Published in final edited form as:
Sci Total Environ. 2023 July 01; 880: 163302. doi:10.1016/j.scitotenv.2023.163302.

Ecological characteristics impact PFAS concentrations in a U.S. North Atlantic food web

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Abstract

This is the first comprehensive study of per- and polyfluoroalkyl substances (PFAS) in a coastal food web of the U.S. North Atlantic, in which we characterize the presence and concentrations of 24 targeted PFAS across 18 marine species from Narragansett Bay, Rhode Island, and surrounding waters. These species reflect the diversity of a typical North Atlantic Ocean food web with organisms from a variety of taxa, habitat types, and feeding guilds. Many of these organisms have no previously reported information on PFAS tissue concentrations. We found significant relationships of PFAS concentrations with respect to various ecological characteristics including species, body size, habitat, feeding guild, and location of collection. Based upon the 19 PFAS detected in the study (5 were not detected in samples), benthic omnivores (American lobsters = 10.5 ng/g ww, winter skates = 5.77 ng/g ww, *Cancer* crabs = 4.59 ng/g ww) and pelagic piscivores (striped bass = 8.50 ng/g ww, bluefish = 4.30 ng/g ww) demonstrated the greatest average ∑PFAS concentrations across all species sampled. Further, American lobsters had the highest concentrations detected in individuals (∑PFAS up to 21.1 ng/g ww, which consisted primarily of long-chain PFCAs). The calculation of field-based trophic magnification factors (TMFs) for the top 8 detected PFAS determined that perfluorodecanoic acid (PFDA), perfluorooctane sulfonic acid (PFOS), and perfluorooctane sulfonamide (FOSA) associated with the pelagic habitat biomagnified, whereas perfluorotetradecanoic acid (PFTeDA) associated with the benthic habitat demonstrated trophic dilution in this food web (calculated trophic levels ranged from 1.65–4.97). While PFAS exposure to these organisms may have adverse implications for ecological impacts via toxicological effects, many of these species are also key recreational and commercial fisheries resulting in potential for human exposure via dietary consumption.

Keywords

trophic magnification; bioaccumulation; wildlife; estuary; fluorochemical

1. Introduction

Per- and polyfluoroalkyl substances (PFAS, which some in the scientific community describe as “forever chemicals”) contain a fluorocarbon backbone attached to a functional group (i.e., C_nF_{2n+1}–R) and have been manufactured since the mid-1900s (Buck et al. 2011, Wang et al. 2017). Their amphiphilic behavior and resistance to degradation are among many unique properties that have led to their sustained and expanding usage in a multitude of consumer, commercial, and industrial applications and products (Glüge et al. 2020, Gaines 2022). After decades of usage, their stability and persistence have resulted in their detection in environmental compartments and biota worldwide, including humans (Domingo and Nadal 2019, Brusseau et al. 2020, De Silva et al. 2021, Kurwadkar et al. 2022). Increasing scientific and medical evidence show adverse effects from PFAS exposure including cancer, liver damage, immune suppression, and endocrine disruption in humans (DeWitt 2015, ATSDR 2021) and a range of effects in aquatic organisms (Giesy et al. 2010, Mahoney et al. 2022). Though the production of certain legacy PFAS has been phased out in many countries, some of the most commonly detected PFAS are the legacy, long-chain perfluoroalkyl acids (PFAAs, i.e., perfluoroalkyl sulfonic acids: PFSAs, and perfluoroalkyl carboxylic acids: PFCAs), and their precursors (e.g., perfluoroalkyl sulfonamides: FASAs) which can degrade into PFAAs (Zhang et al. 2021).

PFAS have been detected in estuarine and marine environments in concentrations ranging from pg/L (parts-per-quadrillion) to hundreds of ng/L (parts-per-trillion), where concentrations in coastal areas tend to be highest due to riverine and coastal urban inputs (Yamashita et al. 2005, Muir and Miaz 2021). Sources to air, water, and soil in such regions include facilities that manufacture and use PFAS in their processes, the use of fire-fighting foams, as well as wastewater treatment facilities and landfills. PFAS then enter the oceans as a final sink due to oceanic and atmospheric transport of precursors and terminal end products (De Silva et al. 2021). In the northern Atlantic Ocean off the US east coast, summed PFAA concentrations have been measured up to 4070 pg/L in surface waters in 2010–2011 (Zhang et al. 2019). In the Narragansett Bay area specifically, scientists report ∑PFAA surface water concentrations at 5800 pg/L from a 2009 survey (Benskin et al. 2012) and from a 2014 survey, ∑PFAA surface water concentrations are reported up to 6721 pg/L with ∑PFAS up to 6786 pg/L (Zhang et al. 2016). After a fuel spill and subsequent fire-fighting foam application in 2018, surface water concentrations of ∑PFAAs ranged 15–26 ng/L in a Providence River Estuary transect leading directly into Narragansett Bay, with ∑PFAS concentrations up to 388 ng/L due to 6:2-FTS contamination (Katz et al. 2022). To our knowledge, sediment concentration data in Narragansett Bay are not yet reported in the scientific literature.

Wildlife exposure to PFAS in aquatic systems occurs via organisms’ uptake from surrounding water/sediments (bioconcentration) as well as through diet (bioaccumulation), where concentrations may subsequently increase through the food web with increasing trophic level (biomagnification). Further, there are indications of adverse effects on aquatic organisms (decrease in body size/growth rate and fecundity, immunotoxicity, and developmental effects, among others), sometimes at environmentally relevant concentrations (McCarthy et al. 2017, Sinclair et al. 2020, Ankley et al. 2021). Processes regulating

the uptake and distribution of PFAS in organisms and across body tissues are not very well-understood, however. For legacy PFAAs, PFSAs tend to be more bioaccumulative than PFCAs of the same chain length, and PFCAs with chain-lengths <8 perfluorinated carbons are not generally bioaccumulative with low biomagnification potential (Conder et al. 2008). Bioaccumulation (Conder et al. 2008, Labadie and Chevreuil 2011, Munoz et al. 2019, Burkhard 2021) and even toxicity (Ankley et al. 2021) have also been shown to increase with increasing chain length within PFAA sub-groups.

Stable isotope analysis can be used as an informative tool to quantify the biomagnification of contaminants such as PFAS. The isotopic carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) compositions of biotic samples allow the primary food sources to food webs and relative trophic levels of organisms therein to be discerned, respectively (Michener and Kaufman 2007). Information obtained from $\delta^{15}N$ values (i.e., calculated trophic levels) can be examined in relation to contaminant concentrations to calculate a trophic magnification factor or “TMF” of a chemical, a descriptor of biomagnification that is commonly used in field-based studies (Conder et al. 2012). Reported marine organism TMFs for PFAS can vary substantially, with TMFs for the most-commonly studied substance, perfluorooctane sulfonic acid (PFOS), ranging from 1.3 to 20 (Miranda et al. 2022). Despite this, TMFs are still considered an important metric for the evaluation of contaminant movement through food webs (Borgå et al. 2012, Conder et al. 2012, Miranda et al. 2022).

Here, we conduct the first comprehensive study of PFAS in a coastal food web of the U.S. North Atlantic by characterizing the presence and concentrations of PFAS across marine fish and invertebrate species collected from Narragansett Bay, Rhode Island, and surrounding waters. These species reflect the diversity of a typical North Atlantic Ocean food web with organisms from a variety of taxa, habitat types, and feeding guilds, many of which have no reported information on PFAS concentrations. This study also extensively utilized an archived sample set for retrospective PFAS analysis as organisms were initially collected and analyzed for previous ecotoxicology and trophic ecology research studies (Piraino and Taylor 2009, Payne and Taylor 2010, Szczebak and Taylor 2011, Taylor et al. 2014, Malek et al. 2016). PFAS were examined with respect to organismal and ecological characteristics (e.g., body size, trophic ecology, and habitat use) to determine their possible influence on concentrations, along with the calculation of TMFs for select PFAS to determine whether trophic magnification was significant in this food web. Though PFAS have been reported for Narragansett Bay surface waters likely associated with urban and industrial sources such as airports and textile mills, among others (Benskin et al. 2012, Zhang et al. 2016, Katz et al. 2022), PFAS have not previously been studied in coastal biota from this region. While PFAS exposure to aquatic organisms has evident implications for ecological impacts, many of these species are also key recreational and commercial fisheries resulting in a pathway for human exposure via seafood consumption.

2. Materials and methods

2.1 Sample collection

Fishes and invertebrates were collected from the Narragansett Bay and Rhode Island Sound/Block Island Sound from April to November (2006–2014) using bottom trawls, hook & line, and by hand (Fig. 1, SI Table 1). Whole body or muscle tissues were extracted, freeze-dried (Labconco FreeZone 4.5-L Benchtop Freeze-Dry System), homogenized with a clean stainless-steel spatula, and stored at room temperature in 40-mL borosilicate vials (more details in Methods section of Supplementary Information).

2.2 PFAS extraction and analysis

The 24 targeted PFAS (Table 1) consist of 11 PFCAs, 7 PFSAs, and precursors including 1 FASA, 2 FASAAs (perfluoroalkane sulfonamido-acetic acids), and 3 n:2 FTSs (fluorotelomer sulfonates). Briefly, freeze-dried tissues were spiked with 10 ng labeled surrogate internal standards and underwent two rounds of alkaline digestion followed by clean-up using Supelclean ENVI-Carb cartridges (Supelco; Bellefonte, PA, USA). Extracts were dried and reconstituted in 1:1 methanol:Milli-Q water with 2 mM ammonium acetate, then spiked with 10 ng labeled PFAS injection internal standards prior to analysis. More detail on chemicals, reagents, and extraction can be found in the Supplementary Information.

Sample extracts were analyzed using an Acquity ultra-high-performance liquid chromatography coupled to a Xevo TQD tandem quadrupole mass spectrometer (Waters Corporation; Milford, MA, USA) in negative electrospray ionization mode using multiple reaction monitoring (further detail in Supplementary Information). PFAS were quantified via isotope dilution; analytes lacking matched, mass-labeled standards were quantified using mass-labeled standards with similar molecular weight and retention time (SI Table 2). Linear and branched isomers were quantified together as single compounds. Method detection limits (MDL) were determined for each compound (SI Table 2) using fortified, freeze-dried fish muscle (Supplementary Information) and applied to data based on initial dry weight concentrations.

2.3 Stable isotope analysis

Duplicate or triplicate subsamples of freeze-dried, homogenized tissue (~2 mg) were analyzed for stable nitrogen ($\delta^{15}N$) and carbon ($\delta^{13}C$) isotopes regardless of tissue type. A portion of the samples had previously been analyzed and quantified on a continuous flow isotope ratio mass spectrometer at the Boston University Stable Isotope Laboratory (BUSIL) (Piraino and Taylor 2009, Payne and Taylor 2010, Szczebak and Taylor 2011, Taylor et al. 2014, Malek et al. 2016). The remainder of the samples were analyzed at the U.S. Environmental Protection Agency's Atlantic Coastal Environmental Sciences Division (ACESD) on an Elementar VisION isotope ratio mass spectrometer coupled with an Elementar Vario Isotope Select elemental analyzer. Replicate analyses of USGS 40, USGS 41, and an in-house working standard (blue mussel homogenate) were used to normalize isotopic results to air ($\delta^{15}N$) and Vienna Pee Dee Belemnite ($\delta^{13}C$) scales via a multi-point linear normalization (Paul et al. 2007). A second working standard (NIST

1547) was used as a quality control. Isotopic ratios are expressed in δ notation following the formula,

$$\delta(‰) = \left(\frac{R_{sample}}{R_{reference}} - 1\right) \times 1000 \quad \text{(Eq. 1)}$$

where R is the ratio of heavy to light C or N isotopes in samples or reference standards.

2.4 Quality Assurance/Quality Control

Quality control samples were also extracted and analyzed to determine the overall accuracy and precision of the PFAS tissue extraction method. Overall method accuracy (recovery of native PFAS spiked into tissues) ranged from 92–171% across organism tissues (SI Table 4). Regarding matrix effects, almost all compounds exhibited signal suppression whereas two exhibited signal enhancement (Supplementary Information; SI Table 2).

For stable isotopic analyses conducted at ACESD, replicate analyses of reference materials USGS 40, USGS 41, and blue mussel homogenate yielded a pooled standard deviation (±1σ) of ±0.10 ‰ for $\delta^{13}C$ and ±0.26 ‰ for $\delta^{15}N$ (n=37). The isotopic quality control NIST 1547 was within 0.2 ‰ of its expected value for both $\delta^{15}N$ and $\delta^{13}C$ (n=17). Standard deviation among sample replicates averaged 0.08 ‰ for $\delta^{13}C$ and 0.11 ‰ for $\delta^{15}N$. A subset of samples run at BUSIL were re-analyzed at ACESD to assess interlaboratory reproducibility and were found to be within 0.7 ‰. Previously published work from the BUSIL is reported to ±0.5 ‰ based on replicate analyses of peptone and glycine.

2.5 Data analysis

All dry weight concentrations were normalized to mass in wet weight based on 73–83% water content of the various tissue types, previously measured for many of the same or similar species (Piraino and Taylor 2009, Payne and Taylor 2010, Taylor and Calabrese 2018). For determination of summed (∑) PFAS and sub-class concentrations, concentrations below MDLs were replaced with 0. For determination of overall trends in PFAS concentrations associated with marine organism variables (habitat, feeding guild, and location of collection), <MDL concentrations were also replaced with 0. For regression analyses of PFAS concentrations and $\delta^{13}C$ or TL, values below MDLs were treated as censored data. Individual PFAS with 0% detects for the entire dataset were completely removed from all statistical analyses (19 of 24 PFAS were detected across the entire study).

2.5.1 PERMANOVA and PCO—Differences in detected PFAS profiles in target fishes and invertebrates as a function of habitat use, geographic location, and feeding guild (Fig. 1, SI Table 1) were examined using a three-way permutational multivariate analysis of variance (PERMANOVA) model, as provided in the PRIMER 7.0 software package (Anderson et al. 2008). A Euclidean distance resemblance matrix of log-transformed data was created using the aforementioned method. If significant results ($p < 0.05$) were obtained using the PERMANOVA model, SIMPER analyses were conducted for each main effect to determine which PFAS contributed to the observed differences across habitats, locations, or feeding guilds. Moreover, to facilitate the interpretation of the PERMANOVA results, principal

coordinates analysis (PCO) was used to visualize the PFAS profile data. This method provides a direct projection of data points in space according to their actual dissimilarities, and PCO axes quantify the amount of variation inherent in the resemblance matrix that is attributable to each successive ordination axis (expressed as percent of total variation) (Anderson et al. 2008). Moreover, using the multiple correlations, vectors of the most commonly detected PFAS (≥10% detection of analyzed samples) were superimposed onto the PCO biplots, which correspond to the monotonic relationships between a PFAS's importance and the ordination axes (Anderson et al. 2008).

2.5.2 PFAS and stable isotope data—$\delta^{15}N$ signatures were used to calculate the trophic level (TL) of each fish and invertebrate sample based the following equations:

$$TL = 2 + \frac{(\delta^{15}N_{Consumer} - \delta^{15}N_{Bivalve})}{\delta^{15}N_{Enrichment}} \quad \text{(Eq. 2)}$$

and

$$\delta^{15}N_{Bivalve} = 6.193 \times \text{Latitude} - 247.17 \quad \text{(Eq. 3)}$$

where "2" is the assumed trophic level of a reference primary consumer (i.e., bivalve), and $\delta^{15}N_{Enrichment}$ is the constant nitrogen isotopic enrichment (‰) per trophic level (also known as "trophic fractionation"), which varied between 2.9 and 3.5 per sample depending on the taxon (fish, squid, crustacean, or bivalve) and tissue-type (muscle or whole body; SI Table 5) (Post 2002, Yokoyama et al. 2005, Sweeting et al. 2007). $\delta^{15}N_{Consumer}$ and $\delta^{15}N_{Bivalve}$ are the respective nitrogen isotopic signatures of the consumer species of interest (sample) and bivalves. Bivalves were selected as the reference primary consumer because of their high site fidelity and consistent phytoplanktivorous diet (Stanley 1985, Newell 1989, Kemp et al. 1990, Naidu 1991). Previous and concurrent investigations in the Narragansett Bay and Rhode Island Sound measured the $\delta^{15}N$ signatures of five bivalve species (Taylor et al. 2012, Malek et al. 2016, this study): blue mussel (*Mytilus edulis*; n = 74, 22–56 mm shell length, SL), ribbed mussel (*Geukensia demissa*; n = 43, 36–89 mm SL), horse mussel (*Modiolus modiolus*; n = 45, 33–79 mm SL), hard shell clam (*Mercenaria mercenaria*; n = 23, 30–95 mm SL), and Atlantic sea scallop (*Placopecten magellanicus*; n = 64, 45–125 mm SL). The geographic locations from which these bivalves were collected in the Bay and Sound are comparable to this study (40.9680 °N to 41.8156 °N), and *a posteriori* analysis revealed that the bivalve $\delta^{15}N$ signatures differed significantly across latitudes (linear regression: $F_{1,248}$ = 1016.5, R^2 = 0.805, $p < 0.001$). More specifically, pronounced ^{15}N depletion occurred over a north-south gradient in the study area, which is ostensibly unrelated to the bivalve's trophic status (Pruell et al. 2006). Thus, the $\delta^{15}N_{Bivalve}$ equation (Eq. 3; "Latitude" unit = decimal degrees north) was used to account for spatial variations in $\delta^{15}N$ values in fish and invertebrate samples that were potentially affected by factors other than their respective trophic positioning.

The $\delta^{13}C$ signature of each fish and invertebrate sample was corrected to account for trophic fractionation, such that:

$$\delta^{13}C_{corrected} = \delta^{13}C_{raw} - 0.39(TL - 1) \quad \text{(Eq. 4)}$$

where $\delta^{13}C_{corrected}$ is the carbon isotopic signature corrected for trophic fractionation, $\delta^{13}C_{raw}$ is the uncorrected (original) isotope value, TL is the trophic level of a sample, and 0.39 is the carbon isotope enrichment (‰) per trophic level (Post 2002).

Corrected $\delta^{13}C$ and TL were then analyzed in relation to individual PFAS concentrations for those PFAS with ≥20% detection above MDL (Eq. 5 & 6, respectively), using the NADA2 package (Helsel 2011) in R v. 4.1.0 (R Core Team 2021) to account for the presence of censored data (i.e. concentrations <MDL). Four samples were removed from these analyses due to missing stable isotope data (n = 340 vs. 344). Regression by Maximum Likelihood Estimation (MLE; lognormal) was utilized if the parametric assumption of normality was met (Shapiro-Francia W test). Otherwise, relationships were assessed using nonparametric Akritas-Theil-Sen line (ATS). Relationships of PFAS concentration and corrected $\delta^{13}C$ or TL were assessed as

$$\ln(\text{PFAS concentration}) = x(\delta^{13}C) + y \quad \text{(Eq. 5)}$$

and

$$\ln(\text{PFAS concentration}) = x(TL) + y \quad \text{(Eq. 6)}$$

where x is the slope and y is the intercept for each model. TMFs were then calculated as

$$e^{x} \quad \text{(Eq. 7)}$$

where x is the slope of the MLE or ATS relationship between PFAS concentration and TL (i.e., from Eq. 6).

3. Results & discussion

3.1 General PFAS trends

Nineteen of the 24 targeted PFAS were detected across samples; 5 PFAS that were not detected in this study were 4:2-, 6:2-, and 8:2-FTS, along with PFHpS and PFNS. Further, 90.7% of samples contained at least one PFAS in concentrations above the MDL. When examining average (± standard deviation) ∑PFAS concentrations across species, American lobster (10.5 ±4.52 ng/g ww) > striped bass (8.50 ±4.47 ng/g ww) > winter skate (5.77 ±4.74 ng/g ww) > *Cancer* crab (4.59 ±3.57 ng/g ww) > bluefish (4.30 ±1.83 ng/g ww; Fig. 2). Rankings change slightly when examining maximum ∑PFAS concentrations in individuals (American lobster: 21.1 > butterfish: 19.7 > winter skate: 18.3 > striped bass: 17.6 ng/g ww; SI Table 6). The most frequently detected compounds were the long-chain PFCAs PFTrDA (82% of samples), PFUnDA (74%), and PFTeDA (64%), which were detected across all species (Table 1; SI Table 7). This was followed by FOSA (49% of samples), the most frequently detected precursor and detected in all species except little skates, winter skates, and smooth dogfish. The most frequently detected PFSA was PFOS

(31% of samples), which was the sixth most frequently detected PFAS overall and was detected in all species except Atlantic herring, scup, and tautog.

PFAS detected at the highest concentrations in individual samples were PFTrDA (≤16.6 ng/g ww in American lobster), PFOS (≤7.48 ng/g ww in striped bass), PFUnDA (≤3.91 ng/g ww in winter skates), and FOSA (≤3.71 ng/g ww in striped bass; SI Table 7). Generally, long-chain PFAAs were detected more frequently and at higher concentrations vs. short-chain PFAAs. Concentrations and specific PFAS detected are similar to other studies of marine or estuarine biota, though PFOS is often the dominant PFAS detected (Houde et al. 2006, Thompson et al. 2011, Naile et al. 2013, Gebbink et al. 2016, Munoz et al. 2019, Ali et al. 2021). Many of these studies did not examine PFTrDA or PFTeDA, however. Unfortunately, due to the limited sample set available for retrospective analysis, we were unable to assess some key prey organisms present at low trophic levels (e.g., planktonic organisms and benthic macroinvertebrates, such as polychaete worms) but recommend that future studies incorporate such organisms when possible.

PFOS concentrations were significantly correlated to ∑Precursor concentrations in organism tissues (Spearman's $\rho = 0.32$, $p < 0.001$; SI Table 8). This may indicate similar exposure sources for these compounds or the biodegradation of precursors to PFOS within organisms (Martin et al. 2010). Whether exposure to precursors indeed led to increased PFOS concentrations in organism tissues in Narragansett Bay would need to be explored further; however, other studies have noted similar PFOS-precursor trends in both freshwater and marine systems (Gebbink et al. 2016, Munoz et al. 2017, Chen et al. 2018). This also has implications for the interpretation of biological data in terms of PFAS bioaccumulation and trophic transfer, as field-based bioaccumulation factors (BAFs) and TMFs for PFOS may be overestimated due to biological precursor degradation (Martin et al. 2010, Miranda et al. 2022).

3.2 Organism body size

Spearman's Rank Correlation was used to examine total body length or width (cm) in relation to total PFAS concentrations for each species (SI Fig. 1). Correlations were conducted across all locations grouped together for each species due to low sample sizes for many locations, though the analysis of size-concentration correlations for different collection locations would be of interest for future studies. Black sea bass, tautog, and *Cancer* crabs demonstrate a positive correlation in ∑PFAS with increasing body size. Black sea bass and tautog are both demersal crustacivores that feed upon *Cancer* crabs, among other organisms. Conversely, *Alosa* sp., Atlantic herring, and little skates demonstrate a negative correlation in ∑PFAS with body size. This may be a result of more efficient PFAS metabolism with size/age, ontogenetic diet shifts to prey with lower PFAS burdens as these organisms grow, or possibly in the case of *Alosa* sp., the relocation of larger/older organisms away from inland, coastal PFAS sources. Another study also found ∑PFAS in Australian rays decreased significantly with increasing age and body size, which was attributed to temporal variation in PFAS associated with a flood event (Baduel et al. 2014). Other studies report mixed results on the association of body size with PFAS concentrations. For example, body size was not related to ∑PFAS concentrations in marine fish from the southeastern coast of the USA (Fair

et al. 2019). A similar result was seen in the analyses of PFAAs in freshwater fish from North America (Gewurtz et al. 2012, Gewurtz et al. 2013). Body size was also not related to ∑PFAS in freshwater fish from Europe (Åkerblom et al. 2017, Babut et al. 2017), though the influence of size on specific PFAS concentrations in one species of fish did exist (e.g., PFOS, PFNA, PFDA, and PFTeDA) likely due to ontogenetic diet shift (Babut et al. 2017).

3.3 Habitat, feeding guild, and collection location

PFAS profiles were analyzed with respect to ecological characteristics of the organisms, including habitat, feeding guild, and location of collection. Habitat-feeding guild and habitat-location interaction effects were significant ($p < 0.001$ and $p = 0.006$, respectively), thereby precluding contrasts across the main effects (Table 2). To address the interaction effects, habitat data were subsequently isolated for pelagic, demersal, and benthic types and 2-way PERMANOVA models were used to examine the effect of feeding guild and location on PFAS profiles.

Accordingly, PFAS composition in pelagic and benthic habitats significantly varied across feeding guilds, though feeding guild demonstrated no effect in the demersal habitat (Table 2). Further, principal coordinate analysis revealed that habitat most closely corresponded to the first PCO axis (PCO1) and accounted for 56.7% of the explainable variation in PFAS contaminant profiles, and feeding guild was associated with the PCO2 axis and described 27.8% of the differences in PFAS composition (Fig. 3). Vectors of the dominant PFAS compounds superimposed on PCO biplot affirmed that total concentrations were maximal in pelagic piscivores (e.g., striped bass and bluefish) and benthic omnivores (e.g., lobster and crabs), and, conversely, reduced in benthic piscivores and demersal species (Fig. 3; Fig. 2). Moreover, FOSA, PFOS, and PFDA were closely associated with the pelagic habitat, whereas benthic taxa had higher concentrations of the long-chain PFCAs PFTrDA and PFTeDA. Such habitat and feeding guild trends in overall PFAS composition could be related to local inputs, PFAS water-sediment partitioning, varying metabolic capabilities of different organisms, or an interplay of these and needs to be studied further. However, our results are similar to PFAS compositions across aquatic habitats seen in other studies, though many of these do not examine the impact of feeding guild. For example, lower PFOS concentrations have been noted in benthic vs. pelagic fishes (Lanza et al. 2017), higher PFNA:L-PFOS ratios were found in benthic fish and invertebrates vs. pelagic species (Munoz et al. 2017), and other studies found lower ∑PFSA: ∑PFCA ratios in benthic vs. pelagic organisms (Martin et al. 2004, Ren et al. 2022).

Location of collection also significantly affected the PFAS profiles of pelagic taxa (Table 2; Fig. 1). Pelagic organisms varied primarily in their relative proportions of PFOS, PFTrDA, PFUnDA, and FOSA depending on location. The effect appears to be mainly driven by a latitudinal decrease in the concentration of these PFAS from the upper, northern reaches Narragansett Bay to offshore, where PFOS shows the most dramatic decrease (SI Fig. 2). Assuming the pelagic organisms sampled exhibit some degree of site fidelity, this PFAS gradient may be driven by upstream sources of PFAS to the bay (e.g., wastewater treatment facilities, industrial, and manufacturing sources from a highly urbanized and previously industrialized area) and subsequent dilution as they are transported farther

offshore. Interestingly, PFAS profiles of less-mobile demersal and benthic species did not significantly vary by collection location (Table 2); further research is needed to clarify these discrepancies. For instance, SI Fig. 2 indicates that in the benthic habitat, concentrations of PFTrDA increase and FOSA decrease from the upper reaches of the bay to offshore; this effect was not statistically significant, however.

PFAS can differentially partition into various tissues, therefore it is often preferred if the same tissue types are analyzed and compared across organisms (reviewed by Burkhard 2021, Miranda et al. 2022). Our study used a mixture of whole body and muscle tissues for analyses (SI Table 5) based on availability of samples for retrospective analysis (total n = 344). Muscle tissue was analyzed for a majority of the organisms in our study (muscle n = 282). *Alosa* sp., lobster, squid, and butterfish samples comprised both muscle and whole body samples (SI Fig. 3), and *Cancer* crab and blue mussel samples consisted of whole bodies only (whole body n = 62). Though concentrations and proportions of some PFAS vary slightly in muscle vs. whole body for those organisms in which both were analyzed (SI Fig. 3), we were unable to determine whether partitioning is the cause as opposed to other factors (e.g. dates/locations of collection, low sample size, etc.). To determine whether the analysis of mixed tissue types impacted our overall results, we repeated PERMANOVA analysis on the subset of samples consisting of muscle tissues only (SI Table 9) and PCO analyses on muscle tissues and whole bodies only (SI Fig. 4a & b). The relatively low sample size of whole body samples and their limited habitat/guild designation did not allow for robust statistical analysis via PERMANOVA. Results of this secondary analysis indicate that overall outcomes remain the same whether the entire dataset or only muscle samples were analyzed (i.e. Table 2 vs. SI Table 9; Fig. 3 vs. SI Fig. 4).

Carbon isotope analysis can serve as a key indicator of primary producers as diet sources to food webs, which are also related to habitats. For instance in marine coastal areas, phytoplankton serve as the primary carbon source to pelagic food webs with negative $\delta^{13}C$ values (−22 ±3 ‰), whereas benthic algae are comparatively enriched (−17 ±4 ‰) (Peterson and Fry 1987, France 1995). Our analyses of 8 individual PFAS with the highest detection frequencies (≥20% detection above MDL) indicate the prevalence of different PFAS in different portions of the Narragansett Bay food web (Table 3a). FOSA demonstrates a significant, negative relationship with $\delta^{13}C$ (p = 0.0048), indicating increased concentrations in organisms with more negative $\delta^{13}C$ values, reflective of organisms with pelagic food sources/trophic linkages. Conversely, the long-chain C11-C14 PFCAs demonstrate significant, positive relationships with $\delta^{13}C$ ($p < 0.001$ in all cases), indicating their increased prevalence in benthic consumers. Whether these PFAS are taken up directly by pelagic and benthic algae as sources of PFAS to the food web would need to be evaluated further, considering primary producers were not examined in this study. However, these results support the aforementioned PFAS- and habitat-related trends determined via PERMANOVA and PCO. One caveat however, is the isotopic variation associated with biochemical heterogeneity and turnover rates in different organism tissues (Michener and Kaufman 2007), along with interspecies variability in growth and subsequently, isotopic turnover rates (Hesslein et al. 1993). Muscle tissue was analyzed for a majority of the organisms in our study, though some organisms were assessed as whole bodies (SI Table 5).

Yet when examining only taxa for which muscle tissue was analyzed, overall trends between PFAS and $\delta^{13}C$ remained the same (i.e., negative relationship between $\delta^{13}C$ and FOSA and positive relationships between $\delta^{13}C$ and long-chain C9 & C11-C14 PFCAs; SI Table 10a). To our knowledge, one other study has examined the relationship between $\delta^{13}C$ and PFAS concentrations, finding positive relationships between $\delta^{13}C$ and C10-C14 PFCAs as well as PFOS in freshwater fish, indicating those feeding from autochthonous carbon sources like periphyton and biofilm were more exposed to PFAS (Macorps et al. 2022).

3.4 Trophic relationships

Examining the relationships of the same 8 PFAS with trophic level allowed us to determine their trophic magnification factors, a proxy for biomagnification across an entire food web of field-collected organisms (there was no indication of multiple, separate food webs according to stable isotope/trophic level biplots; SI Fig. 5). TMFs >1 indicate biomagnification. Statistical evaluation demonstrates that PFDA, PFOS, and FOSA concentrations significantly increase with increasing trophic level for marine organisms in Narragansett Bay and immediately offshore, i.e., TMFs are significantly > 1 (Table 3b; SI Fig. 6). PFDA demonstrates the largest degree of bioaccumulation potential (TMF = 1.7, $p < 0.001$), followed by PFOS (TMF = 1.4, $p = 0.020$) and then FOSA (TMF = 1.3, $p = 0.042$). For the longest-chain PFCA, PFTeDA (TMF = 0.77, $p = 0.051$), concentrations significantly decrease with increasing trophic level indicating trophic dilution (Table 3b). The C9 and C11-C13 PFCAs do not demonstrate statistically significant trophic magnification or dilution in this study (Table 3b). In other marine and estuarine field-based studies, PFOS has been shown to biomagnify (Houde et al. 2006, Kelly et al. 2009, Tomy et al. 2009, Munoz et al. 2017, Gao et al. 2019, Miranda et al. 2021), as well as PFDA and FOSA to biomagnify (Houde et al. 2006, Kelly et al. 2009, Tomy et al. 2009, Munoz et al. 2017), though Miranda et al. (2021) show biodilution of L-FOSA but they did not analyze branched isomers. PFTeDA has been shown to biomagnify in a study with a wide range of trophic levels (1.0–5.5) including marine mammals (Kelly et al. 2009), but biodilute in studies with more limited ranges of trophic levels (approximately 1.5 or 2.0–4.5) (Munoz et al. 2017, Miranda et al. 2021). Due to the range of sampling dates in this retrospective study, the TMFs reported herein are time integrated. Low sample sizes for specific dates did not allow for the assessment of potential temporal trends in TMFs, though this is of interest as significant shifts in PFAS production and use have occurred over the past two decades with phase-outs and reductions in legacy PFAS in favor of alternatives, including novel and short-chain PFAS (Glüge et al. 2020, ITRC 2020).

Due to differential partitioning of PFAS into various tissues, it is recommended that the same tissue types be used across organisms for TMF calculations, with preference for the use of whole body concentrations (Miranda et al. 2022), or via the use of conversion factors if available (e.g., Tomy et al. 2009, Munoz et al. 2017, Valsecchi et al. 2021, Kaboré et al. 2022). Our study used a mixture of whole body and muscle tissues for PFAS analysis (SI Table 5) based on availability of samples for retrospective analysis. Therefore, we also derived TMFs by limiting the dataset to a “muscle-only” food web as this was the predominant tissue analyzed and due to a lack of data on muscle to whole

body conversion factors for the specific organisms analyzed. As a result, PFDA, PFOS, and FOSA still demonstrate significant trophic magnification (TMFs = 2.3, 2.0, and 3.4, respectively), along with the significant trophic dilution in PFTeDA (TMF = 0.72; SI Table 10b). However, PFUnDA and PFDoDA also demonstrate significant magnification in the "muscle-only" food web (TMFs = 1.3 for both PFAS; SI Table 10b). It is recommended that TMF calculations be based upon a minimum of 3 trophic levels (Borgå et al. 2012); in limiting the Narragansett Bay food web to analysis of only muscle tissues however, we limit the number of trophic levels in the food web: using all tissues, TLs range 1.65–4.97; using muscle tissue only, TLs range 2.55–4.97. While general trends in magnification/dilution are similar for TMFs calculated using all tissues vs. muscle samples only, the range of TMFs reported for the "muscle-only" food web is larger. More data would be needed to determine whether this is in fact due to differences in observed PFAS concentrations as a result of tissue partitioning or simply the removal of lower trophic level organisms from the TMF analysis.

TMF values calculated for PFAS appear relatively variable across studies in marine and estuarine systems (reviewed by Miranda et al. 2022); however, TMFs may not necessarily be directly comparable without recalculation as TMFs are calculated in two different manners, i.e., the use of natural logarithm vs. base 10 logarithm results in TMFs with different scales. Additionally, because TMF is a relatively simple metric based on the assumption that concentrations are solely related to trophic level with diet as the major exposure route, confounding factors can greatly influence calculated TMFs for PFAS (Borgå et al. 2012, Conder et al. 2012, Franklin 2016, Kidd et al. 2019), many of which have been shown to impact PFAS concentrations both in this study and the broader scientific literature. Examples include study-design-based factors such as the method for the treatment of values below detection limits (Munoz et al. 2017), inclusion/exclusion of organisms analyzed (Martin et al. 2004), and variation in locations sampled (Macorps et al. 2022), as well as factors inherent to organisms sampled such as organism size (Babut et al. 2017), sex (Gewurtz et al. 2012), and feeding guild (Yang et al. 2012). Further, the presence of metabolizable PFAS precursors may result in inflated TMF values for their subsequent end-products (Franklin 2016, Miranda et al. 2022). For instance, we see significant magnification of both FOSA and PFOS in the present study along with correlations in their concentrations (SI Table 8). We therefore suggest that while statistically evaluated TMF metrics are useful in the broad weight of evidence for the overall occurrence of PFAS biomagnification (Franklin 2016) and possibly for relative ranking of multiple PFAS' bioaccumulation potentials within a study (taking biotransformation into account), the caveat remains that absolute TMF values for individual PFAS may vary considerably when analyzing subsets of food webs, comparing metrics across different food webs, and comparing metrics across studies.

3.5 Conclusions

In Narragansett Bay, PFDA, PFOS, and FOSA associated with the pelagic habitat biomagnify, whereas PFTeDA associated with the benthic habitat demonstrates trophic dilution. Based on our data, benthic omnivores (lobster, winter skate, and *Cancer* crab) and pelagic piscivores (striped bass and bluefish) collected between 2006–2014 demonstrate the

greatest PFAS concentrations in their tissues; comparing the species measured in this study, their consumption would result in the greatest potential for human exposure. Some species also demonstrate a positive correlation in ∑PFAS concentrations with increasing body size (black sea bass, tautog, and *Cancer* crabs), again with implications for human exposure. The consumption of seafood containing PFAS has indeed been shown to contribute to human body burdens (Falandysz et al. 2006, Haug et al. 2010) with subsequent implications for toxic effects (Sunderland et al. 2019, Fenton et al. 2021). Whether the concentrations detected in this study were large enough to adversely impact these organisms directly is not known; in addition to the possible effects from individual PFAS, the potential mixture effects with other PFAS, chemical contaminants, and environmental stressors must also be considered (Sinclair et al. 2020). For instance, in marine laboratory studies PFOS and PFOA decreased normal larval development in mussels at concentrations as low as 0.1 μg/L (NOEC 0.01 μg/L) (Fabbri et al. 2014), and PFOS was shown to exacerbate negative effects of elevated temperature on corals at 0.1 μg/L (Bednarz et al. 2022). Further, it remains to be seen whether the trends in PFAS concentrations and organisms' ecological characteristics hold for Narragansett Bay organisms sampled more recently (i.e., due to changes in overall production and use of legacy PFAS to shorter-chain and novel PFAS sub-classes), or for other North Atlantic coastal food webs overall. However, we suggest that researchers and stakeholders account for key ecological characteristics to more fully understand the presence and trophic transfer of PFAS in local food webs.

Supplementary Material

Refer to Web version on PubMed Central for supplementary material.

Acknowledgements

We would like to thank Laura Coiro, Richard McKinney, and Maya Morales-McDevitt for their assistance, and Tara Burke, Michaela Cashman, and Marissa Giroux for their initial review of the manuscript. This project was supported under the U.S. EPA 2020 Chemical Safety for Sustainability Research Program and in part by the Roger Williams University Foundation Fund Based Research Grant. This document has been reviewed by the U.S. Environmental Protection Agency, Office of Research and Development, and approved for publication. The views expressed in this article are those of the authors and do not necessarily represent the views or policies of the U.S. Environmental Protection Agency.

Data availability

The dataset will be made available upon publication in ScienceHub at ScID: D-h717; DOI: 10.23719/1528286.

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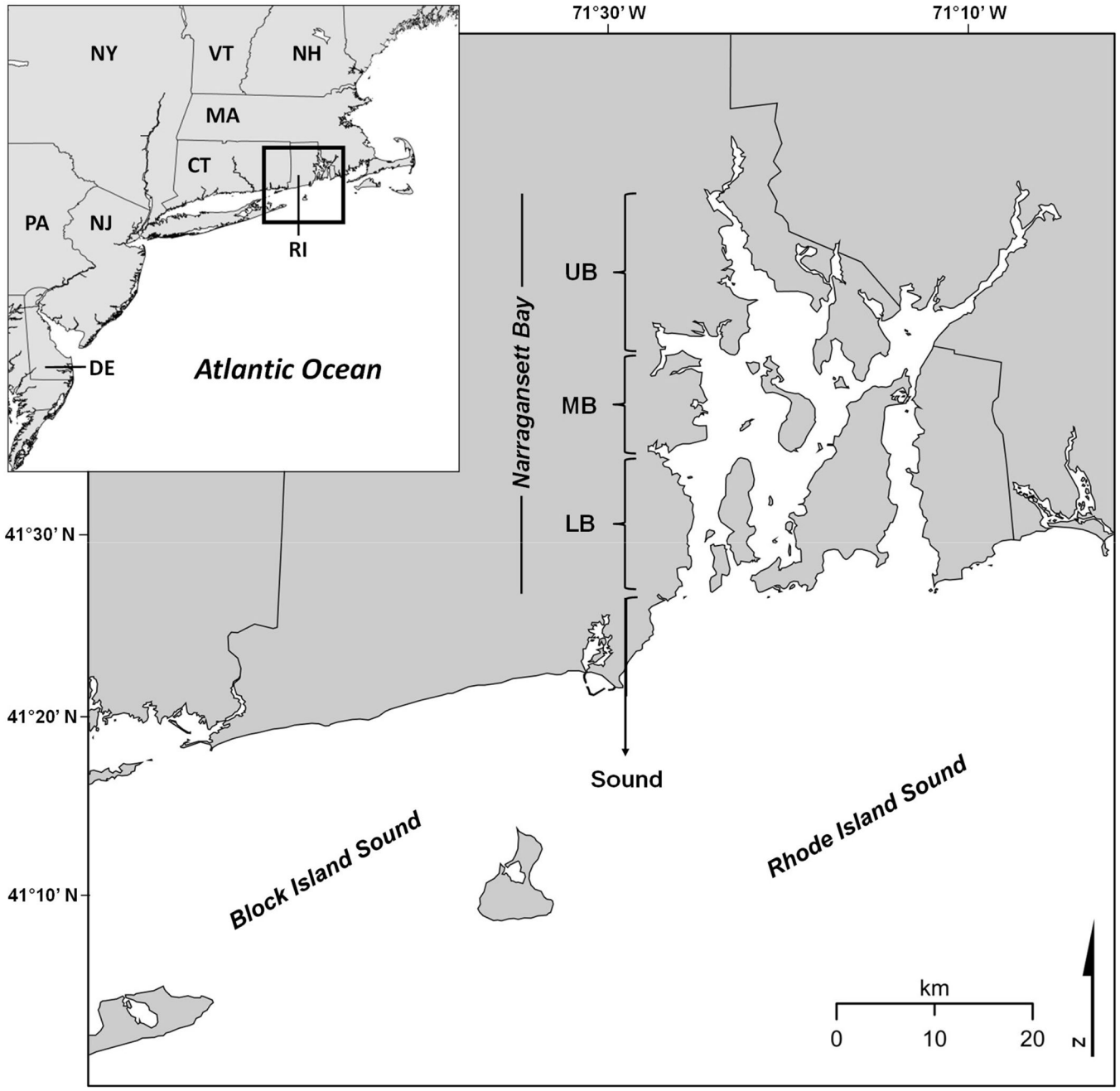

Figure 1.

Collection locations of marine species from Narragansett Bay, Rhode Island, USA. Locations were divided as follows: UB = Upper Bay, MB = Mid Bay, LB = Lower Bay, Sound = offshore including Block Island and Rhode Island Sounds.

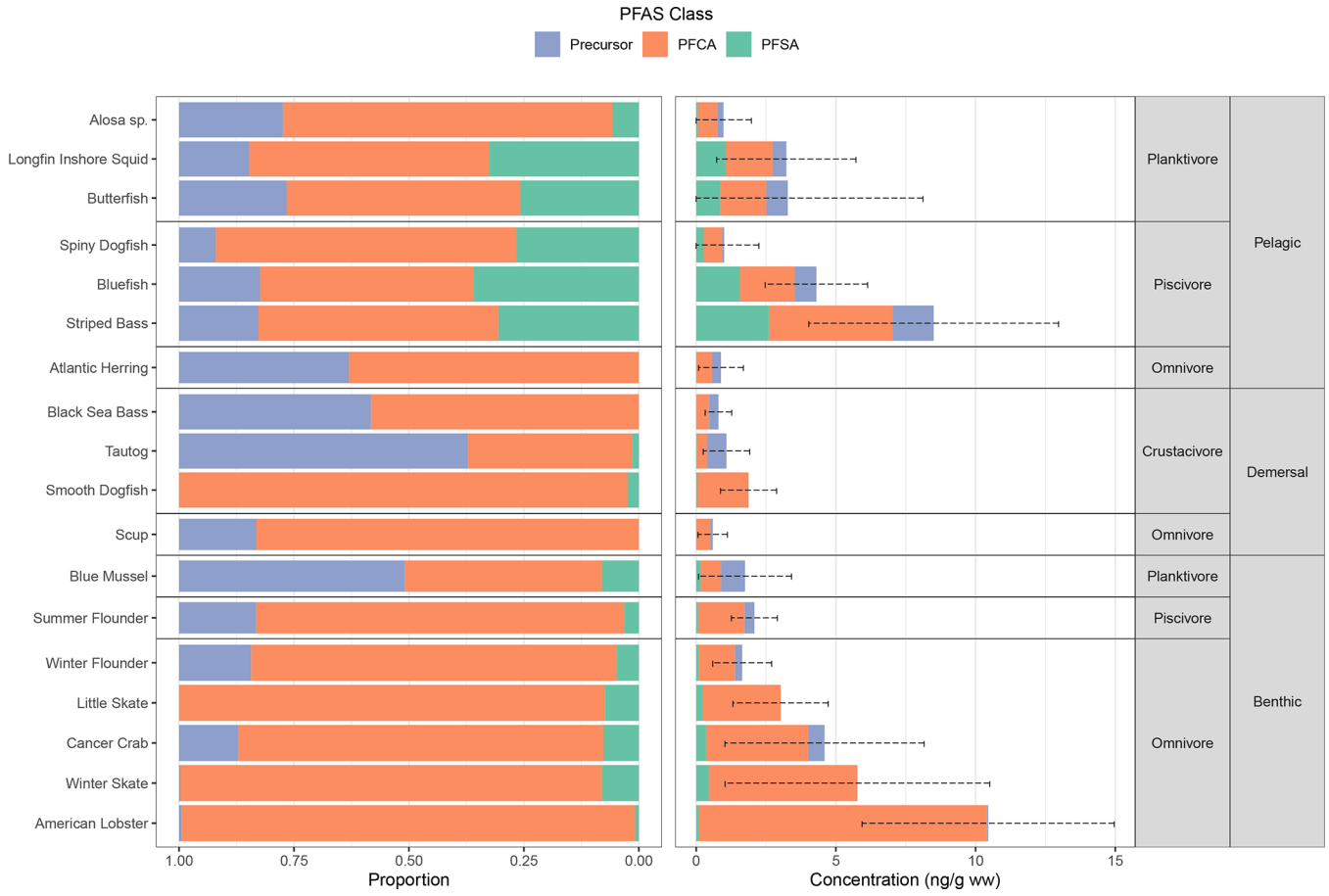

Figure 2.

Proportions and average concentrations summed for PFAS sub-classes in marine species categorized by feeding guild and habitat. Error bars in the right panel report standard deviation for total, ∑PFAS values and are not specific to PFAS sub-classes.

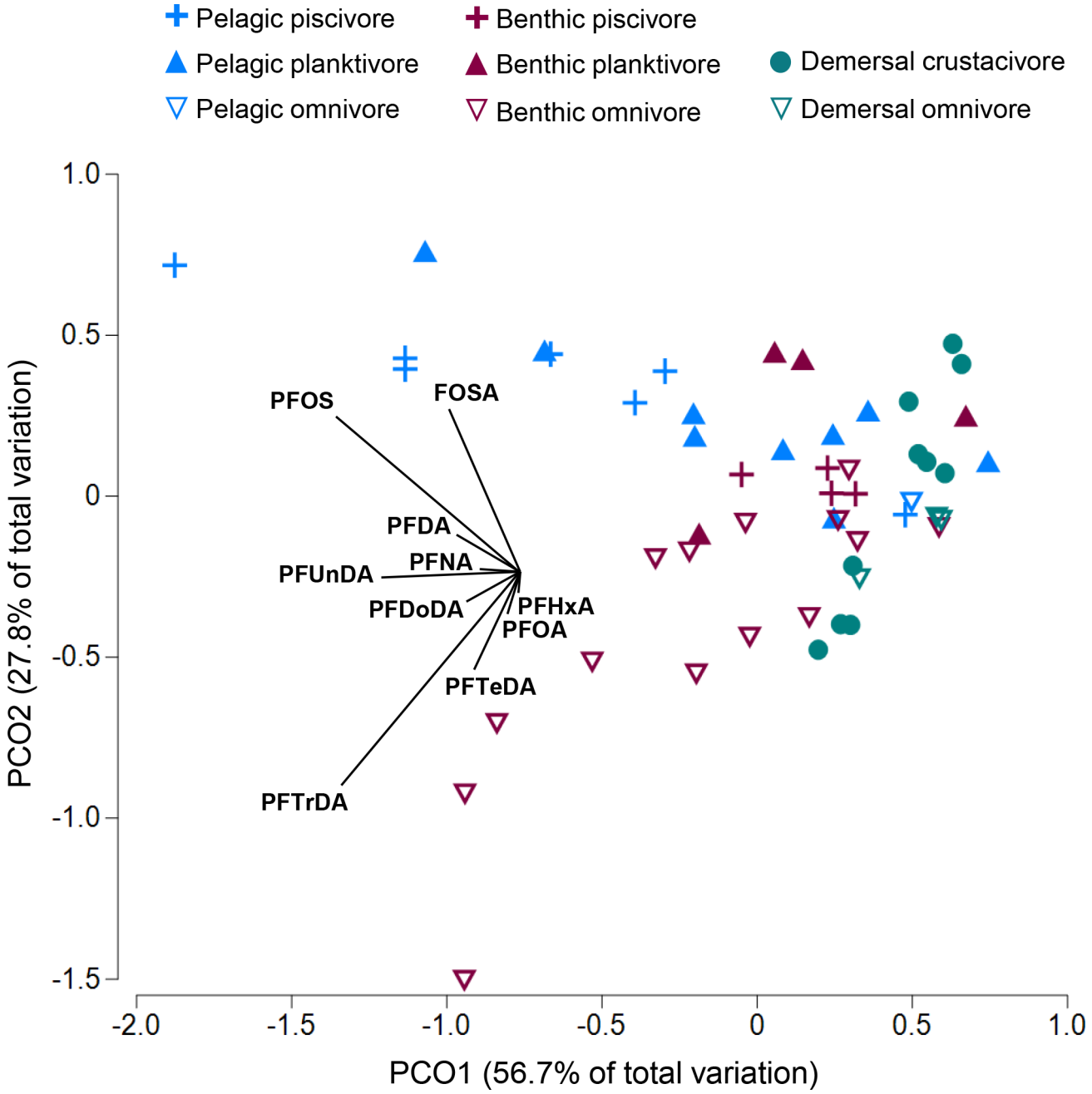

Figure 3.

Principal coordinates (PCO) configuration plot that represents the PFAS-profile dissimilarities among fish and invertebrate species. Each data element reflects PFAS profiles as a function of species and location of collection. Each species, in turn, is defined by its habitat use (pelagic benthic, and demersal) and feeding guild (piscivore, planktivore, omnivore, or crustacivore). Solid lines superimposed on the PCO plot represent vectors of the most commonly detected PFAS (≥10% detection of analyzed samples), which

correspond to the monotonic relationships between a PFAS's importance and the ordination axes.

Table 1.

List of 24 targeted PFAS analyzed in marine organism tissues along with sub-class designation (PFCA = perfluoroalkyl carboxylic acid, PFSA = perfluoroalkyl sulfonic acid, FASA = perfluoroalkane sulfonamide, FASAA = perfluoroalkane sulfonamidoacetic acid, FTS = n:2 fluorotelomer sulfonate, short = short-chain, long = long-chain), CAS registry number (CAS RN), concentration ranges (ng/g wet weight), and rates of detection at/above method detection limits (MDL) for all samples (*n* = 344).

Native PFAS name	Abbreviation	PFAS sub-class	CAS RN	Concentration range (ng/g wet weight)	% Detection at/above MDL
Perfluorobutanoic acid	PFBA	PFCA (short)	375-22-4	<MDL – 1.01	4%
Perfluoropentanoic acid	PFPeA	PFCA (short)	2706-90-3	<MDL – 0.81	2%
Perfluorohexanoic acid	PFHxA	PFCA (short)	307-24-4	<MDL – 2.34	15%
Perfluoroheptanoic acid	PFHpA	PFCA (short)	375-85-9	<MDL – 0.88	7%
Perfluorooctanoic acid	PFOA	PFCA (long)	335-67-1	<MDL – 1.39	16%
Perfluorononanoic acid	PFNA	PFCA (long)	375-95-1	<MDL – 1.40	30%
Perfluorodecanoic acid	PFDA	PFCA (long)	335-76-2	<MDL – 1.73	25%
Perfluoroundecanoic acid	PFUnDA	PFCA (long)	2058-94-8	<MDL – 3.91	74%
Perfluorododecanoic acid	PFDoDA	PFCA (long)	307-55-1	<MDL – 1.48	38%
Perfluorotridecanoic acid	PFTrDA	PFCA (long)	72629-94-8	<MDL – 16.64	82%
Perfluorotetradecanoic acid	PFTeDA	PFCA (long)	376-06-7	<MDL – 2.47	64%
Perfluorobutane sulfonic acid	PFBS	PFSA (short)	375-73-5	<MDL – 0.64	4%
Perfluoropentane sulfonic acid	PFPeS	PFSA (short)	2706-91-4	<MDL – 0.12	<1%
Perfluorohexane sulfonic acid	PFHxS	PFSA (long)	355-46-4	<MDL – 1.15	4%
Perfluoroheptane sulfonic acid	PFHpS	PFSA (long)	375-92-8	<MDL	0%
Perfluorooctane sulfonic acid	PFOS	PFSA (long)	1763-23-1	<MDL – 7.48	31%
Perfluorononane sulfonic acid	PFNS	PFSA (long)	68259-12-1	<MDL	0%
Perfluorodecane sulfonic acid	PFDS	PFSA (long)	335-77-3	<MDL – 0.96	9%
Perfluorooctane sulfonamide	FOSA	Precursor (FASA)	754-91-6	<MDL – 3.71	49%
N-methylperfluorooctane sulfonamidoacetic acid	N-MeFOSAA	Precursor (FASAA)	2355-31-9	<MDL – 0.46	7%
N-ethylperfluorooctane sulfonamidoacetic acid	N-EtFOSAA	Precursor (FASAA)	2991-50-6	<MDL – 1.17	2%
1H, 1H, 2H, 2H-perfluorohexane sulfonic acid	4:2-FTS	Precursor(FTS)	757124-72-4	<MDL	0%
1H, 1H, 2H, 2H-perfluorooctane sulfonic acid	6:2-FTS	Precursor(FTS)	27619-97-2	<MDL	0%
1H, 1H, 2H, 2H-perfluorodecane sulfonic acid	8:2-FTS	Precursor(FTS)	39108-34-4	<MDL	0%

Table 2.

Summary statistics for three-way permutational multivariate analysis of variance model used to examine differences in fish and invertebrate PFAS chemical profiles as a function of feeding guild, habitat, and location. Bold text indicates statistical significance at $\alpha = 0.05$.

Factor	pseudo-*F* (df)	*p*-value
Combined habitats (3-way PERMANOVA)		
Habitat × Feeding guild × Location	1.02 (2)	0.392
Habitat × Feeding guild	**5.79 (2)**	**< 0.001**
Habitat × Location	**2.67 (5)**	**0.006**
Feeding guild × Location	0.91 (8)	0.539
Habitat	**3.99 (1)**	**0.021**
Feeding guild	1.02 (2)	0.379
Location	**3.18 (2)**	**0.011**
Pelagic habitat (2-way PERMANOVA)		
Feeding guild × Location	1.21 (2)	0.253
Feeding guild	**3.29 (2)**	**0.014**
Location	**10.8 (3)**	**< 0.001**
Demersal habitat (2-way PERMANOVA)		
Feeding guild × Location	1.43 (2)	0.223
Feeding guild	2.18 (1)	0.105
Location	1.26 (3)	0.257
Benthic habitat (2-way PERMANOVA)		
Feeding guild × Location	0.78 (6)	0.696
Feeding guild	**2.50 (2)**	**0.034**
Location	1.58 (3)	0.129

Table 3.

Relationship of individual PFAS concentration vs. a.) corrected $\delta^{13}C$, and b.) trophic level (TL) using either parametric regression by Maximum Likelihood Estimation (MLE) or nonparametric regression by Akritas-Theil-Sen line (ATS). For b.), trophic magnification factor (TMF) was derived as e^x. Models were only fit to PFAS with ≥20% detection rate for the dataset where data below method detection limits were treated as censored; $n = 340$. TLs range 1.65–4.97.

a.)

Compound	% detect (ALL)	Method: MLE *or* ATS	Model parameters: ln(PFAS) = $x(\delta^{13}C)+y$		MLE: Rescaled likelihood ratio *or* ATS: Kendall's Tau	*p*-value (significant at α = 0.1*, 0.05**, 0.01***)
			x	*y*		
FOSA	49%	MLE	−0.11	−3.6	−0.16	0.0048***
PFNA	31%	MLE	0.065	−1.3	0.084	0.17
PFDA	26%	MLE	−0.012	−2.3	−0.018	0.78
PFUnDA	74%	MLE	0.14	1.6	0.24	<0.001***
PFDoDA	38%	MLE	0.17	1.2	0.26	<0.001***
PFTrDA	82%	ATS	0.29	5.1	0.30	<0.001***
PFTeDA	64%	ATS	0.32	4.0	0.25	<0.001***
PFOS	32%	MLE	−0.014	−1.7	−0.016	0.79

b.)

Compound	% detect (ALL)	Method: MLE *or* ATS	Model parameters: ln(PFAS) = x(TL)+y		MLE: Rescaled likelihood ratio *or* ATS: Kendall's Tau	*p*-value (significant at α = 0.1*, 0.05**, 0.01***)	TMF = e^x
			x	*y*			
FOSA	49%	MLE	0.24	−2.3	0.11	0.042**	1.3
PFNA	31%	MLE	0.12	−3.0	0.053	0.39	1.1
PFDA	26%	MLE	0.55	−4.0	0.29	<0.001***	1.7
PFUnDA	74%	MLE	0.096	−1.4	0.053	0.35	1.1
PFDoDA	38%	MLE	0.18	−2.6	0.094	0.12	1.2
PFTrDA	82%	ATS	−0.15	0.12	−0.052	0.15	0.86
PFTeDA	64%	ATS	−0.26	−1.0	−0.071	0.051*	0.77
PFOS	32%	MLE	0.36	−2.7	0.14	0.020**	1.4