Are Chinese mitten crabs (**Eriocheir sinensis**) suitable as biomonitor or bioindicator of per- and polyfluoroalkyl substances (PFAS) pollution?

Are Chinese mitten crabs *(Eriocheir sinensis)* suitable as biomonitor or bioindicator of per- and polyfluoroalkyl substances (PFAS) pollution?

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Abstract

Per- and polyfluoroalkyl substances (PFAS) are ubiquitous in the environment. In Flanders, the bioaccumulation in aquatic organisms is currently being monitored using European perch and European eel. Since both are native species, there is an ethical need to search for other suitable biomonitors. This study aims to investigate whether the invasive Chinese mitten crab could be used in biomonitoring programs by assessing PFAS accumulation in hepatopancreas, muscle tissue, and carapace. Furthermore, we correlated accumulated concentrations to those in the local abiotic environment. Concentrations in the crabs (highest average ∑PFAS concentration of 688 ± 505 ng/g ww) were often higher than those in crab species from other regions across the globe, confirming that Flanders is highly polluted with PFAS. Concentrations in the crabs did not reflect those in the abiotic environment. This implies that biomonitoring is necessary to investigate the impact of PFAS pollution on organisms in aquatic ecosystems, as important data is missing when only the abiotic environment is monitored. The accumulation profiles differed between the invasive crab and the native European perch and European eel, potentially due to a different ecology and trophic position. Since all three species provide complementary information on the PFAS pollution, a multi-species approach in biomonitoring is recommended. Overall, our results show that the crabs can be used as biomonitor, but more information is necessary to confirm their suitability as bioindicator.

Keywords: Aquatic environment; invasive species; bioaccumulation; PFAS; spatial distribution

Statement of environmental implication

Per- and polyfluoroalkyl substances (PFAS) are persistent and bioaccumulative chemicals that are ubiquitously present in the environment and in biota. Exposure to PFAS may lead to toxic effects and some are already included as hazardous substance under the EU Water Framework Directive. In Flanders, biomonitoring programs use indigenous and often endangered fish species to investigate PFAS bioaccumulation. Hence, from an ethical perspective, there is a need to identify alternative biomonitors. Our study shows that the invasive Chinese mitten crab, that is being caught anyway to control populations and reduce their environmental impact, can be used complementary to the fish in biomonitoring programs. However, our data is insufficient to draw conclusions on their suitability as bioindicator.

1. Introduction

Per- and polyfluoroalkyl substances (PFAS) are a group of man-made fluorinated organic chemicals that have been used in a large variety of both industrial and commercial applications, such as waterproofing materials, non-stick cookware, and firefighting foams (Buck et al., 2011). Their production and use has led to a global distribution of PFAS in the environment (Giesy and Kannan, 2001; Herzke et al., 2012; Groffen et al., 2018; Padilha et al., 2022), where they are shown to be persistent, bioaccumulative, and toxic (Cousins et al., 2020; Ankley et al., 2021; Fenton et al., 2021; Dickman and Aga, 2022). As a result, PFAS are considered to be chemicals of global scientific and public concern (Ji et al., 2020).

There are many sources from which PFAS can spread through the environment, including manufacturing plants, wastewater treatment plants, and landfills (Ji et al., 2020). Once in the ecosystem, PFAS can spread further through streams and rivers (Sinclair et al., 2020), ocean currents (Miranda et al., 2021) and via the atmosphere (Sinclair et al., 2020; Miranda et al., 2021). Many PFAS bioaccumulate and biomagnify in food webs (Domingo and Nadal, 2019), causing PFAS to be detected in every environmental matrix (Saleeby et al., 2021).

Within the aquatic environment, short-chained PFAS are more likely to partition to the water due to their higher water solubility (Ji et al., 2020), whereas long-chained PFAS tend to sorb to sediment (Lenka et al., 2021) through both hydrophobic and electrostatic interactions with various sediment characteristics (Shahsavari et al., 2020). However, the exact role of this sorption to sediment, and the subsequent bioavailability, is still unclear (Li et al., 2018).

Recently, PFAS pollution in Belgium has attracted widespread media and public attention after increased environmental and human serum PFAS levels were detected around a hotspot close to the city of Antwerp (Government of Flanders, 2021). Previous studies in the terrestrial environment around this hotspot reported some of the highest PFAS concentrations ever detected in the terrestrial environment (Groffen et al., 2019a, 2019b; Lopez-Antia et al., 2019). However, there is still a general

lack of understanding of PFAS distribution in the Belgian aquatic environment. A study by Teunen et al. (2021) measured various PFAS in the European perch (*Perca fluviatilis*), the European eel (*Anguilla anguilla*), quagga mussels (*Dreissena bugensis*), and Asiatic clams (*Corbicula fluminea*) at 44 locations in Flanders (Belgium). They reported that perfluorooctane sulfonate (PFOS) concentrations in the European perch and European eel exceeded the European Biota Quality Standards (EQS_{biota}) at approximately half of the sampled locations.

The measurement of the body burden of chemicals in organisms can be determined through biomonitoring. This approach has several advantages over environmental monitoring, as it provides useful information on the bioavailability, mobility, and fate of contaminants in the environment. Thus, organisms that are exposed to pollutants in their natural habitat provide a time-integrated measure of environmental concentrations, taking into account spatiotemporal fluctuations in the environment. The terms biomonitor and bioindicator are interchangeable among the general public, but scientists differentiate between both by specifying that bioindicators qualitatively assess biotic responses to environmental stress, whereas biomonitors quantitatively determine a response (Holt and Miller, 2010). Good bioindicators often share specific characteristics (Holt and Miller, 2010): 1) they should be able to survive in contaminated areas in which they provide a measurable response (e.g., bioaccumulation of pollutants) which reflects the whole population/community/ecosystem response and is proportional to the degree of contamination or degradation; 2) they should be commonly present with an adequate population density and their populations should be stable despite environmental variability; 3) their ecology and life history should be well understood; and 4) the species should be easy and cheap to survey and ideally the species is already caught for other purposes. Currently, in Flanders the native European perch and European eel are used as biomonitoring species for PFAS in the aquatic environment (Teunen et al., 2020). However, from an ethical perspective, and considering that European eel is currently listed as critically endangered by the IUCN (Jacoby and Gollock, 2014), an alternative biomonitoring species would be preferred.

An example of such alternative biomonitoring species could be the Chinese mitten crab (*Eriocheir sinensis* H. Milne Edwards 1853), an invasive species in Europe (European Union, 2016). This crab is a well-established species that has been reported in Belgium since 1933 (Ewers et al., 2023). They are opportunistic omnivores (Dittel and Epifanio, 2009) that are known to cause considerable ecological and economic damages (Dittel and Epifanio, 2009; Czerniejewski et al., 2010; Schoelynck et al., 2019). They are tolerant to a wide range of environmental conditions (Veilleux and de Lafontaine, 2007), although preferences for salinity and temperature ranges may differ depending on their developmental stages (Wang et al., 2019). Juveniles are born in estuaries and migrate to freshwater habitats in spring. After being resident for 2 to 5 years, adults return to the sea in autumn to reproduce. Chinese mitten crabs are expected to be suitable biomonitoring species, for multiple reasons including: 1) they are present along the entire river continuum and in various habitats, such as estuaries, big rivers, small rivers, lakes, ponds, etc.; 2) they tolerate high pollution levels (Veilleux and de Lafontaine, 2007; Jabbar et al., 2019); and 3) catching Chinese mitten crabs is required to control the populations of this invasive species, as obligated by the European Union (European Union, 2014). The species has been previously used in studies on persistent organic pollutants (Van Ael et al., 2012; Hoogenboom et al., 2015; Brust et al., 2018; Leenders et al., 2021) and metals (Van Ael et al., 2017). However, to the best of our knowledge, studies using Chinese mitten crabs to monitor PFAS pollution are scarce (see Brust et al. (2018) and Leenders et al. (2021) for the only available literature data).

In the present study, we investigated the PFAS concentrations in hepatopancreas, muscle tissue, and carapace of Chinese mitten crabs in Flanders, and correlated accumulated concentrations in the crabs to concentrations present in the abiotic environment.

2. Materials and methods

2.1 Sampling locations and sample collection

Twenty-six sampling locations across Flanders were chosen, Chinese mitten crabs could be caught at 23 locations (Figure 1, Table A1). In total, 96 individuals were caught with two fyke nets, that were placed for 24h per site, in September 2020 and from May to November 2021. These periods are outside

the migration periods, and so crabs are considered resident. The crabs at Grobbendonk (site 9, Figure 1) and Lippenbroek (site 21, Figure 1) were caught using a customized crab trap (Schoelynck et al., 2021). At the same sampling sites three replicates of water and sediment samples were collected between August and December 2021. Water was sampled using a pre-cleaned polyethylene bucket and subsamples of 50 mL were stored in polypropylene (PP) tubes. For sediment, we either used a Van Veen grab sampler, a plastic tube, or a trowel. Three sediment grabs were pooled in one bucket, from which three replicate samples were taken and stored in 50 mL PP tubes. The pH and electrical conductivity (EC) of the water were measured using a pH/conductometer (Metrohm 914 pH/DO/Conductometer and WTW MultiLine 3430 IDS). The sampling locations covered the various habitats of Chinese mitten crabs and included rivers, canals, creeks in a tidal marsh, and a pond. A detailed overview of the sampling locations can be found in Table A1. All samples and crabs were stored at -20 °C prior to further analysis.

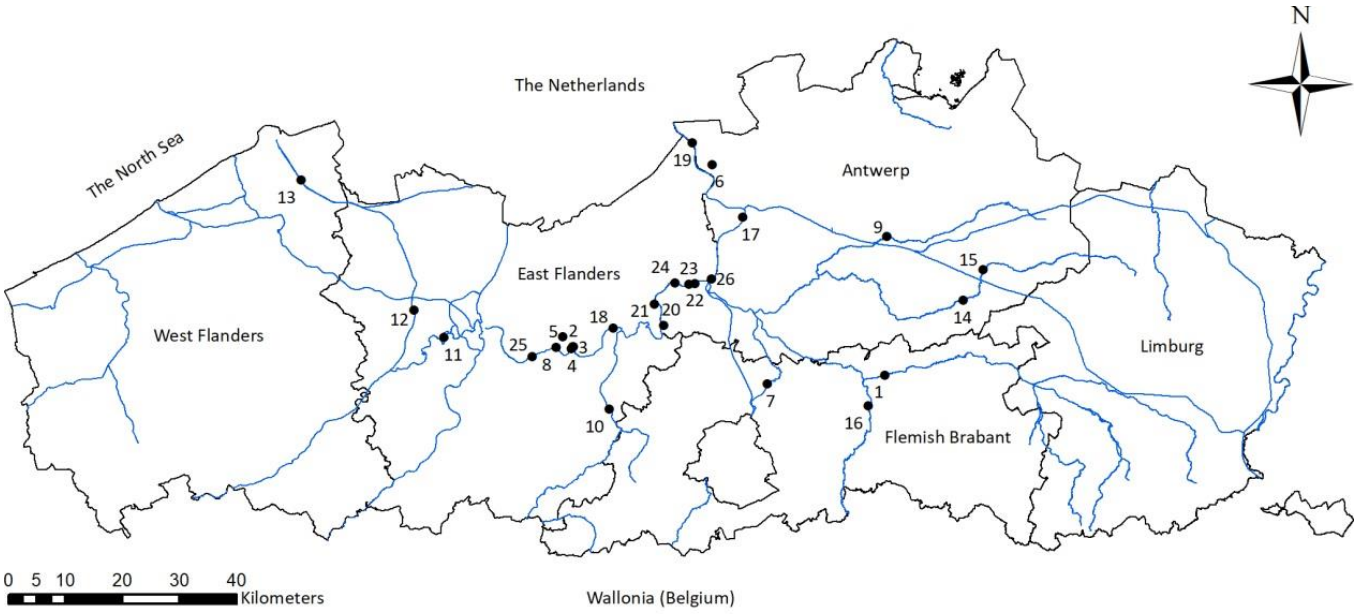


Figure 1. Overview of the 26 sampling locations. Details on the individual locations are provided in Table A1. Made with ArcMap 10.7.1 (projection: Lambert 72).

2.2 Extraction and chemical analyses

2.2.1 Sample pretreatment

The crabs were weighed (± 0.01 g; Sartorius TE1502S) and their carapace width was determined, as a measure of crab size, by measuring the width just behind the tips of the fourth bilateral spines using a caliper (± 0.1 mm). All crabs were bigger than 2.5 cm (Table A12), so considered resident adults (Panning, 1938; Schoelynck et al., 2021). The sex was determined by inspecting their abdomen shape, as described by Veilleux and de Lafontaine (2007). The crabs were dissected to collect the hepatopancreas, white muscle tissue from the legs, and carapace for PFAS analyses. The carapace was rinsed with Milli-Q water (MQ; pore size = 0.22 μm, Merck Millipore, Darmstadt, Germany) to remove dirt. The hepatopancreas samples were homogenized by mixing thoroughly with a stainless steel rod, the muscle samples by using a TissueLyser LT (Qiagen, Germany) with stainless steel beads, and the carapace by cutting into small pieces (± 1-2 mm) with stainless steel scissors. The soft tissues were stored in 2 mL PP tubes, whereas carapaces were stored in 50 mL PP tubes. The sediment samples were thoroughly mixed with a stainless steel spatula, and a subsample of the sediment was oven-dried at 60 °C prior to PFAS extraction. Water samples were extracted without any pretreatment.

2.2.2 Chemical extraction

After weighing the samples (± 0.2 g of crab tissue, 0.2-0.3 g of dried sediment and 10 mL of water), the samples were spiked with 10 ng of an isotopically mass-labelled internal standard mixture (ISTD, MPFAC-MXA, Wellington Laboratories, Guelph, Canada). Hereafter, 10 mL of acetonitrile (ACN) was added to the hepatopancreas, muscle tissue, and sediment samples, whereas 10 mL of methanol (MeOH) was added to the carapace. No solvents were added to the water samples. After vortex-mixing, all samples were sonicated (3 x 10 min with vortex-mixing in between) and left overnight on a shaking plate (135 rpm) at room temperature. All samples were then centrifuged (4 °C, 10 min, 2400 rpm, Eppendorf 5804R with A-4-44 rotor) and the extracts were treated differently depending on the matrix.

The crab samples were further extracted following a protocol described by Powley et al. (2005). The supernatants were dried to approximately 0.5 mL in a rotational vacuum concentrator (30 °C, Martin

Christ RVC 2-25 and Eppendorf Concentrator 5301), after which the concentrated extracts were added to a 1.5 mL PP tube containing 50 mg of graphitized carbon powder (Supelclean ENVI-Carb) and 50 μL of glacial acetic acid. The tubes were rinsed twice with 250 μL of ACN (or MeOH in case of the carapace samples), which was also transferred to the 1.5 mL PP tube. After vortex-mixing for at least 1 min, the samples were centrifuged (4 °C, 10000 rpm, 10 min, Eppendorf Centrifuge 5415R with F-45-24-11 rotor) and the supernatant was dried completely under vacuum.

The water and sediment samples were analyzed according to a protocol described by Groffen et al. (2019c). The supernatants of the extracts were loaded onto Chromabond HR-XAW SPE cartridges (Macherey-Nagel, Germany) that were pre-conditioned and equilibrated using 5 mL of ACN and 5 mL of MQ, respectively. Hereafter, the cartridges were washed with 5 mL of a 25 mM ammonium acetate solution (dissolved in MQ), followed by 2 mL of ACN. Finally, the PFAS were eluted from the cartridges using 2 x 1 mL of a 2% ammonium hydroxide solution (dissolved in ACN).

The dried supernatants of all samples were finally reconstituted with 200 μL of a 2% ammonium hydroxide solution (dissolved in ACN), vortex-mixed, and filtrated through a 13 mm Ion Chromatography Acrodisc Syringe Filter with a 0.2 μm Supor polyethersulfone (PES) membrane into a PP auto-injector vial.

2.2.3 UPLC-TQD analysis

The samples were analyzed using ultra-performance liquid chromatography-tandem mass spectrometry (UPLC-MS/MS, ACQUITY TQD, Waters, Milford, MA, USA), using negative electrospray ionization (ES(-)). An ACQUITY BEH C18 column (2.1 x 50 mm; 1.7 μm, Waters, Milford, MA, USA) was used to separate the target analytes. To retain any PFAS contamination originating from the system, an ACQUITY BEH C18 pre-column (2.1 x 30 mm; 1.7 μm, Waters, Milford, MA, USA) was inserted between the solvent mixer and the injector. As mobile phase solvents a 0.1% formic acid in water solution and a 0.1% formic acid in ACN solution were used. The injection volume was set at 6 μL, with a flow rate of 450 μL/min. The solvent gradient started at 65% of 0.1% formic acid in water, went to

0% in 3.4 min and back to 65% at 4.7 min. Multiple reaction monitoring (MRM) of two diagnostic transitions per analyte or ISTD was used to identify and quantify the 29 targeted PFAS. The diagnostic transitions were validated by Groffen et al. (2019c, 2021) and an overview of the targeted PFAS and instrumental settings is provided in Table A2.

2.2.4 Quality control

One procedural blank, consisting of 10 mL of ACN for biota (except carapace, then 10 mL of MeOH was used) and sediment, or 10 mL of MQ water for water samples, was included per batch of 15 samples to detect any contamination that may have occurred during the extraction and analyses. Any contamination present in these blanks were subtracted from the levels in the samples within the same batch. In addition, ACN was injected regularly as instrumental blank to prevent cross-over contamination between injections. The limit of quantification (LOQ) and limit of detection (LOD) of each analyte was determined in matrix as the concentration corresponding to a signal-to-noise ratio of 10 or 3, respectively and are shown in Table A3.

2.3 Determination of sediment physicochemical characteristics

The total organic carbon (TOC) content of the sediment was determined using a protocol based on the loss on ignition (LOI) method described by Heiri et al. (2001). Briefly, porcelain crucibles were oven-dried at 105 °C for 2 h, cooled down in a desiccator and weighted. The crucibles were then filled with oven-dried (70 °C, 2 days) sediments, and further oven-dried at 105 °C for 2 h, cooled down in a desiccator, and weighted. In the last step, the sediment samples were incinerated in a muffle furnace (550 °C, 5 h). After cooling down samples were set in the desiccator again, the weight loss was determined and the TOC was calculated using Formula 1.

$$TOC\ (\%) = ((DW_{105} - DW_{550})/DW_{105}*100)/1.742 \quad (1)$$

With DW_{105} and DW_{550} being the dry weight (g) after drying at 105 °C and 550 °C, and 1.724 being the van Bemmelen factor, assuming that 58% of the organic matter consists of carbon (Nelson and Sommers, 1996). The dry weights are corrected for the weights of the crucibles.

A Malvern Mastersizer 2000 and Hydro 2000G were used to determine the clay content (particles with a size < 2 μm) of the sediment. Approximately 2 g of sediment was incubated overnight with 10 mL of 30% hydrochloric acid and 15 mL of 33% hydrogen peroxide to break down sediment aggregates and remove the organic fraction from the samples. Hereafter, 25 mL of hydrogen peroxide was added and the samples were boiled to speed up the removal of the organic fraction. Finally, the samples were sieved over a 1 mm test sieve prior to analyses.

To determine the cation exchange capacity (CEC), the sediment samples were incubated at 40 °C for 2 days. To 2.5 g of sediment, 25 mL of 1 M ammonium acetate (adjusted with 25% ammonium hydroxide to a pH of 7) was added. Three procedural blanks, containing 25 mL of 1 M ammonium acetate, were used as quality control. After three-dimensional shaking for 1 h, the pH of the extract was measured (WTW MultiLine 3430 IDS), and the extracts were filtered through a syringe filter with 0.45 μm mixed cellulose ester (MCE) membrane. Two titration curves were made by adding a total volume of 10 mL of 0.1 N acetic acid to 25 mL of 1 M ammonium acetate in steps of 0.1 mL, measuring the pH after each addition of acetic acid. The exchangeable acidity of the samples was determined based on these titration curves. The concentrations of aluminum, calcium, iron, magnesium, manganese, potassium, and sodium were analyzed using inductive coupled plasma-optical emission spectrometry (ICP-OES, iCAP 6300 Duo, Thermo Scientific) in order to determine the exchangeable bases and acidic cations. The CEC was calculated as the sum of the exchangeable acidity, exchangeable bases and acidic cations.

2.4 Statistical analyses

Statistical analyses were performed using Rstudio (version 2022.02.2, R version 4.1.2). The level of significance was set at $p \leq 0.05$. PFAS concentrations that were < LOQ were assigned a replacement concentration following a maximum likelihood estimation (MLE) method (Villanueva, 2005; de Solla et

al., 2012) prior to statistical analyses. Since this method assumes a normal distribution of the data, we only included PFAS with a detection frequency of ≥ 30% in the statistical analyses. The MLE method is not appropriate for datasets with insufficient measurements above the LOQ, because of the lack of evidence to know whether the assumed normal distribution fits the data well (Helsel, 2006). Furthermore, with insufficient detected values, those values that are above the LOQ would have likely been identified as outliers in the statistical analyses (Helsel, 2006). In the calculation of ∑PFAS concentrations, values < LOQ were replaced by zero. Compounds that were not detected in any of the samples (i.e. PFHpA, PFHxS, PFHpS, PFDS, 4:2 FTS, HFPO-DA, 9Cl-PF3ONS, 11Cl-PF3OudS, PF4OpeA, PF5OhxA, 3,6-OPFHpA, and PFEESA) were excluded from the results and further analyses. The validity of the models' assumptions were examined using Shapiro-Wilk tests. If the data did not follow these assumptions, non-parametric tests were used. Outliers were identified using the Grubbs test and were, if present, removed prior to the analyses. Outliers were only identified in Spearman/Pearson correlations between the PFAS concentrations in the abiotic environment and those in the crab tissues, where 4 datapoints (17.4%) were considered outliers. Principal component analysis (PCA) was conducted to find underlying correlations between the different PFAS and abiotic characteristics as well as among the abiotic characteristics (Figure 7). The first principal component (PC) explained 28% of the variance, whereas PC2 and PC3 explained 24% and 22%, respectively. Factor loadings of these three principal components are displayed in Table A14. Furthermore, multiple regression analyses were performed to relate the environmental concentrations to those accumulated in the crabs, taking into account sediment and water characteristics (both as individual parameters, as well as two-way interactions between these characteristics). Model selection was based on the Akaike information criterion (AIC). Correlation tests were used to assess the influence of water and sediment characteristics on PFAS concentrations in water and sediment, respectively. Differences in PFAS concentrations between crab tissue types were investigated using repeated measures ANOVA with pairwise paired t-tests as post hoc analysis (or paired t-tests in case only two tissues were investigated). Correlation tests were used to correlate PFAS concentrations among the three types of crab tissue.

3. Results and discussion

3.1 PFAS concentrations in water

In water, only PFBA, PFPeA, PFHxA, PFOA, PFDA, PFDoDA, and PFOS were detected at concentrations above the LOQ, with PFHxA being the most frequently detected (at 25 out of 26 sites), followed by PFOA (17 sites), and PFDoDA (14 sites). The highest mean ∑PFAS concentration in water was measured in the samples from a pond at Lillo (388 ng/L; site 6 on Figure 1), a location in the Port of Antwerp (Figure 2). At this site PFBA contributed over 90% to the ∑PFAS concentration (Figure 3). Details on mean ∑PFAS concentrations as well as concentrations of individual PFAS are provided in Table A4.

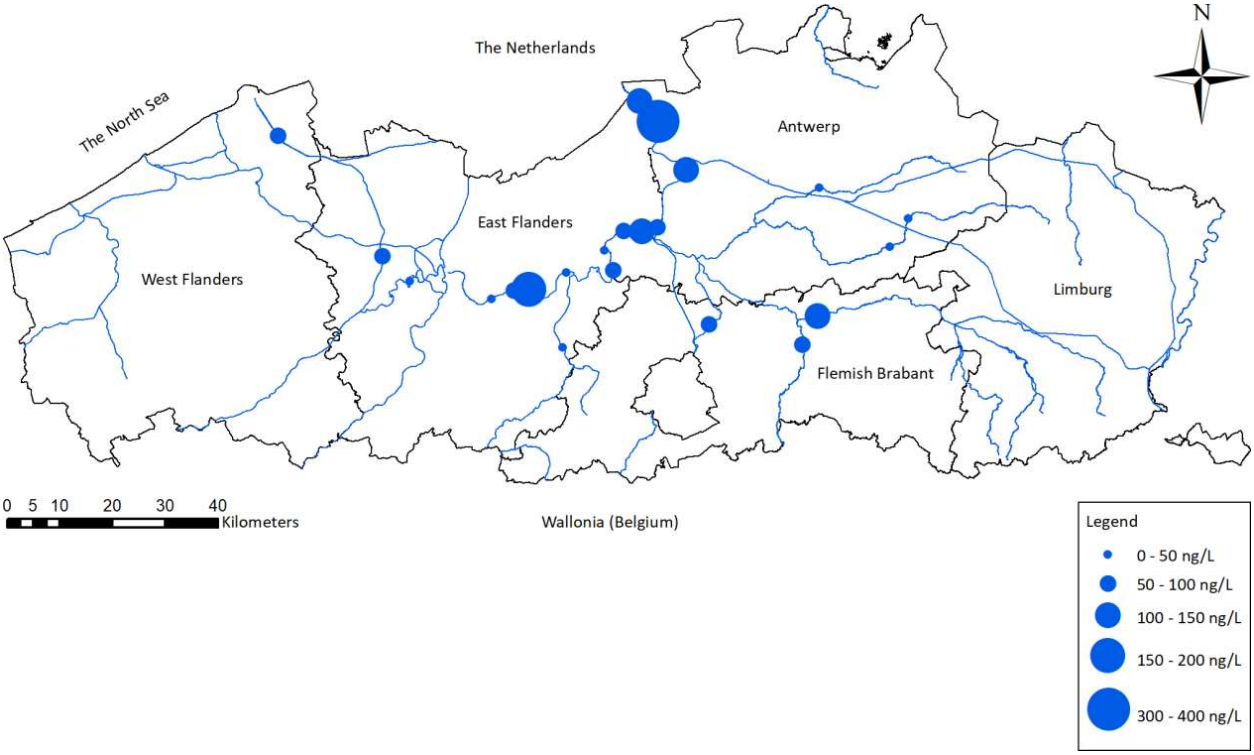


Figure 2. Mean ∑PFAS concentrations in water (ng/L). The main waterways are represented by blue lines. Some sampling sites overlap due to the size of the circles. A detailed overview of the ∑PFAS concentrations and concentrations of individual PFAS per site is provided in Table A4. Made with ArcMap 10.7.1 (projection: Lambert 72).

In samples where PFDoDA was detected, these concentrations contributed most to the ∑PFAS at these sites. This was unexpected, since PFDoDA is a long-chained PFAS. Long-chained PFAS are hydrophobic and are therefore expected to adsorb to solid matrices such as sediment (Li et al., 2018). The high PFAS concentrations detected in the northwestern part of the Antwerp province could be linked to

wastewater discharge in the Scheldt coming from multiple companies in the Antwerp harbor. A second site with prominent PFAS concentrations is in the branch from the Scheldt River in East Flanders province. Possible important sources for this site could include fire stations and multiple firefighting training sites as well as pollution coming from the industries in the Port of Ghent.

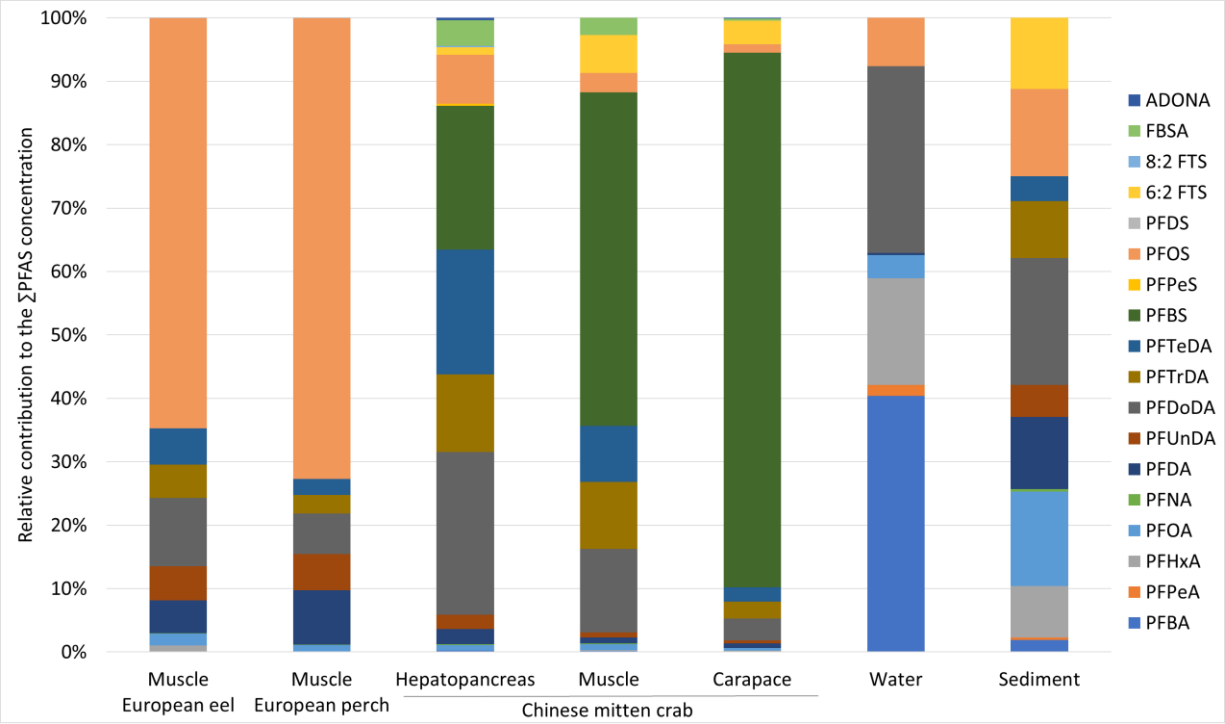


Figure 3. Relative contribution of individual PFAS to the 100% PFAS profile. Compounds that were <LOQ in all samples were omitted from the Figure. Data from Teunen et al. (2020) was used to create the PFAS profiles of European eel and European perch.

The most frequently occurring PFAS in surface water across the globe are PFHpA, PFOA, PFNA, PFBS and PFOS, although PFBA, PFPeA and PFNA have also been detected sporadically (Podder et al., 2021). In Europe, PFOA concentrations typically dominate the PFAS profiles in water (Podder et al., 2021), although an increase in PFBS concentrations has been observed after 2009, likely due to the replacement of long-chained PFAS by short-chained alternatives. The patterns observed in the present study deviate from the profiles typically occurring in European waters. This could be due to differences in partitioning between water and sediment and different sources and requires further examination. Concentrations of PFOS and PFOA in European surface waters have remained in the 10–100 ng/L range for over 20 years (Podder et al., 2021). The PFOA concentrations observed in the present study were

often lower than 10 ng/L, whereas the PFOS concentrations, when detected, fit within this range (Table A4).

The pH and EC of the water (Table A5) were not significantly correlated to the concentrations of PFHxA, PFOA, PFDoDA, and PFOS, although a positive correlation was found between the EC and ∑PFAS ($p < 0.001$, $r = 0.680$; Table A15 shows p and r values of the correlation tests). A high EC value indicates presence of more electrolytes in the water, including cations. The anionic head of PFAS can bind with cations (Wu et al., 2020), possibly explaining this positive correlation between EC and PFAS concentrations. Correlations between pH, EC, and concentrations of other PFAS could not be investigated due to too low detection frequencies of the other PFAS compounds. The absence of correlations with pH are possibly due to the range of pH values observed in the present study (6.71–7.84), which is unlikely to cause major changes in net charge of the PFAS or sediment particles. Changes in pH are known to affect partitioning of PFAS to solid matrices, as pH changes affect the surface charge of organic carbon and clay minerals (Nguyen et al., 2020). At low pH values, the organic carbon is more protonated, resulting in enhanced adsorption of particularly long-chain PFAS by both electrostatic and hydrophobic interactions (Higgins and Luthy, 2006). Short-chain PFAS are less sensitive to pH changes due to their lower Kd values, and prefer to be partitioned in the aqueous phase at lower pH values (Nguyen et al., 2020).

3.2 PFAS concentrations in sediment

In sediment, we detected PFBA, PFPeA, PFHxA, PFOA, PFNA, PFDA, PFUnDA, PFDoDA, PFTrDA, PFTeDA, PFOS, and 6:2 FTS in at least one sample (Table A6). PFOA was detected most frequently (at all 26 sites), followed by PFDoDA (at 25 sites) and PFUnDA (at 23 sites). The highest sediment concentration was that of 6:2 FTS in the Scheldt River at Steendorp (14.1 ng/g dw; site 23 on Figure 1). Long-chained PFAS were most abundantly detected in sediment (Figure 3), which is consistent with previous studies that reported a stronger adsorption of long-chained PFAS to sediments compared to short-chained hydrophilic PFAS that are more soluble in water (Gagliano et al., 2020; Ji et al., 2020; Lenka et al., 2021).

The presence of PFOA at all sites is likely due to its historical usage and production in Flanders. PFOA was used until 2010 in firefighting foams and was produced by 3M, which has a PFAS-producing factory in Flanders, until it phased-out production in the early 2000s. The adsorption of PFOA to sediments, and thus the possibility to leach out to groundwater or deeper sediment layers, is known to be affected by sediment composition, hydrochemistry, organic matter content, and surfactants (Lyu et al., 2021). It is possible that, in the present study, these factors prevented migration to deeper sediment layers, causing PFOA to be detected in surface sediments at all sites. Additionally, eroded particles from source zones (including soil that eroded due to heavy rainfalls) may have been deposited on the surface sediments (Borthakur et al., 2021).

In more than half of the samples, PFDoDA was predominant in the determination of the ∑PFAS concentrations (Table A6). The highest mean ∑PFAS concentration was measured in sediment from the Scheldt River at Wetteren (25.1 ng/g dw; site 25 on Figure 1) in East Flanders province (Figure 4). Similarly to the ∑PFAS concentration in water, this might be due to local sources such as former and recent fire-training areas and stations, and industry in the Port of Ghent. Sum concentrations in water were in almost all cases higher than those in sediment, which could be due to the presence of resuspended contaminated sediments that were caused by anthropogenic processes such as shipping (Kennish, 2002; Roberts, 2012; Bu et al., 2020). A recent review by Borthakur et al. (2021) reported that suspended particles in surface water can contain significantly higher PFAS concentrations than the sediment below, which was mainly caused by erosion of particles from source zones. They concluded that suspended particles can be a dominant pathway for PFAS transportation in aquatic environments. Since water samples were not filtered prior to extraction, it is possible that PFAS bound to suspended particles were still present in these samples.

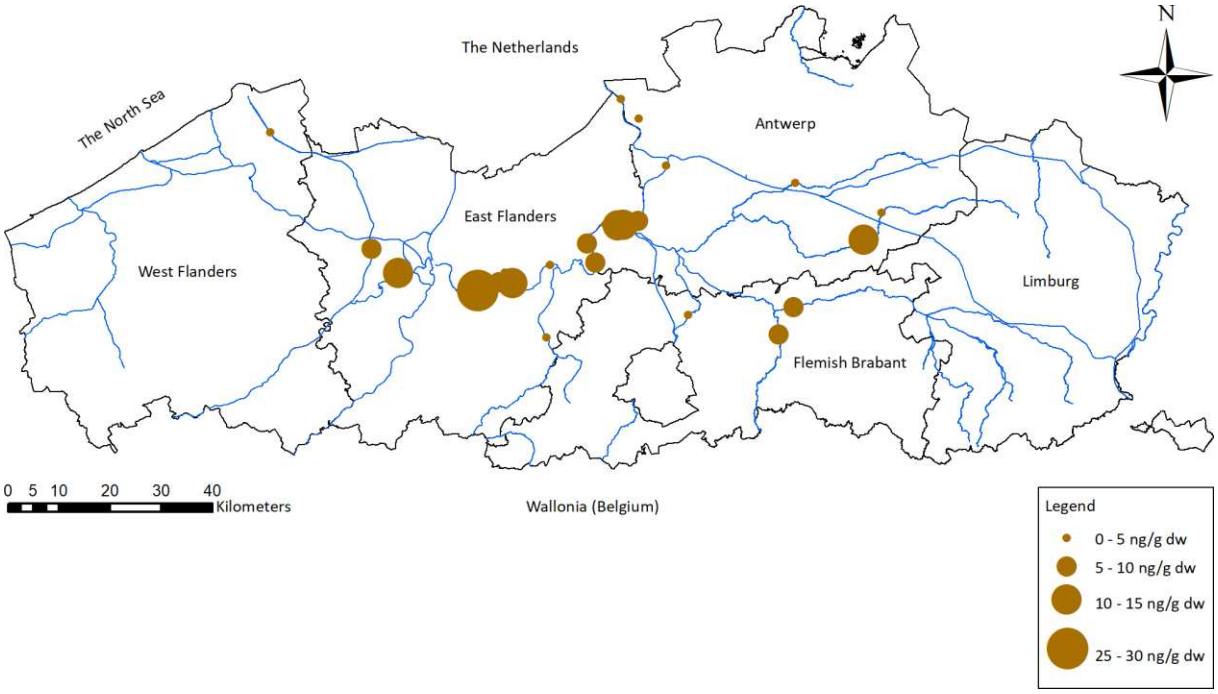


Figure 4. Mean ∑PFAS concentration in sediment (ng/g dw). Some points overlap due to the size of the circles. An overview of mean ∑PFAS concentration in sediment per location is shown in Table A6. Made with ArcMap 10.7.1 (projection: Lambert 72).

Comparable information on sediment PFAS concentrations in regions affected by multiple sources is scarce, though the ∑PFAS concentrations in the present study were higher than those reported in the Tampa Bay area (0.037–2.99 ng/g dw; Pulster et al., 2022), Jiulong River (0.24–1.9 ng/g dw; Wang et al., 2022) and Pensacola Bay System watershed (< 3.89 ng/g dw; Ahmadireskety et al., 2021), which are all three severely affected by multiple potential sources of PFAS contamination. On the other hand, the ∑PFAS concentrations in Flanders appeared to be in the lower part of the range of those detected at Lake Sänksjön in Sweden (3.0–61 ng/g dw; Mussabek et al., 2020), which is impacted by firefighting foams, and those detected in the Jucar River in Spain (14.3 – 75.9 ng/g dw; Campo et al., 2016), which is affected by multiple potential sources, including urban and industrial discharges.

Concentrations of PFHxA, PFOA, PFDoDA ,PFOS, and ∑PFAS were not significantly correlated between water and sediment (Table A15 shows p and r-values), possibly caused by differences among PFAS in their partitioning behavior to sediment (hydrophobic ones) and water (hydrophilic ones). Since these were the only PFAS with detection frequencies ≥ 30% in both matrices, we did not correlate PFAS concentrations in water and sediment for other PFAS. Similarly, besides ∑PFAS concentrations, only

compounds with a detection frequency ≥ 30% in sediment (i.e., PFHxA, PFOA, PFDA, PFUnDA, PFDoDA, PFTrDA, and PFOS) were correlated to the sediment characteristics. Both ∑PFAS concentrations and concentrations of the aforementioned PFAS were positively correlated to TOC, CEC, and clay content (Table A7). In general, the correlations with TOC appeared to be the strongest, followed by clay content and CEC. These observed correlations with TOC and clay content were in concordance with literature about the influence of sediment characteristics on the adsorption of PFAS (Shahsavari et al., 2020). The positive correlations with CEC could be due to divalent cations functioning as a bridge between the negatively charged sorbent surfaces and the negatively charged functional group of PFAS (Li et al., 2018).

3.3 Chinese mitten crabs

3.3.1 PFAS accumulation and tissue distribution

Hepatopancreas samples (Table A8) contained the largest diversity in detected PFAS compared to muscle tissue (Table A9) and carapace (Figure 3; Table A10). The internal standard of PFHxA could not be detected in any of the hepatopancreas samples. However, since all other ISTDs could be detected in this matrix and the ISTD of PFHxA was detected in the procedural blanks, we expect this to be caused by matrix effects specifically affecting PFHxA. The ∑PFAS concentration in the three crab tissues (Figure 5) was calculated as described previously for abiotic matrices (section 3.1). Only sampling sites with three or more analyzed crabs are shown in Figure 5, and details on the ∑PFAS concentration at the other sites are displayed in Tables A8–A10.

The precursor FBSA and the replacement compounds ADONA were detected at some sites, albeit with low relative contributions to the ∑PFAS concentration (Figure 3). ADONA contributed to less than 0.5% of the total PFAS in the hepatopancreas and carapace (and was not detected in muscle tissue), whereas FBSA contributed to 4%, 2% and 0.3% in the hepatopancreas, muscle, and carapace, respectively. FBSA has only been recently reported in environmental samples (Chu et al., 2016; Kaboré et al., 2022; Pickard et al., 2022) and is a degradation product and major metabolite of other precursors in surface

treatment products and aqueous film-forming foams (AFFFs). It is a precursor of PFBS, which is nowadays used as a replacement for long-chained sulfonic acids (Barzen-Hanson et al., 2017; Pickard et al., 2022). ADONA is used as a substitute for long-chain PFAS (Fromme et al., 2017; Munoz et al., 2019), for example as emulsifier in the production of fluoropolymers (Fromme et al., 2017), and is also only sporadically observed in the environment (Fromme et al., 2017; Pan et al., 2018).

Overall the accumulated PFAS concentrations showed significant variability, reflected by high standard errors, in all the tissues (Tables A8-A10). This is likely caused by the sometimes small number of crabs collected and analyzed at each site (Table A1), or other factors such as differences in age or sex. Although all caught crabs were considered adults, their exact age was not investigated. Differences in PFAS accumulation between males and females could also not be assessed due to the relative small sample size. However, both factors are known to cause variation in PFAS accumulation in organisms (Sinclair et al., 2006).

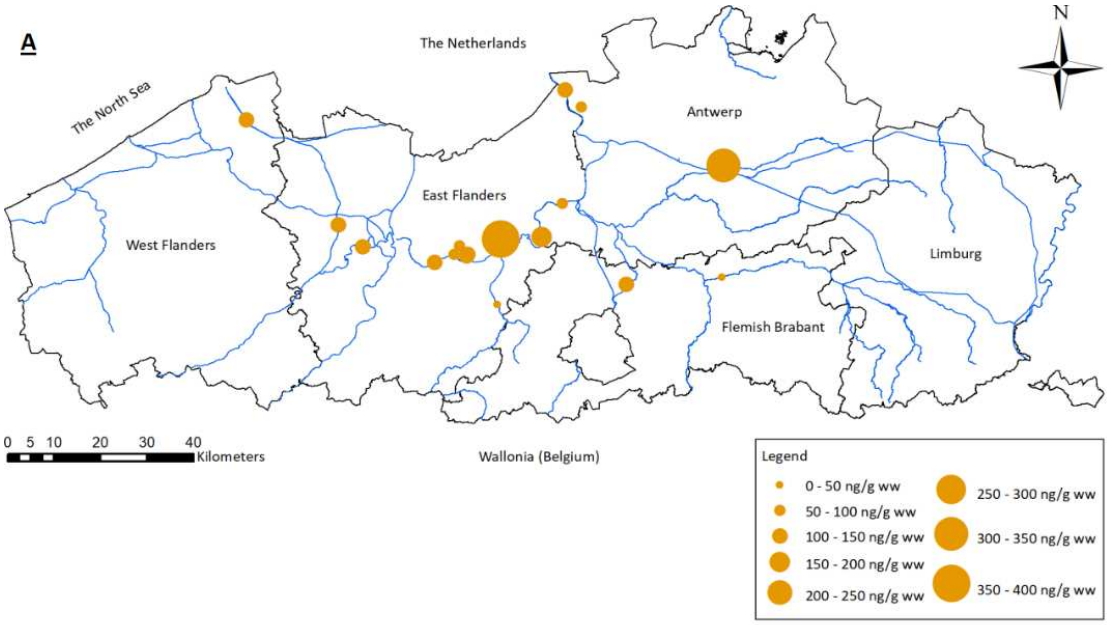
The ∑PFAS concentrations in hepatopancreas were dominated by the long-chained PFDoDA, PFTrDA, and PFTeDA (Figure 3). At some locations, short-chain PFAS such as PFBA, PFBS, and PFPeS, were also detected in the hepatopancreas, albeit in relatively low concentrations. Similarly, PFDoDA and PFTrDA dominated the ∑PFAS concentrations in muscle tissue (Figure 3). However, when PFTeDA and PFBS were detected in muscle tissue, their contribution to the ∑PFAS was generally higher than those of PFDoDA and PFTrDA (Table A9). Thus, despite their lower detection, the concentrations of PFTeDA and PFBS significantly affected PFAS profiles in muscle tissue of the crabs (Figure 3), which were otherwise very similar to those of the hepatopancreas. Contrary to what was observed in hepatopancreas and muscle tissue, PFBS, when detected, dominated the ∑PFAS concentrations in carapace (Figure 3; Table A10). Although we washed the carapace samples with Milli-Q water, we should be aware of the possibility that the PFAS concentrations in the carapace could also be influenced by external contamination. Possible external contamination of PFAS has been reported before for other inert tissues, such as feathers (Groffen et al., 2020). Brust et al. (2018) and Leenders et al. (2021) reported

the dominance of PFOS in Chinese mitten crabs caught in the Netherlands, although both studies analyzed PFAS in meat from the body, which includes hepatopancreas and gonads. Besides these tissue-specific differences, differences in pollution sources in both countries could also attribute to differences in dominance of certain PFAS. Nonetheless, the dominance of long-chained PFAS in hepatopancreas was expected, as long-chained PFAS are known to bind to liver fatty acid binding proteins (Khazaee et al., 2021; Wang et al., 2022). Furthermore, crabs are known to accumulate high concentrations of long-chain PFAS in particular (Choi et al., 2020; Taylor et al., 2021; Byns et al., 2022; Young et al., 2022).

The highest ∑PFAS concentration in hepatopancreas was measured in samples from the Scheldt River at Appels (site 18 on Figure 1), followed by the Kleine Nete River at Grobbendonk (Figure 5a; site 9 on Figure 1). In muscle tissue, the highest concentrations were observed in the Bergenmeersen nature area (Figure 5b, sites 2, 3 and 4 on Figure 1) and in carapace, the highest concentration was detected in the Scheldt River at Kastel/Baasrode (Figure 5c; site 20 on Figure 1). Hong et al. (2015) also reported the presence of PFAS in the carapace of multiple crab species (e.g., beach crab, penicillate shore crab, and flat shore crab) from the Korean west coast, but the ∑PFAS concentrations in those crabs (< 20 ng/g ww) were lower than those reported in the present study (mean ∑PFAS concentration of 115 ng/g ww). To the best of our knowledge, there are no other studies that have examined PFAS accumulation in carapaces. Prosser et al. (2016) have suggested that PFAS could potentially bind to proteins in chitin in the carapace of arthropods. Hence, it is hypothesized that the molting process of crabs could be a potential elimination pathway for PFAS accumulated in crabs (Veilleux and de Lafontaine, 2007). However, more research is necessary to further investigate this.

For some crab tissues, and some PFAS, we found negative correlations between the accumulated concentrations and the crab size (expressed as weight and carapace width) (Table A13). Growth dilution of PFAS has been reported before in animals and humans (e.g., Hoff et al., 2003; Wu et al., 2015), but has, to the best of our knowledge, not been investigated in crabs thus far.

The ∑PFAS concentrations in soft tissues (approximately 35 ng/g ww) and legs (< 10 ng/g ww) of crabs from the Korean west coast (Hong et al., 2015) were lower than those reported in the hepatopancreas (mean ∑PFAS concentration of 123 ng/g ww) and muscle tissue (mean ∑PFAS concentration of 76.3 ng/g ww) of crabs collected in the present study. Similarly, the ∑PFAS concentrations in whole Chinese mitten crabs from the Netherlands (approximately 27 ng/g ww; Leenders et al., 2021) were lower than those reported in the individual tissues of crabs collected in the present study. In addition, mean concentrations in the tissues of various crab species from South Korea, China, Pakistan, India, Australia, UK, Japan, and Norway were often lower than those reported in Flanders (Nakata et al., 2006; Clarke et al., 2010; Langberg et al., 2019; Taylor, 2019; Choi et al., 2020; Ali et al., 2021). Geographical features or climatic conditions may affect the distribution of PFAS in crabs (Habibullah-Al-Mamun et al., 2017). Furthermore, pollution sources and the degree of environmental exposure may differ between regions. Within Europe, Flanders has been recently identified as highly PFAS contaminated area, with numerous already known contaminated sites (Forever Pollution Project, 2023).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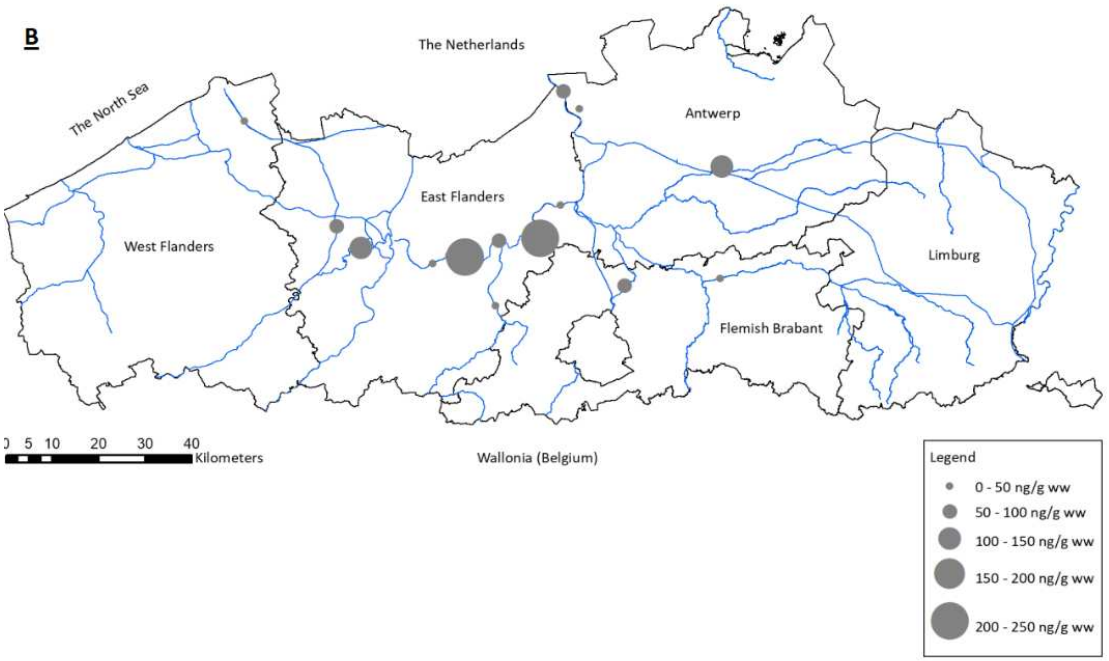
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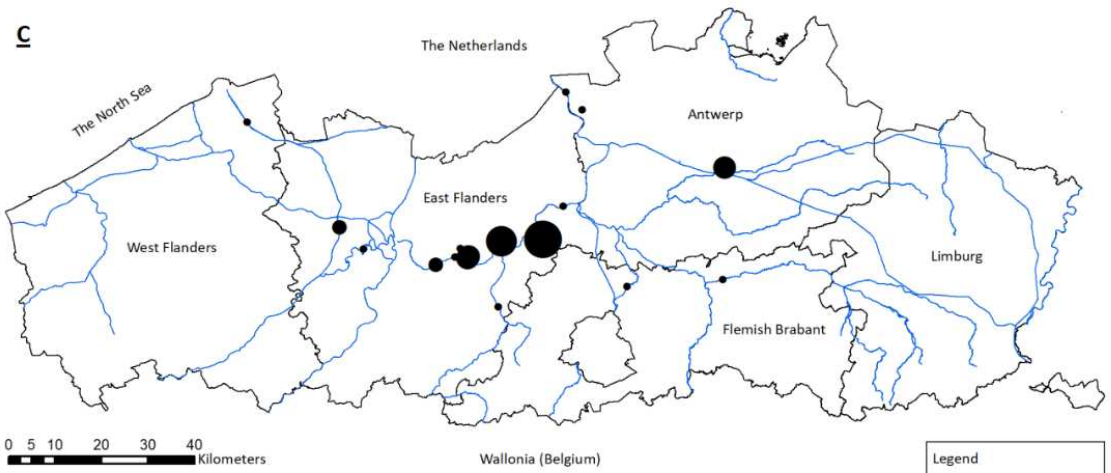
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Figure 5. Mean ∑PFAS concentration in A) hepatopancreas (ng/g ww), B) muscle tissue (ng/g ww), C) carapace (ng/g ww). Only sampling sites with three or more crabs are displayed. Some sampling points overlap and a detailed overview of the mean ∑PFAS concentration per location is displayed in Table A8 for hepatopancreas, Table A9 for muscle tissue and Table A10 for carapace. Made with ArcMap 10.7.1 (projection: Lambert 72).

Since we only took compounds with a detection frequency ≥ 30% in the crab tissues into account, comparisons among the three crab tissues, and correlations between the tissues, were only possible for PFOA, PFDA, PFUnDA, PFDoDA, PFTrDA, PFTeDA, and PFOS. Due to too low detection in one of the tissues, FBSA concentrations were only compared and correlated between hepatopancreas and muscle tissue, NaDONA only between hepatopancreas and carapace, and PFBS between muscle tissue and carapace. In addition, ∑PFAS concentrations were compared and correlated among these tissues (Figure 6).

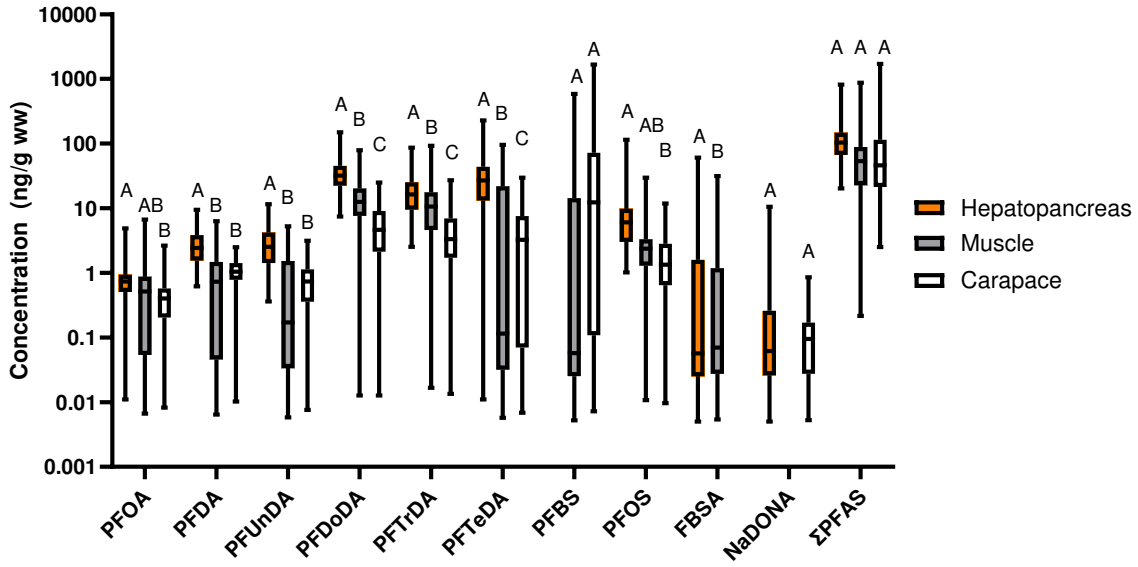

Figure 6. Comparison of PFAS concentrations among the three crab tissues (hepatopancreas, muscle tissue, and carapace). Only PFAS with a detection frequency of ≥ 30% were included. Statistical differences (i.e., p-values ≤ 0.05) among the tissues are indicated by differences in letters. Note that the y-axis has a log-scale.

Although ∑PFAS concentrations did not differ among the three tissues ($p = 0.116$), significantly higher concentrations of PFDA, PFUnDA, PFDoDA, PFTrDA, and PFTeDA were observed in hepatopancreas compared to both muscle tissue and carapace ($p < 0.001$). In addition, PFOA and PFOS concentrations in hepatopancreas were higher than those in the carapace ($p < 0.001$), but did not differ from those in the muscle tissue. FBSA concentrations were higher in hepatopancreas compared to muscle tissue (p

= 0.051) and NaDONA concentrations in hepatopancreas were higher than those in the carapace (p = 0.061). Finally, the PFDoDA, PFTrDA, and PFTeDA concentrations were significantly lower in the carapace than in the muscle tissue ($p < 0.001$), whereas those of PFBS were lower in the muscle tissue (p = 0.053). Those of PFDA and PFUnDA did not differ between muscle and carapace.

A follow-up study with better characterization of lipid and protein content in the crab tissue would allow better insights in whether the observed differences can be attributed to PFAS having different affinities to accumulate in these three tissues. However, as mentioned earlier, long-chained PFAS are known to have a high affinity for liver fatty acid binding proteins (Sinclair et al., 2006; Khazaee et al., 2021; Wang et al., 2022; Zhao et al., 2023), which might explain the higher concentrations in hepatopancreas compared to the other tissues. In addition, especially acidic PFAS are also known to associate with phospholipids rather than storage lipids (Sinclair et al., 2006; Zhao et al., 2023), which could also contribute to tissue-specific accumulation differences of PFAS in the crabs. Furthermore, the molting process of crabs could explain why concentrations differ among the tissues. Hong et al. (2015) also reported higher concentrations in the soft tissues of the crabs compared to the carapace and legs of crabs. However, they reported almost twice as high concentrations in the carapace than in the legs. Similarly, Choi et al. (2020) reported higher concentrations in offal (which include hepatopancreas) compared to the legs of Korean, Chinese, Indian, and Pakistani crabs.

The accumulated PFAS concentrations in the muscle tissue were not correlated to those in the carapace, with exception of a significant correlation for PFBS and ∑PFAS (Table A11). However, they were often positively correlated to those in hepatopancreas. PFAS concentrations in the hepatopancreas were also often significantly correlated (positive) with those in the carapace (Table A11). The hepatopancreas acts primarily to absorb and store nutrients from food and is a primary digestive organ in crustaceans. Stored nutrients are transported to the muscle and other tissues during growth and reproductive stages (Wang et al., 2014). In addition, the hepatopancreas stores large amounts of lipids that are needed for molting, reproduction, etc. (Xu et al., 2020). This might explain

why correlations between the hepatopancreas and other tissues were often observed, whereas correlations between muscle tissue and carapace were often absent.

3.3.2 Correlations with the abiotic environment

To evaluate the possibilities of using the invasive crabs to assess environmental contamination with PFAS, we performed correlation tests between internal concentrations and those present in the water and sediment. Furthermore, we performed multiple regressions taking into account sediment and water characteristics.

We only considered PFAS when their mean concentrations were detected in at least 30% of the sites in the two matrices that were correlated to each other. The ∑PFAS concentrations were always correlated between the different matrices. More specifically, we correlated the ∑PFAS concentrations in water, and concentrations of PFOA, PFDA, PFUnDA, PFDoDA, PFTrDA, PFOS, and ∑PFAS in sediment, to those internally accumulated in the crabs. No significant correlations were observed between environmental concentrations and crab concentrations, which is also clear from the PCA biplot (Figure 7; Table A16). This general lack of correlations suggests that Chinese mitten crabs cannot be used to predict local PFAS pollution in the abiotic environment.

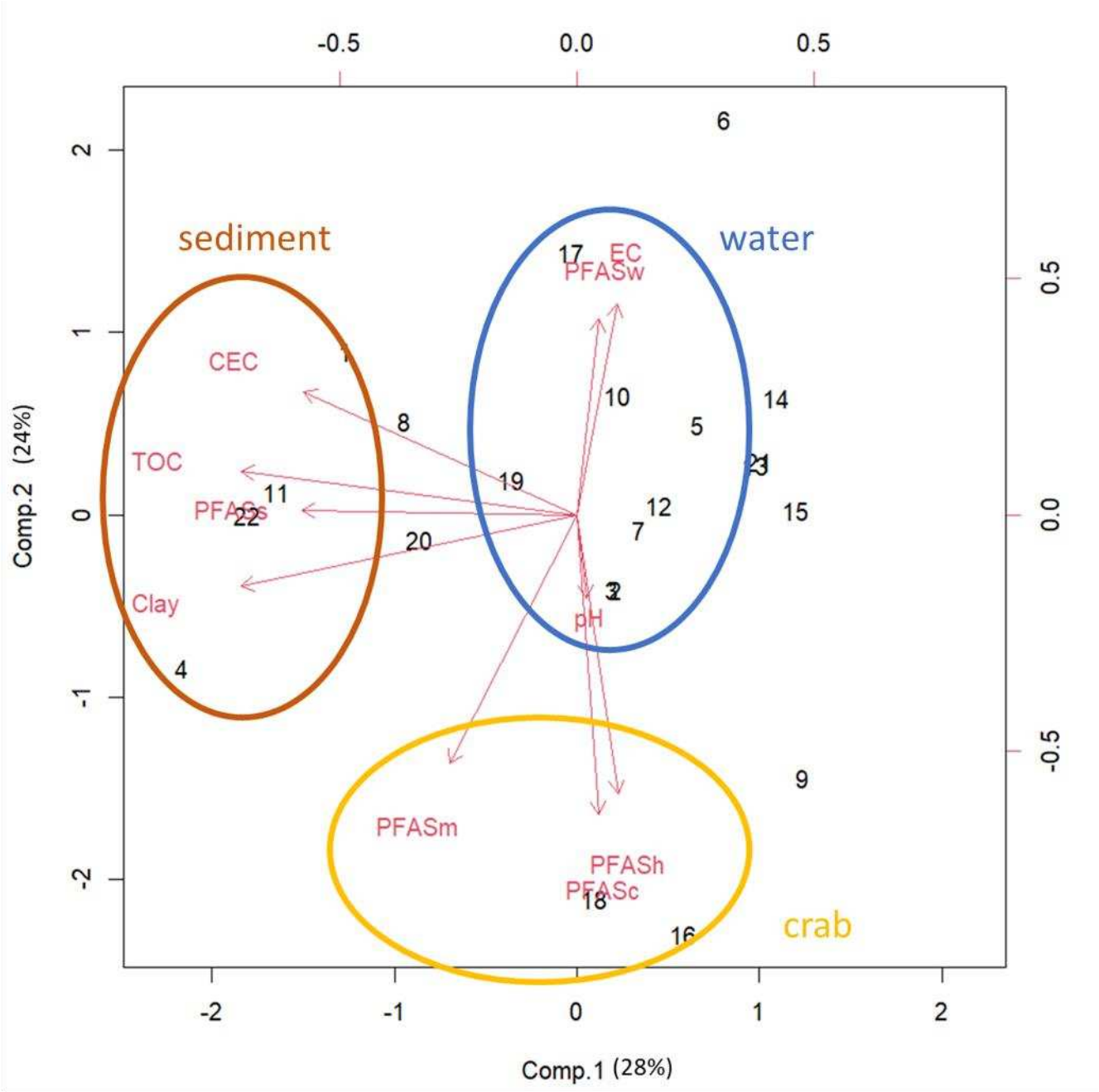

Figure 7. Principal component analysis of the ∑PFAS concentration in water (PFASw), sediment (PFASs), muscle (PFASm), hepatopancreas (PFASh), and carapace (PFASc), as well as the different physicochemical characteristics of water (electrical conductivity (EC) and pH) and sediment (cation exchange capacity (CEC), total organic carbon content (TOC) and clay content (Clay)). The colored circles represent the different groups of samples: brown = sediment, blue = water, and orange = crabs. N = 22. PC1 explained 28% of the variation. PC2 explained 24% of the proportion of variance.

Current PFAS biomonitoring programs in Flanders use indigenous and endangered fish species together with translocated bivalves (Teunen et al., 2020). Our results show that the invasive Chinese mitten crabs can be used simultaneously in biomonitoring studies as they provide complementary information to the fish species and mussels. In general, similar types of PFAS accumulated in all organisms, with some exceptions. However, the dominance of specific PFAS differed among species.

Teunen et al. (2020) reported a dominance of PFOS and PFOA in fish (Figure 3) and clams, respectively, whereas in the present study PFDoDA and PFTrDA were most abundant in the muscle and hepatopancreas of the crabs, and PFBS was dominant in the carapace (Figure 3). Chinese mitten crabs have a different feeding ecology and occupy a different trophic position than the fish and clam species, indicating differences in bioaccumulation but also in potential health risks for top predators through potential biomagnification. This not only shows that species-specific differences in PFAS bioavailability and bioaccumulation occur, but also implies that risks for top predators cannot be examined solely based on fish monitoring, as, by doing so, important information on other PFAS that are bioavailable in the environment would be ignored. A multi-species approach in biomonitoring is therefore recommended (Holt and Miller, 2010).

4. Conclusion

Chinese mitten crab is an invasive species of which populations have to be managed to preserve ecosystem functioning and services. Their presence at highly contaminated sites, as well as the high accumulated concentrations of PFAS in their analyzed tissues, confirm that this species can tolerate high pollution levels. In addition, our results further confirm that the aquatic environment in Flanders is highly contaminated with PFAS. Since the ubiquitously present Chinese mitten crabs do accumulate PFAS, they can be used to quantitatively assess the presence of PFAS in aquatic environments, meaning that they can be used as a biomonitor (quantitative assessment). However, their suitability as bioindicator (qualitative assessment) is not confirmed by this study, as accumulated levels do not reflect those of the abiotic environment. However, since we did not investigate the bioavailable fraction of PFAS, further research is necessary to investigate whether the internal concentrations in the crabs reflect the bioavailable fraction. In addition, PFAS accumulation profiles differed from those in resident fish species, showing that Chinese mitten crab responses to PFAS pollution do not correspond to those of the whole community or ecosystem. Thus, this invasive species can be considered a suitable biomonitor for PFAS pollution, but cannot yet replace conventional

biomonitoring with fish. More research is first needed such as dose-response experiments and a better characterization of lipid and protein content in relation to the PFAS concentration. We do recommend to include Chinese mitten crabs in future biomonitoring programs, because they do provide complementary information to the PFAS concentrations in the currently used indigenous fish species and mussels.

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Statements and declarations

Author contributions

Thimo Groffen: Conceptualization, Methodology, Formal Analysis, Writing, Visualization, Supervision; Funding Acquisition; Heleen Keirsebelik: Conceptualization, Methodology, Writing; Hannes Dendievel: Investigation, Methodology, Formal Analysis, Writing, Visualization; Mathilde Falcou-Préfol: Methodology, Writing; Lieven Bervoets: Conceptualization, Methodology, Writing, Supervision, Funding Acquisition; Jonas Schoelynck: Conceptualization, Methodology, Writing, Supervision, Funding Acquisition

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Disclosure statement

All authors certify that they have no affiliations with or involvement in any organization or entity with any financial interest or non-financial interest in the subject matter or materials discussed in this manuscript. Hence, the authors report there are no competing interests to declare.

Availability of data

The datasets generated and/or analyzed during the current study are not publicly available. The test data is restricted to the relevant personnel of the project and is not allowed to be disclosed to the public but are available from the corresponding author on reasonable request.

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Appendix

Table A1. Overview of sample locations, type of water body, Lambert 72 coordinates, number of crabs collected, and sediment sampling method. The numbers of the water body (Site No.) correspond to the numbers in Figure 1.

Site no.	Water body	Location	Type	X (Lambert 72)	Y (Lambert 72)	#crabs	Sediment sampling method	Remarks
1	Demer	Rotselaar	River	176745	184948	3	Plastic tube	
2	Main branch	Bergenmeersen	Creek in tidal marsh	121861	189941	3	Trowel	
3	Sluice	Bergenmeersen	River	121991	189961	2	Trowel	
4	Ditches 13 & 14	Bergenmeersen	Creek in tidal marsh	121617	189590	13	Trowel	
5	Driesesloot	Berlare	River	120111	191635	4	Trowel	Water and sediment collected at x = 120111, y = 191635 because of limited accessibility
6	Fish spawning pond	Lillo	Pond	146439	221934	5	Trowel	
7	Zenne	Eppegem	River	156107	183353	5	Trowel	
8	Kalkense Vaart	Kalken	River	118914	189775	5	Trowel	
9	Kleine Nete	Grobbendonk	River	177101	209311	5	Trowel	
10	Dender	Aalst	River	128290	178929	4	Plastic tube	
11	Leie	Gent	River	99259	191560	5	Plastic tube	
12	Schipdonk Canal	Nevele/Deinze	Canal	93949	196299	3	Grab sampler	Water and sediment collected at x = 90487, y = 186649 because of limited accessibility
13	Leopold Canal	Damme	Canal	74157	219196	5	Trowel	
14	Grote Laak	Geel	River	190461	198085	2	Plastic tube	
15	Grote Nete	Geel	River	193995	203508	2	Trowel	
16	Dijle	Wijgmaal	River	173809	179531	1	Trowel	
17	Scheldt	Antwerpen	River	151789	212664	0	Grab sampler	
18	Scheldt	Appels	River	129004	193176	4	Trowel	
19	Scheldt	Doel	River	142882	225713	10	Grab sampler	
20	Scheldt	Kastel/Baasrode	River	137834	193644	3	Grab sampler	Water and sediment collected at Baasrode
21	Scheldt	Lippenbroek	Creek in tidal marsh	136219	197361	1	Trowel	
22	Scheldt	Notelaer	Creek in tidal marsh	143406	201056	2	Trowel	
23	Scheldt	Steendorp	River	142284	200891	5	Grab sampler	
24	Scheldt	Temse	River	139876	201169	0	Grab sampler	
25	Scheldt	Wetteren	River	114823	188235	5	Grab sampler	
26	Scheldt	Wintam	River	146212	201883	0	Grab sampler	

Table A2. MRM transitions, internal standards (ISTDs), cone voltages (V) and collision energy (eV) for the target perfluoroalkyl substances and their internal standards. Table adapted from Groffen et al. (2019c, 2021). Blank cells indicate that no second diagnostic product ion was used (and thus no collision energy and cone voltage could be reported).

Name	Acronym	Precursor ion (m/z)	Product ion (m/z)		Collision energy (eV)		Cone voltage (V)		ISTD used for quantification
			Diagnostic product ion 1	Diagnostic product ion 2	Diagnostic product ion 1	Diagnostic product ion 2	Diagnostic product ion 1	Diagnostic product ion 2	
Perfluorobutanoic acid	PFBA	213	169	169	19	50	19	19	[1,2,3,4-$^{13}C_4$]PFBA
Perfluoropentanoic acid	PFPeA	263	219	219	10	45	15	15	[1,2,3,4-$^{13}C_4$]PFBA
Perfluorohexanoic acid	PFHxA	313	269	119	21	65	19	19	[1,2-$^{13}C_2$]PFHxA
Perfluoroheptanoic acid	PFHpA	363	319	169	40	30	24	24	[1,2-$^{13}C_2$]PFHxA
Perfluorooctanoic acid	PFOA	413	369	169	13	60	22	22	[1,2,3,4-$^{13}C_4$]PFOA
Perfluorononanoic acid	PFNA	463	419	169	17	20	28	28	[1,2,3,4,5,-$^{13}C_5$]PFNA
Perfluorodecanoic acid	PFDA	513	469	219	29	29	25	25	[1,2-$^{13}C_2$]PFDA
Perfluoroundecanoic acid	PFUnDA	563	519	169	30	35	18	18	[1,2-$^{13}C_2$]PFUnDA
Perfluorododecanoic acid	PFDoDA	613	569	319	21	30	22	22	[1,2-$^{13}C_2$]PFDoDA
Perfluorotridecanoic acid	PFTrDA	663	619	319	21	30	26	26	[1,2-$^{13}C_2$]PFDoDA
Perfluorotetradecanoic acid	PFTeDA	713	669	169	21	21	28	28	[1,2-$^{13}C_2$]PFDoDA
Perfluorobutane sulfonate	PFBS	299	80	99	65	45	40	40	$^{18}O_2$-PFHxS
Perfluoropentane sulfonate	PFPeS	349	80	99	40	40	40	35	[1,2,3,4-$^{13}C_4$]PFOS
Perfluorohexane sulfonate	PFHxS	399	80	99	30	60	22	22	$^{18}O_2$-PFHxS
Perfluoroheptane sulfonate	PFHpS	449	80	98.5	47	45	40	40	[1,2,3,4-$^{13}C_4$]PFOA
Perfluorooctane sulfonate	PFOS	499	80	99	58	58	60	60	[1,2,3,4-$^{13}C_4$]PFOS
Perfluorodecane sulfonate	PFDS	599	80	99	63	63	29	29	[1,2,3,4-$^{13}C_4$]PFOS
4:2 fluorotelomer sulfonate	4:2 FTS	327	307	80	25	33	20	20	[1,2,3,4-$^{13}C_4$]PFOS
6:2 fluorotelomer sulfonate	6:2 FTS	427	407	80	25	33	20	20	[1,2,3,4-$^{13}C_4$]PFOS
8:2 fluorotelomer sulfonate	8:2 FTS	527	507	81	40	40	36	36	[1,2,3,4-$^{13}C_4$]PFOS
4,8-dioxa-3H-perfluorononanoic acid	NaDONA	376.8	250.7	84.8	35	32	23	23	[1,2,3,4-$^{13}C_4$]PFOA
Hexafluorpropylene oxide-dimer acid	HFPO-DA (GenX)	285	169		20		30		[1,2-$^{13}C_2$]PFHxA
9-chlorohexadecafluoro-3-oxanonane-1-sulfonate	9Cl-PF3ONS	531	350.5	83	32	37	46	40	[1,2,3,4,5,-$^{13}C_5$]PFNA
11-chloroeicosafluoro-3-oxaundecane-1-sulfonate	11Cl-PF3OUdS	631	451	83	40	35	50	40	[1,2-$^{13}C_2$]PFUnDA
Perfluoro-4-oxapentanoic acid	PF4OPeA (PFMPA)	228.8	85		20		20		[1,2,3,4-$^{13}C_4$]PFOA
Perfluoro-5-oxahexanoic acid	PF5OHxA (PFMBA)	279	85		20		20		[1,2-$^{13}C_2$]PFHxA
Perfluoro-3,6-dioxaheptanoic acid	3,6-OPFHpA (NFDHA)	201	85		25		30		[1,2-$^{13}C_2$]PFHxA

Perfluoro(2-ethoxyethane) sulfonate	PFEESA	315	135	69	20	55	30	35	[1,2-$^{13}C_2$]PFDA
Perfluorobutane sulfonamide	FBSA	298	219	78	27	38	34	40	[1,2,3,4-$^{13}C_4$]PFBA
	[1,2,3,4-$^{13}C_4$]PFBA	217	172	172	19	50	19	19	
	[1,2-$^{13}C_2$]PFHxA	315	269	119	21	65	19	19	
	[1,2,3,4-$^{13}C_4$]PFOA	417	372	172	13	60	22	22	
	[1,2,3,4,5,-$^{13}C_5$]PFNA	468	423	172	17	20	28	28	
	[1,2-$^{13}C_2$]PFDA	515	470	220	29	29	25	25	
	[1,2-$^{13}C_2$]PFUnDA	565	520	170	32	35	18	18	
	[1,2-$^{13}C_2$]PFDoDA	615	570	320	21	30	22	22	
	$^{18}O_2$-PFHxS	403	84	103	30	60	22	22	
	[1,2,3,4-$^{13}C_4$]PFOS	503	80	99	58	58	60	60	

Table A3. Limits of detection (LOD) and limits of quantification (LOQ) per PFAS analyte and per matrix. LODs and LOQs were determined in matrix, as the concentration corresponding to a S/N-ratio of 3 and 10, respectively.

Compound	Water (ng/L)		Sediment (ng/g dw)		Hepatopancreas (ng/g ww)		Crab muscle (ng/g ww)		Carapace (ng/g ww)	
	LOD	LOQ	LOD	LOQ	LOD	LOQ	LOD	LOQ	LOD	LOQ
PFBA	1.54	5.12	0.0660	0.219	0.0330	0.110	0.0620	0.206	0.0680	0.227
PFPeA	1.34	4.45	0.0470	0.158	0.116	0.388	0.140	0.466	0.0980	0.327
PFHxA	1.60	5.33	0.225	0.751	0.148	0.492	0.0860	0.285	0.146	0.486
PFHpA	1.79	5.97	0.175	0.584	0.720	2.40	0.119	0.397	0.230	0.765
PFOA	1.73	5.75	0.0420	0.140	0.0370	0.122	0.0510	0.171	0.0490	0.163
PFNA	1.65	5.51	0.0400	0.134	0.0510	0.171	0.193	0.642	0.0340	0.112
PFDA	4.68	15.6	0.0680	0.227	0.123	0.411	0.0640	0.212	0.0600	0.199
PFUnDA	4.83	16.1	0.0620	0.205	0.0980	0.327	0.0930	0.309	0.0470	0.158
PFDoDA	20.5	68.2	0.169	0.564	0.321	1.07	0.351	1.17	0.0890	0.296
PFTrDA	3.24	10.8	0.104	0.346	0.225	0.751	0.161	0.537	0.0600	0.201
PFTeDA	12.7	42.4	0.384	1.28	1.17	3.90	0.726	2.42	0.182	0.606
PFBS	18.9	63.1	1.48	4.94	0.933	3.11	0.333	1.11	0.561	1.87
PFPeS	2.62	8.74	0.0920	0.307	0.188	0.628	1.04	3.48	0.0660	0.221
PFHxS	50.7	169	0.660	2.20	2.78	9.28	1.75	5.84	0.423	1.41
PFHpS	21.7	72.4	0.306	1.02	0.774	2.58	0.945	3.15	0.146	0.486
PFOS	1.62	5.41	0.0380	0.128	0.113	0.378	0.186	0.619	0.0280	0.0950
PFDS	3.39	11.3	0.0960	0.319	0.804	2.68	0.573	1.91	0.0510	0.171
4:2 FTS	2.04	6.80	0.0850	0.284	0.257	0.857	0.639	2.13	0.287	0.957
6:2 FTS	3.66	12.2	0.146	0.485	0.187	0.623	1.35	4.50	0.113	0.375
8:2 FTS	4.14	13.8	0.124	0.412	0.438	1.46	1.07	3.58	0.0700	0.233
NaDONA	4.26	14.2	0.163	0.544	0.0350	0.116	0.0330	0.109	0.0110	0.0380
HFPO-DA (GenX)	5.94	19.8	0.181	0.603	0.0300	0.101	0.115	0.384	0.669	2.23
9Cl-PF3ONS	2.24	7.47	0.0830	0.278	0.531	1.77	0.299	0.997	0.0160	0.0520
11Cl-PF3OUdS	2.50	8.33	0.0900	0.299	1.16	3.85	0.144	0.481	0.0560	0.188
PF4OPeA	5.28	17.6	0.213	0.711	0.435	1.45	0.197	0.655	0.101	0.338
PF5OHxA	5.70	19.0	0.180	0.599	0.567	1.89	0.227	0.758	0.155	0.515
3,6-OPFHpA	7.47	24.9	0.194	0.645	0.705	2.35	0.252	0.840	0.170	0.567
PFEESA	2.74	9.13	0.0940	0.313	0.744	2.48	1.42	4.73	0.0740	0.245
FBSA	2.03	6.75	0.193	0.644	0.064	0.213	0.110	0.367	0.105	0.349

Table A4. Mean ∑PFAS concentrations (ng/L; ± SE) and concentrations (mean (± SE) and range) of individual PFAS (ng/L) in water at the different sampling sites. Sampling sites are numbered according to Table A1. Values < LOQ were substituted according to the MLE method in the calculation of mean concentrations of the individual compounds. In the calculation of ∑PFAS concentrations, values < LOQ were substituted by 0. N = 3 per site, except for site 4 where N = 6.

Site no.		PFBA	PFPeA	PFHxA	PFOA	PFDA	PFDoDA	PFOS	∑PFAS
1	Mean	< LOQ	< LOQ	8.39 ± 5.05	< LOQ	< LOQ	< LOQ	50.9 ± 36.3	114 ± 4.73
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 17.5	< LOQ – 7.04	< LOQ - < LOQ	< LOQ - < LOQ	7.55 - 123	
2	Mean	< LOQ	< LOQ	7.00 ± 4.25	< LOQ	< LOQ	152 ± 77.5	< LOQ	162 ± 74.4
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 14.7	< LOQ – 8.68	< LOQ - < LOQ	< LOQ – 255	< LOQ - < LOQ	
3	Mean	< LOQ	< LOQ	11.4 ± 1.98	< LOQ	< LOQ	< LOQ	< LOQ	11.4 ± 1.98
	Range	< LOQ - < LOQ	< LOQ - < LOQ	7.42 – 13.6	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	
4	Mean	< LOQ	< LOQ	10.5 ± 1.04	< LOQ	< LOQ	< LOQ	< LOQ	48.7 ± 24.0
	Range	< LOQ - < LOQ	< LOQ - < LOQ	7.86 – 14.2	< LOQ – 6.04	< LOQ - < LOQ	< LOQ – 139	< LOQ - < LOQ	
5	Mean	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	9.31 ± 5.79
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 13.5	< LOQ – 7.99	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	
6	Mean	369 ± 40.6	8.50 ± 1.21	15.5 ± 2.12	10.8 ± 0.81	< LOQ	< LOQ	< LOQ	404 ± 43.6
	Range	299 – 435	6.37 – 8.44	11.5 – 18.7	9.42 – 12.2	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	
7	Mean	< LOQ	< LOQ	13.1 ± 2.92	< LOQ	< LOQ	< LOQ	< LOQ	78.4 ± 30.5
	Range	< LOQ - < LOQ	< LOQ - < LOQ	9.77 – 18.9	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 110	< LOQ - < LOQ	
8	Mean	< LOQ	13.2 ± 1.85	13.2 ± 2.70	< LOQ	< LOQ	85.9 ± 44.2	< LOQ	105 ± 47.4
	Range	< LOQ - < LOQ	< LOQ – 5.88	8.26 – 17.5	< LOQ – 7.38	< LOQ - < LOQ	< LOQ – 147	< LOQ - < LOQ	
9	Mean	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	3.32 ± 3.32
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 9.96	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	
10	Mean	< LOQ	< LOQ	7.18 ± 3.79	< LOQ	< LOQ	< LOQ	< LOQ	9.26 ± 1.94
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 12.9	< LOQ – 6.26	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	
11	Mean	< LOQ	< LOQ	31.9 ± 0.82	< LOQ	< LOQ	< LOQ	< LOQ	34.1 ± 2.80
	Range	< LOQ - < LOQ	< LOQ - < LOQ	30.4 – 33.2	< LOQ – 6.36	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	
12	Mean	< LOQ	< LOQ	17.9 ± 3.53	< LOQ	< LOQ	< LOQ	< LOQ	46.1 ± 14.6
	Range	< LOQ - < LOQ	< LOQ - < LOQ	6.51 – 18.0	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 78.5	< LOQ – 6.09	
13	Mean	< LOQ	< LOQ	8.60 ± 2.15	< LOQ	< LOQ	< LOQ	< LOQ	72.0 ± 36.5
	Range	< LOQ - < LOQ	< LOQ - < LOQ	5.57 – 12.8	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 119	< LOQ - < LOQ	
14	Mean	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	17.0 ± 9.42
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 10.9	< LOQ – 7.72	< LOQ – 26.1	< LOQ - < LOQ	< LOQ - < LOQ	
15	Mean	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	11.3 ± 7.35
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 12.8	< LOQ – 6.51	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 8.73	
16	Mean	< LOQ	< LOQ	8.40 ± 0.967	< LOQ	< LOQ	77.7 ± 39.5	< LOQ	88.3 ± 71.1
	Range	< LOQ - < LOQ	< LOQ - < LOQ	6.73 – 10.1	< LOQ – 6.37	< LOQ - < LOQ	< LOQ – 129	< LOQ - < LOQ	
17	Mean	44.4 ± 6.49	< LOQ	9.40 ± 1.88	7.37 ± 3.88	< LOQ	< LOQ	16.1 ± 1.33	111 ± 37.4
	Range	31.6 – 52.6	< LOQ - < LOQ	7.08 – 13.2	< LOQ – 13.3	< LOQ - < LOQ	< LOQ – 102	13.5 – 17.6	
18	Mean	< LOQ	< LOQ	10.8 ± 5.49	< LOQ	< LOQ	< LOQ	< LOQ	10.8 ± 5.51
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 18.2	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	
19	Mean	64.1 ± 15.5	< LOQ	14.0 ± 3.46	6.20 ± 3.12	< LOQ	< LOQ	14.6 ± 3.63	157 ± 45.9
	Range	42.7 – 94.2	< LOQ - < LOQ	7.35 – 18.9	< LOQ – 10.3	< LOQ - < LOQ	< LOQ – 95.2	7.49 – 19.3	
20	Mean	< LOQ	< LOQ	12.0 ± 1.22	< LOQ	< LOQ	< LOQ	< LOQ	79.4 ± 36.0
	Range	< LOQ - < LOQ	< LOQ - < LOQ	9.81 – 14.0	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 116	< LOQ - < LOQ	
21	Mean	< LOQ	< LOQ	6.97 ± 3.85	< LOQ	< LOQ	< LOQ	7.30 ± 3.68	17.1 ± 8.67
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 13.4	< LOQ – 8.51	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 12.2	
22	Mean	< LOQ	12.5 ± 6.18	< LOQ	22.8 ± 8.47	< LOQ	< LOQ	87.0 ± 36.8	122 ± 51.1
	Range	< LOQ - < LOQ	< LOQ – 19.2	< LOQ - < LOQ	7.31 – 36.5	< LOQ - < LOQ	< LOQ - < LOQ	18.6 – 144	
23	Mean	< LOQ	< LOQ	6.75 ± 1.44	< LOQ	< LOQ	< LOQ	< LOQ	43.2 ± 18.9
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 7.81	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 97.7	< LOQ - < LOQ	
24	Mean	< LOQ	< LOQ	8.46 ± 0.393	< LOQ	< LOQ	79.4 ± 70.8	< LOQ	87.8 ± 40.5
	Range	< LOQ - < LOQ	< LOQ - < LOQ	7.95 – 9.23	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 136	< LOQ - < LOQ	
25	Mean	< LOQ	< LOQ	6.53 ± 5.62	< LOQ	< LOQ	< LOQ	9.99 ± 1.01	19.4 ± 0.424
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 10.7	< LOQ – 8.70	< LOQ - < LOQ	< LOQ - < LOQ	8.06 – 11.5	
26	Mean	< LOQ	< LOQ	15.5 ± 2.31	< LOQ	< LOQ	85.5 ± 45.8	6.98 ± 3.41	110 ± 51.9
	Range	< LOQ - < LOQ	< LOQ - < LOQ	11.0 – 18.8	< LOQ – 5.89	< LOQ - < LOQ	< LOQ – 157	< LOQ – 11.2	

Table A5. pH and electrical conductivity (EC) of the water samples, and organic carbon content (TOC), clay content and cation exchange capacity (CEC) of the sediment samples collected at the different sampling sites. Sampling sites are numbered according to Table A1.

Site no.	Water		Sediment	
	pH	EC (µs/cm)	TOC (%)	Clay content (%)	CEC (meq/100 g)
1	7.29	573	6.99	4.44	29.9
2	7.64	702	2.35	2.52	22.4
3	7.61	722	2.18	3.26	24.9
4	7.58	709	5.67	5.68	40.6
5	7.10	485	2.80	0.985	23.9
6	7.56	11940	1.66	0.893	34.1
7	7.62	965	1.98	3.96	18.6
8	7.28	798	4.25	5.68	34.1
9	7.23	515	1.56	1.79	6.43
10	6.97	643	2.83	3.05	24.8
11	6.90	677	5.64	5.34	35.7
12	6.95	642	1.84	2.57	20.7
13	7.57	716	1.54	0.913	21.4
14	6.91	850	6.81	2.90	24.3
15	6.92	363	3.32	0.553	8.83
16	7.82	906	0.864	0.679	13.5
17	6.71	9873	0.392	0	18.5
18	7.74	737	1.99	3.59	21.9
19	7.84	13390	2.76	2.49	46.1
20	7.75	676	2.21	2.58	24.8
21	7.44	685	2.93	4.40	24.3
22	7.66	1193	4.83	4.13	33.0
23	7.75	780	0.411	0.289	18.9
24	6.92	522	0.189	0	9.95
25	7.72	729	5.07	5.03	32.5
26	7.06	511	0.36	0	17.2

Table A6. Mean ∑PFAS concentrations (ng/g dw; ± SE) and concentrations (mean (± SE) and range) of individual PFAS (ng/g dw) in sediment at the different sampling sites. Sampling sites are numbered according to Table A1. Values < LOQ were substituted according to the MLE method in the calculation of mean concentrations of the individual compounds. In the calculation of ∑PFAS concentrations, values < LOQ were substituted by 0. N = 3 per site, except for site 4 where N = 6.

Site no.		PFBA	PFPeA	PFHxA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA	PFTeDA	PFOS	6:2 FTS	∑PFAS
1	Mean	< LOQ	< LOQ	0.850 ± 0.462	0.914 ± 0.152	< LOQ	1.28 ± 0.078	0.523 ± 0.058	3.40 ± 0.252	0.854 ± 0.082	2.05 ± 0.031	0.732 ± 0.076	< LOQ	10.6 ± 0.867
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 1.60	0.644 – 1.17	< LOQ - < LOQ	1.15 – 1.42	0.439 – 0.634	3.10 – 3.91	0.771 – 1.02	2.00 – 2.10	0.583 – 0.828	< LOQ - < LOQ	
2	Mean	< LOQ	< LOQ	< LOQ	0.814 ± 0.026	< LOQ	0.940 ± 0.122	0.358 ± 0.041	1.84 ± 0.034	0.515 ± 0.024	< LOQ	0.658 ± 0.072	< LOQ	5.80 ± 0.446
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 1.09	0.765 – 0.852	< LOQ - < LOQ	0.796 – 1.18	0.279 – 0.418	1.78 – 1.88	0.468 – 0.539	< LOQ - < LOQ	0.549 – 0.794	< LOQ - < LOQ	
3	Mean	< LOQ	< LOQ	< LOQ	0.579 ± 0.234	< LOQ	0.750 ± 0.101	0.448 ± 0.086	2.07 ± 0.191	0.588 ± 0.033	< LOQ	0.542 ± 0.020	< LOQ	4.98 ± 0.511
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	0.312 – 1.05	< LOQ - < LOQ	0.549 – 0.862	0.342 – 0.619	1.69 – 2.28	0.532 – 0.646	< LOQ - < LOQ	0.508 – 0.578	< LOQ - < LOQ	
4	Mean	< LOQ	0.237 ± 0.114	0.970 ± 0.312	1.42 ± 0.369	0.252 ± 0.099	1.42 ± 0.307	0.899 ± 0.182	4.00 ± 1.00	1.54 ± 0.475	1.44 ± 0.592	3.34 ± 1.23	< LOQ	15.3 ± 4.43
	Range	< LOQ - < LOQ	< LOQ – 0.694	< LOQ – 1.74	0.605 – 3.16	< LOQ – 0.627	0.657 – 2.63	0.468 – 1.42	1.03 – 6.71	< LOQ – 2.73	< LOQ – 3.29	< LOQ – 6.48	< LOQ - < LOQ	
5	Mean	< LOQ	< LOQ	< LOQ	0.789 ± 0.120	< LOQ	0.550 ± 0.040	0.338 ± 0.059	0.720 ± 0.059	< LOQ	< LOQ	< LOQ	< LOQ	2.45 ± 0.118
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	0.561 – 0.968	< LOQ - < LOQ	0.512 – 0.634	0.225 – 0.421	0.602 – 0.798	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 0.145	< LOQ - < LOQ	
6	Mean	1.58 ± 0.436	< LOQ	< LOQ	0.739 ± 0.151	< LOQ	0.710 ± 0.074	0.220 ± 0.053	0.970 ± 0.094	< LOQ	< LOQ	< LOQ	< LOQ	4.47 ± 1.03
	Range	0.863 – 2.37	< LOQ - < LOQ	< LOQ – 0.889	0.534 – 1.03	< LOQ - < LOQ	0.609 – 0.852	< LOQ – 0.299	0.815 – 1.14	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	
7	Mean	< LOQ	< LOQ	< LOQ	0.691 ± 0.037	< LOQ	0.820 ± 0.065	0.351 ± 0.018	1.85 ± 0.110	< LOQ	< LOQ	0.314 ± 0.135	< LOQ	4.56 ± 0.484
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 1.03	0.617 – 0.731	< LOQ - < LOQ	0.738 – 0.952	0.322 – 0.383	1.66 – 2.03	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 0.559	< LOQ - < LOQ	
8	Mean	< LOQ	< LOQ	< LOQ	1.17 ± 0.279	< LOQ	1.15 ± 0.047	0.589 ± 0.026	1.77 ± 0.279	0.716 ± 0.050	< LOQ	0.388 ± 0.188	< LOQ	5.78 ± 0.235
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	0.753 – 1.70	< LOQ - < LOQ	1.06 – 1.22	0.552 – 0.638	1.33 – 2.29	0.631 – 0.802	< LOQ - < LOQ	0.194 – 0.763	< LOQ - < LOQ	
9	Mean	< LOQ	< LOQ	< LOQ	0.642 ± 0.246	< LOQ	0.540 ± 0.090	0.282 ± 0.034	< LOQ	< LOQ	< LOQ	0.198 ± 0.046	< LOQ	2.10 ± 0.598
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	0.154 – 0.945	< LOQ - < LOQ	0.399 – 0.708	0.216 – 0.326	< LOQ – 0.793	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 0.246	< LOQ - < LOQ	
10	Mean	< LOQ	< LOQ	< LOQ	1.03 ± 0.153	< LOQ	0.960 ± 0.089	< LOQ	1.26 ± 0.078	< LOQ	< LOQ	0.155 ± 0.077	< LOQ	3.69 ± 0.553
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 0.881	0.864 – 1.34	< LOQ - < LOQ	0.787 – 1.09	< LOQ - < LOQ	1.15 – 1.41	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 0.267	< LOQ - < LOQ	
11	Mean	< LOQ	< LOQ	1.07 ± 0.106	1.40 ± 0.212	< LOQ	0.900 ± 0.204	0.612 ± 0.073	3.65 ± 0.227	1.98 ± 0.298	2.35 ± 0.396	1.13 ± 0.070	< LOQ	13.1 ± 1.11
	Range	< LOQ - < LOQ	< LOQ - < LOQ	0.924 – 1.28	1.08 – 1.80	< LOQ - < LOQ	0.498 – 1.13	0.494 – 0.744	3.30 – 4.08	1.55 – 2.55	1.56 – 2.77	1.03 – 1.27	< LOQ - < LOQ	
12	Mean	< LOQ	< LOQ	< LOQ	0.781 ± 0.128	< LOQ	0.740 ± 0.074	0.291 ± 0.030	1.33 ± 0.181	0.370 ± 0.169	< LOQ	0.247 ± 0.120	1.70 ± 1.31	6.05 ± 0.735
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 1.03	0.599 – 1.03	< LOQ - < LOQ	0.669 – 0.890	0.249 – 0.349	1.05 – 1.67	< LOQ – 0.579	< LOQ - < LOQ	< LOQ – 0.395	< LOQ – 4.28	
13	Mean	< LOQ	< LOQ	< LOQ	0.850 ± 0.283	< LOQ	0.720 ± 0.0509	0.487 ± 0.130	1.62 ± 0.276	< LOQ	< LOQ	0.658 ± 0.205	< LOQ	4.62 ± 1.01
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 0.848	0.343 – 1.32	< LOQ - < LOQ	0.660 – 0.823	0.284 – 0.729	1.08 – 1.97	< LOQ - < LOQ	< LOQ - < LOQ	0.264 – 0.953	< LOQ - < LOQ	
14	Mean	< LOQ	< LOQ	< LOQ	1.61 ± 0.495	< LOQ	1.16 ± 0.224	0.572 ± 0.061	3.52 ± 0.469	1.01 ± 0.108	2.78 ± 0.354	1.75 ± 0.193	< LOQ	12.9 ± 2.10
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 1.47	0.618 – 2.14	< LOQ - < LOQ	0.830 – 1.59	0.450 – 0.641	2.59 – 4.10	0.803 – 1.17	2.07 – 3.14	1.47 – 2.12	< LOQ - < LOQ	
15	Mean	< LOQ	< LOQ	< LOQ	0.555 ± 0.084	< LOQ	0.650 ± 0.125	0.265 ± 0.123	0.790 ± 0.394	< LOQ	< LOQ	0.262 ± 0.055	< LOQ	2.78 ± 0.665
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	0.434 – 0.716	< LOQ – 0.322	0.547 – 0.900	< LOQ – 0.398	< LOQ – 1.27	< LOQ – 0.484	< LOQ - < LOQ	0.161 – 0.348	< LOQ - < LOQ	
16	Mean	< LOQ	< LOQ	< LOQ	0.677 ± 0.073	< LOQ	0.610 ± 0.142	0.284 ± 0.132	< LOQ	< LOQ	< LOQ	< LOQ	4.79 ± 4.08	7.18 ± 4.70
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 0.894	0.552 – 0.806	< LOQ - < LOQ	0.324 – 0.768	< LOQ – 0.442	< LOQ – 0.855	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 12.9	

17	Mean	< LOQ	< LOQ	< LOQ	0.622 ± 0.090	< LOQ	0.470 ± 0.069	0.361 ± 0.023	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	2.21 ± 0.744
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 1.45	0.460 – 0.772	< LOQ - < LOQ	0.329 – 0.547	0.328 – 0.405	< LOQ – 0.837	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	
18	Mean	< LOQ	< LOQ	< LOQ	0.351 ± 0.316	< LOQ	< LOQ	< LOQ	< LOQ	0.751 ± 0.369	< LOQ	0.470 ± 0.039	< LOQ	1.54 ± 0.663
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 0.982	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 1.24	< LOQ - < LOQ	0.400 – 0.535	< LOQ - < LOQ	
19	Mean	< LOQ	< LOQ	< LOQ	0.827 ± 0.290	< LOQ	0.730 ± 0.030	0.267 ± 0.025	0.630 ± 0.327	< LOQ	< LOQ	< LOQ	< LOQ	2.75 ± 0.395
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 0.916	0.264 – 1.23	< LOQ - < LOQ	0.681 – 0.785	0.224 – 0.309	< LOQ – 1.17	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	
20	Mean	< LOQ	< LOQ	< LOQ	0.935 ± 0.153	< LOQ	0.710 ± 0.084	0.360 ± 0.030	2.05 ± 0.406	0.616 ± 0.065	< LOQ	0.831 ± 0.090	< LOQ	6.21 ± 0.817
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 1.07	0.605 – 1.34	< LOQ - < LOQ	0.618 – 0.880	0.301 – 0.402	1.29 – 2.68	0.496 – 0.719	< LOQ - < LOQ	0.675 – 0.987	< LOQ - < LOQ	
21	Mean	< LOQ	< LOQ	0.885 ± 0.0439	0.942 ± 0.153	0.136 ± 0.0513	1.01 ± 0.162	0.470 ± 0.030	1.81 ± 0.204	0.742 ± 0.031	< LOQ	2.81 ± 0.090	< LOQ	9.83 ± 0.733
	Range	< LOQ - < LOQ	< LOQ - < LOQ	0.797 – 0.933	0.664 – 1.19	< LOQ – 0.189	0.711 – 1.27	0.419 – 0.522	1.41 – 2.04	0.709 – 0.803	< LOQ – 1.73	2.66 – 2.97	< LOQ - < LOQ	
22	Mean	< LOQ	< LOQ	< LOQ	1.34 ± 0.158	< LOQ	0.860 ± 0.440	< LOQ	1.48 ± 1.43	2.40 ± 0.334	< LOQ	4.42 ± 0.762	< LOQ	10.4 ± 2.54
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	1.16 – 1.66	< LOQ - < LOQ	< LOQ – 1.59	< LOQ - < LOQ	< LOQ – 4.33	1.77 – 2.90	< LOQ - < LOQ	2.90 – 5.32	< LOQ - < LOQ	
23	Mean	< LOQ	< LOQ	< LOQ	0.661 ± 0.085	< LOQ	0.75 ± 0.079	0.375 ± 0.068	1.31 ± 0.240	< LOQ	< LOQ	0.160 ± 0.131	8.61 ± 4.35	12.3 ± 3.66
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 0.778	0.541 – 0.826	< LOQ - < LOQ	0.632 – 0.900	0.293 – 0.510	1.04 – 1.79	< LOQ – 0.609	< LOQ - < LOQ	< LOQ – 0.421	< LOQ – 14.1	
24	Mean	< LOQ	< LOQ	< LOQ	0.662 ± 0.186	< LOQ	0.550 ± 0.097	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	0.628 ± 0.578	2.52 ± 0.895
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 0.789	0.369 – 1.01	< LOQ - < LOQ	0.357 – 0.672	< LOQ – 0.262	< LOQ – 0.851	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 1.78	
25	Mean	< LOQ	< LOQ	< LOQ	1.37 ± 0.239	< LOQ	1.91 ± 0.131	1.80 ± 0.401	6.02 ± 0.860	9.47 ± 1.64	< LOQ	4.48 ± 0.895	< LOQ	25.1 ± 3.78
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	1.11 – 1.85	< LOQ - < LOQ	1.72 – 2.16	< LOQ – 2.21	4.76 – 7.66	6.36 – 11.9	< LOQ - < LOQ	2.71 – 5.55	< LOQ - < LOQ	
26	Mean	< LOQ	< LOQ	< LOQ	0.544 ± 0.037	< LOQ	0.620 ± 0.043	< LOQ	0.748 ± 0.169	< LOQ	< LOQ	< LOQ	3.25 ± 2.40	5.21 ± 2.34
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	0.491 – 0.615	< LOQ - < LOQ	0.534 – 0.667	< LOQ – 0.230	0.573 – 1.09	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 7.96	

Table A7. Pearson correlations (p-values and r values) between PFAS concentrations in sediment and TOC, CEC and clay content. R values of significant correlations are displayed in bold.

		TOC	CEC	Clay content
PFHxA	p	0.007	0.026	0.037
	r	**0.300**	**0.247**	**0.232**
PFOA	p	< 0.001	< 0.001	< 0.001
	r	**0.526**	**0.446**	**0.456**
PFDA	p	< 0.001	< 0.001	< 0.001
	r	**0.579**	**0.429**	**0.530**
PFUnDA	p	< 0.001	< 0.001	< 0.001
	r	**0.475**	**0.359**	**0.459**
PFDoDA	P	< 0.001	< 0.001	< 0.001
	r	**0.669**	**0.449**	**0.599**
PFTrDA	P	< 0.001	0.003	< 0.001
	r	**0.438**	**0.321**	**0.446**
PFOS	P	< 0.001	< 0.001	< 0.001
	r	**0.563**	**0.395**	**0.536**
∑PFAS	P	< 0.001	< 0.001	< 0.001
	r	**0.543**	**0.381**	**0.464**

Table A8. Mean ∑PFAS concentrations (ng/g ww; ± SE) and concentrations (mean (± SE) and range) of individual PFAS (ng/g ww) in hepatopancreas at the different sampling sites. Sampling sites are numbered according to Table A1. The number of replicates varied per site and are shown in Table A1. Values < LOQ were substituted according to the MLE method in the calculation of mean concentrations of the individual compounds. In the calculation of ∑PFAS concentrations, values < LOQ were substituted by 0. Recoveries for PFHxA were too low to quantify these concentrations. [a]The mean value represents a single value, since N = 1. No crabs were caught at sites 17, 24 and 26.

Site no.		PFBA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA	PFTeDA	PFBS	PFPeS	PFOS	6:2 FTS	8:2 FTS	FBSA	NaDONA	∑PFAS
1	Mean	< LOQ	0.208 ± 0.126	< LOQ	1.17 ± 0.244	0.975 ± 0.147	19.3 ± 4.14	5.26 ± 2.69	12.3 ± 4.15	< LOQ	< LOQ	2.20 ± 0.661	< LOQ	< LOQ	< LOQ	< LOQ	41.4 ± 9.73
	Range	< LOQ - < LOQ	< LOQ – 0.399	< LOQ - < LOQ	0.759 – 1.58	0.750 – 1.20	12.3 – 26.2	3.57 – 12.8	11.8 – 24.7	< LOQ - < LOQ	< LOQ - < LOQ	1.07 – 3.33	< LOQ – 0.948	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 0.264	
2	Mean	< LOQ	0.400 ± 0.195	< LOQ	2.38 ± 0.795	3.66 ± 1.04	61.0 ± 19.2	25.7 ± 6.71	29.1 ± 4.83	< LOQ	0.718 ± 0.704	6.88 ± 2.89	< LOQ	< LOQ	< LOQ	< LOQ	130 ± 34.5
	Range	< LOQ - < LOQ	< LOQ – 6.10	< LOQ - < LOQ	1.16 – 3.87	1.67 – 5.16	23.9 – 88.3	14.3 – 37.6	20.1 – 36.5	< LOQ - < LOQ	< LOQ – 2.13	2.23 – 12.2	< LOQ – 1.59	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 0.141	
3	Mean	< LOQ	0.593 ± 0.020	< LOQ	2.23 ± 0.580	2.49 ± 0.622	43.7 ± 5.36	16.3 ± 0.447	61.2 ± 4.77	< LOQ	< LOQ	5.41 ± 1.06	< LOQ	< LOQ	< LOQ	0.261 ± 0.244	132 ± 8.54
	Range	< LOQ - < LOQ	0.573 – 0.613	< LOQ - < LOQ	1.65 – 2.81	1.87 – 3.11	38.3 – 49.0	15.8 – 16.7	56.5 – 66.0	< LOQ - < LOQ	< LOQ - < LOQ	4.35 – 6.47	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 0.505	
4	Mean	< LOQ	0.556 ± 0.076	< LOQ	3.35 ± 0.572	4.32 ± 0.808	59.8 ± 9.73	23.8 ± 2.96	30.7 ± 3.73	< LOQ	< LOQ	9.86 ± 2.38	< LOQ	< LOQ	< LOQ	< LOQ	133 ± 19.0
	Range	< LOQ - < LOQ	0.224 – 1.10	< LOQ – 0.371	1.33 – 8.60	1.18 – 11.5	24.4 – 149	12.3 – 44.7	12.4 – 50.2	< LOQ - < LOQ	< LOQ – 1.20	1.31 – 28.2	< LOQ – 2.67	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 0.306	
5	Mean	< LOQ	0.556 ± 0.076	0.198 ± 0.027	3.63 ± 0.555	3.35 ± 0.617	26.3 ± 6.93	16.5 ± 5.25	7.17 ± 1.24	< LOQ	< LOQ	2.72 ± 0.399	< LOQ	1.71 ± 0.252	< LOQ	< LOQ	60.1 ± 14.4
	Range	< LOQ - < LOQ	0.295 – 0.917	< LOQ – 0.280	2.59 – 5.05	2.35 – 5.08	10.7 – 44.3	4.47 – 29.6	< LOQ – 9.31	< LOQ - < LOQ	< LOQ - < LOQ	2.14 – 3.90	< LOQ - < LOQ	< LOQ – 2.47	< LOQ - < LOQ	< LOQ - < LOQ	
6	Mean	< LOQ	0.691 ± 0.039	< LOQ	1.53 ± 0.294	1.12 ± 0.090	21.6 ± 1.27	12.2 ± 2.05	23.8 ± 4.31	< LOQ	1.99 ± 1.93	6.21 ± 0.791	0.762 ± 0.429	< LOQ	16.6 ± 11.1	0.294 ± 0.138	86.7 ± 9.83
	Range	< LOQ - < LOQ	0.542 – 0.772	< LOQ - < LOQ	1.03 – 2.60	0.864 – 1.36	16.9 – 24.0	8.63 – 20.1	12.5 – 34.1	< LOQ - < LOQ	< LOQ – 9.72	4.59 – 9.14	< LOQ – 1.89	< LOQ - < LOQ	< LOQ – 60.2	< LOQ – 0.831	
7	Mean	0.227 ± 0.086	0.848 ± 0.086	< LOQ	1.86 ± 0.289	1.52 ± 0.393	42.3 ± 4.97	9.32 ± 0.916	65.5 ± 6.99	< LOQ	0.966 ± 0.537	7.41 ± 2.15	< LOQ	< LOQ	< LOQ	0.157 ± 0.129	130 ± 9.76
	Range	< LOQ – 0.430	0.702 – 1.18	< LOQ – 0.291	1.32 – 2.59	0.802 – 2.65	29.2 – 60.0	6.38 – 11.3	48.0 – 86.2	< LOQ - < LOQ	< LOQ – 2.97	1.82 – 14.2	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 0.673	
8	Mean	< LOQ	0.435 ± 0.150	< LOQ	5.84 ± 0.320	5.32 ± 0.846	26.6 ± 6.81	13.7 ± 4.45	8.06 ± 4.93	< LOQ	< LOQ	6.50 ± 0.401	< LOQ	< LOQ	< LOQ	< LOQ	66.6 ± 15.5
	Range	< LOQ - < LOQ	< LOQ – 0.802	< LOQ – 0.534	4.66 – 6.55	3.28 – 8.23	18.6 – 53.7	5.83 – 29.4	< LOQ – 20.3	< LOQ - < LOQ	< LOQ - < LOQ	5.04 – 7.22	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 0.295	
9	Mean	0.206 ± 0.120	0.779 ± 0.224	< LOQ	4.48 ± 0.673	3.61 ± 0.731	66.6 ± 11.0	44.5 ± 10.7	118 ± 28.1	69.1 ± 63.2	< LOQ	15.3 ± 3.05	< LOQ	< LOQ	1.71 ± 0.470	0.191 ± 0.167	327 ± 70.4
	Range	< LOQ – 0.614	< LOQ – 1.46	< LOQ – 0.360	3.00 – 6.86	2.06 – 5.72	41.7 – 110	27.9 – 86.1	81.1 – 228	< LOQ – 321	< LOQ - < LOQ	7.44 – 23.8	< LOQ - < LOQ	< LOQ – 5.11	< LOQ – 2.78	< LOQ – 0.859	
10	Mean	< LOQ	0.647 ± 0.194	< LOQ	1.15 ± 0.170	0.807 ± 0.082	11.8 ± 1.76	4.08 ± 0.423	10.5 ± 1.22	< LOQ	< LOQ	2.18 ± 0.464	1.35 ± 0.803	< LOQ	< LOQ	< LOQ	32.5 ± 2.70
	Range	< LOQ - < LOQ	< LOQ – 0.920	< LOQ - < LOQ	0.697 – 1.47	0.638 – 1.03	7.43 – 15.9	3.07 – 5.13	7.09 – 12.9	< LOQ - < LOQ	< LOQ - < LOQ	1.24 – 3.45	< LOQ – 3.33	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 0.275	
11	Mean	< LOQ	0.593 ± 0.082	< LOQ	1.59 ± 0.181	3.79 ± 0.443	30.0 ± 2.93	19.0 ± 2.32	51.1 ± 8.21	< LOQ	< LOQ	1.92 ± 0.297	< LOQ	< LOQ	0.221 ± 0.183	< LOQ	109 ± 12.2
	Range	< LOQ - < LOQ	0.386 – 0.648	< LOQ - < LOQ	0.951 – 2.05	2.47 – 4.95	22.9 – 40.0	11.5 – 25.1	35.1 – 74.5	< LOQ - < LOQ	< LOQ – 0.656	1.02 – 2.82	< LOQ – 2.24	< LOQ - < LOQ	< LOQ – 0.954	< LOQ – 0.291	
12	Mean	< LOQ	0.631 ± 0.305	< LOQ	2.51 ± 0.190	5.97 ± 0.369	47.1 ± 1.71	21.8 ± 1.58	35.6 ± 2.73	< LOQ	< LOQ	10.1 ± 1.09	< LOQ	< LOQ	0.934 ± 0.889	< LOQ	125 ± 5.47
	Range	< LOQ - < LOQ	< LOQ – 1.10	< LOQ - < LOQ	2.29 – 2.89	5.44 – 6.68	45.3 – 50.5	18.8 – 24.2	30.2 – 39.2	< LOQ - < LOQ	< LOQ - < LOQ	8.22 – 12.0	< LOQ – 1.01	< LOQ - < LOQ	< LOQ – 2.71	< LOQ – 0.195	
13	Mean	< LOQ	0.922 ± 0.170	< LOQ	5.45 ± 1.53	3.38 ± 0.692	37.1 ± 10.4	21.3 ± 10.3	27.4 ± 11.0	7.49 ± 5.75	< LOQ	5.03 ± 0.807	4.00 ± 3.94	< LOQ	< LOQ	0.405 ± 0.209	112 ± 33.6
	Range	< LOQ - < LOQ	0.437 – 1.45	< LOQ – 0.466	1.42 – 9.44	1.68 – 5.39	18.3 – 75.5	2.53 – 58.7	7.66 – 69.4	< LOQ – 29.8	< LOQ - < LOQ	2.86 – 7.02	< LOQ – 19.8	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 1.22	
14	Mean	0.489 ± 0.359	0.249 ± 0.213	< LOQ	0.930 ± 0.313	1.19 ± 0.208	21.2 ± 1.58	9.01 ± 2.08	36.3 ± 11.8	< LOQ	< LOQ	3.97 ± 1.30	< LOQ	< LOQ	< LOQ	0.141 ± 0.118	73.4 ± 13.2
	Range	0.130 – 0.849	< LOQ – 0.462	< LOQ - < LOQ	0.617 – 1.24	0.982 – 1.40	19.6 – 22.8	6.93 – 11.1	24.4 – 48.1	< LOQ - < LOQ	< LOQ - < LOQ	2.68 – 5.26	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 0.259	
15	Mean	< LOQ	0.683 ± 0.177	< LOQ	1.25 ± 0.057	0.789 ± 0.430	8.29 ± 0.356	3.09 ± 0.334	8.47 ± 8.45	< LOQ	< LOQ	3.82 ± 1.19	< LOQ	< LOQ	< LOQ	0.328 ± 0.290	26.7 ± 6.75
	Range	< LOQ - < LOQ	0.506 – 0.860	< LOQ - < LOQ	1.19 – 1.30	0.360 – 1.22	7.94 – 8.65	2.76 – 3.43	< LOQ – 16.9	< LOQ - < LOQ	< LOQ - < LOQ	2.63 – 5.00	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 0.618	
16[a]	Mean	< LOQ	0.862	0.287	1.63	1.65	41.5	14.6	31.9	< LOQ	< LOQ	1.04	< LOQ	< LOQ	0.366	< LOQ	93.8
18	Mean	< LOQ	0.911 ± 0.098	< LOQ	2.57 ± 0.202	2.08 ± 0.354	28.8 ± 4.56	23.7 ± 7.67	28.2 ± 3.79	275 ± 217	< LOQ	5.06 ± 1.06	0.903 ± 0.834	< LOQ	< LOQ	< LOQ	367 ± 230
	Range	< LOQ - < LOQ	0.725 – 1.06	< LOQ - < LOQ	2.17 – 2.78	1.65 – 2.79	20.4 – 36.2	11.2 – 37.7	20.6 – 32.6	< LOQ – 702	< LOQ - < LOQ	2.95 – 6.35	< LOQ – 2.57	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 0.142	
19	Mean	0.525 ± 0.338	2.20 ± 0.407	0.613 ± 0.177	3.33 ± 0.473	3.34 ± 0.413	32.9 ± 2.54	19.5 ± 2.74	23.6 ± 6.21	7.29 ± 3.36	< LOQ	13.2 ± 2.78	6.70 ± 5.65	< LOQ	14.7 ± 2.75	1.90 ± 0.997	127 ± 9.65
	Range	< LOQ – 3.32	0.603 – 4.88	< LOQ – 1.89	1.55 – 5.53	1.49 – 5.21	14.1 – 41.4	6.59 – 30.9	< LOQ – 48.0	< LOQ – 30.3	< LOQ - < LOQ	4.80 – 28.8	< LOQ – 57.3	< LOQ – 1.61	2.59 – 29.9	0.117 – 10.5	
20	Mean	< LOQ	0.737 ± 0.156	0.301 ± 0.115	2.25 ± 0.570	1.71 ± 0.434	24.5 ± 1.92	18.8 ± 2.47	30.2 ± 2.80	83.4 ± 83.3	< LOQ	12.2 ± 7.04	< LOQ	< LOQ	< LOQ	0.159 ± 0.104	174 ± 80.0

	Range	< LOQ - < LOQ	0.431 – 0.937	< LOQ – 0.451	1.61 – 3.38	1.25 – 2.58	21.7 – 28.1	14.1 – 22.4	25.1 – 34.8	< LOQ – 250	< LOQ - < LOQ	3.68 – 26.1	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 0.368	
21[a]	Mean	< LOQ	1.19	< LOQ	3.75	1.95	28.5	14.1	3.79	< LOQ	< LOQ	15.5	< LOQ	< LOQ	1.72	< LOQ	70.4
22	Mean	< LOQ	1.35 ± 0.290	0.445 ± 0.344	4.73 ± 0.521	4.69 ± 0.648	50.9 ± 6.39	35.3 ± 3.60	41.4 ± 4.74	< LOQ	< LOQ	79.7 ± 34.8	< LOQ	< LOQ	1.54 ± 0.078	< LOQ	220 ± 50.9
	Range	< LOQ - < LOQ	1.06 – 1.64	< LOQ – 0.789	4.21 – 5.26	4.04 – 5.34	44.5 – 57.3	31.7 – 38.9	36.7 – 46.2	< LOQ - < LOQ	< LOQ - < LOQ	44.9 – 114	< LOQ – 0.780	< LOQ - < LOQ	1.46 – 1.62	< LOQ - < LOQ	
23	Mean	0.162 ± 0.084	1.05 ± 0.250	< LOQ	2.48 ± 0.635	2.17 ± 0.70	20.8 ± 3.37	10.3 ± 1.84	23.6 ± 1.88	< LOQ	< LOQ	6.77 ± 1.91	< LOQ	< LOQ	4.66 ± 1.45	0.926 ± 0.523	73.1 ± 9.50
	Range	< LOQ – 0.307	0.458 – 1.96	< LOQ – 0.386	0.905 – 2.60	0.705 – 4.75	13.1 – 32.5	7.24 – 17.4	17.1 – 26.6	< LOQ - < LOQ	< LOQ – 0.735	2.24 – 13.3	< LOQ - < LOQ	< LOQ - < LOQ	1.14 – 9.57	< LOQ – 2.68	
25	Mean	< LOQ	0.740 ± 0.039	< LOQ	1.84 ± 0.279	3.04 ± 0.396	46.6 ± 4.66	32.7 ± 9.44	33.5 ± 13.5	< LOQ	< LOQ	8.03 ± 2.49	< LOQ	< LOQ	0.326 ± 0.224	< LOQ	127 ± 26.0
	Range	< LOQ - < LOQ	0.644 – 0.828	< LOQ - < LOQ	1.11 – 2.81	2.06 – 4.11	32.5 – 60.8	13.5 – 64.7	< LOQ – 76.3	< LOQ - < LOQ	< LOQ - < LOQ	3.23 – 17.0	< LOQ – 1.32	< LOQ - < LOQ	< LOQ – 1.22	< LOQ - < LOQ	

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Table A9. Mean ∑PFAS concentrations (ng/g ww; ± SE) and concentrations (mean (± SE) and range) of individual PFAS (ng/g ww) in muscle tissue at the different sampling sites. Sampling sites are numbered according to Table A1. The number of replicates varied per site and are shown in Table A1. Values < LOQ were substituted according to the MLE method in the calculation of mean concentrations of the individual compounds. In the calculation of ∑PFAS concentrations, values < LOQ were substituted by 0. [a]The mean value represents a single value, since N = 1. The analysis failed for four crabs of site 4, resulting in N = 9 at this site, and both crabs of site 14. Hence, site 14 was omitted from this Table. No crabs were caught at sites 17, 24 and 26.

Sampling site no.		PFBA	PFHxA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA	PFTeDA	PFBS	PFOS	6:2 FTS	FBSA	∑PFAS
1	Mean	< LOQ	< LOQ	0.676 ± 0.475	< LOQ	< LOQ	< LOQ	5.28 ± 2.64	4.06 ± 2.03	6.68 ± 6.67	< LOQ	< LOQ	< LOQ	< LOQ	17.6 ± 11.0
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 1.60	< LOQ - < LOQ	< LOQ – 0.555	< LOQ – 0.346	< LOQ – 8.14	< LOQ – 6.44	< LOQ – 20.0	< LOQ - < LOQ	< LOQ – 0.906	< LOQ - < LOQ	< LOQ – 0.916	
2	Mean	< LOQ	< LOQ	0.434 ± 0.418	< LOQ	0.748 ± 0.733	0.525 ± 0.509	16.3 ± 0.910	16.9 ± 1.65	21.4 ± 3.35	106 ± 63.3	2.16 ± 0.692	< LOQ	< LOQ	164 ± 67.4
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 1.27	< LOQ - < LOQ	< LOQ – 2.21	< LOQ – 1.54	14.9 – 18.0	14.3 – 20.0	16.1 – 27.6	18.1 – 228	1.28 – 3.52	< LOQ - < LOQ	< LOQ - < LOQ	
3	Mean	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	< LOQ	10.8 ± 0.518	9.71 ± 2.40	29.0 ± 14.9	31.4 ± 31.4	2.71 ± 0.342	< LOQ	< LOQ	83.6 ± 13.2
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	10.3 – 11.3	7.31 – 12.1	14.1 – 43.9	< LOQ – 62.8	2.37 – 3.05	< LOQ - < LOQ	< LOQ - < LOQ	
4	Mean	< LOQ	< LOQ	0.978 ± 0.508	< LOQ	1.12 ± 0.675	0.367 ± 0.237	33.0 ± 7.15	30.1 ± 8.23	33.6 ± 10.7	143 ± 69.7	4.54 ± 1.79	< LOQ	< LOQ	246 ± 91.6
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 4.27	< LOQ - < LOQ	< LOQ – 6.27	< LOQ – 1.71	13.8 – 78.6	< LOQ – 92.2	< LOQ – 95.6	< LOQ – 584	1.08 – 18.4	< LOQ - < LOQ	< LOQ – 0.975	
5	Mean	< LOQ	0.370 ± 0.085	0.457 ± 0.078	< LOQ	2.57 ± 0.444	2.33 ± 0.703	14.8 ± 6.15	12.5 ± 5.29	8.15 ± 3.15	< LOQ	1.66 ± 0.401	5.45 ± 0.946	< LOQ	44.5 ± 17.8
	Range	< LOQ - < LOQ	< LOQ – 0.623	0.237 – 0.593	< LOQ - < LOQ	1.26 – 3.15	0.776 – 3.57	4.34 – 29.0	3.12 – 25.3	2.47 – 15.7	< LOQ - < LOQ	< LOQ – 2.45	< LOQ – 8.28	< LOQ - < LOQ	
6	Mean	< LOQ	< LOQ	0.705 ± 0.109	< LOQ	0.728 ± 0.304	0.729 ± 0.199	8.06 ± 3.12	6.41 ± 2.90	< LOQ	< LOQ	2.26 ± 0.844	< LOQ	3.34 ± 1.18	25.6 ± 8.44
	Range	< LOQ - < LOQ	< LOQ - < LOQ	0.449 – 1.04	< LOQ - < LOQ	< LOQ – 1.86	< LOQ – 1.19	< LOQ – 17.7	< LOQ – 16.6	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 5.15	< LOQ – 6.64	1.24 – 7.48	
7	Mean	< LOQ	< LOQ	0.490 ± 0.110	< LOQ	0.859 ± 0.157	0.665 ± 0.390	19.6 ± 4.17	6.99 ± 1.45	30.1 ± 8.02	< LOQ	3.51 ± 1.28	< LOQ	< LOQ	63.5 ± 13.0
	Range	< LOQ - < LOQ	< LOQ - < LOQ	0.348 – 0.841	< LOQ - < LOQ	0.475 – 1.33	< LOQ – 2.11	11.5 – 34.2	4.20 – 10.7	11.0 – 59.5	< LOQ - < LOQ	< LOQ – 7.90	< LOQ – 6.20	< LOQ - < LOQ	
8	Mean	< LOQ	< LOQ	0.353 ± 0.223	< LOQ	0.445 ± 0.419	< LOQ	7.80 ± 2.00	8.31 ± 3.54	< LOQ	44.4 ± 44.4	1.80 ± 0.590	< LOQ	< LOQ	66.6 ± 39.5
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 1.15	< LOQ - < LOQ	< LOQ – 2.12	< LOQ - < LOQ	< LOQ – 10.9	< LOQ – 21.4	< LOQ – 10.4	< LOQ – 222	< LOQ – 3.51	< LOQ – 7.39	< LOQ - < LOQ	
9	Mean	< LOQ	< LOQ	1.57 ± 0.808	< LOQ	0.654 ± 0.381	0.337 ± 0.307	5.35 ± 3.52	14.0 ± 2.60	11.6 ± 7.24	38.5 ± 18.6	1.79 ± 0.825	50.8 ± 50.7	< LOQ	124 ± 51.6
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 4.42	< LOQ - < LOQ	< LOQ – 1.66	< LOQ – 1.57	< LOQ – 17.5	9.81 – 23.9	< LOQ – 33.7	< LOQ – 108	< LOQ – 4.18	< LOQ - 254	< LOQ - < LOQ	
10	Mean	< LOQ	< LOQ	0.440 ± 0.266	< LOQ	0.687 ± 0.426	< LOQ	9.29 ± 1.22	4.41 ± 0.467	< LOQ	< LOQ	1.38 ± 0.306	< LOQ	< LOQ	18.6 ± 2.95
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 1.16	< LOQ - < LOQ	< LOQ – 1.84	< LOQ – 0.376	6.50 – 11.9	3.39 – 5.47	< LOQ - < LOQ	< LOQ - < LOQ	0.793 – 2.24	< LOQ – 9.00	< LOQ – 0.428	
11	Mean	< LOQ	< LOQ	1.69 ± 1.24	< LOQ	1.17 ± 0.265	3.08 ± 0.615	30.1 ± 5.89	28.4 ± 6.37	44.1 ± 8.75	< LOQ	1.75 ± 0.222	< LOQ	< LOQ	114 ± 25.7
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 6.67	< LOQ – 0.795	0.554 – 2.12	1.69 – 5.24	18.8 – 48.6	17.3 – 48.0	27.0 – 73.3	< LOQ - < LOQ	1.29 – 2.58	< LOQ – 18.9	< LOQ - < LOQ	
12	Mean	< LOQ	< LOQ	1.08 ± 0.479	< LOQ	1.62 ± 0.082	3.86 ± 0.069	35.2 ± 2.31	25.3 ± 1.44	< LOQ	< LOQ	3.74 ± 0.596	< LOQ	1.36 ± 0.193	72.1 ± 3.28
	Range	< LOQ - < LOQ	< LOQ - < LOQ	0.569 – 2.04	< LOQ - < LOQ	1.50 – 1.78	3.72 – 3.94	30.5 – 37.7	23.4 – 28.2	< LOQ - < LOQ	< LOQ - < LOQ	3.15 – 4.94	< LOQ - < LOQ	0.976 – 1.58	
13	Mean	< LOQ	< LOQ	< LOQ	< LOQ	1.22 ± 0.502	< LOQ	10.4 ± 2.02	6.93 ± 2.51	< LOQ	< LOQ	1.85 ± 0.400	5.20 ± 5.15	0.402 ± 0.153	26.2 ± 7.38
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 0.450	< LOQ - < LOQ	< LOQ – 2.42	< LOQ – 0.737	5.32 – 17.0	0.831 – 4.6	< LOQ - < LOQ	< LOQ - < LOQ	0.940 – 3.09	< LOQ – 25.8	< LOQ – 0.830	
15	Mean	< LOQ	< LOQ	2.08 ± 1.49	< LOQ	0.476 ± 0.438	< LOQ	2.54 ± 0.671	0.753 ± 0.709	2.63 ± 2.61	< LOQ	< LOQ	< LOQ	< LOQ	8.43 ± 5.97
	Range	< LOQ - < LOQ	< LOQ - < LOQ	0.588 – 3.56	< LOQ - < LOQ	< LOQ – 0.914	< LOQ - < LOQ	1.87 – 3.21	< LOQ – 1.46	< LOQ – 5.25	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	
16[a]	Mean	< LOQ	< LOQ	0.765	< LOQ	1.15	0.593	12.5	7.84	18.7	12.1	0.711	< LOQ	< LOQ	54.3
18	Mean	< LOQ	< LOQ	0.213 ± 0.146	< LOQ	0.304 ± 0.230	0.878 ± 0.430	20.9 ± 3.50	17.5 ± 5.62	< LOQ	36.8 ± 8.77	4.37 ± 0.882	6.34 ± 6.27	< LOQ	87.1 ± 17.8
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 0.505	< LOQ - < LOQ	< LOQ – 0.764	< LOQ – 1.60	15.1 – 27.2	7.52 – 27.0	< LOQ - < LOQ	19.3 – 46.5	2.97 – 6.00	< LOQ – 18.9	< LOQ - < LOQ	
19	Mean	0.267 ± 0.152	< LOQ	0.791 ± 0.179	< LOQ	1.43 ± 0.378	1.24 ± 0.393	13.3 ± 2.79	11.3 ± 2.61	12.9 ± 5.17	< LOQ	4.08 ± 0.657	4.77 ± 1.49	9.87 ± 2.75	58.1 ± 11.8
	Range	< LOQ – 1.62	< LOQ – 0.831	< LOQ – 1.74	< LOQ - < LOQ	< LOQ – 3.30	< LOQ – 3.50	1.46 – 29.3	2.32 – 29.2	< LOQ – 43.8	< LOQ - < LOQ	1.32 – 7.63	< LOQ – 15.3	1.99 – 31.3	
20	Mean	< LOQ	< LOQ	1.18 ± 0.864	< LOQ	0.451 ± 0.361	1.28 ± 0.206	15.9 ± 2.21	13.4 ± 1.96	< LOQ	181 ± 157	3.41 ± 0.218	4.77 ± 4.70	1.06 ± 0.187	223 ± 158
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 2.88	< LOQ - < LOQ	< LOQ – 1.17	0.899 – 1.60	11.5 – 18.6	10.6 – 17.2	< LOQ - < LOQ	16.5 – 495	3.07 – 3.82	< LOQ – 14.2	0.699 – 1.32	
21[a]	Mean	< LOQ	< LOQ	0.864	< LOQ	2.13	1.46	11.7	8.66	16.2	6.22	2.49	< LOQ	< LOQ	49.8
22	Mean	< LOQ	< LOQ	0.791 ± 0.659	< LOQ	0.851 ± 0.698	0.946 ± 0.756	22.6 ± 7.10	24.2 ± 8.65	< LOQ	< LOQ	18.9 ± 10.7	< LOQ	0.671 ± 0.574	68.7 ± 29.5
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 1.45	< LOQ - < LOQ	< LOQ – 1.55	< LOQ – 1.70	15.5 – 29.7	15.5 – 32.8	< LOQ - < LOQ	< LOQ - < LOQ	8.17 – 29.7	< LOQ - < LOQ	< LOQ – 1.25	
23	Mean	< LOQ	< LOQ	0.596 ± 0.097	< LOQ	0.582 ± 0.143	< LOQ	4.84 ± 1.27	3.26 ± 0.479	5.29 ± 1.61	< LOQ	1.54 ± 0.245	< LOQ	1.78 ± 0.328	18.0 ± 3.35
	Range	< LOQ - < LOQ	< LOQ - < LOQ	0.426 – 0.954	< LOQ - < LOQ	< LOQ – 0.824	< LOQ – 0.771	< LOQ – 7.45	1.87 – 4.43	< LOQ – 8.75	< LOQ - < LOQ	1.01 – 2.46	< LOQ - < LOQ	0.997 – 2.58	
25	Mean	< LOQ	< LOQ	0.342 ± 0.151	< LOQ	0.547 ± 0.207	0.692 ± 0.287	18.1 ± 3.83	17.6 ± 5.83	< LOQ	2.53 ± 1.65	2.33 ± 0.625	< LOQ	< LOQ	44.3 ± 10.0
	Range	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 0.754	< LOQ - < LOQ	< LOQ – 1.22	< LOQ – 1.69	8.40 – 31.2	5.08 – 39.7	< LOQ - < LOQ	< LOQ – 8.37	0.949 – 4.65	< LOQ – 6.37	< LOQ - < LOQ	

Table A10. Mean ∑PFAS concentrations (ng/g ww; ± SE) and concentrations (mean (± SE) and range) of individual PFAS (ng/g ww) in carapace at the different sampling sites. Sampling sites are numbered according to Table A1. The number of replicates varied per site and are shown in Table A1. Values < LOQ were substituted according to the MLE method in the calculation of mean concentrations of the individual compounds. In the calculation of ∑PFAS concentrations, values < LOQ were substituted by 0. [a]The mean value represents a single value, since N = 1. No crabs were caught at sites 17, 24 and 26.

Sampling site no.		PFHxA	PFOA	PFNA	PFDA	PFUnDA	PFDoDA	PFTrDA	PFTeDA	PFBS	PFOS	6:2 FTS	FBSA	NaDONA	∑PFAS
1	Mean	< LOQ	0.167 ± 0.126	0.162 ± 0.098	0.867 ± 0.094	5.94 ± 2.35	5.94 ± 2.35	3.38 ± 0.451	5.39 ± 1.38	< LOQ	0.725 ± 0.345	18.1 ± 10.4	< LOQ	0.137 ± 0.072	37.5 ± 7.17
	Range	< LOQ - < LOQ	< LOQ – 0.425	< LOQ – 0.309	0.761 – 1.08	< LOQ – 1.03	2.10 – 9.78	2.60 – 4.16	3.01 – 7.78	< LOQ – 8.81	0.145 – 1.30	< LOQ – 36.1	< LOQ - < LOQ	< LOQ – 0.261	
2	Mean	< LOQ	0.484 ± 0.009	< LOQ	1.43 ± 0.254	1.48 ± 0.508	15.1 ± 5.89	9.28 ± 3.48	8.60 ± 2.77	97.8 ± 41.8	3.23 ± 1.85	< LOQ	< LOQ	0.097 ± 0.034	138 ± 28.6
	Range	< LOQ - < LOQ	0.466 – 0.493	< LOQ - < LOQ	1.01 – 1.89	0.510 – 2.22	3.72 – 23.4	2.52 – 14.1	3.40 – 12.9	54.8 – 181	0.982 – 6.90	< LOQ - < LOQ	< LOQ - < LOQ	0.047 – 0.162	
3	Mean	< LOQ	0.552 ± 0.037	< LOQ	0.866 ± 0.147	< LOQ	1.23 ± 0.394	0.450 ± 0.120	0.635 ± 0.622	123 ± 120	0.917 ± 0.148	< LOQ	< LOQ	< LOQ	128 ± 119
	Range	< LOQ – 0.741	0.514 – 0.589	< LOQ - < LOQ	0.719 – 1.01	< LOQ - < LOQ	0.832 – 1.62	0.330 – 0.570	< LOQ – 1.26	3.64 – 243	0.769 – 1.06	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	
4	Mean	< LOQ	0.407 ± 0.050	< LOQ	1.30 ± 0.170	1.40 ± 0.236	10.8 ± 2.57	9.48 ± 2.64	7.94 ± 2.27	109 ± 20.1	2.95 ± 0.608	< LOQ	< LOQ	0.136 ± 0.060	144 ± 20.6
	Range	< LOQ – 1.02	0.169 – 0.658	< LOQ – 1.35	0.480 – 2.49	0.272 – 2.63	0.858 – 24.9	< LOQ – 26.4	< LOQ – 25.1	31.6 – 253	0.409 – 8.03	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 0.746	
5	Mean	< LOQ	< LOQ	< LOQ	1.00 ± 0.313	0.700 ± 0.249	3.35 ± 1.55	3.71 ± 1.93	1.05 ± 0.934	< LOQ	0.192 ± 0.131	< LOQ	< LOQ	< LOQ	9.87 ± 4.71
	Range	< LOQ - < LOQ	< LOQ – 0.404	< LOQ - < LOQ	< LOQ – 1.50	< LOQ – 1.22	< LOQ – 6.79	< LOQ – 9.09	< LOQ – 3.85	< LOQ - < LOQ	< LOQ – 5.85	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ - < LOQ	
6	Mean	< LOQ	0.386 ± 0.179	< LOQ	0.720 ± 0.065	0.681 ± 0.080	2.05 ± 0.382	1.99 ± 0.383	1.40 ± 0.843	< LOQ	1.01 ± 0.273	28.5 ± 9.40	1.92 ± 1.86	0.233 ± 0.052	40.5 ± 8.66
	Range	< LOQ - < LOQ	< LOQ – 0.932	< LOQ - < LOQ	0.525 – 0.874	0.515 – 0.981	1.24 – 3.36	0.901 – 3.27	< LOQ – 4.00	< LOQ – 8.80	0.416 – 1.91	7.55 – 57.4	< LOQ – 9.34	0.123 – 0.376	
7	Mean	0.534 ± 0.142	0.431 ± 0.105	< LOQ	1.21 ± 0.238	0.652 ± 0.216	9.08 ± 3.03	2.51 ± 0.899	7.29 ± 3.07	3.77 ± 1.60	4.14 ± 1.42	5.51 ± 1.76	< LOQ	0.056 ± 0.019	35.2 ± 8.40
	Range	< LOQ – 0.885	0.205 – 0.778	< LOQ - < LOQ	0.785 – 2.08	< LOQ – 1.30	1.81 – 16.1	0.353 – 5.27	1.38 – 18.4	< LOQ – 9.64	0.815 – 7.95	1.16 – 10.1	< LOQ - < LOQ	< LOQ – 0.116	
8	Mean	< LOQ	0.344 ± 0.048	< LOQ	1.78 ± 0.192	1.22 ± 0.174	4.48 ± 1.19	3.14 ± 1.12	1.66 ± 0.575	9.54 ± 6.29	1.61 ± 0.285	6.09 ± 1.65	< LOQ	0.112 ± 0.014	30.2 ± 8.50
	Range	< LOQ – 1.11	0.262 – 0.523	< LOQ - < LOQ	1.25 – 2.19	0.818 – 1.83	2.44 – 8.98	1.39 – 7.58	< LOQ – 3.61	< LOQ – 34.4	0.986 – 2.31	< LOQ – 9.88	< LOQ - < LOQ	0.062 – 0.139	
9	Mean	0.661 ± 0.192	0.438 ± 0.038	< LOQ	1.32 ± 0.095	0.808 ± 0.125	7.61 ± 0.908	4.11 ± 0.585	7.53 ± 1.25	97.8 ± 23.1	2.06 ± 0.454	1.98 ± 0.779	< LOQ	0.177 ± 0.076	124 ± 23.4
	Range	< LOQ – 1.15	0.345 – 0.554	< LOQ – 0.143	0.984 – 1.51	0.397 – 1.14	4.90 – 9.87	2.07 – 5.24	3.73 – 10.8	44.1 – 156	1.14 – 3.22	< LOQ – 4.27	< LOQ - < LOQ	0.074 – 0.479	
10	Mean	< LOQ	0.187 ± 0.124	< LOQ	0.870 ± 0.106	0.169 ± 0.140	2.32 ± 1.16	1.36 ± 0.573	0.785 ± 0.761	16.2 ± 3.11	0.775 ± 0.314	< LOQ	< LOQ	0.168 ± 0.130	22.7 ± 3.91
	Range	< LOQ - < LOQ	< LOQ – 0.543	< LOQ - < LOQ	0.571 – 1.07	< LOQ – 0.588	< LOQ – 5.34	< LOQ – 2.64	< LOQ – 3.07	9.33 – 22.0	0.333 – 1.67	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 0.557	
11	Mean	0.628 ± 0.166	0.374 ± 0.081	< LOQ	0.903 ± 0.105	0.885 ± 0.176	4.59 ± 1.35	3.87 ± 1.16	4.57 ± 1.67	26.5 ± 17.8	0.636 ± 0.138	4.64 ± 1.55	< LOQ	0.056 ± 0.018	47.6 ± 17.7
	Range	< LOQ – 0.962	0.240 – 0.687	< LOQ - < LOQ	0.524 – 1.09	0.346 – 1.36	0.947 – 9.24	1.03 – 7.92	< LOQ – 9.76	4.35 – 97.3	0.229 – 1.03	0.497 – 8.80	< LOQ - < LOQ	< LOQ – 0.108	
12	Mean	< LOQ	0.241 ± 0.128	< LOQ	1.10 ± 0.585	1.06 ± 1.04	8.76 ± 8.00	12.3 ± 7.68	9.91 ± 9.89	14.1 ± 14.1	3.11 ± 2.70	< LOQ	< LOQ	0.119 ± 0.100	51.4 ± 44.4
	Range	< LOQ - < LOQ	< LOQ – 0.456	< LOQ - < LOQ	< LOQ – 2.01	< LOQ – 3.13	< LOQ – 24.7	1.24 – 27.1	< LOQ – 29.7	< LOQ – 42.2	< LOQ – 8.48	< LOQ - < LOQ	< LOQ – 2.32	< LOQ – 0.318	
13	Mean	< LOQ	0.418 ± 0.293	< LOQ	0.828 ± 0.482	0.427 ± 0.230	5.35 ± 1.84	4.66 ± 2.67	< LOQ	22.4 ± 11.9	0.735 ± 0.437	13.4 ± 6.02	< LOQ	0.114 ± 0.066	48.3 ± 7.39
	Range	< LOQ - < LOQ	< LOQ – 1.54	< LOQ - < LOQ	< LOQ – 2.08	< LOQ – 1.10	< LOQ – 11.3	< LOQ – 14.6	< LOQ - < LOQ	< LOQ – 67.2	< LOQ – 1.84	< LOQ – 30.4	< LOQ - < LOQ	< LOQ – 0.364	
14	Mean	0.525 ± 0.506	0.605 ± 0.031	< LOQ	1.09 ± 0.263	1.03 ± 0.255	11.0 ± 4.56	7.52 ± 4.23	14.0 ± 8.64	126 ± 77.8	2.56 ± 1.59	3.99 ± 1.96	< LOQ	0.259 ± 0.126	169 ± 55.6
	Range	< LOQ – 1.03	0.574 – 0.637	< LOQ - < LOQ	0.829 – 1.36	0.773 – 1.28	6.48 – 15.6	3.29 – 11.7	5.31 – 22.6	48.3 – 204	0.966 – 4.15	2.03 – 5.95	< LOQ - < LOQ	0.133 – 0.385	
15	Mean	< LOQ	0.412 ± 0.093	< LOQ	0.636 ± 0.134	0.620 ± 0.215	2.64 ± 0.034	1.30 ± 0.022	2.29 ± 0.673	< LOQ	0.704 ± 0.208	4.52 ± 1.29	< LOQ	0.085 ± 0.006	13.7 ± 0.824
	Range	< LOQ – 0.919	0.319 – 0.504	< LOQ - < LOQ	0.502 – 0.770	0.405 – 0.836	2.60 – 2.67	1.27 – 1.32	1.62 – 2.97	< LOQ - < LOQ	0.496 – 0.911	3.23 – 5.81	< LOQ - < LOQ	0.079 – 0.091	
16[a]	Mean	< LOQ	< LOQ	< LOQ	0.964	< LOQ	< LOQ	0.923	< LOQ	73.8	< LOQ	< LOQ	< LOQ	< LOQ	75.7
18	Mean	< LOQ	0.417 ± 0.207	< LOQ	0.951 ± 0.530	0.636 ± 0.283	7.54 ± 2.89	8.82 ± 3.66	3.35 ± 1.65	562 ± 416	2.08 ± 1.03	5.28 ± 3.68	< LOQ	0.481 ± 0.075	592 ± 417
	Range	< LOQ - < LOQ	< LOQ – 0.759	< LOQ - < LOQ	< LOQ – 1.89	< LOQ – 0.957	4.05 – 13.3	3.77 – 15.9	< LOQ – 5.16	92.3 – 1392	< LOQ – 2.94	1.35 – 12.6	< LOQ - < LOQ	0.403 – 0.631	
19	Mean	< LOQ	0.703 ± 0.242	< LOQ	0.687 ± 0.189	0.464 ± 0.120	3.05 ± 0.587	2.94 ± 0.595	2.45 ± 1.88	1.95 ± 1.88	1.82 ± 0.448	4.26 ± 1.47	1.47 ± 0.571	0.188 ± 0.080	19.8 ± 4.31
	Range	< LOQ - < LOQ	< LOQ – 2.64	< LOQ - < LOQ	< LOQ – 1.49	< LOQ – 0.912	< LOQ – 4.84	< LOQ – 5.28	< LOQ – 7.09	< LOQ – 18.9	< LOQ – 5.23	< LOQ – 14.4	< LOQ – 5.69	< LOQ – 0.852	
20	Mean	< LOQ	0.379 ± 0.097	< LOQ	1.40 ± 0.258	0.729 ± 0.173	8.48 ± 1.12	7.68 ± 0.487	8.40 ± 0.275	657 ± 502	3.12 ± 1.07	< LOQ	< LOQ	0.409 ± 0.163	688 ± 505
	Range	< LOQ - < LOQ	0.184 – 0.477	< LOQ - < LOQ	0.976 – 1.87	0.428 – 1.03	6.34 – 10.6	7.05 – 8.64	7.98 – 8.92	149 – 1661	1.74 – 5.22	< LOQ - < LOQ	< LOQ - < LOQ	0.221 – 0.734	
21[a]	Mean	< LOQ	0.328	< LOQ	0.887	0.741	4.52	4.27	4.47	7.65	1.37	< LOQ	< LOQ	< LOQ	24.2
22	Mean	< LOQ	0.895 ± 0.031	< LOQ	1.86 ± 0.040	1.29 ± 0.117	10.4 ± 3.01	14.5 ± 4.83	10.2 ± 4.25	34.7 ± 3.72	11.5 ± 0.301	< LOQ	< LOQ	0.082 ± 0.054	85.5 ± 8.87
	Range	< LOQ - < LOQ	0.864 – 0.926	< LOQ - < LOQ	1.82 – 1.90	1.17 – 1.41	7.41 – 13.4	9.64 – 19.3	5.95 – 14.5	31.0 – 38.5	11.2 – 11.8	< LOQ - < LOQ	< LOQ - < LOQ	< LOQ – 0.136	
23	Mean	< LOQ	0.701 ± 0.145	< LOQ	1.46 ± 0.213	0.601 ± 0.268	5.24 ± 0.910	2.96 ± 0.585	4.11 ± 0.663	26.9 ± 20.0	1.86 ± 0.599	5.36 ± 3.27	< LOQ	0.110 ± 0.048	49.7 ± 17.7
	Range	< LOQ – 1.15	0.176 – 0.999	< LOQ - < LOQ	0.903 – 2.19	< LOQ – 1.43	3.47 – 8.51	1.67 – 4.90	2.82 – 6.29	2.21 – 106	0.837 – 4.11	< LOQ – 13.4	< LOQ - < LOQ	< LOQ – 0.274	
25	Mean	< LOQ	0.375 ± 0.119	< LOQ	0.824 ± 0.192	0.651 ± 0.200	8.29 ± 1.84	8.99 ± 2.90	8.77 ± 2.85	45.3 ± 19.6	1.72 ± 0.650	< LOQ	< LOQ	0.140 ± 0.017	75.0 ± 21.6
	Range	< LOQ - < LOQ	< LOQ – 0.762	< LOQ - < LOQ	< LOQ – 1.14	< LOQ – 1.18	3.41 – 13.8	2.40 – 18.0	< LOQ – 17.2	< LOQ – 113	< LOQ – 4.02	< LOQ – < LOQ	< LOQ - < LOQ	0.097 – 0.186	

Table A11. Pearson correlations (p-values and r values) between PFAS concentrations in hepatopancreas, muscle tissue and carapace. R values of significant correlations are displayed in bold. NA = not assessed due to too low detection frequency in a certain tissue.

		Hepatopancreas	Muscle
PFOA	Muscle	p = 0.431 (r = 0.084)	-
	Carapace	p = 0.201 (r = 0.136)	p = 0.580 (r = -0.059)
PFDA	Muscle	p = 0.301 (r = 0.110)	-
	Carapace	**p < 0.001 (r = 0.384)**	p = 0.652 (r = -0.048)
PFUnDA	Muscle	**p = 0.021 (r = 0.243)**	-
	Carapace	**p < 0.001 (r = 0.375)**	p = 0.385 (r = 0.093)
PFDoDA	Muscle	**p < 0.001 (r = 0.373)**	-
	Carapace	**p < 0.001 (r = 0.432)**	p = 0.757 (r = 0.033)
PFTrDA	Muscle	**p < 0.001 (r = 0.477)**	-
	Carapace	**p < 0.001 (r = 0.420)**	p = 0.140 (r = 0.157)
PFTeDA	Muscle	p = 0.179 (r = 0.143)	-
	Carapace	**p = 0.050 (r = 0.208)**	p = 0.579 (r = -0.059)
PFBS	Carapace	NA	**p < 0.001 (r = 0.500)**
PFOS	Muscle	p = 0.823 (r = 0.024)	-
	Carapace	**p < 0.001 (r = 0.701)**	p = 0.830 (r = -0.023)
FBSA	Muscle	**p < 0.001 (r = 0.493)**	-
NaDONA	Carapace	**p < 0.001 (r = 0.349)**	NA
∑PFAS	Muscle	p = 0.370 (r = 0.096)	-
	Carapace	**p < 0.001 (r = 0.477)**	**p < 0.001 (r = 0.407)**

Table A12. Carapace width (cm) and crab weight (g) at the different sites. Values represent mean values ± SE. The number of replicates varied per site and are shown in Table A1. [a]No mean values were calculated as N = 1 at these sites. No crabs were caught at sites 17, 24 and 26.

Site no.	Carapace width (cm)	Weight (g)
1	5.13 ± 0.571	66.8 ± 24.8
2	4.32 ± 0.117	34.1 ± 2.76
3	5.61 ± 0.970	79.0 ± 40.7
4	3.87 ± 0.268	29.5 ± 4.12
5	6.04 ±0.135	85.1 ± 5.95
6	3.99 ± 0.159	26.8 ± 2.90
7	4.36 ± 0.106	39.6 ± 2.92
8	5.52 ± 0.226	72.5 ± 8.29
9	3.45 ± 0.085	16.1 ± 1.43
10	4.69 ± 0.186	43.0 ± 4.57
11	5.31 ± 0.165	64.7 ± 4.77
12	5.71 ± 0.070	69.5 ± 1.56
13	4.30 ± 0.087	32.6 ± 2.15
14	3.64 ± 1.15	24.9 ± 17.7
15	5.17 ± 0.435	50.7 ± 7.12
16[a]	2.55	6.44
18	2.99 ± 0.147	11.0 ± 1.86
19	5.19 ± 0.281	55.1 ± 9.08
20	3.26 ± 0.307	15.0 ± 4.60
21[a]	2.69	8.89
22	2.47 ± 0.035	6.42 ± 0.725
23	4.38 ± 0.182	32.4 ± 2.03
25	3.58 ± 0.071	19.0 ± 0.882

Table A13. Pearson correlations (p-values and r values) between PFAS concentrations in the three crab tissues and the carapace width and crab weight. R values of significant correlations are displayed in bold.

		Carapace width	Weight
Hepatopancreas	PFOA	p = 0.874 (r = 0.017)	p = 0.534 (r = -0.066)
	PFDA	p = 0.519 (r = - 0.069)	p = 0.594 (r = -0.057)
	PFUnDA	p = 0.240 (r = 0.125)	p = 0.119 (r = 0.166)
	PFDoDA	**p = 0.009 (r = -0.273)**	**p = 0.033 (r = -0.226)**
	PFTrDA	**p = 0.012 (r = -0.263)**	**p = 0.039 (r = -0.218)**
	PFTeDA	**p = 0.030 (r = -0.229)**	**p = 0.044 (r = -0.212)**
	PFOS	**p < 0.001 (r = -0.354)**	**p = 0.005 (r = -0.294)**
	FBSA	p = 0.655 (r = 0.048)	p = 0.872 (r = -0.017)
	NaDONA	p = 0.973 (r = -0.003)	p = 0.613 (r = -0.052)
	∑PFAS	**p < 0.001 (r = -0.359)**	**p = 0.004 (r = -0.301)**
Muscle	PFOA	p = 0.559 (r = -0.062)	p = 0.759 (r = -0.033)
	PFDA	**p = 0.017 (r = 0.251)**	**p = 0.022 (r = 0.242)**
	PFUnDA	**p = 0.004 (r = 0.303)**	**p = 0.004 (r = 0.302)**
	PFDoDA	p = 0.799 (r = -0.027)	p = 0.868 (r = -0.018)
	PFTrDA	p = 0.638 (r = -0.050)	p = 0.878 (r = -0.016)
	PFTeDA	p = 0.266 (r = 0.119)	p = 0.103 (r = 0.173)
	PFBS	**p = 0.036 (r = -0.221)**	**p = 0.049 (r = -0.208)**
	PFOS	p = 0.473 (r = -0.077)	p = 0.458 (r = -0.079)
	FBSA	**p = 0.031 (r = 0.227)**	p = 0.072 (r = 0.191)
	∑PFAS	p = 0.107 (r = -0.171)	p = 0.175 (r = -0.144)
Carapace	PFOA	p = 0.387 (r = -0.092)	p = 0.204 (r = -0.135)
	PFDA	p = 0.404 (r = -0.089)	p = 0.799 (r = -0.027
	PFUnDA	p = 0.640 (r = -0.050)	p = 0.883 (r = -0.016)
	PFDoDA	p = 0.192 (r = -0.139)	p = 0.175 (r = -0.144)
	PFTrDA	p = 0.228 (r = -0.128)	p = 0.312 (r = -0.108)
	PFTeDA	p = 0.149 (r = -0.153)	p = 0.162 (r = -0.149)
	PFBS	**p < 0.001 (r = -0.362)**	**p = 0.005 (r = -0.294)**
	PFOS	**p = 0.004 (r = -0.304)**	**p = 0.010 (r = -0.269)**
	NaDONA	**p = 0.009 (r = -0.266)**	**p = 0.013 (r = -0.253)**
	∑PFAS	**p < 0.001 (r = -0.374)**	**p = 0.003 (r = -0.307)**

Table A14. Principal component analysis factor loadings, proportion of variance of each principal component (PC) and cumulative proportion of variance. TOC = total organic carbon, CEC = cation exchange capacity, EC = electrical conductivity

	PC1	PC2	PC3
PFAS water		0.336	0.475
PFAS sediment	-0.437		
PFAS muscle	-0.202	-0.426	0.208
PFAS hepatopancreas		-0.477	0.236
PFAS carapace		-0.513	0.258
TOC sediment	-0.533		-0.124
Clay content sediment	-0.534	-0.123	
CEC sediment	-0.435	0.211	0.332
pH water		-0.143	0.482
EC water		0.361	0.496
Proportion of Variance	0.277	0.239	0.223
Cumulative proportion of variance	0.277	0.516	0.739

Table A15. Pearson correlations (p-values and r values) between PFAS concentrations in water and the pH and electrical conductivity (EC) of the water, as well as between water PFAS concentrations and concentrations of the same analyte in sediment.

	PFHxA		PFOA		PFDoDA		PFOS		∑PFAS	
	p-value	r-value	p-value	r-value	p-value	r-value	p-value	r-value	p-value	r-value
pH	0.669	-0.088	0.448	0.156	0.840	0.042	0.583	0.113	0.291	0.214
EC	0.338	0.196	0.072	0.358	0.444	-0.157	0.803	0.052	**<0.001**	0.680
Sediment concentration of same analyte	0.532	0.128	0.335	0.197	0.439	-0.159	0.465	0.150	0.422	-0.164

Table A16. Pearson correlations (p-values and r values) between PFAS concentrations in the abiotic environment and those accumulated in the different crab tissues. For water only ∑PFAS concentrations were correlated.

		Water	Sediment
PFOA	Hepatopancreas		p = 0.351 (r = -0.204)
	Muscle		p = 0.468 (r = 0.163)
	Carapace		p = 0.341 (r = 0.208)
PFDA	Hepatopancreas		p = 0.621 (r = -0.109)
	Muscle		p = 0.562 (r = -0.131)
	Carapace		p = 0.776 (r = 0.063)
PFUnDA	Hepatopancreas		p = 0.452 (r = 0.165)
	Muscle		p = 0.871 (r = -0.037)
	Carapace		p = 0.638 (r = 0.104)
PFDoDA	Hepatopancreas		p = 0.662 (r = 0.096)
	Muscle		p = 0.119 (r = 0.342)
	Carapace		p = 0.085 (r = 0.367)
PFTrDA	Hepatopancreas		p = 0.063 (r = 0.394)
	Muscle		p = 0.104 (r = 0.356)
	Carapace		p = 0.063 (r = 0.393)
PFOS	Hepatopancreas		p = 0.167 (r = 0.298)
	Muscle		p = 0.088 (r = 0.373)
	Carapace		p = 0.095 (r = 0.356)
∑PFAS	Hepatopancreas	p = 0.286 (r = -0.232)	p = 0.403 (r = -0.183)
	Muscle	p = 0.351 (r = -0.209)	p = 0.590 (r = 0.122)
	Carapace	p = 0.556 (r = -0.130)	p = 0.602 (r = -0.115)