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Occurrence of perfluoroalkyl substances (PFASs) in a large number of wild and farmed aquatic animals collected in the Netherlands

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HIGHLIGHTS

- PFASs levels in 246 fish and fishery product samples is presented.
- PFAS levels in eel ≫ bivalves and crustaceans > marine fish > farmed fish.
- ΣPFASs levels up to 172 ng/g ww were detected in eels.

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ABSTRACT

A range of perfluoroalkyl substances (PFASs) was analysed in marine fish, farmed fish, crustaceans, bivalves and European eel caught in (mostly) Dutch waters, or purchased at Dutch markets (approximately 250 samples, collected between 2012 and 2018). ΣPFAS levels were highest in eels collected from rivers and lakes (average 43.6 ng/g and max 172 ng/g), followed by shrimps collected near the Dutch coast (average 6.7 and max. 33 ng/g ww), and seabass (average 4.5 and max. 9.4 ng/g ww). Most of the farmed fish (e.g. trout, catfish, turbot, salmon, tilapia, pangasius) were among the lowest contaminated samples in this study (averages ranged from 0.06 to 1.5 ng/g ww). Geographically, levels in marine fish from the northern North Sea (e.g. haddock, whiting, herring) were lower than in the central and southern North Sea (e.g. cod and flatfish). Concerning eel, no substantial geographical differences were found (apart from two distinct locations). The contamination pattern was similar in all species, where PFOS mostly dominated the profile, and other long-chain PFASs being frequently detected. Short-chain PFASs were rarely found. PFOS concentrations in eel varied from 3.3 ng/g (close to the North Sea) to 67 ng/g ww in eel caught from Ghent-Terneuzen canal. The majority of detected PFOS levels in eels (93%) and 1 shrimp sample from Eems-Dollard exceeded the EU Environmental Quality Standard (EQS) for surface water of 9.1 μg/kg ww. Other samples (e.g. shrimps, bivalves, flounder), subject to the EQS, did not exceed this level.

1. Introduction

Perfluoralkyl substances (PFASs) are a class of widespread pollutants of which some may present a potential risk for human health. A recent risk assessment by the European Food Safety Authority (EFSA) implied that effects of perfluorooctanoic acid (PFOA) and perfluorooctane sulfonate (PFOS) may be detected at levels observed in at least part of the human population (EFSA, 2018). Most of the exposure to PFASs (and other persistent organic pollutants, POPs) comes from animal derived products, including fish, shellfish and crustaceans. In the Netherlands, levels of POPs are routinely measured in marine and farmed fish, shellfish and

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crustaceans. Concerning freshwater fish, commercial eel fishing has been ongoing for many centuries but the safety of consuming wild eels is under debate since many rivers and lakes are polluted and eels have a high potential for accumulating POPs, as it is a long-living benthic predator residing in the same location for most of its life (de Boer et al., 2010; Guhl et al., 2014; Kwadijk et al., 2010). Several studies have reported on levels of PCDD/Fs, PCBs and BFRs in European eels (*Anguilla anguilla*) (Gotz et al., 2017; Malarvannan et al., 2014; de Boer et al., 2010; Van Leeuwen and de Boer, 2008). High levels of PCDD/Fs and PCBs in eel from particularly the main Dutch rivers like Rhine, Meuse, IJssel and their delta resulted in a ban on commercial eel fishing from highly polluted fishing areas in the Netherlands from 2011 onwards (http://wetten.overheid.nl/BWBR0024539/2015-09-22#Bijlage15). Other areas like the Lake IJssel are open for commercial eel fishery since PCDD/F and PCB levels in eel are lower than the current maximum levels.

PFASs have been reported in earlier studies in different marine fish species sampled in The Netherlands, including herring, mackerel, cod, plaice, common dab, haddock and farmed species like shrimps, salmon, trout, tilapia and pangasius (Hoff et al., 2003; Van Leeuwen et al., 2009, 2013; Noorlander et al., 2011). However, most of these studies are snapshots rather than a systematic investigation of PFASs levels in frequently consumed aquatic species. In a broader perspective, the recent EFSA opinion (2018) reported on samples collected throughout Europe, showing that these contaminants accumulate in a wide range of edible wild fish, farmed fish, mollusk and shellfish species. Several European studies over the last years showed that PFASs also accumulate in freshwater eels (Kwadijk et al., 2010; Hölzer et al., 2011; Guhl et al., 2014; Couderc et al., 2015; Giari et al., 2015; Pignotti et al., 2017). Furthermore, a recent study investigating the accumulation of POPs in men consuming eel from Dutch polluted rivers (Van den Dungen et al., 2016) showed that these men have higher serum levels of PFASs compared to men consuming eel from aquaculture or from relatively clean areas. In that study, no PFAS levels in eels from polluted Dutch areas were investigated, which remains a knowledge gap to date (with the exception of PFOS) (Kwadijk et al., 2010).

There is quite some debate on the human risk of PFASs. Recently, the EFSA re-evaluated its Tolerable Daily Intakes (TDIs) established in 2008 for PFOS and also PFOA of 150 and 1500 ng/kg bw/day, respectively (EFSA, 2008). EFSA derived much lower Tolerable Weekly Intakes (TWI) of 13 and 6 ng/kg bw/week for PFOS and PFOA (EFSA, 2018), respectively. These TWIs are based on associations of human serum levels with increased serum cholesterol, but also protect against other adverse effects associated with relatively low serum levels, like reduced vaccination response in children, liver damage (indicated by abnormal serum ALT levels), and reduced birth weight. The critical PFOS levels in serum were clearly lower than those observed in consumers of eel from polluted rivers (Van den Dungen et al., 2016). These new TWIs are lower than most established health based guidance values (HBGVs) based on effects in laboratory animals, but similar to the provisional minimal risk levels which were recently proposed by the US-ATSDR for PFOS (2 ng/kg bw/day) and PFOA (3 ng/kg bw/day) (https://www.atsdr.cdc.gov/mrls/mrllist.asp#237tag). In the Netherlands, RIVM derived a TDI for PFOA of 12.5 ng/kg bw/day based on liver effects in rats (Zeilmaker et al., 2016). RIVM did not establish TDIs for PFOS or other PFASs, but based on similar effects, RIVM recently proposed a set of relative potency factors (RPFs) for various PFASs, thereby linking the sum of these PFASs to the TDI for PFOA (Zeilmaker et al., 2018). EFSA is currently reviewing the possibility to derive an HBGV for the group of PFASs, including PFOS and PFOA. There are currently no maximum levels for PFASs in fish or other food. However, in the EU Directive 2013/39/EU (EU 2013), an environmental quality standard (EQS) for PFOS in biota on the basis of human exposure was set, being 9.1 μg/kg ww. This EQS is one of the tools to monitor surface water quality.

Considering these much lower HBGVs and the lack of data on PFASs in fish from Dutch marine and fresh waters, it was appropriate to collect more data to enable a human exposure assessment through the consumption of aquatic animals in the Netherlands. Therefore, the aim of the present study was to investigate the occurrence of PFASs in various fish, bivalves and crustaceans, collected in the Netherlands between 2010 and 2018. To our knowledge this is the first study reporting levels of not only PFOS but also other PFASs in a large number of eel samples (n = 86) collected from rivers and canals covering a large part of the Netherlands, as well as a large set of marine fish (n = 78), farmed fish (n = 52) and bivalves and crustaceans (n = 30).

2. Materials and methods

2.1. Sample collection

All samples were collected by Wageningen Marine Research (WMR). The marine fish, farmed fish, bivalves and crustacean samples were collected from 2012 to 2018 (160 samples). The marine fish investigated in this study were caught in the North Sea (ICES/FAO areas IVa to IVc), the English Channel (ICES/FAO area VII) and the Atlantic ocean (ICES/FAO area V). Shrimps and brown crab were caught in the Dutch Wadden Sea or close the Dutch west coast. Mussels originated from the Eastern Scheldt and Wadden Sea. All marine fish samples were obtained from commercial fishermen or sampled by the Tridens research vessel. Details on the sampling were, whenever possible, recorded (including date and coordinates; see Supplementary Data Table S2). Investigated fish species were herring, mackerel, cod, hake, whiting, common sole, plaice, flounder, dab and seabass. Farmed fish were obtained from whole sale traders or directly from aquaculture companies, and sample details were recorded (type of sample, country of origin). Investigated farmed species included salmon, trout, turbot, catfish, tilapia, pangasius and eel. In principle, for each sample, 25 individuals (whenever available) were pooled in order to reduce variability from biological origin. In some cases, a lower number of individuals was pooled if a lower number was available that met the sampling criteria. In the case of aquaculture samples, the variance was assumed to be less pronounced and therefore in some cases a lower number of individuals was allowed. Upon arrival at the laboratory, intestines were removed (if not yet removed at sea) and the samples were frozen at −20 °C until further processing. For shrimps, 3 kg of unprocessed shrimps were collected. Mussels were processed directly, without depuration. Three kg of mussel meat was collected from the bivalves. The collected fillets of individual fish, the collected brown crab meat, collected white meat, the mussel meat and the collected whole shrimps were pooled per location, year and (if applicable) size class (Supplementary Data Table S2). Each pooled sample was ground and homogenized and kept into polyethylene or glass bottles in order to avoid PFAS contamination of the sample, or absorption of the PFASs present in the sample to the bottle. Finally, all the fish samples were stored in the freezer (−20 °C) till the analysis.

The 86 investigated eel samples were collected by WMR from different rivers, canals and lakes in the Netherlands in the period May–June, during the years 2010–2016. The eel samples are thought to represent the local state of contamination. The eels were collected by electric fishery or, in brackish waters, by fykes. For

every sample, multiple animals (preferably 25) were targeted to ultimately result in a single pooled eel sample (as discussed below). In specific cases, the target number of 25 could not be reached, despite extensive fishing, probably due to low eel densities. The collection and treatment of animals was subjected to ethical approvals, which was granted. Eels were distributed over two size ranges, i.e. 30–40 cm and >45 cm (nose tip to tail fin end). Often only one size range was collected, in some cases two size ranges. The eels were transported to the WMR laboratory, where sizes (tail end to nose tip) and weights of individual eels were recorded. The sampling locations of the eel samples are illustrated in Fig. 1, and details of the samples can be found in the supporting information (Supplementary Data Table S3). The eels were filleted and the collected fillets were pooled per location, year and size class. Each pooled sample was ground and homogenized and stored in the freezer (−20 °C). The frozen samples were sent to RIKILT for analysis.

2.2. Chemicals and materials

In the current study 16 PFASs were included: perfluorobutanoic acid (PFBA), perfluoropentanoic acid (PFPeA), perfluorohexanoic acid (PFHxA), perfluoroheptanoic acid (PFHpA), perfluorooctanoic acid (PFOA), perfluorononanoic acid (PFNA), perfluorodecanoic acid (PFDA), perfluoroundecanoic acid (PFUnDA), perfluorododecanoic acid (PFDoDA), perfluorotridecanoic acid (PFTrDA), perfluorotetradecanoic acid (PFTeDA), perfluorobutanesulfonic acid (PFBS), perfluorohexanesulfonic acid (PFHxS), perfluoroheptanesulfonic acid (PFHpS), PFOS and perfluorodecanesulfonic acid (PFDS). The analysis of eel samples was conducted at RIKILT, whereas the analysis of the marine and farmed fish, mussels and shellfish was conducted at WMR. The details of the chemicals and materials (e.g. origin, purity) used throughout the sample analysis are provided in paragraphs S1 and S2.

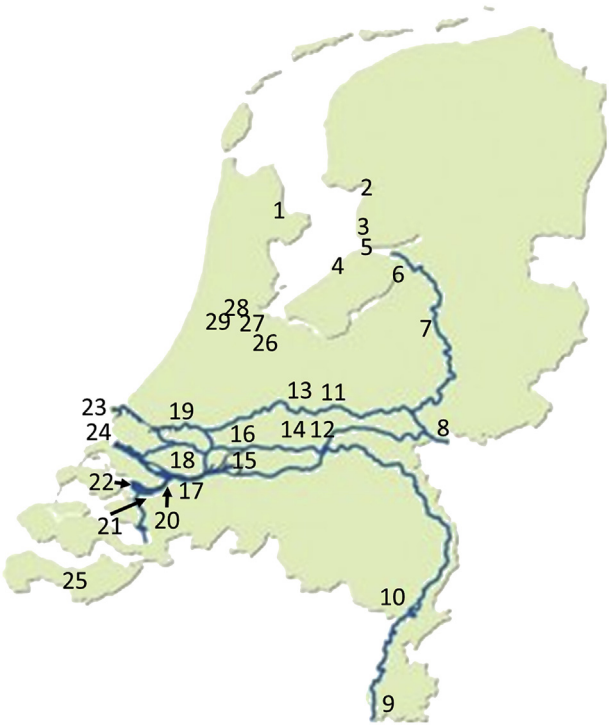

Fig. 1. Eel sampling areas in Dutch surface water locations. Each location has a unique number, but per location, multiple samples could be taken (e.g. several years or eel sizes). Details of the samples are provided in Table S3.

2.3. Sample preparation and analysis of marine fish, farmed fish, crustaceans and bivalve samples

The samples were analysed by WMR according to the method previously published by Kwadijk et al. (2010). Details are provided in Paragraph S2. Briefly, the sample was fortified with mass labeled PFOS, PFOA and PFBA and subsequently extracted with acetonitrile by shaking, followed by centrifugation. The extract was dried over sodium sulphate, washed with hexane for removal of interferences and transferred to Eppendorf tubes containing ENVIcarb for additional clean-up. After centrifugation, the acetonitrile layer was concentrated and an equal volume of demineralised water was added prior to injection. Instrumental analysis was carried out using a HPLC system coupled with a Thermo Finnigan LCQ advantage Ion-Trap MS instrument with electrospray (ESI-MS/MS). 10 µl of extract was injected onto a 100 × 2.10 mm (5 µm) Fluorphase RP column. The MS/MS mode was used to determine the carboxylic acids. Because of the limitations of the ion trap mass spectrometer, it was not possible to use the MS/MS mode for the assessment of the sulfonates. However, in order to remove some interference MS/MS mode is still used by setting the target-ion to the mass of the required compound with 20% fragmentation energy and subsequently monitoring the same fragment (*m*/*z* 499) (see paragraph S2 for all details).

The WMR method was validated according to NEN7777 and accredited under ISO17025 for PFOS and PFOA. Repeatability was tested by duplicate analysis (2 replicates) of the same 8 fish samples on the same day. While reproducibility of the method was tested by multiple analyses (8 replicates sample) of the same fish sample on eight different days. The calculated interday RSD% were 2.4% and 7.7% for PFOS and PFOA respectively. Calibration curves, covering concentrations from 0.5 ng/ml to 300 ng/ml (8 points excluding 0 ng/ml), were used for the quantification of the PFAS concentrations in the samples. The r^2 was greater than 0.99 for all the calibration curves. Limit of quantification (LOQ) for each compound is calculated using sample intake and the used standard in the calibration curve. All PFOS isomers (branched + linear) were summarised and quantified against a linear-PFOS standard. Any sample detected at a lower concentration than the lowest standard is reported as < LOQ. Sample intake varied between years resulting in a lower or higher LOQ per sample. With each set of samples blank and an internal reference sample (pike perch) are analysed. No PFASs were detected in the blanks during analysis while results for the internal reference sample were all within normal limits (<2s). WMR also takes part in the Quasimeme proficiency tests (http://www.quasimeme.org/) where satisfactory results are obtained.

2.4. Sample preparation and instrumental analysis of eel samples

The samples were analysed by RIKILT in consecutive years. During those years, the method of analysis was adapted. The analysis of eel samples of 2010–2015 was performed according to the method first published by Vassiliadou et al., (2015). Detailed information can be found in the Paragraph S1, but briefly, the method starts with 1 g of sample, to which the mass labeled internal standard mixture was added. The sample was extracted with pressurized liquid extraction (PLE) with MeOH, cleaned-up over a glass column filled with 1.5 g florisil, 1 g basic aluminum and 1 g of sodium sulphate. The sample was eluted with MeOH, concentrated and subsequently analysed by LC-MS/MS. The samples of 2016 were fortified with internal standard mixture, sodium hydroxide was added for alkaline digestion and extraction was performed with ACN, followed by clean-up using weak anion exchange Oasis WAX cartridges. The final extract was analysed by LC-MS/MS. Detailed information on the instruments used and their settings is presented

in the supporting information (Paragraph S1; Table S4). In order to assess the comparability of both extraction methods, a selection of 7 eel samples was analysed by both approaches, and showed reasonable comparability as shown in Table S5.

The methods were validated for repeatability, reproducibility, specificity, recovery and sensitivity according to the Eurachem guide (Magnusson B. and Örnemark U., 2014) (Paragraph S1).

For the analysis of the samples, an isotope dilution method was applied, using mass-labeled internal standard (Table S4). The recoveries of the mass labeled internal standards, added prior to extraction, were monitored and ranged between 60 and 115% for all the mass-labeled compounds for the samples up to 2015 (ASE-Silicagel method) and 62–103% on average for the different PFASs for the 2016 samples (Oasis WAX method) (Table S6). Quality-control (QC) standards (one blank and one eel sample spiked at 10 ng/g) were analysed in every batch of samples, controlling in this way the repeatability of the analytical method. The recovery of the spike varies from 76 to 104% on average for the different PFASs. In addition, for the identification of the analytes, the ion ratio of the secondary mass transition response relative to the primary mass transition response and the retention time were recorded for each compound. The response of the instrument was also monitored by adding $^{13}C_8$-PFOS and $^{13}C_8$-PFOA into the vial just before the injection. The recovery of $^{13}C_8$-PFOS and $^{13}C_8$-PFOA ranged from 90 to 120% in all the samples, verifying the absence of matrix effects and the sufficient ionisation of the compounds. All PFOS isomers (branched + linear) in sample extracts were summarised and quantified against a linear-PFOS standard. Background contamination was also monitored by the analysis of blank samples in every sequence. No PFASs were detected in any of the blank samples. The limit of detection (LOD) and the limit of quantification (LOQ) for both methods were determined as 3 and 10 times the signal to noise ratio, respectively. A reporting limit was set at 0.3 ng/g for all the compounds except for PFBA and PFPeA, where it was set at 5 ng/g. RIKILT participated in the QUASIMEME proficiency testing scheme for PFASs in fish matrices.

3. Results and discussion

3.1. PFAS levels and patterns in marine fish, farmed fish, bivalves and crustaceans

Fig. 2 shows the average concentrations in the samples for the most abundant PFASs (PFOS and C_8-C_{13} PFCAs). Other PFASs, particularly the short chain-length compounds were rarely detected. Detailed information on individual samples with calculated average, median, maximum values and detection frequencies can be found in Table S7. The averages presented in Fig. 2 were calculated based on the lowerbound principle, meaning that < LOQ values were replaced by 0 and not by the LOQ value itself (upperbound principle). This was considered the best approach as LOQ values varied over the years, which would have considerably influenced the average value, presumably leading to an overestimation of the average PFAS levels. In some cases, the detection frequency was low (particularly in the case of farmed fish and some marine fish) or the PFAS results were non-normally distributed. Despite these limitations, we consider the average values presented in Fig. 2 the best representation of the PFAS concentrations in edible fish over the 2012–2018 period.

PFOS was generally detected at higher frequency than other PFASs, and often at higher levels. In the crustaceans/mussels group, highest PFOS levels (up to 25 ng/g ww) were found in shrimps taken from the Eems-Dollard location (Table S7), suggesting a local contamination source, or contaminated effluent from the Eems river. Similar levels of PFOS (13.9 ng/g ww) in shrimps have been reported in a previous study analyzing samples from China (Gulkowska et al., 2006). However, in the current study, shrimps from other sampling locations along the Dutch coast showed lower levels of PFOS (<0.6–4.6 ng/g ww), that are in accordance with a previous study presenting PFAS levels in shrimps from Greece, where the detected PFOS concentration was 5.15 ng/g ww (Vassiliadou et al., 2015). For mussels, the 4 samples were collected from the Wadden Sea and Eastern Scheldt. One of the shrimps showed high levels of PFASs (ΣPFASs: 14.9 ng/g ww) and especially of PFUnDA (9.6 ng/g ww), while the other did not (ΣPFASs: 0.5 ng/g ww). However, the number of samples is limited, which hampers drawing solid conclusions. Low levels of ΣPFASs and PFOS have been reported previously in mussels from Spain (Zalabeta et al., 2015; Gómez et al., 2011; Fernandez-Sanjuan et al., 2010), France (Munschy et al., 2015), Greece (Vassiliadou et al., 2015), Denmark (Bossi et al., 2008), California coast (Dodder et al., 2014) and the Mediterranean Sea (Nania et al., 2009). Brown crab meat (body tissues; n = 6) contained higher levels (ΣPFASs: up to 8.2 ng/g ww) than the white meat (i.e. muscle tissue from the legs and claws, 'appendages', n = 7) (ΣPFASs: <0.06–0.8 ng/g ww). Nevertheless, these levels of PFASs were lower compared to a previous study (Clarke et al., 2010), in which the ΣPFAS concentration in the brown crab body tissue was 14 ng/g ww.

In the marine fish group, the highest levels were observed in seabass (Fig. 2), presumably as a result of the predatory nature of this species. ΣPFAS concentrations ranged up to 9.4 ng/g ww, primarily caused by PFOS. The PFOS levels in seabass were higher compared to the ones reported in previous studies (Paiano et al., 2013; Berger et al., 2009; Nania et al., 2009). Most flatfish samples (flounder, dab, sole and plaice) were taken close to the Dutch coast (6–12 miles off the coast). The river effluent of the rivers Meuse and Rhine, when entering the North Sea, is transported by sea currents in a northern direction along the Dutch coast and through the Wadden Sea. As a result, the contaminated water and sediments from the rivers are also transported along the Dutch coast, and that may influence the contamination of the fish and shellfish caught in this 'plume'. Cod and haddock showed higher PFAS levels (ΣPFASs up to 2.3 ng/g ww) than other *Gadidea* family members like whiting (ΣPFASs up to 0.4 ng/g ww), and hake (ΣPFASs up to 0.4 ng/g ww). Most likely, the catchment area plays a role here as cod was caught mostly in the south and central North Sea, which is more polluted than the northern North Sea (as shown in earlier studies on other POPs; de Boer and Brinkman, 1994) where most whiting and hake samples were collected. Haddock was sampled south-west of Ireland. The current results agree with previous studies in the same marine fish. In particular, cod and whiting showed low PFASs levels (Ericson et al., 2008; Schecter et al., 2010; Clarke et al., 2010; Haug et al., 2010). On the other hand, Vassiliadou et al. (2015) have reported ΣPFAS levels (2.33 ng/g ww) higher than the ones of the current study.

In farmed fish, the PFAS levels were generally low (ΣPFAS concentration below 1.3 ng/g ww). This agrees also with other studies where farmed fish showed lower PFAS contamination than freshwater or marine fish (Koponen et al., 2015; Mwakalapa et al., 2018; Brambilla et al., 2015). A substantial variation was observed among the individual farmed eel samples and the number of analysed samples was low (n = 4, ΣPFASs: 0.36–2.5 ng/g ww). Thus, more data on farmed eel are needed before solid conclusions can be drawn. On the other hand, farmed eel samples are clearly less contaminated than wild eel samples as presented below. Also, for other farmed fish (mostly n = 6 or 7) variability among various samples was observed. We assume that this relates to the levels in fish feed rather than sampling location or origin, but this study did

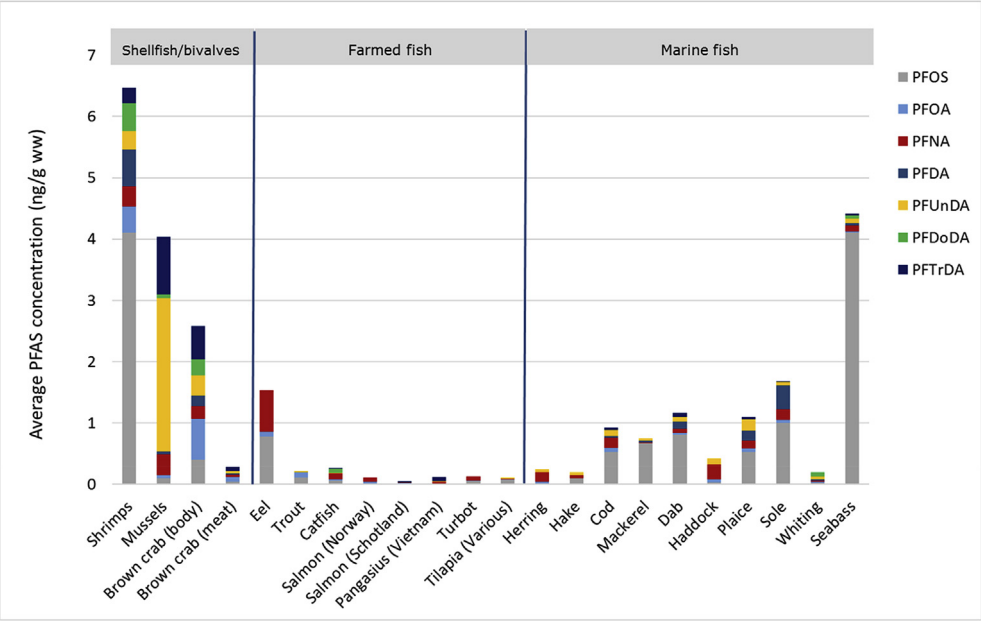

Fig. 2. Average PFAS levels (ng/g ww) in bivalves, crustaceans, marine and farmed fish. In case levels were <LOQ, the LOQ value was replaced by a 0 (lowerbound principle). Detailed information on individual samples, median and maximum values can be found in the supporting information.

not aim for fish feed investigation and therefore no conclusions can be drawn on this.

The PFAS pattern was established for a selection of marine species where a sufficient number of compounds was detected (i.e. low number of <LOQ values), allowing establishing such pattern (see Fig. 3). The patterns are limited to PFOS, PFOA, PFNA, PFDA and PFUnDA), except for shrimps, where also PFDoDA and PFTrDA were frequently detected. These compounds together accounted for >95% of the ∑PFAS concentrations for nearly all species. The pattern is dominated by PFOS in most fish (42–92%), followed by the longer chain perfluorocarboxylic acids. Seabass showed the most pronounced PFOS accumulation, and other PFASs contributed (on average) less than 5%. In mussels, PFUnDA dominates the profile. The PFOS dominance is also observed in other studies on fish (Van Leeuwen et al., 2009; Kumar et al., 2009; Labadie and Chevreuil, 2011; Zhao et al., 2011; Koponen et al., 2015). The error bars indicate that the profile can be variable among samples, which can have several reasons, such as sampling at different locations among the different years, as discussed above.

3.2. PFAS concentrations and patterns in eels

Eighty-six eel fillet samples from the Netherlands were analysed for 16 PFASs. This included eels two size ranges, 30–40 cm and >45 cm. PFOS (100%), PFDA (94%), PFUnDA (93%), PFTrDA (87%), PFTeDA (65%) and PFDoDA (61%) showed the highest detection frequency. In addition, PFHxA, PFOA, PFNA, PFBS, PFHxS, and PFDS were detected in one or more samples but at a frequency below 40%. The concentrations of each individual PFAS and the ΣPFASs for each eel sample are presented in Table S8. ΣPFAS concentrations ranged between 4.7 and 172 ng/g ww. PFOS was detected in all eel samples and concentrations ranged from 3.3 to 67 ng/g ww. Long-chain PFASs ($C \geq 8$) were the next most frequently detected compounds, while the short-chain PFASs were rarely found.

When eel was sampled at the same location for multiple years, data were grouped and averages were calculated. The results were grouped irrespective of the eel's size class (30–40 cm or >45 cm), as it turned out that this does not affect the PFAS levels in the fillets. This was confirmed by one-way ANOVA, where no statistically significant difference ($p > 0.05$) was observed. These results are in agreement with previous studies supporting that the size of freshwater fish, including eel, does not influence the bioaccumulation of PFASs (Hoff et al., 2005; Ye et al., 2008; Quinete et al., 2009; Giari et al., 2015; Couderc et al., 2015). This is due to a rapid uptake (and depletion) of PFASs in fish (Martin et al., 2003; Falk et al., 2015), showing that, presumably independent of fish size and age, PFASs levels in fish are rapidly equilibrated with their surrounding water.

The average of ΣPFAS concentrations in eels was found to fluctuate among the different sampling locations (6.2–133 ng/g ww). In Fig. 4 the average levels at all locations are presented for the most frequently detected compounds (i.e. PFOS, PFNA to PFTeDA). No specific trend as regards location or sampling year was observed. This corresponds to PFASs levels in surface waters of the Rhine and Meuse river basin (several locations) and lake IJssel (Andijk), where nearly no fluctuation in levels was observed (PFOS 4–6 ng/L; PFOA 2–4 ng/L; PFNA <1 ng/L; PFDA <1 ng/L and PFUnDA <1 ng/L, data from 2017) (RIWA, 2018a, b). In another study by Gebbink et al. (2017), river water measurements (upstream of a hotspot, at Merwede, Waal and Rhine) showed similar levels. The most contaminated location, both in terms of average concentration of PFOS or ΣPFASs (67 ng/g ww and 133 ng/g ww respectively), was the Ghent-Terneuzen Canal (location 25, Fig. 1). The elevated PFOS levels can probably be attributed to (former) activities of a fluorochemical plant located in Zwijndrecht near Antwerp (Hoff et al., 2003; Van de Vijver et al., 2003). The industrial discharges of the plant may have contributed to PFOS contamination of the Scheldt river, and downstream to the Western Scheldt, resulting in the high contamination of eel from the Ghent-Terneuzen Canal. In the sample from this location, the level of PFUnDA is also higher compared to other locations, but the origin of that elevated level is unclear as to the best of our knowledge this fluorochemical plant did not produce this PFAS.

The lowest ΣPFAS concentrations were measured in eel collected from marine environments, like the Maasvlakte (location 23, 6.2 ng/g ww) and at the seaside of the Haringvlietdam (location

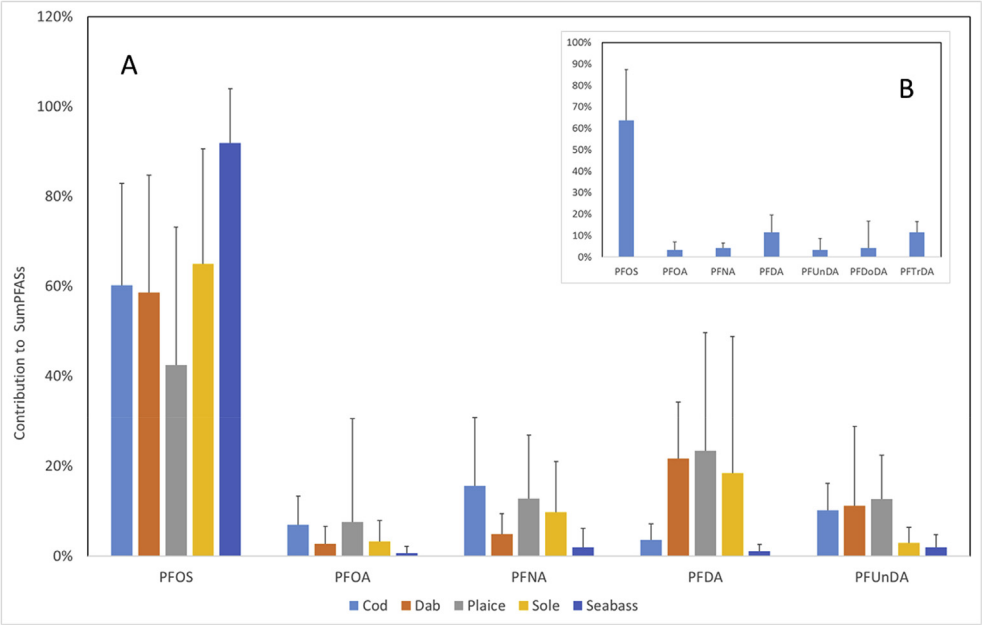

Fig. 3. Average relative contribution (mean and standard deviation) of major PFASs to the ∑PFAS concentrations in fish from the Netherlands (Figure A), based on average concentrations shown in Table S7. Figure B shows the PFAS pattern in shrimps.

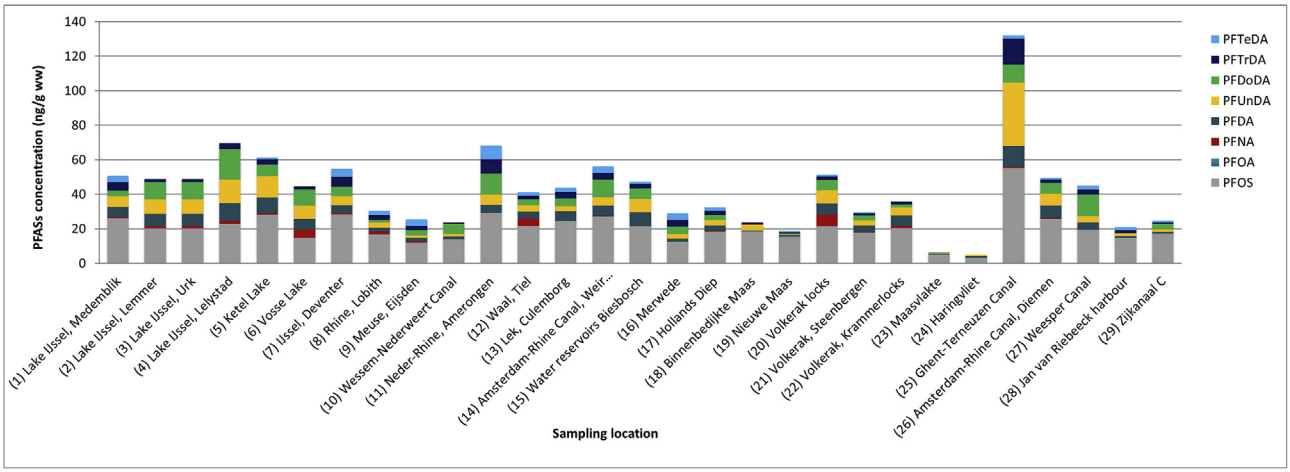

Fig. 4. Concentrations of PFASs in eel samples from various Dutch locations. When multiple samples were available per location, the average result is included. Results of individual samples are provided in Table S8. The location number cross-references to Fig. 1.

24, 12.9 ng/g ww), despite the fact that they were close to the mouth of highly polluted rivers. The river effluent is highly diluted due to tidal influences and sea currents, which explains the lower contamination level of the locally caught eel.

PFOS concentrations (3.3–67.2 ng/g ww) observed in the present study, were consistent with previous studies on eels collected from the Netherlands and other European countries. According to Kwadijk et al. (2010), PFOS concentrations in eel from Dutch rivers ranged from 7 to 58 ng/g ww in retrospectively analysed eel muscle samples from 1978 to 2008 at several locations similar to those in our study. Comparable PFOS concentrations were also reported in eels collected from the river Mohne in Germany (37–83 ng/g ww) (Hölzer et al., 2011) and from the Loire estuary in France (17.9–39 ng/g ww) (Couderc et al., 2015). In contrast, lower PFOS levels were found in eel muscle tissues from Italy (<0.4–2.47 ng/g ww) (Giari et al., 2015) and Spain (highest PFOS concentration: 21.6 ng/g ww) (Pignotti et al., 2017).

In the present study PFOA levels were in general below the LOQ (0.3 ng/g ww) and detected in only five out of eighty-six samples and at very low concentrations (range: <LOQ – 0.9 ng/g ww). PFOA in eel muscle tissue has often been reported to be low, with maximum concentrations equal to 2.3 (Hölzer et al., 2011) and 0.3 ng/g ww (Schuetze et al., 2010). Giari et al. (2015) detected PFOA in eel muscle tissue samples from Italy at levels from <0.4 to 24.7 ng/g ww (detection frequency 17%). Average PFOA concentrations in blood, kidney, liver, and gonad were 13.90, 12.85, 7.27 and 8.99 ng/g ww, respectively.

Considering all PFASs, the general pattern of contamination observed in this study was also similar with previous studies, presenting long–chain PFASs (>C8) as the most frequently detected compounds after PFOS and short-chain PFASs being rarely detected (Couderc et al., 2015; Kwadijk et al., 2010). The PFAS levels in the

Loire study (Couderc et al., 2015) were somewhat lower than those in the current study. According to Conder et al. (2008), short-chain PFASs possess a very low or non–existent potential of bioaccumulation, and this may explain their low detection frequency. Although short-chain PFASs are present in Dutch river water, as recently demonstrated by, among others, Gebbink et al. (2017), they hardly accumulate. This was also shown by Kwadijk et al. (2010) who determined water-eel bioaccumulation factors, finding that the shorter the fluorinated alkyl chain, the lower the bioaccumulation potential. Gebbink et al. (2017) also detected several emerging PFASs in water of Dutch rivers, and further studies are needed to determine to what degree those emerging PFASs can accumulate in eel or other fish.

Figure S1 shows the contribution of the individual PFASs to the sum concentration (limited to the most predominant PFASs). As mentioned before, PFOS is the most predominant compound with a 33–82% contribution to the sum. The highest contribution was observed at those locations where PFAS levels were low (e.g. Haringvliet and Maasvlakte). Correlations of the five predominantly detected PFASs (PFDA, PFUnDA, PFDoDA, PFTrDA, PFTeDA and PFOS) were investigated using all individual samples (Fig. S2). Significant positive correlations were generally found between concentrations of all five PFASs. However, PFTeDA was not significantly correlated to PFDA, PFUnDA or PFOS. The positive correlations between the individual PFASs indicate that the eels have a similar exposure to these PFASs regardless of the location. However, elevated PFUnDA and PFOS levels were observed at the Ghent-Terneuzen location, resulting in a deviating profile, as discussed earlier.

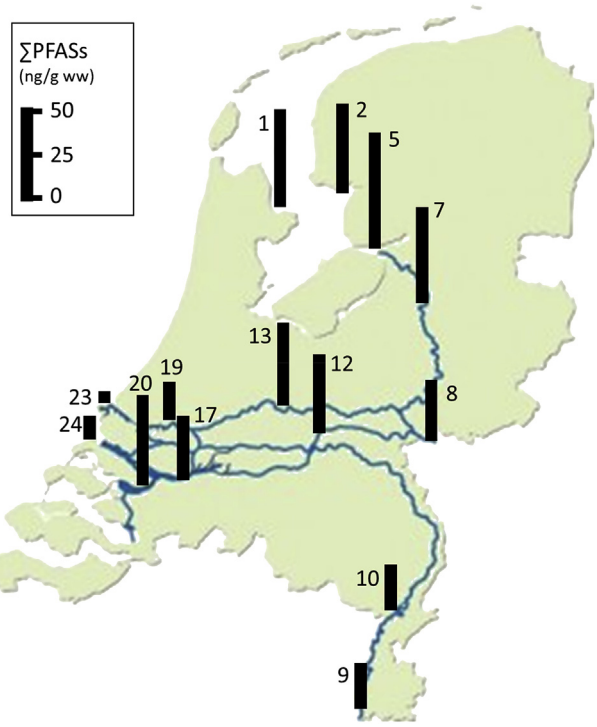

Fig. 5. Sum PFAS concentrations in eel for several samples in the river basin, the main rivers and lake IJssel in the Netherlands. The numbers refer to the sampling locations (see also Fig. 1).

3.3. Spatial and temporal trends of PFASs in eel

The ∑PFAS concentrations for several locations are plotted in Fig. 5. This is based on the data presented in Fig. 3, but presented in a geographical way, which allows to assess the concentrations in the river basin of the main rivers. Along the Meuse river, entering the Netherlands in the south (loc 9, 25.4 ng/g ww), levels remain similar (loc 10) and rise slightly to 33.2 ng/g ww at Hollands-Diep (loc 17) and enter the North Sea at much lower concentration (12.9 ng/g ww, loc 24). Along the Rhine trajectory, concentrations change from 31.2 (Dutch-German border, loc 8) to 42.4 (loc 12) and finally 6.2 ng/g ww (loc 23) when entering the sea at the 2e Maasvlakte. Along the river IJssel, lake Ketel and lake IJssel in Fig. 5, ∑PFAS concentrations are very stable from 51.1 (loc7) to 50.9 (loc1). This lack of spatial trend on this trajectory (IJssel, lake Ketel and lake IJssel) is in contrast to the sum-TEQ levels for PCDD/Fs and dl-PCBs in these samples (presented in Fig. S3 for PFOS), being higher in the river Rhine (Lobith), IJssel and in the Ketelmeer, than in eel from the Lake IJssel (Van Leeuwen et al., 2013). PCDD/Fs and PCBs are less mobile compared to PFASs, and associated to suspended particulate matter, which starts to precipitate after the Ketelmeer due to decreased flow rate (increased residence time). Therefore, the PCDD/F and especially PCB levels in sediment gradually drop after that border, and consequently also the levels in eel. On the other hand, the PFASs investigated in this study are water soluble compounds and consequently disperse throughout the Lake IJssel. As a result, PFAS concentrations in eel from river Rhine, river IJssel, and the lakes Ketelmeer and Lake IJssel are similar.

In this study, we also evaluated if a time trend could be observed in PFAS concentrations in eel for some of the sampling locations. PFOS concentrations from the eel samples of the locations Hollands Diep (location 17, 2010–2016), Lake IJssel (location 1, 2011–2016) and Rijn (location 8, 2010–2016) were evaluated (Fig. S4). The trend-lines at the three locations suggest no clear time-trend. Kwadijk et al. retrospectively analysed PFOS in eels collected between 1978 and 2008 from Hollands Diep, Rijn and Haringvliet (Kwadijk et al., 2010). The PFOS levels in their samples were slightly higher than the current ones from the same locations during the last six years (2010–2016) (current study). The average PFOS concentration in samples from Hollands Diep and Rijn during the 6 years in the current study was 18.3 and 16.8 ng/g ww respectively. In the 30-year time-trend presented in the Kwadijk et al. study, PFOS concentrations showed an increase until the mid-1990s, followed by a decline in the years after until levels similar as those observed in the late 1970s. More specifically, PFOS levels during 2006 and 2008 were approximately 20–30 ng/g ww in eel from both Hollands Diep and Rijn. The current results may indicate a modest decline of PFOS levels over the last six years compared to Kwadijk et al. but no strong conclusions can be drawn on that.

3.4. Comparison of PFOS levels with the EU EQS

There are no maximum levels for PFASs in food. However, in 2013, the European Commission set Environmental Quality Standards (EQSs) in the Water Framework Directive (WFD) for certain contaminants, including PFOS (EQS for PFOS = 9.1 μg/kg ww in biota) (European Commission, 2013; European Union, 2014). These standards were derived to monitor the surface water quality but also include human safety. The EQS for PFOS is based on the previous EFSA TDI of 150 ng/kg bw/day, assuming a body weight of 70 kg, a fish consumption of 115 g/day and an acceptable contribution to the PFOS intake via fish of 10%. In the present study, 93% of the eel samples exceeded this value, while the highest PFOS concentration, detected in eel caught from Kanaal Ghent-Terneuzen (location 25), was approximately 7 times the EU EQS. Also the average PFOS concentration in eels from all sampling locations was above 9.1 μg/kg ww. Only at the Maasvlakte (location 23) and Haringvliet (location 24) PFOS concentrations were below the EU EQS. These findings are in agreement with a previous study

conducted in eels from France (Couderc et al., 2015), where 75% of the analysed eels exceeded the EQS for PFOS, with the highest PFOS concentration being approximately 14 times higher. Among the bivalves (mussels, oysters), crustaceans (shrimps) and marine fish (e.g. flounder), caught in coastal waters that are subject to the EQS, none of the average PFOS results exceeded the EQS. Only a single shrimp sample from Eems-Dollard had a PFOS level of 25 ng/g, thereby exceeding the EQS. It is unclear whether the EQS for PFOS will be changed based on the new much lower TWIs established by EFSA. It should, however, be stressed that European maximum levels for contaminants in food are normally based on the principal "strict but feasible", and may as such differ from quality standards for surface water.

4. Conclusions

The present study shows that, among all investigated species, average ΣPFAS levels were highest in eel from Dutch rivers and lakes (up to 48.8ng/g ww), followed by shrimps collected at the Dutch coast (6.7 ng/g ww) and by seabass (4.4 ng/g ww). The farmed fish (e.g. trout, catfish, turbot, salmon, tilapia, pangasius) were among the lowest contaminated samples in this study. Geographically, PFASs levels in marine fish from the northern North Sea (e.g. haddock, whiting, herring) were lower than in the central and southern North Sea (e.g. cod and flatfish). Concerning eel, levels were generally in the same range throughout the country. The contamination pattern is mostly dominated by PFOS, but also long–chain PFCAs ($C \geq 8$) were detected frequently. Short-chain PFASs were rarely found.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2019.05.200.

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