

SCIENTIFIC REPORT OF EFSA

Results of the monitoring of perfluoroalkylated substances in food in the period 2000 - 2009¹

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ABSTRACT

Perfluoroalkylated substances (PFASs) are a large group of compounds consisting of a fully or partially fluorinated hydrophobic alkyl chain and a hydrophilic end group. Due to their thermal and chemical stability they are used in e.g. cleaning agents, impregnation agents for textiles, carpets, paper, packaging, furniture, paint and varnish, fire-extinguishing liquids, wax, floor polishing agents. Several PFASs are recognised as generally persistent in the environment and are associated with a broad spectrum of health effects. Human exposure may result through e.g. consumption of contaminated food, beverages and inhalation. The European Commission issued the Commission Recommendation 2010/161/EU on the monitoring of PFASs in food in the Member States. Member States were recommended to monitor the presence of PFASs in food during the years 2010 and 2011. Data obtained, as well as data collected in the previous years, was required to be submitted to EFSA for assessment. A total of 4,881 samples collected in previous years (2000 – 2009) in seven Member States was considered for a detailed data analysis. Data were reported on different sets of 17 PFASs resulting in 24,204 single observations. Overall, only 11.8 % of the results were quantifiable results. Perfluorooctane sulfonate was the most frequent compound (31 %). Across food groups, PFASs were mostly found in ‘Fish offal’ (68 %), ‘Edible offal, game animals’ (64%), ‘Meat, game mammals’ (22 %), ‘Water molluscs’ (20 %), ‘Crustaceans’ (17 %) and ‘Fish meat’ (9.7 %). The highest contamination both in terms of frequency and mean level was found in meat and edible offal of game animals, fish and seafood, whereas meat and edible offal of farmed animals resulted less contaminated. To ensure an accurate assessment of the presence of PFASs in food and beverages, further improvement of the analytical methods, sampling and data reporting are recommended.

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KEY WORDS

Perfluoroalkylated substances, PFAS, PFOS, PFOA, occurrence, food

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SUMMARY

Perfluoroalkylated substances (PFASs) are a large group of compounds consisting of a fully or partially fluorinated hydrophobic alkyl chain and a hydrophilic end group. They were manufactured for over 50 years and are still used in a broad spectrum of products and processes. Due to their thermal and chemical stability and surface activity, they are widely used in various products e.g. cleaning agents, paint and varnish, wax, floor polishing agents, impregnation agents for textiles, carpets, paper, packaging, furniture, shoes, fire-extinguishing liquids, photo paper and insecticide formulations. This has led to a global distribution of PFASs into the environment and the human body, thus raising public health concern.

Negative health effects as hepatotoxicity, developmental toxicity, neurobehavioral toxicity, immunotoxicity, reproductive toxicity, lung toxicity, hormonal effects and a weak genotoxic and carcinogenic potential have been described in animal studies in relation to PFASs.

In 2008, in its Scientific Opinion on perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA), the EFSA Panel on Contaminants in the food chain recommended that more PFASs occurrence data in different foodstuffs and human body should be collected, particularly with respect to exposure assessment. The European Commission issued the Recommendation 2010/161/EU on the monitoring of perfluoroalkylated substances in food⁴. Member States were recommended to monitor during 2010 and 2011 the presence of PFASs in food and submit to EFSA the data obtained together with data from previous years. These data are needed by the Commission as a basis for deciding on any possible risk management measures.

In 2010, data from previous years were submitted to EFSA. The present evaluation is based on a set of 4,881 samples collected between 2000 and 2009 in seven Member States. Data were reported on different sets of compounds from a total of 17 PFASs resulting in 24,204 single analytical results. All results were expressed in $\mu\text{g}/\text{kg}$ whole weight (wet weight). Most of the LOQs (79 %) reported for each of the 17 compounds across all food groups were below or equal to the value recommended by the Commission Recommendation 2010/161/EC of $1 \mu\text{g}/\text{kg}$. This demonstrates that the suggested maximum LOQ for the analysis of PFASs ($1 \mu\text{g}/\text{kg}$) is in most cases achievable. Regarding recovery, high variation was observed for the individual PFASs with median values ranging between 41 % and 75 %. In total, results above LOD or above LOQ were reported for only 11.8 % of the results across the 17 PFASs.

The most frequently found PFASs were PFOS (31.1 %), PFTriDA (17.2 %), PFOSA (16.6 %), PFOA (11.5 %), PFDA (11 %), PFDoDA (9.8 %), PFNA (9.3 %) and PFUnDA (7 %). PFBA, PFPA and PFHpS were not detected in any of the samples analysed. Across food groups, PFASs were mostly found in 'Fish offal' (68 %), 'Edible offal, game animals' (64 %), 'Meat, game mammals' (22 %), 'Water molluscs' (20 %), 'Crustaceans' (17 %) and 'Fish meat' (9.7 %). In other food groups PFASs were detected with a much lower frequency (below 5 %). However, for several food groups and compounds only a limited number of samples was analysed and thus it was difficult to draw a clear conclusion on the contamination levels of these food groups.

The highest mean contamination for PFOS ($216 \mu\text{g}/\text{kg}$), PFNA ($10.3 \mu\text{g}/\text{kg}$), PFOA ($7.1 \mu\text{g}/\text{kg}$), PFDA ($6.0 \mu\text{g}/\text{kg}$) and PFDoDA ($3.7 \mu\text{g}/\text{kg}$) was found in 'Edible offal, game animals'. Lower mean concentration of PFOS and PFOA was observed in meat of game animals (both mammals and birds). Compared to the corresponding matrices of game animals, meat of farmed animals and their edible offal were less contaminated with PFASs.

In 'Fish offal', the highest mean concentration was found for PFOS ($47 \mu\text{g}/\text{kg}$) and PFOSA ($15 \mu\text{g}/\text{kg}$). The same compounds had the highest mean concentrations also in 'Fish meat', though at

⁴ Commission Recommendation 2010/161/EU of 17 March 2010 on the monitoring of perfluoroalkylated substances in food. OJ, L 68, 18.3.2010, p.22-23.

lower level, with 4.9 µg/kg for PFOS and 2.7 µg/kg for PFOSA. Only a limited number of crustacean samples was analysed but the levels found were similar to those observed in fish meat.

Different samples were analysed for different sets of PFASs. Therefore, calculating and comparing sum of PFASs was not possible. This prevented also from comparing the total contamination levels across food groups. A harmonisation effort to define a minimum standardised set of PFASs to be analysed in all samples would be required to allow a better comparability of the contamination across food groups. For this purpose, more research (e.g. total diet studies, biomonitoring, toxicological studies) is needed to establish the most representative PFASs.

In view of a more realistic exposure assessment which might follow at the end of the 2010-2011 monitoring it would be advantageous to increase the analytical performances of the methods applied in the analysis of PFASs in order to reduce the proportion of left-censored data. Both literature data and data provided for this report prove that this goal is achievable for all PFASs in all food groups.

The present report includes samples from both random and targeted monitoring, even if the latter is not always specifically stated. The results should therefore be interpreted with caution. Targeted samples and in particular samples taken from “hot-spots” may lead to an overestimation of the contamination levels.

For a more clear analysis of the background contamination and in view of an exposure assessment more data should be collected, in particular for food groups where the number of samples was limited but the frequency of contamination was high (crustaceans, water molluscs) and for food groups with low contamination levels but with high intake (drinking water and other beverages, foods for infants and small children). Also, taking into account that PFASs may migrate into food from containers in which food is stored, prepared or served it would be important to collect more data on ready-to-eat food (cooked food) and packaged food.

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BACKGROUND AS PROVIDED BY THE EUROPEAN COMMISSION

Commission Recommendation 2010/161/EU on the monitoring of perfluoroalkylated substances in food of 17 March 2010 requires the Member States to perform during the years 2010 and 2011 monitoring on the presence of perfluoroalkylated substances in food. It also recommends the transmission to EFSA of the data obtained, including data available from previous years, in the electronic reporting format set out by EFSA. These data are needed by the Commission as a basis for deciding on any possible risk management measures. In addition to Member State monitoring, EFSA is requested to launch a call for data on the presence of perfluoroalkylated substances in food.

TERMS OF REFERENCE AS PROVIDED BY THE EUROPEAN COMMISSION

Monitoring data and additional data, obtained through a call for data will be collated, analysed and summarised by EFSA. An intermediate report delivered in January 2011 could allow for adjustment of the monitoring performed by the Member States. The final report will be delivered in May 2012.

ASSESSMENT

1. Introduction

Perfluoroalkylated substances (PFASs) are a large group of compounds consisting of a hydrophobic alkyl chain of varying length (typically C₄ to C₁₆) and a hydrophilic end group, F(CF₂)_n-R. The hydrophobic part may be fully or partially fluorinated (Figure 1). When fully fluorinated, the molecules are also called perfluorinated substances. Partially fluorinated compounds that contain a -CH₂CH₂- moiety between the hydrophilic part and the fully fluorinated remaining carbon chain – F(CF₂)_n-CH₂CH₂-X – have been suggested as precursors of PFASs found in the environment. The compounds containing the -CH₂CH₂- moiety are called telomer substances and derive their name from the telomerisation production process (de Voogt et al., 2006).

The hydrophilic end group can be neutral (e.g., -OH and -SO₃NH₂), or negatively charged (carboxylates (-COO⁻), sulfonates (-SO₃⁻) and phosphonates (-PO₃⁻) or positively charged (e.g., quaternary ammonium group). Thus, the resulting compounds are non-ionic, anionic or cationic surface active agents due to their amphiphilic character. Many of the neutral PFAS are considered to be potential precursors of PFOS (e.g., perfluorooctane sulfonamide (PFOSA), N-ethyl perfluorooctane sulfonamidoethanol (NETFOSE)) or PFOA (e.g. 8:2 fluorotelomer alcohol, PFOSA and N-EtFOSE).

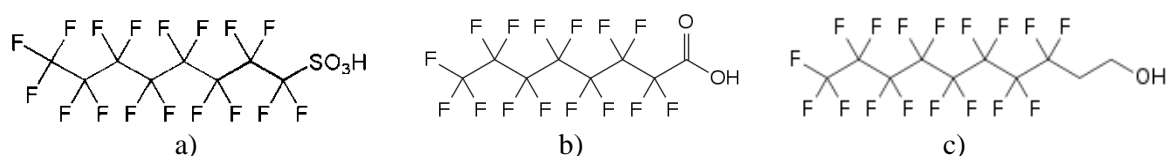


Figure 1: Structural formula of a) PFOS and b) PFOA and c) 8:2 fluorotelomer alcohol.

PFASs are produced by two major processes: electrochemical fluorination (EF) and telomerisation (TM). The EF process entails electrolysis of a solution of an organic compound in a solution of hydrogen fluoride. All of the hydrogen atoms in the molecule are replaced with fluorine atoms. In the telomerisation process, many fluorotelomers, such as fluorotelomer alcohols, are fluorocarbon-based because they are synthesized from tetrafluoroethylene. Other telomerisation processes are: fluorotelomer iodide oxidation, fluorotelomer olefin oxidation, and fluorotelomer iodide carboxylation (Prevedouros et al., 2006). The TM process results in compounds consisting of only a linear alkyl chain with an even number of carbon atoms.

After the major world producer using the EF process, announced the termination of the EF production process by 2002, attention was directed towards fluorotelomers. It has been proven that fluorotelomers and fluorotelomer-based compounds are a source of environmentally persistent PFASs. For example, 8:2 fluorotelomer alcohol can be degraded by bacteria from soil and wastewater treatment plants to PFOA. Similarly, 2- N-ethyl(perfluorooctane sulfonamido)ethanol can be degraded by wastewater treatment to PFOS (Parson et al., 2008).

PFASs were manufactured for over 50 years and are still used in a broad spectrum of products and processes. Due to their extreme thermal and chemical stability, and surface activity, PFASs have found wide applicability in various products: certain cleaning agents, paint and varnish, wax, floor polishing agents, impregnation agents for textiles, carpets, paper, packaging, furniture, shoes, fire-extinguishing liquids, photo paper, and insecticide formulations (Prevedouros et al., 2006). These consumer and industrial applications have led to the global distribution of PFASs into the environment including humans. PFOS and its salts were recently included as persistent organic pollutants (POPs) in Annex B of the Stockholm Convention (Stockholm Convention on Persistent Organic Pollutants, 2010) meaning that their use is accepted only for a defined list of applications.

Within the last ten years, PFASs have attracted public concern because of the findings that PFOS and PFOA are present in the environment and in the human body. Hepatotoxicity, developmental toxicity, neurobehavioral toxicity, immunotoxicity, reproductive toxicity, lung toxicity, hormonal effects, and also a weak genotoxic and carcinogenic potential have been described in animal studies as main endpoints of health concern for PFASs (Lau et al., 2007; Zhang et al., 2008; Shi et al., 2009; Peden-Adams et al., 2009; Eriksen et al., 2010; Pinkas et al., 2010).

PFASs are ubiquitously found in various environmental matrices as river waters and sediments (Loos et al., 2008; Yeung et al., 2009; Clara et al., 2009; Bao et al., 2010; Lin et al., 2010; Pan and You, 2010; Möller et al. 2010), marine waters (Sanchez-Avila et al., 2010) rain-water (Dreyer et al., 2010), drinking water (Wilhelm et al., 2010, Eschauzier et al. 2010), wastewater (Loganathan et al., 2007; Ahrens et al., 2009; Murakami et al., 2009; Clara et al., 2009; Lin et al., 2010), landfill leachates (Busch et al., 2010; Eggen et al., 2010), and air (Murakami et al., 2008). Several studies demonstrated that besides the industrial discharges wastewater, sewage water and sewage sludge represent an important source of contamination for the environment (Ahrens et al., 2009; Clara et al., 2009; Murakami et al., 2009; Lin et al., 2010).

Due to the global ocean and atmospheric circulation PFASs have reached regions without anthropogenic activities as the arctic environment. They have been detected in arctic waters and in the body of arctic animals (Schivone et al., 2009; Butt et al., 2010; Nash et al., 2010).

Unlike other persistent organic pollutants, PFASs do not preferentially accumulate in adipose tissue. Due to their amphiphilic properties, they rather accumulate in organisms by binding to proteins (Jones et al., 2003). Bioaccumulation in biota and biomagnification in the food chain have also been demonstrated for PFAS (Haukås et al., 2007; Meyer et al., 2009; Herzke et al., 2009). Some compounds do not accumulate as such in the organism but they constitute precursors for other PFASs, in some case with even higher persistence and bioaccumulative potential (Brandsma et al., 2010).

Many studies reported the presence of PFASs in fish (Berger et al., 2009; Ye et al., 2008; Delinsky et al., 2009), but concentrations found in fish from various locations can be highly variable (Nania et al., 2009, Schuetze et al., 2010). It was thus suggested that possible critical contamination location (hot-spots) exist and this must be taken into account when assessing the human exposure to PFASs. Concentrations of PFASs in farmed fish are generally lower compared to wild fish (van Leeuwen et al., 2009).

PFASs have been frequently detected in human blood (Kärrman et al., 2006; Ericson et al., 2007; Ingelido et al., 2010; Vassiliadou et al., 2010) and plasma (Longnecker et al., 2008; Wilhelm et al., 2009). The widespread human exposure is of concern due to their persistence and toxic potential. Some PFASs have half-lives of several years in humans (Olsen et al., 2007; Genuis et al., 2010).

Dietary exposure has been suggested as the main exposure route to PFASs in the general population. Seafood, in particular, is considered a major source of PFAS in humans (Haug et al., 2010a) but contaminated drinking water can also be an important source of exposure in contaminated areas (Fromme et al. 2009). Besides food and drinking water, exposure via inhalation may result from outdoor air and indoor air (Shoeib et al., 2005; Fromme et al. 2009; D'Hollander et al., 2010).

Jogsten et al., (2009) evaluated the exposure to PFASs through consumption of various raw and cooked foodstuffs, including packaged food. It was not sufficiently clear whether cooking with non-stick cookware, or packaged foods, could contribute to a higher human exposure to PFASs. Nelson et al. (2010) found a positive association between the serum concentration of PFOA and PFNA and the fast food consumption. The authors suggest that PFASs may enter the food chain both through bioaccumulation and contact with packaging. The French Food Safety Agency has evaluated in 2009 the potential human health risks related to the residual presence of PFOA in non-stick coatings for cookware (AFSSA, 2009). It has been concluded that the consumer health risk related to residues of PFOA in non-stick coating for cookware is negligible.

Table 1: Selected perfluoroalkylated substances

Compound	Abbreviation	Molecular formula	Molecular weight	CAS No.
Fluorotelomer alcohols				
4:2 fluorotelomer alcohol	4:2 FTOH	C ₆ H ₅ F ₉ O	264.09	2043-47-2
6:2 fluorotelomer alcohol	6:2 FTOH	C ₈ H ₅ F ₁₃ O	364.11	647-42-7
8:2 fluorotelomer alcohol	8:2 FTOH	C ₁₀ H ₅ F ₁₇ O	464.12	678-39-7
10:2 fluorotelomer alcohol	10:2 FTOH	C ₁₂ H ₅ F ₂₁ O	564.13	865-86-1
Fluorotelomer sulfonates				
6:2 fluorotelomer sulfonate	6:2 FTS	C ₈ H ₅ F ₁₃ O ₃ S	428.17	27619-97-2
8:2 fluorotelomer sulfonate	8:2 FTS	C ₁₀ H ₅ F ₁₇ O ₃ S	528.18	39108-34-4
Fluorotelomer carboxylates				
6:2 fluorotelomer unsaturated carboxylate	6:2 FTUCA	C ₈ H ₂ F ₁₂ O ₂	358.08	70887-88-6
8:2 fluorotelomer unsaturated carboxylate	8:2 FTUCA	C ₁₀ H ₂ F ₁₆ O ₂	458.09	70887-84-2
10:2 fluorotelomer unsaturated carboxylate	10:2 FTUCA	C ₁₂ H ₂ F ₂₀ O ₂	558.11	70887-94-4
6:2 fluorotelomer carboxylate	6:2 FTCA	C ₈ H ₃ F ₁₃ O ₂	378.09	53826-12-3
8:2 fluorotelomer carboxylate	8:2 FTCA	C ₁₀ H ₃ F ₁₇ O ₂	478.10	27854-31-5
10:2 fluorotelomer carboxylate	10:2 FTCA	C ₁₂ H ₃ F ₂₁ O ₂	578.11	53826-13-4
Perfluoroalkyl sulfonates				
Perfluorobutane sulfonate (Perfluorobutanesulfonic acid)	PFBS	C ₄ HF ₉ O ₃ S	300.1	375-73-5
Perfluorohexane sulfonate (Perfluorohexanesulfonic acid)	PFH _x S	C ₆ HF ₁₃ O ₃ S	400.11	355-46-4
Perfluoroheptane sulfonate	PFHpS	C ₇ HF ₁₅ O ₃ S	450.12	375-92-8
Perfluorooctane sulfonate (Perfluorooctane sulfonic acid)	PFOS	C ₈ HF ₁₇ O ₃ S	500.13	1763-23-1
Perfluorodecane sulfonate (Perfluorodecane sulfonic acid)	PFDS	C ₁₀ HF ₂₁ O ₃ S	600.14	335-77-3
Perfluoroalkyl carboxylates				
Perfluorobutanoic acid	PFBA	C ₄ HF ₇ O ₂	214.04	375-22-4
Perfluoropentanoic acid	PFPA	C ₅ HF ₉ O ₂	264.05	2706-90-3
Perfluorohexanoic acid	PFH _x A	C ₆ HF ₁₁ O ₂	314.05	307-24-4
Perfluoroheptanoic acid	PFHpA	C ₇ HF ₁₃ O ₂	364.06	375-85-9
Perfluorooctanoic acid	PFOA	C ₈ HF ₁₅ O ₂	414.07	335-67-1
Perfluorononanoic acid	PFNA	C ₉ HF ₁₇ O ₂	464.08	375-95-1
Perfluorodecanoic acid	PFDA	C ₁₀ HF ₁₉ O ₂	514.08	335-76-2
Perfluoroundecanoic acid	PFUnDA	C ₁₁ HF ₂₁ O ₂	564.09	4234-23-5

Compound	Abbreviation	Molecular formula	Molecular weight	CAS No.
Perfluorododecanoic acid	PFD _o DA	C ₁₂ HF ₂₃ O ₂	614.1	307-55-1
Perfluorotridecanoic acid	PFTriDA	C ₁₃ HF ₂₅ O ₂	664.11	72629-94-8
Perfluorotetradecanoic acid	PFTeDA	C ₁₄ HF ₂₇ O ₂	714.11	376-06-7
Perfluoroalkyl sulfonamides/ sulfonamidoethanols/ethylacrylate				
Perfluorooctane sulfonamide (Perfluorooctanesulfonamide)	PFOSA	C ₈ H ₂ F ₁₇ NO ₂ S	499.14	754-91-6
N-Methylheptadecafluorooctane sulfonamide	N-Me-FOSA	C ₁₂ H ₅ F ₂₁ O	564.13	31506-32-8
N-Ethylheptadecafluorooctane sulfonamide	N-Et-FOSA	C ₁₀ H ₆ F ₁₇ NO ₂ S	527.19	4151-50-2
N-methyl perfluorooctane sulfonamidoethanol	N-MeFOSE	C ₁₁ H ₈ F ₁₇ NO ₃ S	557.23	24448-09-7
N-ethyl perfluorooctane sulfonamido ethylacrylate	N-EtFOSEA	C ₁₅ H ₁₂ F ₁₇ NO ₄ S	625.30	423-82-5

A number of assessments have been conducted in recent years to evaluate the level of exposure of humans to PFASs. In its Scientific Opinion of 2008, the EFSA Panel on Contaminants in the food chain (CONTAM Panel) concluded that it is unlikely that adverse effects of PFOS and PFOA are occurring in the general population but due to lack of data only a limited exposure assessment was possible (EFSA, 2008; Johansson et al. 2009). The CONTAM Panel has established tolerable daily intakes (TDI) for PFOS and PFOA of 150 ng/kg body weight and 1.5 µg kg/ body weight, respectively (EFSA, 2008). The CONTAM Panel noted that the indicative dietary exposure to PFOS and PFOA was below the TDI within the general population. In a 2008 statement, the Federal Institute for Risk Assessment (BfR, 2008) concluded that there is no health risk arising from dietary exposure to PFOS and PFOA at levels found in food. To a similar conclusion came the UK Committee on Toxicity of Chemicals in Food, Consumer Products and the Environment in its statement on the tolerable daily intake for perfluorooctanoic acid (COT, 2006) and the Office for Risk Assessment in The Netherlands in its recommendation on perfluoroalkyl compounds in Dutch food (VWA, 2010). Fromme et al. (2009) assessed the overall exposure of the general population in western countries to PFASs taking into consideration all the potential exposure routes. The total estimated intake of PFOS and PFOA were well below the lowest recommended TDI values. However these studies recommended further monitoring on a larger number of compounds in a broader spectrum of foodstuffs for obtaining representative data for exposure assessment.

The CONTAM Panel recommended in its Scientific Opinion of 2008 that more occurrence data of PFASs in different foodstuffs and humans should be collected, particularly with respect to monitoring trends in exposure. As a follow-up of the recommendation, the European Commission issued the Commission Recommendation 2010/161/EU on the monitoring of perfluoroalkylated substances in food in the Member States. Member States are recommended to monitor during 2010 and 2011 the presence of PFASs in food. The monitoring should include a wide variety of foodstuffs reflecting consumption habits including food of animal origin such as fish, meat, eggs, milk and derived products and food of plant origin in order to enable an accurate estimation of exposure. Additionally, since 2009, a three years EU research project (PERFOOD) has been carried out with the aim to improve the analytical tools for the determination of PFASs in food items, to contribute to the understanding of PFASs transfer from the environment into dietary items and to quantify the possible contribution of food/beverage contact materials and food and water processing to the overall PFASs levels in the diet.

2. Objectives

As requested by the European Commission the report should focus on:

1. Checking the accuracy of data transmitted and building a database on PFASs in food.
2. Analyse and summarise the collected data.
3. Evaluate the contamination levels of the individual compounds per food groups.
4. Make recommendations to adjust the on-going monitoring for PFASs in food.

The final report to be issued in 2012 after having collected all data from the on-going monitoring may address more complex questions.

3. Material and Methods

3.1. Sampling and analytical procedure

Commission Recommendation 2010/161 recommends to the Member States to carry out the analysis of PFASs in accordance with Annex III to Regulation (EC) No 882/2004 of the European Parliament and of the Council of 29 April 2004⁵ on official controls performed to ensure the verification of compliance with feed and food law, animal health and animal welfare rules by making use of a method of analysis that has been proven to generate reliable results. Ideally, the recovery rates should be in the 70-120 % range, with limits of quantification of 1 µg/kg. In 2010, EFSA issued a call for data on PFASs in food. Seven Member States sent data and more is expected to be collected in 2011. Data included in the present report were generated before the aforementioned Regulation was issued and thus the specified requirements have not been fulfilled in all cases. To ensure a better comparability, data has been handled as detailed in section 3.2. A list of 17 PFASs for which results were provided is presented in Table 2.

Samples were collected randomly, targeted or the sampling strategy was not known or not reported. Analytical methods used to generate the results were Liquid Chromatography single Mass Spectrometry (LC-MS) or Liquid Chromatography tandem Mass Spectrometry (LC-MS/MS).

The report includes data obtained on samples taken from 2000 onwards with most of the data collected between 2006 and 2009. Inclusion of older data in the analysis entails comparability issues because the analytical methods for the substances considered have improved over time. Also, older data may not reflect the contamination levels found in recent years. However, these data could be of use in the future for trend analysis.

3.2. Data management and validation

The food samples were classified according to the FoodEx⁶ classification system. Examples of foods covered in the present assessment are presented in Annex A.

As data included in this report was obtained by the Member States before the Commission Recommendation 2010/161/EU and before the call for data was issued, data providers could not supply all the information requested. Therefore, in certain cases a conservative approach in handling the data has been applied.

⁵ Regulation (EC) No 882/2004 of the European Parliament and of the Council of 29 April 2004 on official controls performed to ensure the verification of compliance with feed and food law, animal health and animal welfare rules. OJ L 191, 28.5.2004, p. 1-59.

⁶ FoodEx is a provisional food classification system developed by the DATEX Unit in 2009 with the objective of simplify the analysis of chemical occurrence data in food and to allow the linkage between occurrence and food consumption data when assessing the exposure to hazardous substances. It contains about 1,800 food names or generic food names which can be grouped according to the needs for statistical analysis.

Table 2: List of PFASs on which data was reported by the data providers.

Compound
Perfluorobutanoic acid
Perfluorobutane sulfonate (Perfluorobutanesulfonic acid)
Perfluoropentanoic acid
Perfluorohexane sulfonate (Perfluorohexanesulfonic acid)
Perfluorohexanoic acid
Perfluoroheptanoic acid
Perfluoroheptane sulfonate
Perfluorooctanoic acid
Perfluorooctane sulfonamide (Perfluorooctanesulfonamide)
Perfluorooctane sulfonate (Perfluorooctane sulfonic acid)
Perfluorononanoic acid
Perfluorodecane sulfonate (Perfluorodecane sulfonic acid)
Perfluorodecanoic acid
Perfluorododecanoic acid
Perfluoroundecanoic acid
Perfluorotridecanoic acid
Perfluorotetradecanoic acid

EFSA received 25,600 individual results (4,996 samples) from seven Member States. A list of validation steps was applied to the data set:

- Non-food samples were excluded from the evaluation.
- Data set was checked for duplicates (same samples transmitted twice or repeated analysis of the same sample); all duplicates were excluded.
- Results from Total Diet Study received from one Member State were not included in the assessment as food samples were aggregated at high level and could not be matched with the food classification applied in this report.
- Results obtained on samples collected before 2000 (360 results on samples collected from 1988 to 1996) were not included in the final dataset (see 3.1) as they may not reflect the contamination level found in recent years. These data may be used in the future for trend analysis.
- The remaining records have been considered for correction on recovery of the method. A lot of data was transmitted by the data providers as corrected for recovery. Where results were not corrected for recovery by the data provider a correction has been applied by using the reported recovery. Where recovery was not available, no correction has been applied.
- Although Commission Recommendation 2010/161 recommends limits of quantification of 1 µg/kg for the monitoring to be carried out in 2010 and 2011, the present report is based on backlog data, generated before 2010. Hence, to give a realistic view on the PFAS monitoring performed before 2010, it has been considered useful to include in the evaluation all results obtained, independently of the performance of the analytical methods used. However, in case

an exposure assessment should be carried out in the future, a critical selection of data should be applied.

- Left-censored data were treated by the substitution method as specified in the “Principles and Methods for the Risk Assessment of Chemicals in Food” (WHO, 2009). Two scenarios were considered. The lower bound (LB) is obtained by assigning a value of zero (minimum possible value) to all samples reported as lower than the LOD (<LOD) or LOQ (<LOQ). The upper bound (UB) is obtained by assigning the numerical value of LOD to values reported as <LOD and LOQ to values reported as <LOQ (maximum possible value), depending on whether LOD or LOQ is reported by the laboratory.
- To enable the comparison of the LOQs between the individual compound and across food groups, where LOQ was not reported by the data providers, it has been generated by multiplying LOD by three.

After applying the validation criteria a set of 24,204 results (4,881 samples) was retained for the final evaluation. All results were expressed in µg/kg whole weight (wet weight).

3.3. Statistical analysis

Frequency tables and summary statistics were produced to describe the PFASs data by year of collection, country of testing, substance and food group. Means and percentiles were computed for each substance and all food groups where sufficient data was available. For the statistical analysis per food groups data was aggregated in 27 groups taking into account the source of food and data availability for a specific group.

All analyses were run using the SAS Statistical Software (SAS software, 1999).

4. Results and discussion

4.1. Number of samples by country and food groups

The final data set obtained after applying the validation criteria included 4,881 food samples analysed for several PFASs (24,204 results) from seven Member States. Most of the data were submitted by Germany followed by France and United Kingdom (Table 3). The data set covers results on samples collected from 2000 to 2009 with the majority of samples taken between 2006 and 2009.

Table 3: Number of samples submitted for each sampling year by Member States.

Member State	Sampling Year								Total	
	2000	2001	2002	2005	2006	2007	2008	2009	N	%
Belgium	29	33	62	1.3
France	142	395	62	599	12
Germany	6	9	8	2	331	1,787	774	745	3,662	75
Ireland	42	17	41	.	100	2.0
Italy	65	53	118	2.4
Spain	35	10	.	45	0.9
United Kingdom	199	75	21	295	6.0
Total	6	9	8	2	373	2,180	1,389	914	4,881	

Results were reported for 17 compounds but samples collected were not analysed for a defined set of substances. Most of the results were available for PFOS and PFOA (20.1 % for each) while results for the other 15 compounds were less represented (2.1 to 6.3 %) (Table 4).

Table 4: Number of country-specific results for the individual perfluoroalkylated substances.

PFAS	Member State							Total results	
	Belgium	France	Germany	Ireland	Italy	Spain	United Kingdom	N	%
PFBA	.	584	584	2.4
PFBS	62	597	286	100	.	.	295	1,340	5.5
PFPA	.	599	599	2.5
PFHxA	62	599	508	100	.	.	295	1,564	6.5
PFHxS	62	597	373	100	.	.	295	1,427	5.9
PFHpA	62	599	54	100	.	.	295	1,110	4.6
PFHpS	.	597	597	2.5
PFOA	62	599	3,650	100	118	40	295	4,864	20.1
PFOS	62	597	3,657	100	118	40	295	4,869	20.1
PFOSA	62	.	54	100	.	.	295	511	2.1
PFNA	62	599	474	100	.	.	295	1,530	6.3
PFDA	62	599	475	100	.	.	295	1,531	6.3
PFDS	.	597	54	651	2.7
PFUnDA	62	582	54	100	.	.	295	1,093	4.5
PFDoDA	62	583	403	100	.	.	295	1,443	6.0
PFTriDA	.	242	54	296	1.2
PFTeDA	.	195	195	0.8
Total	682	8,765	10,096	1,100	236	80	3,245	24,204	100
Percentage	2.8	36.2	41.7	4.5	1.0	0.3	13.4	100	

The sampled food groups and the number of samples by country and food group are illustrated in Table 5. The most frequent analysed food groups were “Edible offal, farmed animals” (23.6 %) followed by “Edible offal, game animals” (17.9 %), “Fish meat” (16.9 %) and “Meat of game mammals” (10.7 %). The frequency of samples analysed within the other food groups were much lower (0.1 to 5.3 %). Food groups were not evenly sampled in the Member States and the monitoring was more focused on food of animal origin.

A general caution is in place in relation to the sampling strategy. Some results were reported as originating from testing randomly taken samples while other results were obtained on targeted samples or the sampling strategy was not known or not reported. Thus, the overall results might overestimate the true European background levels of PFASs in food.

4.2. Performance of analytical methods

Results included in the final data set were obtained by LC-MS or LC-MS/MS methods. The LOQs for the analysis varied with the compound analysed. Median LOQs for the individual PFASs were in the range of 0.13 to 1 µg/kg (Figure 2). Most of the results were obtained by methods having LOQs ≤ 5 µg/kg (97 % of results) or even LOQs ≤ 1 µg/kg (79 %) (Figure 3).

LOQs also varied with the food matrix analysed. Most of the LOQs (57 %) reported for the 17 PFASs across food groups were in the range of 0.1 to 1 µg/kg. Higher LOQs were reported in particular for ‘Edible offal, farmed animals’, ‘Edible offal, game animals’, ‘Crustaceans’, and ‘Food for infants and small children’. The lowest LOQs for the individual PFASs were achieved for water (0.0003 – 0.005 µg/litre) (Figure 4). The results demonstrate that LOQs below 1 µg/kg, as recommended by the Commission Recommendation 2010/161/EC, are achievable in the analysis of PFASs in all food

groups and for all compounds. LOQs at low nanogram/kg in food and in picogram/litre in beverages have been reported in recent years by several authors (Ericson et al., 2008; Berger et al., 2009; Kärman et al., 2009; Ballesteros-Gómez et al., 2010; Haug et al., 2010b).

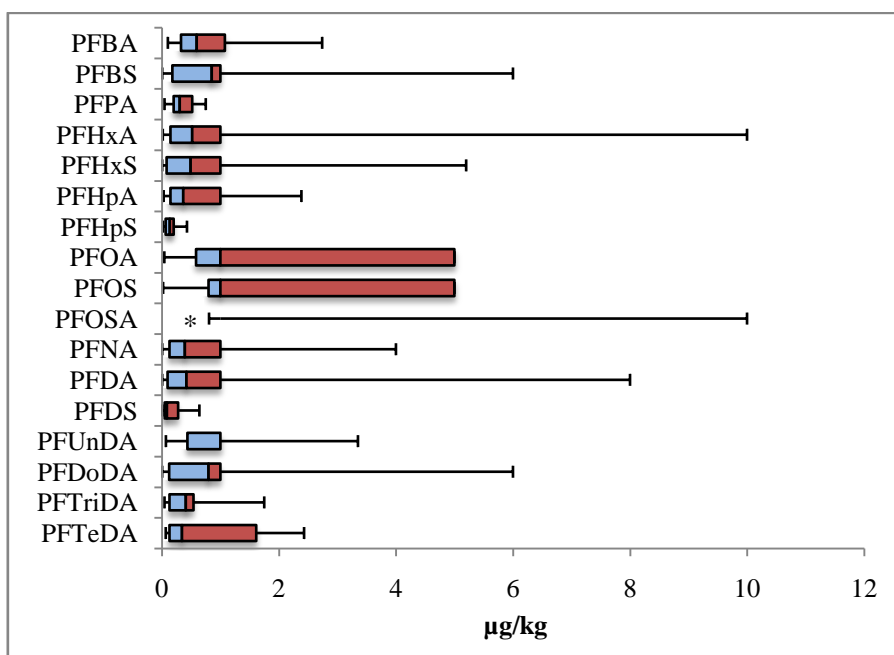
Table 5: Number of samples by Member State and food group.

Food group	Member State							Total samples	
	Belgium	France	Germany	Ireland	Italy	Spain	United Kingdom	N	%
Grains and grain-based products	12	45	.	4	.	.	13	74	1.5
Vegetables and vegetable products	.	46	78	5	.	.	33	162	3.3
Potatoes and potatoes products	.	1	227	2	.	.	20	250	5.1
Legumes, nuts and oilseeds	.	9	.	1	.	.	7	17	0.3
Fruit and fruit products	.	.	1	.	.	.	4	5	0.1
Meat, livestock animals	3	79	22	15	.	.	8	127	2.6
Meat, poultry	.	39	4	6	.	.	4	53	1.1
Meat, game mammals	.	.	521	.	.	.	1	522	10.7
Meat, game birds	.	.	8	8	0.2
Edible offal, farmed animals	.	18	1,089	12	.	.	35	1,154	23.6
Edible offal, game animals	.	.	871	.	.	.	3	874	17.9
Meat products (ham, sausages)	.	77	4	2	.	.	5	88	1.8
Fish meat	11	46	553	12	58	40	103	823	16.9
Fish offal	.	.	38	38	0.78
Crustaceans	24	16	4	.	7	.	10	61	1.2
Water molluscs	.	21	24	5	.	5	2	57	1.2
Milk, liquid	.	38	30	15	28	.	10	121	2.5
Fermented milk products	12	75	87	1.8
Cheese	.	32	.	1	.	.	10	43	0.9
Eggs and egg products	.	30	29	15	.	.	12	86	1.8
Sugar and confectionary	.	5	1	6	0.1
Honey	.	.	5	.	25	.	.	30	0.6
Animal and vegetable fats and oils	.	1	1	5	.	.	9	16	0.3
Drinking water	.	6	141	147	3.0
Food for infants and small children	.	.	10	10	0.2
Composite food	.	8	2	.	.	.	1	11	0.2
Other foods	.	7	4	11	0.2
Total samples	62	599	3,662	100	118	45	295	4,881	100

The methods should be selective in discriminating the presence of interfering substances and be able to separate linear PFOS from its branched isomers. There is evidence that taurodeoxycholic acid (TDCA), a bile acid present in foods of animal origin, can interfere with the PFOS determination by LC-MS and LC-MS/MS (Benskin et al, 2007). Therefore, PFOS levels in food of animal origin, mainly in eggs and offals (liver), may be overestimated by TDCA. Thus, it is recommended to monitor for any known interference that may yield false positive results or overestimation.

The Commission Recommendation 2010/161/EC also specifies that, ideally, the recovery rate of the method of analysis should be in the range of 70 – 120 %. As regards to the data included in the present

report, not all data providers collected and transmitted information on recovery. This information was available only for a limited number of results (n = 6,929). Median values for reported recoveries ranged between 41 % and 75 % but high variation was observed for the individual PFASs (Figure 5). The lowest recovery rate was reported for PFTeDA.



*P25, P50 and P75 equal to 1 µg/kg

Figure 2: Distribution of the limit of quantification reported for the individual perfluoroalkylated substances (n = 24,240) (Box-plot: whiskers at P5 and P95, box at P25 and P75 with line at P50).

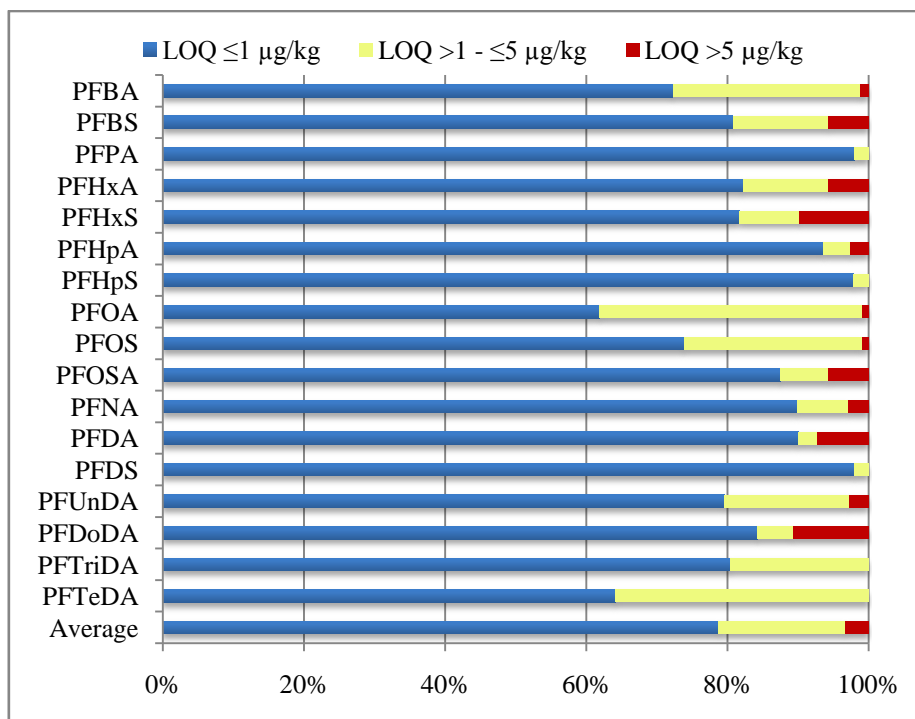


Figure 3: Frequency of results obtained for the individual perfluoroalkylated substances in relation to the limit of quantifications (n = 24,240 results).

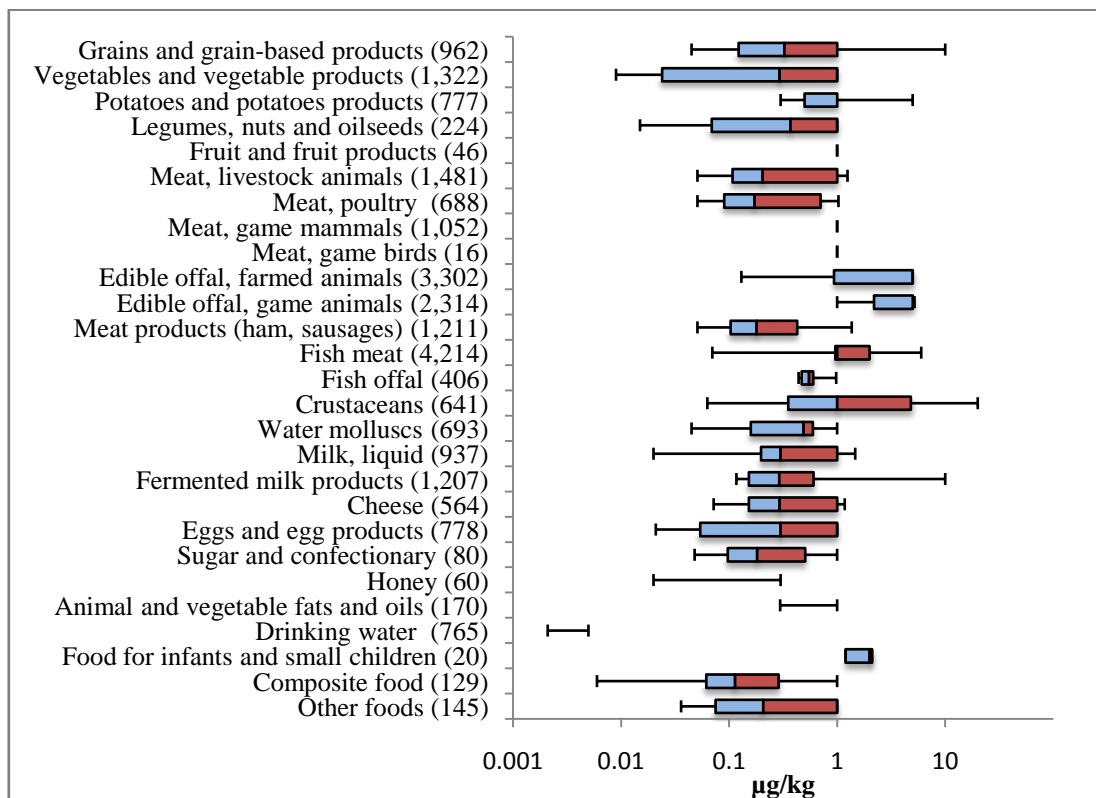


Figure 4: Distribution of the limits of quantification of single compounds across food groups (n = 24,240) (Box-plot: whiskers at P5 and P95, box at P25 and P75 with line at P50). Honey: P5, P25, P50 and P75 = 0.02 µg/kg; Animal and vegetable fats and oils: P25, P50, P75 and P95 = 1 µg/kg; Water: P25, P50, P75 and P95 = 0.005 µg/l.

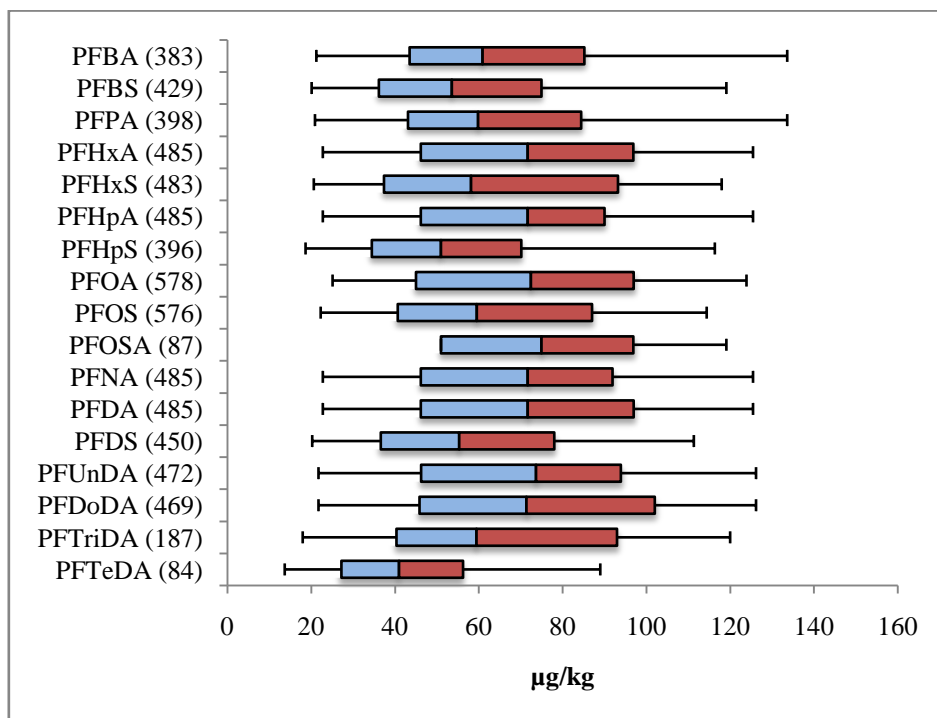


Figure 5: Distribution of the recovery rates reported for the individual perfluoroalkylated substances (n = 6,929) (Box-plot: whiskers at P5 and P95, box at P25 and P75 with line at P50).

4.3. Frequency of left-censored data

The frequency of left-censored data (analytical results reported as non-detects) was 88.2 % out of the 24,240 observations across the 17 PFASs. PFBA, PFPA and PFHpS were not detected in any of the samples analysed. The most frequently found PFASs were PFOS (31.1 %), PFTriDA (17.2 %), PFOSA (16.6 %), PFOA (11.5 %), PFDA (11 %), PFDoDA (9.8 %), PFNA (9.3 %) and PFUnDA (7 %). PFBS, PFHxA, PFHxS, PFDS and PFTeDA were detected only in a limited number of samples (Figure 6).

Across food groups, the frequency of left-censored data ranged between 80 and 100 % apart from “Edible offal, game animals”, Fish offal”, Meat, game mammals” where the percentage of non-detects was lower (Figure 7). A more detailed discussion on the results within the food groups is presented in the section 4.4.

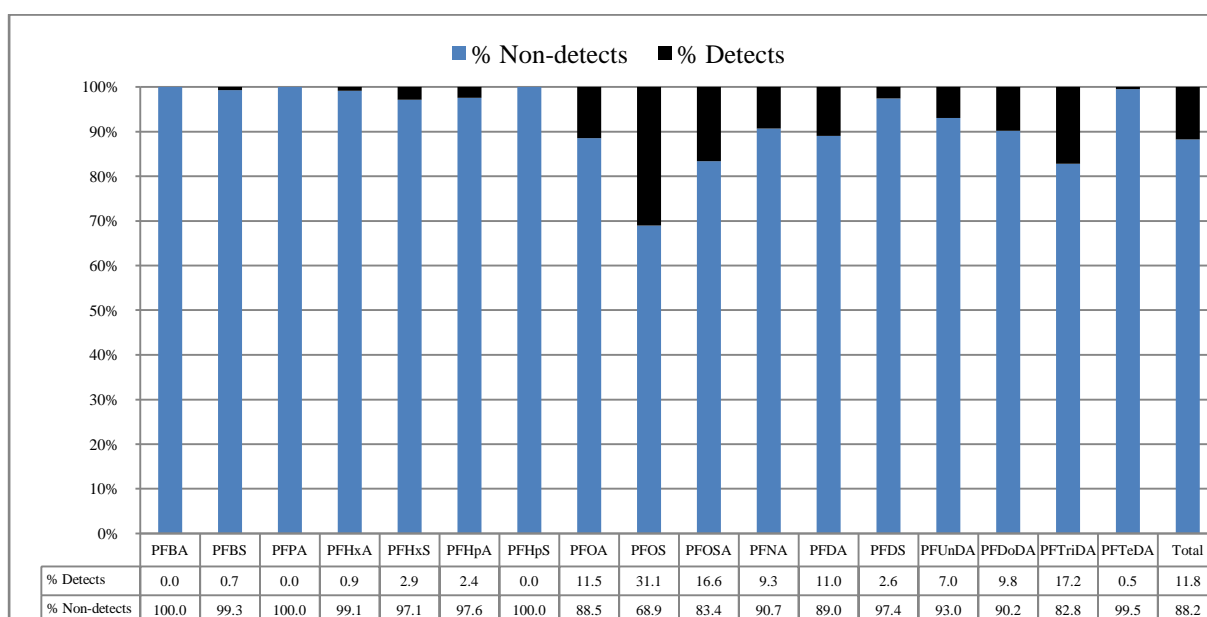


Figure 6: Frequency of results below and above the LOD or LOQ obtained for the individual PFASs (n = 24,240 results).

4.4. Contamination level across food groups

Across food groups, PFASs were mostly found in “Edible offal, game animals”, Fish offal”, Meat, game mammals”, Water molluscs”, “Fish meat”. In other food groups PFASs were detected with a very low frequency (below 5 %). No quantifiable results were obtained for any of the PFASs analysed in ‘Fruit and fruit products’, ‘Meat, game birds’, ‘Fermented milk products’, ‘Cheese’, ‘Other dairy products’, ‘Sugar and confectionary’ and ‘Food for infants and small children’ (Figure 7). However, the high percentage of results below the limit of quantification (up to 100% in some food groups) should be interpreted taking into account that for certain food groups only a limited number of samples was analysed and not all PFASs were analysed in all samples. An overview on the total number of samples analysed for the individual PFASs within each food category is given in Table 6.

In the following sub-sections only the contamination levels for the food groups where quantifiable results were reported will be discussed. Lower-bound and upper-bound values will be consistently presented only for the PFASs with at least 20% quantifiable results. Where the proportion of left-censored data was above 80 %, the estimation of UB and LB was mainly driven by the reported LODs or LOQs. The estimated occurrence values in those cases were more reflecting the variation of LOD and LOQ values and the performance of the analytical methods rather than the actual PFAS levels. However, for orientation purpose only, the mean values will be given for food groups with lower proportion of quantified results.

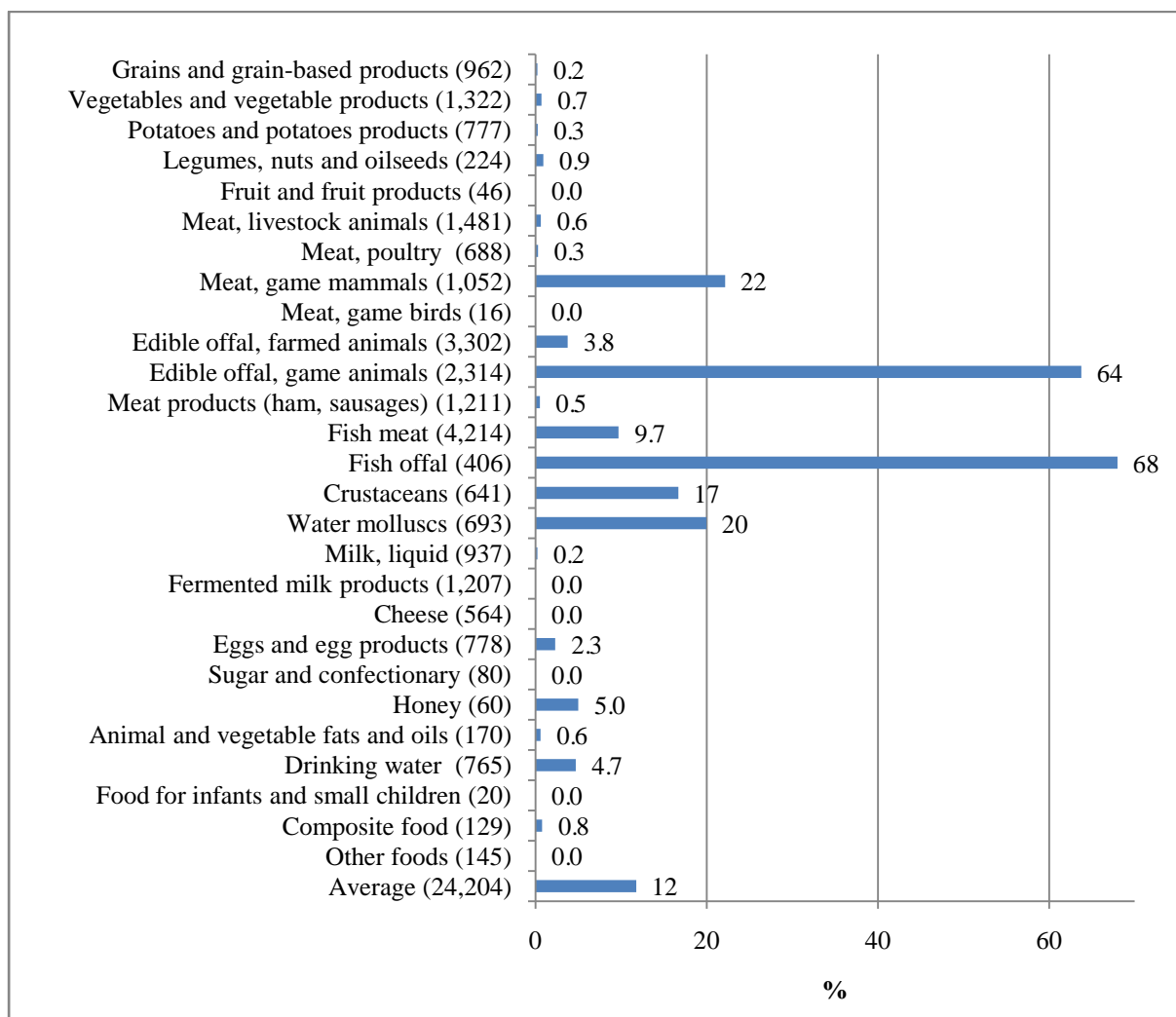


Figure 7: Frequency of results above the LOD or LOQ for the individual PFASs across food groups (n = 24,240).

Table 6: Number of samples analysed for the individual PFASs within food groups and the frequency of results above the LOD or LOQ.

Food group	Number of samples analysed (% results above the LOD or LOQ)																
	PFBA	PFBS	PFPA	PFHxA	PFHxS	PFHpA	PFHpS	PFOA	PFOS	PFOSA	PFNA	PFDA	PFDS	PFUnDA	PFDoDA	PFTriDA	PFTeDA
Grains and grain-based products	45(0)	74(1.4)	45(0)	74(0)	74(0)	74(1.4)	45(0)	74(0)	74(0)	29(0)	74(0)	74(0)	45(0)	73(0)	74(0)	7(0)	7(0)
Vegetables and vegetable products	46(0)	92(0)	46(0)	113(0)	92(0)	84(0)	46(0)	159(2.5)	160(1.3)	38(0)	92(1.1)	92(0)	46(0)	84(0)	94(2)	19(0)	19(0)
Potatoes and potatoes products	1(0)	32(3.1)	1(0)	44(0)	32(0)	23(0)	1(0)	250(0)	250(0.4)	22(0)	32(0)	32(0)	1(0)	23(0)	32(0)	1(0)	0(0)
Legumes, nuts and oilseeds	9(0)	17(5.8)	9(0)	17(0)	17(0)	17(0)	9(0)	17(5.9)	17(0)	8(0)	17(0)	17(0)	9(0)	17(0)	17(0)	5(0)	5(0)
Fruit and fruit products	0(0)	4(0)	0(0)	4(0)	4(0)	4(0)	0(0)	5(0)	5(0)	4(0)	4(0)	4(0)	0(0)	4(0)	4(0)	0(0)	0(0)
Meat, livestock animals	79(0)	105(0)	79(0)	105(0)	105(0)	105(1)	79(0)	127(2.4)	127(3.9)	26(0)	105(0)	105(0)	79(0)	104(0)	105(0)	24(0)	22(0)
Meat, poultry	39(0)	49(0)	39(0)	49(0)	49(0)	49(0)	39(0)	53(1.9)	53(0)	10(0)	49(0)	49(2)	39(0)	48(0)	49(0)	14(0)	11(0)
Meat, game mammals	0(0)	1(0)	0(0)	1(0)	1(0)	1(0)	0(0)	521(10)	522(34)	1(0)	1(0)	1(0)	0(0)	1(0)	1(0)	0(0)	0(0)
Meat, game birds	0(0)	0(0)	0(0)	0(0)	0(0)	0(0)	0(0)	8(0)	8(0)	0(0)	0(0)	0(0)	0(0)	0(0)	0(0)	0(0)	0(0)
Edible offal, farmed animals	18(0)	63(0)	18(0)	167(2.4)	63(0)	65(0)	16(0)	1153(1)	1151(7)	47(6.4)	166(3)	167(9)	16(0)	65(9.2)	109(0)	18(0)	0(0)
Edible offal, game animals	0(0)	93(0)	0(0)	93(0)	93(0)	3(0)	0(0)	873(42)	874(97)	3(0)	93(96)	93(94)	0(0)	3(0)	93(95)	0(0)	0(0)
Meat products (ham, sausages)	62(0)	84(0)	77(0)	84(0)	84(0)	84(0)	77(0)	88(1.1)	88(5.7)	7(0)	84(0)	84(0)	77(0)	83(0)	84(0)	35(0)	29(0)
Fish meat	46(0)	287(0.7)	46(0)	312(2.2)	312(0.3)	172(1.7)	46(0)	818(4.5)	819(32)	126(16)	312(0)	312(9.6)	46(0)	172(13)	296(4)	46(13)	46(0)
Fish offal	0(0)	0(0)	0(0)	33(3)	33(82)	33(36)	0(0)	38(87)	38(100)	33(100)	33(88)	33(73)	33(48)	33(97)	33(29)	33(55)	0(0)
Crustaceans	16(0)	50(2)	16(0)	51(0)	51(2)	50(0)	16(0)	61(26)	61(48)	34(24)	51(14)	51(22)	16(0)	50(20)	51(24)	16(75)	0(0)
Water molluscs	21(0)	28(0)	21(0)	52(0)	52(13)	49(14)	21(0)	56(27)	55(60)	28(75)	52(23)	52(0)	42(2.4)	49(12)	52(23)	42(36)	21(4.8)
Milk, liquid	38(0)	63(0)	38(0)	63(0)	63(0)	63(0)	38(0)	121(0)	121(1.9)	25(0)	63(0)	63(0)	38(0)	63(0)	55(0)	11(0)	11(0)
Fermented milk products	75(0)	87(0)	75(0)	87(0)	87(0)	87(0)	75(0)	87(0)	87(0)	12(0)	87(0)	87(0)	75(0)	86(0)	81(0)	16(0)	16(0)
Cheese	32(0)	43(0)	32(0)	43(0)	43(0)	43(0)	32(0)	43(0)	43(0)	11(0)	43(0)	43(0)	32(0)	40(0)	41(0)	0(0)	0(0)
Eggs and egg products	30(0)	57(0)	30(0)	57(0)	57(0)	57(0)	30(0)	86(5.8)	85(14)	27(0)	57(0)	57(0)	30(0)	50(0)	57(1.8)	6(0)	5(0)
Sugar and confectionary	5(0)	6(0)	5(0)	6(0)	6(0)	6(0)	5(0)	6(0)	6(0)	1(0)	6(0)	6(0)	5(0)	5(0)	6(0)	0(0)	0(0)
Honey	0(0)	0(0)	0(0)	0(0)	0(0)	0(0)	0(0)	30(10)	30(0)	0(0)	0(0)	0(0)	0(0)	0(0)	0(0)	0(0)	0(0)
Animal and vegetable fats and oils	1(0)	15(0)	1(0)	15(0)	15(0)	15(0)	1(0)	16(0)	16(6.2)	14(0)	15(0)	15(0)	1(0)	15(0)	15(0)	0(0)	0(0)
Drinking water	6(0)	70(5.7)	6(0)	74(2.7)	74(6.8)	6(50)	6(0)	142(7.7)	147(7.5)	0(0)	74(0)	74(0)	6(0)	6(0)	74(0)	0(0)	0(0)
Food for infants and small children	0(0)	0(0)	0(0)	0(0)	0(0)	0(0)	0(0)	10(0)	10(0)	0(0)	0(0)	0(0)	0(0)	0(0)	0(0)	0(0)	0(0)
Composite food	8(0)	9(0)	8(0)	9(0)	9(0)	9(0)	8(0)	11(9)	11(0)	1(0)	9(0)	9(0)	8(0)	9(0)	9(0)	1(0)	1(0)
Other foods	7(0)	11(0)	7(0)	11(0)	11(0)	11(0)	7(0)	11(0)	11(0)	4(0)	11(0)	11(0)	7(0)	10(0)	11(0)	2(0)	2(0)

4.4.1. Foods of plant origin

Foods of plant origin (n = 508) have been analysed for a broad range of PFASs (Table 6) but only a very limited number of positive results were found in the food groups ‘Grain and grain-based products’, ‘Vegetables and vegetable products’, ‘Potatoes and potatoes products’, and ‘Legumes, nuts and oil seeds’. Contamination levels ranged between 0.01 and 2 µg/kg for the various PFASs detected. Number of positive samples and the compounds found are presented in Table 7.

Table 7: Contamination levels (µg/kg) in food of plant origin.

Food group/PFASs	N	N detects	Min – Max
<i>Grain and grain-based products</i>			
PFBS	74	1	2
PFHpA	74	1	1
<i>Vegetables and vegetable products</i>			
PFOA	159	4	0.01 - 0.02
PFOS	160	2	0.02 - 1.1
PFNA	92	1	0.03
PFDoDA	94	2	2
<i>Potatoes and potatoes products</i>			
PFBS	32	1	1
PFOS	250	1	1.2
<i>Legumes, nuts and oil seeds</i>			
PFBS	17	1	2
PFOA	17	1	0.03

4.4.2. Meat and edible offal

PFASs found in meat and edible offal and levels of contamination are presented in Table 8. The food groups ‘Meat, livestock animals’ and ‘Meat, poultry’ were analysed for 17 compounds but only PFOS, PFOA and PFHpA were detected sporadically and at levels below 1 µg/kg.

In the category ‘Edible offal, farmed animals’, analyses were carried out for 16 PFASs. Positive results were reported for seven PFASs but only a few results for PFOS and PFOA reached levels above 1 µg/kg.

Samples in the food group ‘Meat, game mammals’ were analysed almost exclusively for PFOS and PFOA. The frequency of positive samples and contamination levels for PFOS and PFOA were substantially higher compared to ‘Meat, farmed animals’. There were three-times more positive samples for PFOS than for PFOA by a similar sampling size. Since only one sample was analysed for other compounds, no conclusion can be drawn on their presence in ‘Meat, game mammals’.

The highest contamination frequency and levels were found in ‘Edible offal, game animals’. It should be noted that 96 % of the analyses carried out in this group were on wild boar liver and thus the results strongly reflect this matrix. From the eleven PFASs for which analyses were carried out within this group, PFOS and PFOA were the most analysed compounds. PFOS, PFOA, PFNA, PFDA and PFDoDA were found in over 90 % of the samples. The highest concentrations were reported for PFOS with mean value of 216 µg/kg in both lower-bound and upper-bound. Compared to PFOS, the frequency of positive results for PFOA was roughly 2.5-times lower; the mean values in both lower-bound and upper-bound were 50-30 times lower. Although the sample size analysed for PFNA, PFDA and PFDoDA was relatively limited, and the levels found were lower than for PFOS, the high frequency of positive samples (> 90%) is an indicator that PFASs are frequent contaminants in ‘Edible

offal, game animals' and specifically in wild boar liver. However, as detailed in section 4.2., overestimation may occur in products of animal origin, notably in liver, due to interference with bile acids. It could not be ruled out whether all methods applied for obtaining the data included in this report were selective enough to completely discriminate between PFASs and the interfering compounds.

In the food category 'Meat products' analyses were carried out for 17 PFASs. Only PFOS and PFOA were found in a few sausage samples with liver content.

Table 8: Contamination levels ($\mu\text{g}/\text{kg}$) in meat and edible offal.

PFASs	N	N detects	Min – Max	Lower bound				Upper-bound			
				Mean	P50	P75	P95	Mean	P50	P75	P95
<i>Meat, livestock animals</i>											
PFHpA	105	1	0.06
PFOA	127	3	0.05 - 1
PFOS	127	5	0.04 - 1
<i>Meat, poultry</i>											
PFOA	53	1	0.05
PFDA	49	1	0.01
<i>Meat, game mammals</i>											
PFOA	521	54	1 - 11	0.25	.	.	.	1.33	.	.	.
PFOS	522	179	1 - 641	3.45	0	1.5	5	4.1	1	1.5	5
<i>Meat, game birds</i>											
PFOA	8	0
PFOS	8	0
<i>Edible offal, farmed animals</i>											
PFHxA	167	4	0.892 – 0.966	0.02	.	.	.	0.36	.	.	.
PFOA	1115	11	0.27 – 4.2	0.01	.	.	.	1.86	.	.	.
PFOSA	47	3	1
PFOS	1151	80	1 - 11	0.39	.	.	.	2.34	.	.	.
PFNA	166	5	0.08 - 1
PFDA	167	15	0.06 - 1	0.019	.	.	.	0.45	.	.	.
PFUnDA	65	6	0.03 - 1	0.03	.	.	.	0.7	.	.	.
<i>Edible offal, game animals</i>											
PFOA	873	363	0.5 - 161	4.37	0	6	20	7.09	5	6	20
PFOS	874	849	0.002 – 3,480	215.9	113	291	770	216.0	113	291	770
PFNA	93	89	1 - 30	10.22	8.9	13.9	22	10.27	8.9	13.9	22
PFDA	93	87	1.4 – 14.5	5.97	5.3	8.1	13.6	6.02	5.3	8.1	13.6
PFDoDA	93	88	0.8 – 9.2	3.62	3.4	5.2	7.3	3.67	3.4	5.2	7.3
<i>Meat products (ham, sausages)</i>											
PFOA	88	1	0.12
PFOS	88	5	0.08 – 16.5 ^(a)

^(a): liver sausage

4.4.3. Fish and fish offal

In the category 'Fish meat', analyses were carried out for 17 PFASs with the largest sampling size for PFOS and PFOA. Among the eleven PFASs identified, the highest frequency (32 %) and highest mean

contamination levels (LB: 4.18 µg/kg; UB: 4.9 µg/kg) were found for PFOS (Table 9). Other compounds found were PFBS, PFHxA, PFHxS, PFHpA, PFOA, PFOSA, PFDA, PFUnDA, PFDoDA and PFTriDA (Table 6).

The sample size for the category ‘Fish offal’ was relatively small and most of the samples were eelpout liver. Positive results were reported for all the twelve PFASs which were analysed. The highest contamination levels were found for PFOS and PFOSA with mean values of 47.1 and 14.9 µg/kg, respectively. Mean contamination levels of other analysed PFASs were generally below 1 µg/kg.

The presence of PFASs in fish was reported in several scientific papers (Berger et al., 2009; Ye et al., 2008; Delinsky et al., 2009), but it has also been pointed out that concentrations found in fish in different locations can be highly variable (Nania et al., 2009; Schuetze et al., 2010).

Table 9: Contamination levels (µg/kg) in fish and fish offal.

PFASs	N	N detects	Min – Max	Lower bound				Upper-bound			
				Mean	P50	P75	P95	Mean	P50	P75	P95
<i>Fish meat</i>											
PFBS	287	2	1 - 3
PFHxA	312	7	0.17 - 23
PFHxS	312	1	1
PFHpA	172	3	0.35 - 8
PFOA	818	37	0.02 – 18.2	0.06	.	.	.	0.86	.	.	.
PFOSA	126	20	1 - 27	0.73	.	.	.	2.7	.	.	.
PFOS	819	265	0.03 - 153	4.18	0	1	27.4	4.9	1	1	27.4
PFDA	312	30	1 - 11	0.27	.	.	.	2.2	.	.	.
PFUnDA	172	22	0.02 – 2	0.08	.	.	.	1.5	.	.	.
PFDoDA	296	14	1 - 16	0.27	.	.	.	2	.	.	.
PFTriDA	46	6	0.11 – 0.51	0.03	.	.	.	0.08	.	.	.
<i>Fish offal</i>											
PFHxA	33	1	0.53
PFHxS	33	27	0.1 – 4.9	0.98	0.19	1.68	4.7	0.99	0.19	1.68	4.7
PFHpA	33	12	0.11 – 0.22	0.06	0.00	0.11	0.22	0.09	0.06	0.11	0.22
PFOA	38	33	0.1 – 2.42	0.26	0.21	0.21	1.03	0.34	0.21	0.41	1.03
PFOSA	33	33	0.4 – 50	14.9	9.6	21.2	42.3	14.9	9.6	21.2	42.3
PFOS	38	38	1.05 – 282	47.1	10.2	27.6	222	47.1	10.2	27.6	222
PFNA	33	29	0.12 – 2.17	0.33	0.24	0.36	0.96	0.33	0.24	0.36	0.96
PFDS	33	16	0.11 – 3.3	0.57	0.00	0.12	3.10	0.60	0.06	0.12	3.10
PFDA	33	24	0.09 – 7.8	0.96	0.18	0.71	3.93	0.98	0.18	0.71	3.93
PFUnDA	33	32	0.11 – 5.51	1.05	0.57	1.03	3.56	1.05	0.57	1.03	3.56
PFDoDA	33	13	0.09 – 8.9	0.86	0.00	0.18	4.87	0.89	0.04	0.18	4.87
PFTriDA	33	18	0.1 – 3.9	0.65	0.11	0.97	3.23	0.67	0.11	0.97	3.23

4.4.4. Crustaceans and water molluscs

In crustaceans, positive results were reported for ten PFASs out of the 16 for which analyses were carried out (Table 6). The highest mean contamination was found for PFOS and PFOA (Table 10). The highest frequency of quantified results was found for PFTriDA but only 16 samples were analysed for this compound.

Water molluscs were analysed for 17 PFASs and positive results were obtained for eleven of them. The highest frequency of contamination was observed for PFOSA and PFOS (Table 6) but the contamination levels were relatively low. With the exception of a few results for PFOS and PFOSA all values were below 1 µg/kg (Table 10).

Although the sample size for crustaceans and water molluscs was small, the high frequency of positive results can be regarded as an indicator of the accumulation potential of PFASs in these organisms. Also, taking into account that seafood is considered a major source of PFAS in humans (Haug et al., 2010a) an intensified monitoring of PFASs in these food groups is recommended.

Table 10: Contamination levels (µg/kg) in crustaceans and water molluscs.

PFASs	N	N detects	Min – Max	Lower bound				Upper-bound				
				Mean	P50	P75	P95	Mean	P50	P75	P95	
<i>Crustaceans</i>												
PFBS	50	1	54
PFHxS	51	1	2
PFOA	61	16	0.02 - 8	0.6	0	0.02	5	3.3	1.9	5.1	10	
PFOSA	34	8	1 - 42	4.1	.	.	.	8.1
PFOS	61	29	0.09 - 46	3.8	0	1	31.8	8	1	20	32	
PFNA	51	7	0.16 - 3	0.18	.	.	.	3.2
PFDA	51	11	0.03 – 0.33	0.03	0	0	0.15	3	1	2.8	10	
PFUnDA	50	10	0.04 - 1	0.05	0	0	0.25	3	1	2.8	10	
PFDODA	51	12	0.03 - 38	0.81	0	0	1	3.5	1	2.8	10	
PFTriDA	16	12	0.02 – 0.28	0.08	0.08	0.14	0.28	0.09	0.075	0.14	0.28	
<i>Water molluscs</i>												
PFHpA	49	7	0.1 - 1
PFHxA	52	7	0.1
PFOA	56	15	0.03 – 0.98	0.04	0	0.04	0.1	0.26	0.1	0.2	1	
PFOSA	28	21	0.1 – 3.1	0.8	0.4	1.3	2.9	1.1	1	1.3	2.9	
PFOS	55	33	0.02 – 2.92	0.2	0.07	0.23	0.57	0.56	0.23	0.57	1.7	
PFNA	52	12	0.1 – 0.2	0.02	0	0	0.1	0.24	0.06	0.1	1	
PFDS	42	1	0.1
PFUnDA	49	6	0.1 – 0.3
PFDODA	52	12	0.01 – 0.08	0.006	0	0	0.04	0.22	0.05	0.05	1	
PFTriDA	42	15	0.01 – 0.17	0.016	0	0.02	0.06	0.043	0.04	0.04	0.06	
PFTeDA	21	1	0.1

4.4.5. Dairy products

Out of the 251 dairy product samples analysed for individual PFASs (Table 6) only two sheep milk samples were found positive for PFOS at the level of 0.14 µg/l and 0.26 µg/l (LOQs = 0.02 µg/l). The two samples were part of a set of 18 sheep milk samples originated from animals grazing in the vicinity of industrial settlements. To clarify the relationship between outdoor grazing sheep and the presence of PFASs in their milk, more data on sheep milk should be collected.

4.4.6. Eggs and egg products

Although the results provided (n = 778) for the 86 samples of eggs and egg products covered the 17 PFASs (Table 6), positive results were reported only for PFOS, PFOA and PFDODA (Table 11).

Considering that the number of samples analysed for the individual compounds was relatively low it could be worthwhile to further monitor this food group.

Table 11: Contamination levels in eggs and egg products ($\mu\text{g}/\text{kg}$).

PFASs	N	N detects	Min – Max	Mean	
				LB	UB
PFOS	85	12	0.06 – 6.4	0.19	0.57
PFOA	86	5	2.1 – 21.5	0.56	0.97
PFDoDA	57	1	1	.	.

4.4.7. Honey

Honey samples ($n = 30$) were analysed only for PFOS and PFOA. Three samples were found positives for PFOA at levels between 0.25 and 0.47 $\mu\text{g}/\text{kg}$.

4.4.8. Fats and oils

In the category ‘Animal fats and oils’ ($n = 16$), one sample (boar fat tissue) was found positive for PFOS at level of 45 $\mu\text{g}/\text{kg}$. The sampling size of this group was too small to draw a firm conclusion on the contamination with PFASs.

4.4.9. Drinking water

In drinking water, six PFASs were detected but only in few of the 147 samples analysed and at lower levels compared to food (Table 12). It should be noted that the results of 141 water samples were submitted by a single Member State and therefore it may not reflect the background contamination in Europe. Fromme et al. (2009), suggested that contaminated drinking water can also be an important source of exposure in contaminated areas. Therefore, it is recommended to monitor the contamination of drinking water at larger scale in Europe.

Table 12: Contamination levels in drinking water ($\mu\text{g}/\text{l}$).

PFASs	N	N detects	Min – Max
PFBS	70	4	0.0015 – 0.24
PFHpA	6	3	0.001 – 0.01
PFHxA	74	2	0.002 – 0.02
PFHxS	74	5	0.002 – 0.1
PFOA	142	11	0.0003 – 0.084
PFOS	147	11	0.001 – 0.012

CONCLUSIONS AND RECOMMENDATIONS

CONCLUSIONS

- The present evaluation is based on a set of 4,881 samples collected between 2000 and 2009 in seven Member States. Data were reported on different sets of compounds from a total of 17 PFASs resulting in 24,204 single analytical results. Overall, only for 11.8 % of the results a value above LOD or above LOQ was reported.
- Data were obtained by methods with heterogeneous analytical performances: high variation of the limits of quantification and of the recovery rates. 79 % of the reported LOQs were below or equal to the value recommended by the Commission Recommendation 2010/161/EC of 1 µg/kg. Median values for reported recoveries ranged between 41 % and 75 %.
- The most frequently found PFASs were PFOS (31.1 %), PFTriDA (17.2 %), PFOSA (16.6 %), PFOA (11.5 %), PFDA (11 %), PFDoDA (9.8 %), PFNA (9.3 %) and PFUnDA (7 %). By contrast, PFBA, PFPA and PFHpS were not detected in any of the samples analysed.
- Across food groups, PFASs were mostly found in certain food groups of animal origin: 'Fish offal' (68 %), 'Edible offal, game animals' (64%), 'Meat, game mammals' (22 %), 'Water molluscs' (20 %), 'Crustaceans' (17 %) and 'Fish meat' (9.7 %). In other food groups PFASs were detected with lower frequency (below 5 %). However, for several food groups and compounds only a limited number of samples was analysed and thus it was difficult to draw a clear conclusion on the contamination levels of those food groups.
- The highest mean contaminations for PFOS (216 µg/kg), PFNA (10.3µg/kg), PFOA (7.1 µg/kg), PFDA (6.0 µg/kg) and PFDoDA (3.7 µg/kg) were found in 'Edible offal, game animals'. Lower mean concentrations of PFOS and PFOA were observed in meat of game animals (both mammals and birds). Compared to the corresponding matrices of game animals, meat of farmed animals and their edible offal were less contaminated with PFASs.
- In fish, the highest mean concentrations were found for PFOS (47 µg/kg) and PFOSA (15 µg/kg) in 'Fish offal'. The same compounds had the highest mean concentrations also in 'Fish meat', though at lower level (PFOS: 4.9 µg/kg; PFOSA: 2.7 µg/kg).
- Although only a limited number of crustacean samples was analysed, there was a relatively high frequency of contamination with levels similar to those observed in fish meat.
- The current assessment includes results from analyses performed for different sets of individual compounds. Therefore, calculating and comparing sum of PFASs in samples was not possible.
- The present report includes results from both random and targeted monitoring, and therefore it should be interpreted with caution. Targeted samples and in particular samples taken from "hot-spots" may lead to an overestimation of the contamination levels.

RECOMMENDATIONS

- A harmonisation effort to define a minimum standardised set of PFASs to be analysed in all samples would be required to allow a better comparability of the contamination across food groups. For this, more research (e.g. total diet studies, biomonitoring, toxicological studies) is needed to establish the most representative PFASs.
- To enable an accurate data handling and interpretation of results, the collection and transmission of all information required in the EFSA reporting format is highly recommended.
- Considering the high percentage of non-detects in the dataset included in this report and the relatively high LOQs reported in some cases it is suggested to improve the analytical performances of the methods applied in the monitoring of PFASs in food. LOQs at low ng/kg in food and in pg/litre in beverages have been reported in recent years by several authors. Low LOQs would reduce the proportion of left-censored data thus allowing a more realistic exposure assessment. The methods applied should be selective in discriminating the presence of interfering substances, such as TDCA.
- For a more clear analysis of the background contamination and in view of an exposure assessment more data should be collected in particular for food groups where the number of samples was limited but the frequency of contamination was high (crustaceans, water molluscs) and for food groups with low contamination levels but with high intake (drinking water and other beverages, foods for infants and small children). Also, taking into account that contamination with PFASs can occur during storage, preparation and serving of food it would be important to collect more data on packaged food and ready-to-eat food (composite foods).
- As data were provided by only seven Member States and in some food groups the majority of results was generated in a single Member State, it would be beneficial to perform a larger European monitoring for all food groups, on a risk analysis basis.

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APPENDIX

A. FOOD GROUPS AND FOOD ITEMS INCLUDED IN THE ASSESSMENT

Food group	Food items (examples)
Grains and grain-based products	Cereal grains and milling products, bread and rolls, breakfast cereals, biscuits, pastry;
Vegetables and vegetable products	Root vegetables, leafy vegetables, bulb vegetable, stem vegetables, Brassica;
Potatoes and potatoes products	Mai-crop potatoes, new-potatoes, French fries, potato baked,
Legumes, nuts and oilseeds	Beans, broad bean, peas, lentils, tree nuts;
Fruit and fruit products	Raspberries, Strawberries, Avocado, Jam, Canned fruit;
Meat, livestock animals	Beef, pork, mutton/lamb meat;
Meat, poultry	Chicken meat, turkey meat, duck meat, ostrich meat;
Meat, game mammals	Wild boar meat, venison meat;
Meat, game birds	Pheasant meat, quail meat, mallard meat;
Edible offal, farmed animals	Beef liver, chicken liver, pork kidney, pork liver, turkey liver, mutton/lamb liver, mutton/la kidney, lamb heart;
Edible offal, game animals	Wild boar liver, venison liver;
Meat products (ham, sausages)	Sausages, ham, bacon, pastes, pâtés;
Fish meat	Includes samples of fish meat or samples described only as “fish” of several species;
Fish offal	Eelpout liver, bream liver;
Crustaceans	Shrimps, crabs, prawns, crayfish, lobster;
Water molluscs	Mussel, oyster, scallop, cuttlefish;
Milk, liquid	Cow milk, goat milk, sheep milk;
Fermented milk products	Yoghurts, Crème fraîche
Cheese	Different types of cheese;
Eggs and egg products	Chicken eggs, duck eggs, fresh or boiled, omelette;
Sugar and confectionary	Sugar, chocolate, caramel
Honey	Different types of honey
Animal and vegetable fats and oils	Butter, vegetable oil, fish oil;
Drinking water	Tap water, carbonated mineral water, well water, bottled water;
Food for infants and small children	Ready-to-eat meal for children, meat-based;
Composite food	Potato-based dishes, meat-based dishes, cereal-based dishes, sandwiches;
Other foods	Fruit juice, soft drinks, herbs, spices, dressing, snack, desserts;

ABBREVIATIONS

10:2 FTCA	10:2 fluorotelomer carboxylate
10:2 FTOH	10:2 fluorotelomer alcohol
10:2 FTUCA	10:2 fluorotelomer unsaturated carboxylate
4:2 FTOH	4:2 fluorotelomer alcohol
6:2 FTOH	6:2 fluorotelomer alcohol
6:2 FTS	6:2 fluorotelomer sulfonate
6:2 FTUCA	6:2 fluorotelomer unsaturated carboxylate
6:2FTCA	6:2 fluorotelomer carboxylate
8:2 FTCA	8:2 fluorotelomer carboxylate
8:2 FTOH	8:2 fluorotelomer alcohol
8:2 FTS	8:2 fluorotelomer sulfonate
8:2 FTUCA	8:2 fluorotelomer unsaturated carboxylate
LC-MS	Liquid Chromatography Mass Spectrometry
LC-MS/MS	Liquid Chromatography Tandem Mass Spectrometry
LOD	Limit of detection
LOQ	Limit of quantification
N-Et-FOSA	N-Ethylheptadecafluorooctane sulfonamide
N-EtFOSE	N-ethyl perfluorooctane sulfonamidoethanol
N-EtFOSEA	N-ethyl perfluorooctane sulfonamido ethylacrylate
N-Me-FOSA	N-Methylheptadecafluorooctane sulfonamide
N-MeFOSE	N-methyl perfluorooctane sulfonamidoethanol
PFASs	Perfluoroalkylated substances
PFBA	Perfluorobutanoic acid
PFBS	Perfluorobutane sulfonate (Perfluorobutanesulfonic acid)
PFDA	Perfluorodecanoic acid
PFDoDA	Perfluorododecanoic acid
PFDS	Perfluorodecane sulfonate (Perfluorodecane sulfonic acid)
PFHpA	Perfluoroheptanoic acid
PFHpS	Perfluoroheptane sulfonate
PFHxA	Perfluorohexanoic acid
PFHxS	Perfluorohexane sulfonate (Perfluorohexanesulfonic acid)
PFNA	Perfluorononanoic acid

PFOA	Perfluorooctanoic acid
PFOS	Perfluorooctane sulfonate (Perfluorooctane sulfonic acid)
PFOSA	Perfluorooctane sulfonamide (Perfluorooctanesulfonamide)
PFPA	Perfluoropentanoic acid
PFTeDA	Perfluorotetradecanoic acid
PFTriDA	Perfluorotridecanoic acid
PFUnDA	Perfluoroundecanoic acid
TDCA	Taurodeoxycholic acid