	12 PFCs, including PFOS and PFOA	PFOA was the most abundant compound, while PFOS showed the greatest concentrations	Mean for high consumers (95th percentile): PFOS: 1.00. PFOA: 0.35	D'Hollander et al.[27]
	Dietary exposure to PFCs according to data from the PERFOOD EU project	7 selected PFCs	Foods of plant origin are most important for the dietary exposure to PFOA, while food of animal and plant origin contributes with equal importance to PFOS intake.	Estimated average dietary intake for adults and children is generally below or close to 1	Klenow et al.[26]
Italy	Study of the presence of PFCs in meals served weekly at lunch time in Italian school canteens	7 PFCs, 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.[28]
	Analysis of PFCs 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	n.a.	Guerranti et al.[29]

	PFC 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.[30]
	Monitoring the presence of PFCs in Italian cow milk	PFOS and PFOA	Contamination by PFOS was frequent, but at low concentrations	n.a.	Barbarossa et al.[32]
Greece	Analysis of PFCs in raw and cooked samples of several species of Mediterranean finfish and shellfish	12 PFCs, including PFOS and PFOA	PFOS was the predominant compound. FPC levels usually higher after cooking	PFOS: Max in picarel: 10.48 PFOA: Max in shrimp: 0.20	Vassiliadou et al.[33]
Greece and The Netherlands	PFC analysis in home produced and commercially produced chicken eggs	11 PFCs, including PFOS and PFOA	Median levels of PFCs 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 PFCs in various edible fish species	13 PFCs, including PFOS and PFOA	PFCs were present in all Baltic Sea and freshwater species, but not in farmed fish. PFOS was the most abundant compound	n.a.	Koponen et al.[36]
Faroe Islands	Analysis of PFCs in food samples and purified drinking water.	15 PFCs, including PFOS and PFOA	PFOS was most frequently detected compound	n.a.	Eriksson et al.[37]
Greenland	Contamination levels of PFCs and other POPs in traditional seafood items, and assessment of the dietary intake	14 PFCs, including PFOS and PFOA	PFCs 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 PFC exposure through consumption of freshwater and marine fish from Hong Kong and Wiamen	10 PFCs, including PFOS and PFOA	The risks and potential effects of PFCs for the health of the coastal population in the Pearl River Delta are of concern	Mean: PFOS (Hong Kong): 2.4 PFOS (Xiamen): 5.1 PFOA (Hong Kong): 3.3 PFOA (Xiamen): 3.0	Zhao et al.[39]
	Contribution of food to the exposure to PFCs and comparison with the ingestion through drinking water and indoor dust	10 PFCs, 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.[40]
	Determination of PFC levels in samples of fatty fish and shellfish from Chinese coastal areas	13 PFCs, including PFOS and PFOA	PFOS was the dominant compound in fatty fish, whereas was the most abundant PFC in shellfish	PFOS: 0.037-0.694 PFOA: 0.008-0.914	Wu et al.[41]
	PFAS determination in seafood samples collected in Bohai Bay	14 PFCs, including PFOS and PFOA	PFOS and PFOA were the most frequent compounds. Species at higher trophic levels accumulate more PFCs than benthic invertebrates	PFOS: 2.44 PFOA: 0.50	Yang et al.[42]

	PFC determination in a number of tissues of popular farmed freshwater fish from Beijing	10 PFCs, including PFOS and PFOA	Overall tissue distribution of PFOS: fish blood > liver > brain > muscle	PFOS: 0.24 Total PFCs: 0.44	Shi et al.[43]
	Residues of PFCs were investigated in fish samples from Danjiangkou reservoir and Hamjiang River	8 PFCs, including PFOS and PFOA	The frequent consumption of yellow coraker 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.[44]
South Korea	Analysis of PFCs in 397 food items, of 66 types, and evaluation of the dietary intake	16 PFCs, 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.[46]
	Correlation study of PFCs in blood serum and composite diet samples	16 PFCs, including PFOS and PFOA	Because of the low number of samples, no correlations between PFC levels in diet and serum were found.	n.a.	Kim et al.[47]
Canada	PFC contamination in sport fish species collected downstream of the Hamilton International Airport, Ontario, Canada	10 PFCs, including PFOS and PFOA	PFOS dominated the PFC profile in the different fish species and sampling locations.	n.a.	Gewurtz et al.[48]
	The role of cooking for reducing exposure to PFCs was studied by analyzing raw, baked, broiled and fried fish fillets	12 PFCs, 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.	n.a.	Bhavsar et al.[49]
USA	Greenhouse study to investigate the potential uptake of PFCs by lettuce and strawberry	9 PFCs, including PFOS and PFOA	PFCs can enter and bioaccumulate in food crops irrigated with reclaimed water	n.a.	Blaine et al.[52]
	Characterization of PFCs in freshwater fish on a national scale	13 PFCs, including PFOS and PFOA	PFOS was the most frequently detected compound	n.a.	Stahl et al.[53]
Four countries: Brazil, Saudi Arabia, Spain and Serbia	Analysis of PFCs in 283 food items from representative countries of the diet in South America, Western Asia, Mediterranean area and South-Eastern Europe	21 PFCs, including PFOS and PFOA	Fish and seafood were identified as the major PFC contributors to the diet in all the countries.	Total PFCs: 33-54	Pérez et al.[54]

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

TOC GRAPHIC


Number of scientific publications on "perfluorinated compounds" and "human health risks" according to Scopus database
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