Fluorinated, Chlorinated and Brominated Contaminants in Fish for Human Consumption

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2009

4.3 Perfluorinated compounds
concentrations and dietary exposure to PFOS from fish products[11]

Abstract
Seven perfluorinated compounds (PFCs), including perfluorooctane sulfonate (PFOS) and perfluorooctanoic acid (PFOA) were determined in a variety of fish and shellfish that are frequently consumed in The Netherlands. PFOS was detected in 35 of the 45 analysed samples (78%) at concentrations from 2 to 230 ng/g ww (in edible parts). Concentrations in livers were higher, up to 730 ng/g ww in flounder livers from the river Western Scheldt. PFOA was found in ten samples, although at lower concentrations, 2-53 ng/g ww. In several samples longer chain PFCs (perfluoroundecanoic and –dodecanoic acid) were also found. Generally, PFC concentrations increase in the following order: open sea and ocean ≈ coastal water < freshwater. The highest concentrations were found in samples from the Western Scheldt, which may be related to use and former production along the river Scheldt.
The fish related human exposure to PFOS was estimated at 73 ng/day. Herring was the major contributor to the exposure (52%), followed by fish from the Gadidae family (incl. cod and haddock) (17%) and plaice (14%). This is substantially lower than the recent estimate by EFSA for human exposure to PFOS in The Netherlands (3410 ng/day). The difference is explained by lower fish consumption figures and lower PFOS concentrations in Dutch fish species.

Introduction
Perfluorinated compounds (PFCs) have been found in a wide variety of environmental matrices including fish (*1-4*), marine mammals, polar bears (*5-7*), and bird (eggs) (*1,4,5,8-10*). Until recently, the focus was on evaluation of ecotoxicological effects of PFCs. The human exposure to PFCs received little attention apart from several studies on PFCs in human blood and serum (*11-14*). The body burden results from several exposure pathways, including air, drinking water and food. Little attention was paid to characterization of exposure through food. Perfluoroctane sulfonate (PFOS) has several adverse effects, such as developmental effects, changes in the thyroid hormone regulation and high lipoprotein concentrations, while the liver is the major target organ for most effects *(15-17)*. Perfluoroctanoic acid (PFOA) may show e.g. developmental, reproductive and carcinogenic effects *(16,17)*.

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[11] Adapted from S.P.J. van Leeuwen, I. van der Veen, P.E.G. Leonards and J. de Boer (2006), Perfluorinated compounds in edible Dutch fish: a source for human exposure *Organohalogen compound* 68, 535-538, 2006.

The European Food Safety Authority (EFSA) recently published a human risk evaluation of PFOS and PFOA. They determined that the mean Dutch population is exposed (diet) to 58 ng PFOS/kg body weight (bw) per day *(17)*. This estimate was based on fish and drinking water. Other foods were not included in that exposure assessment due to the lack of data. The exposure (58 ng PFOS/kg bw per day) was slightly below the tolerable daily intake (TDI) of 150 ng PFOS/kg bw per day *(17)*, leaving only a small margin of safety. High fish consumers exceed the PFOS TDI. Fish was the main contributor whereas drinking water contributed less than 0.5% (14.2 ng/day). The mean dietary exposure to PFOA is 2 ng/kg bw per day, which is well below the TDI of 1.5 µg/kg bw per day *(17)*. For evaluation of human exposure in relation to fish consumption, there is a need for data on PFC concentrations in edible fish tissues. This study presents PFC concentrations in fish tissues of popular fish and shellfish consumed in The Netherlands.

Materials and methods

The investigated species are given in Table 4.9. The majority of fish and shellfish were sampled between April and October 2004. Marine fish was mostly sampled during surveys of the research vessel Tridens. Remaining samples were obtained directly from fishermen, from fish auctions or from wholesale traders. Eel was caught by electric fishery.

After transportation to the laboratory, lengths and weights of the individual fishes were measured (except for tuna, mussels, oysters and shrimps). Each sample consisted of multiple individuals (17-25 in the case of wild fish, 5-22 for farmed fish, 22 for the oyster sample; 500 grams of shrimps and 3 kg of mussels). The fish was filleted (except for farmed salmon and tuna which were purchased as parts of fillets). Each sample consisted of equal amounts of fillet per individual fish. The pooled samples were homogenized in a Waring blender. Each mussel sample was prepared by taking the meat from the shells of a 3 kg sample. Subsequently, 100 g mussel meat was pooled and homogenised. The oyster sample was prepared by pooling the meat from the individual oysters and subsequent homogenisation. Pooled shrimp samples were prepared by homogenization of approx. 500 g unpeeled and uncooked whole shrimps.

The seven compounds analysed in this study are PFOS, perfluorohexane sulfonate (PFHxS), PFOA, perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDcA), perfluoroundecanoic acid (PFUnA) and perfluorododecanoic acid (PFDoA). PFCs were extracted according to an adapted method by Hansen *et al.* *(18)*. Briefly, the method is as follows: 10 g of homogenised sample was extracted 3 times with methyl-*tert*-butylether (MTBE) in the presence of the ion pairing agent tetrabutylammonium hydrogen sulfate (TBA). The extracts were pooled and MTBE was concentrated to a final volume of 1 ml. Clean up was performed by neutral silica column chromatography (0.6 mm internal diameter glass column with 1.8 gram, 1.5% deactivated silica). The lipids were removed from the extract by (elution by 15 ml dichloromethane), followed by 30 ml acetone for elution of the target compounds. The acetone was removed by evaporation and replaced by 0.7 ml methanol, after which the extracts were ready for analysis. The extracts were injected on a Thermo Electron Surveyer high pressure liquid chromatography (HPLC) system, coupled with an LCQ-Advantage ion trap mass spectrometric system (MS) and electrospray ionisation interface (ESI).

Table 4.9 PFC concentrations in fish and shellfish in ng/g wet weight (data taken from (21)).

	Species name	**PFOA**	**PFNA**	**PFDcA**	**PFUnA**	**PFDoA**	**PFHxS**	**PFOS**
Shellfish and crustaceans								
Mussels Eastern Scheldt	*Mytilus edulis*	<2	<2	<2	<2	<4	<3	<2
Mussels, Wadden Sea (East)	*Mytilus edulis*	<2	<2	<2	<2	<4	<4	4
Mussels, Wadden Sea (West)	*Mytilus edulis*	<2	<2	<2	<2	<3	<3	<2
Shrimps, North Sea (Rijnmond)	*Crangon crangon*	<2	<2	<2	<2	<3	<6	8
Shrimps, Wadden Sea	*Crangon crangon*	<2	<2	<2	<2	<3	<6	30
Oysters, Eastern Scheldt (Yerseke)	*Ostrea edulis*	<2	<2	<2	<2	<4	<4	2
Western Scheldt and freshwater								
Flounder A liver, Western Scheldt	*Platichthys flesus*	15	5	62	33	4	<3	730
Flounder A, Western Scheldt	*Platichthys flesus*	2	<2	<2	<2	<3	<6	230
Flounder B liver, Western Scheldt	*Platichthys flesus*	53	6	50	52	<3	27	540
Flounder B, Western Scheldt	*Platichthys flesus*	3	<2	<2	8	<3	<6	93
Pike-perch liver, Hollands Diep	*Stizostedion lucioperca*	<2	<2	27	15	<3	<3	270
Pike-perch, Hollands Diep	*Stizostedion lucioperca*	<2	<2	2	3	<4	<3	40
Pike-perch, IJssel Lake	*Stizostedion lucioperca*	2	<2	<2	<2	<4	<4	150
Eel, Nieuwe Merwede	*Anguilla anguilla*	<2	<2	<2	<2	<3	<3	30
Eel, Ketel Lake	*Anguilla anguilla*	<2	3	30	57	<3	<3	57
Eel, Haringvliet (West)	*Anguilla anguilla*	<2	<2	6	8	<3	<3	37
Eel, IJssel Lake (Medemblik)	*Anguilla anguilla*	<2	<2	<2	<2	<3	<3	52
Eel, Meuse (Keizersveer)	*Anguilla anguilla*	<2	<2	<2	<2	<3	<3	5.9
Eel, Rhine (Lobith)	*Anguilla anguilla*	<2	<2	4	5	<3	<3	44
Marine fish								
Herring liver, Southern North Sea	*Clupea harengus*	<2	<2	<2	<2	<3	<3	67
Herring, Southern North Sea	*Clupea harengus*	<1	<1	<2	<1	<3	<5	8
Herring, English Channel	*Clupea harengus*	<2	<2	<2	<2	<3	<6	<1
Herring, central North Sea	*Clupea harengus*	<2	<2	<2	<2	<3	<6	51
Plaice liver, Southern North Sea	*Pleuronectes platessa*	<2	<2	2	<2	<3	<3	35
Plaice, Southern North Sea	*Pleuronectes platessa*	<2	<2	<2	<2	<4	<4	20
Mackerel, North Sea	*Scomber scombrus*	<2	<2	<2	<2	<3	<5	7
Cod, Central North Sea	*Gadus morhua*	<1	<1	<2	<1	<3	<5	<1

Table 4.9 Continued.

	Species name	PFOA	PFNA	PFDcA	PFUnA	PFDoA	PFHxS	PFOS
Haddock, Central North Sea	*Melanogrammus aeglefinus*	<1	<2	<2	<1	<3	<5	5
Sole liver, Southern North Sea	*Solea solea*	3	4	6	<2	<3	<3	130
Sole, Southern North Sea	*Solea solea*	3	<2	<2	<2	<4	<4	45
Sole, Mouth Western Scheldt	*Solea solea*	<2	<2	<2	<2	<4	<4	10
Sole, Dutch coast, Hoek van Holland	*Solea solea*	2	<2	<3	<2	<4	<4	13
Sole, Dutch coast, IJmuiden	*Solea solea*	2	<2	<2	<2	<4	<4	12
Sole, Dutch coast, Egmond	*Solea solea*	<2	<2	<2	<2	<4	<4	<2
Sole, Dutch coast, Texel	*Solea solea*	<2	<2	<2	<2	<4	<4	<2
Herring, Skagerak	*Clupea harengus*	<2	<2	<2	<2	<3	<6	23
Herring, Shetland Islands	*Clupea harengus*	<2	<2	<2	<2	<3	<6	23
Mackerel, Shetland Islands	*Scomber scombrus*	<2	<2	<2	1	<3	<5	22
Tuna, Mediterranean	*Thunnus thynus*	<2	2	<2	2	<4	<3	<2
Farmed fish								
Salmon, Farmed, Scotland	*Salmo salar*	<2	<2	<2	<2	<4	<4	<2
Salmon, Farmed, Norway	*Salmo salar*	1	<2	<2	<2	<4	<4	<2
Eel liver, Farmed, Italy	*Anguilla anguilla*	<2	<2	<2	<2	<3	<3	14
Eel, Farmed, Italy	*Anguilla anguilla*	<2	<2	<2	<2	<3	<6	<1
Eel liver, Farmed, Netherlands	*Anguilla anguilla*	<2	<2	<2	<2	<3	<3	23
Eel, Farmed, Netherlands	*Anguilla anguilla*	<2	<2	<2	<2	<3	<5	10

For PFOS and PFHxS, 7H-perfluorinated heptanoic acid (7H-PFHpA) was used as internal standard, whereas for PFOA and other perfluorinated acids $^{13}C_2$-labeled PFOA has been used. The sensitivity of 7H-PFHpA was limited at the MS conditions used for PFOS. Therefore, in some cases, $^{13}C_2$-labeled PFOA was used as internal standard for the calculation of PFOS. Admittedly, this may not be the optimum internal standard, but at the time of the study (2004) no mass-labeled PFOS was available.

Results and discussion

Table 4.9 shows that fish from nearly all origins is contaminated with PFOS. PFOA is also found in fish from different origins, although at lower concentrations and in fewer samples. Generally, concentrations increase in the following order: open sea and ocean ≈ coastal water < freshwater. The highest PFC concentrations were found in the river Western Scheldt. This is related to the (historic) production of PFCs in Antwerp and the industrial and domestic use of PFCs in the river Scheldt basin.

PFCs preferentially accumulate in fish liver as compared to muscle tissue, which can be seen in all samples of which both liver and muscle tissue were analysed (farmed eel, herring, sole, plaice and pike perch). Liver concentrations were ca. 5-fold higher than muscle tissue concentrations (mean of 8 values). PFC concentrations in flounder liver from the Western

Scheldt compare well to those reported by de Vijver *et al. (19)* and Hoff *et al. (20)* for this location.
Freshwater fish (eel and pike perch) show in this study the next highest concentrations as compared to the other locations. Hoff *et al.* found very high PFOS concentrations in eel liver samples (up to 9 µg/g ww) from the Ieperlee canal at Boezinge (Belgium) (*2*), being much higher than the levels in eel (incl. farmed eel livers) and pike-perch in our study. The authors suggested that nearby industrial and household discharges may have caused these high concentrations (*2*).
The PFOS concentrations for sole and plaice from the Southern North Sea in our study compare well to those reported for plaice and bib caught off the Belgian North Sea coast *(20)*. PFOS concentrations in other marine fish are in the same order of magnitude as those reported by Kallenborn *et al. (22)*
Other PFCs detected were e.g. PFDcA and PFUnA (up to 60 ng/g ww) in flounder liver from the Western Scheldt. PFDcA and PFUnA were also found in eel from the river Rhine and the Ketel Lake. PFNA was found in four samples (up to 6 ng/g ww) and PFDoA was found in only one flounder liver from the Western Scheldt (4 ng/g ww). PFHxS was detected in one Western Scheldt flounder liver (27 ng/g ww).
Concentrations in Chinese seafood (wild fish) were in the same range as those observed in the present study (PFOS: 0.4-2.9 ng/g ww (fish) and 1.8-14 ng/g ww (shrimp), PFOA: 0.42-0.45 ng/g ww (shrimp) and <LOD (fish), PFNA: all <LOD, PFDcA: 0.3 ng/g ww (shrimp) and <LOD in fish, PFUnA: 0.35-0.65 ng/g ww (fish) and 0.42-0.93 ng/g ww (shrimps) *(23)*). PFOS concentrations in seafood (wild fish) from Cataluna, Spain were lower (0.65 ng/g ww) and PFOA was <LOQ *(24)*. The concentrations in wild fish are higher than those in farmed fish (see Chapter 4.4). PFCs were only detected in approx. half of the farmed fish samples analysed at concentrations from 20 (PFUnA) to 600 pg/g ww *(25)*.

Human exposure

Virtually no detailed information is available on human exposure to PFCs from fish consumption. Therefore, an estimation was made of the fish-related dietary exposure of the mean Dutch citizen. This estimation was limited to PFOS as only for this PFC considerable data on individual species is available (see Table 4.9). This data was combined with consumption data from the Dutch National Food Consumption survey (DNFCS) of 1997/1998 *(26)*. The DNCSF contains files of 6250 people in the age of 1 to 99 years, who recorded the food they consumed during two consecutive days in a food diary. The PFOS exposure calculation is based on the daily fish consumption multiplied with the mean lower bound concentrations (per species): DE = (Qa x Ca + Qb x Cb +…Qz x Cz), in which DE = Daily Exposure, Q = Quantity of daily consumption of fish species and C = mean lower bound concentration of

PFOS in fish species (measured in this study). A to z indicate different fish species.
These estimations are compared with an estimation made by EFSA (Table 4.10) for which the Concise European Food Consumption Database (CEFCD) for the exposure assessment was used *(17,27)*. This database contains concise food consumption data from a variety of European countries for people from 16-64 years. The food consumption data used in the EFSA estimate concerns *consumers-only* data, meaning only data of people that actually consumed fish during the survey were used. The PFOS concentration data used for this estimation is the mean of PFOS concentrations in fish from various European countries *(17)*. The dietary exposure estimated by EFSA therefore results from DE = (Q_{mean} x C_{mean}) in which DE = Daily Exposure, Q_{mean} = mean quantity of daily consumed fish species (*consumers-only*) and C_{mean} = mean PFOS concentration in fish from Europe. The results are presented in Table 4.10.

Table 4.10 Dietary exposure estimations for the Dutch population according to two scenarios.

	EFSA[1]	This study
Food consumption data base:		
Name abbreviation	CEFCD[2]	DNFCS[3]
Population included[4]	Consumers only	Total population
No. individuals / age range	901, 16-64	6250, 1-99
Exposure input data:		
Fish consumption (g/day)	50 (median)	10.4 (mean)[5]
PFOS concentration in fish (ng/g ww)	68.1 (mean)	Range: <1-57 (see Table 4.9)
Results:		
Exposure (ng/day)	3410	72.5
Exposure (ng/ kg bw per day) for a 60 kg person	56.8	1.2
Margin of exposure:	2.6	125

1 *Scenario taken from (17);*
2 *Concise European Food Consumption Database (agregated fish consumption data);*
3 *Dutch National Food Consumption Survey (individual fish consumption data);*
4 *Consumers only: data only based on persons that actually consumed fish during the survey; total population: data based on the entire survey population, also including persons who did not consume fish at all during the survey;*
5 *This mean is based on fish species of which both consumption data (DNFCS) and contaminant data (this study) were available. Other species were left out. The included fish species represent 92% of all consumed species.*

The EFSA estimates of the PFOS exposure from fish and drinking water consumption for Italy, The Netherlands, Sweden and the UK are 58, 57, 45 and 49 ng PFOS/kg bw per day, respectively *(17)*. Fish dominates this exposure. Drinking water contributed only <0.5%, and can, therefore, be neglected. These exposure estimates are much higher than estimates resulting from the present study (1.2 ng/kg bw per day, or 73 ng PFOS per day for a 60 kg person). This is partly due to the high fish consumption estimate used by EFSA

(50 g/day) for the Dutch adult population. This estimate was based on *consumers-only*, which results in a conservative, higher exposure estimate. The data from the DNFCS were taken as *total population* data, which is 5-fold lower (10 g/day), because the frequency (and therefore volume) of fish consumption in The Netherlands is much lower than the *consumers-only* data would suggest. Furthermore, the PFOS concentrations in fish in the EFSA dataset (covering whole Europe) covered 3 to 4 orders of magnitude and contained higher PFC concentrations than are being found in The Netherlands (excluding fish from the Western Scheldt area) *(17)*. The resulting mean value of 68.1 ng/g ww was used for the exposure estimation for the four aforementioned countries, leading to a higher exposure estimate. The lower fish consumption data and the lower PFOS concentration data in this study resulted in a 50-fold lower exposure (72.5 ng/d) and a 50-fold larger margin of exposure compared to EFSA. In case we would consider Dutch consumers-only, the difference with the EFSA would have been less pronounced.

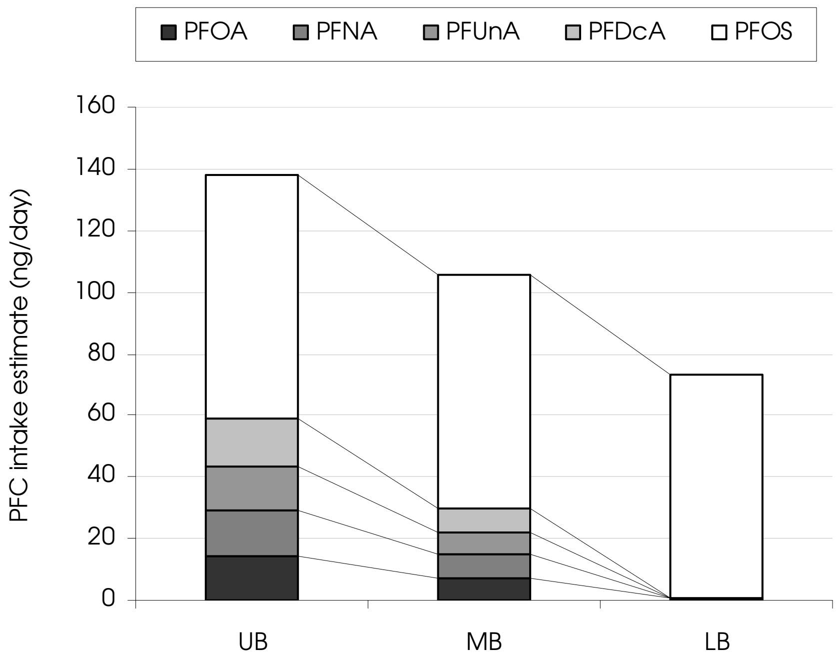

Figure 4.8 PFC exposure estimate for the Dutch population from fish consumption. UB = upperbound, MB = middle bound, LB = lower bound

The mean dietary intake for the general population of Bavaria, Germany was estimated at 1.8 ng/kg bw per day (duplicate diet study, 31 individuals, aged 16-45 years) *(28)*. This is slightly higher than in our study, possibly because Fromme et al. included all food items and beverages whereas the present study only took fish into account. The dietary exposure of the Catalan citizens to PFOS was estimated to be 63 ng/d for food, to which fish contributed 34 ng/d *(24)*. No beverages (except for milk) or drinking water were included. The intake from drinking water in Catalonia was estimated at 0.8-1.7 ng/d *(29)* being 10-20 times lower than the EFSA estimate (14 ng/d) *(17)*.

The human exposure is predominated by PFOS in all three scenarios (lower bound, LB; medium bound, MB and upper bound, UB) (see Figure 4.8). Because PFOS was detected in almost all samples, there is only a small difference between the UB and LB scenario for this PFC (79 vs. 73 ng/d). For the other PFCs, the differences are much larger because of the low detection frequency. Focussing on PFOS, herring contributes most to the intake (52%, Figure 4.9), followed by fish from the Gadidae family (cod, haddock), plaice and shrimps.

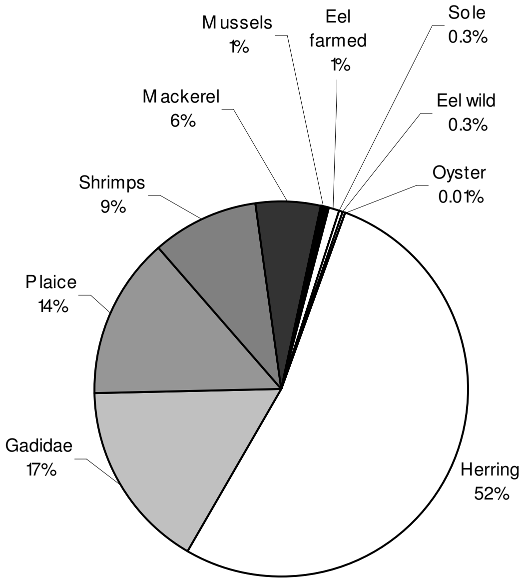

Figure 4.9 Relative contribution of fish species to the fish related human exposure to PFOS (lowerbound). The contribution of salmon and tuna was 0% (not shown in graph).

Acknowledgements

The Dutch Ministry of Agriculture, Nature Management and Food Quality is gratefully acknowledged for providing funding. Furthermore, Dr. M.I. Bakker is acknowledged for making the DNFCS fish consumption data available.

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