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No place to hide: Marine habitat does not determine per- and polyfluoroalkyl substances (PFAS) in odontocetes

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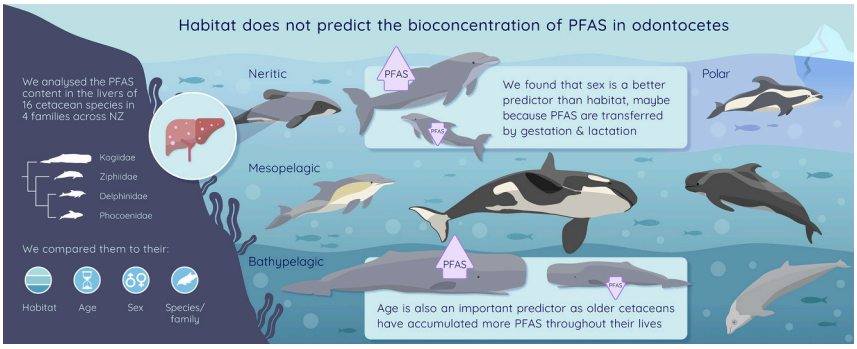
HIGHLIGHTS

- PFAS assessment of 16 odontocetes representing four families
- Comparison of neritic, mesopelagic, bathypelagic and polar species
- Marine habitat is a weak predictor of PFAS in odontocetes
- Sex and age best explain PFAS burden detected across all species

GRAPHICAL ABSTRACT



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ABSTRACT

As meso- and apex predators in food webs, marine mammals can bioconcentrate persistent environmental contaminants like per- and polyfluoroalkyl substances (PFAS). Although the presence of PFAS is widely reported in the marine environment, there is a lack of data for cetaceans in Oceania. We investigated whether ecological habitat influences bioconcentration patterns across a range of odontocete (toothed whale, dolphin and porpoise) species. We measured PFAS in liver samples (n = 127) from 16 cetacean species representing four families inhabiting four marine habitats along the Aotearoa New Zealand coastline. We analysed six perfluoroalkyl carboxylic acids, ten perfluoroalkyl sulphonic acids and four precursor compounds in the context of sex, body index, habitat and species/family using generalized linear mixed models. Results showed that marine habitat remained a weak predictor of PFAS burden. Instead, biological factors including sex and age class best explained the levels of PFAS detected across all species and habitats. We offer first important insights on PFAS levels across several new taxa globally, including endemic endangered species and poorly described polar vagrants. Our findings further highlight how the ubiquitous nature of PFAS pose a higher risk to odontocetes across different seascapes than previously anticipated.

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1. Introduction

Per- and polyfluoroalkyl substances (PFAS) are a class of anthropogenic chemicals (Buck et al., 2011). As emerging contaminants of concern, PFAS pose a risk and are found in the environment, although their impacts on biota and humans remain to be fully characterised (Wang et al., 2024). Based on the latest PFAS definition by the Organisation for Economic Co-operation and Development (OECD), more than 14,000 PFAS chemical structures have been documented to date (Wang et al., 2021; Richard et al., 2023). These include perfluoroalkyl acids (PFAAs) such as perfluoroalkyl carboxylates (PFCAs) and perfluoroalkyl sulfonates (PFSAs), as well as their precursors, a group of polyfluoroalkyl substances that can be transformed to PFAAs. Both PFAAs and their precursors have been detected in various marine environments including seawater, sediment and biota (Ahrens et al., 2009; Ahrens and Bundschuh, 2014). The primary sources and pathways of PFAS released into the marine environment include urban and agricultural runoffs, discharges from industrial and manufacturing processes, effluents from wastewater treatment plants and atmospheric deposition into the oceans (Sunderland et al., 2019), with highest levels typically reported near industrial areas, wastewater discharge points, and urbanized coastal regions (Zhou et al., 2013; Ahrens and Bundschuh, 2014). Due to the persistence of PFAS, oceans are assumed to be the final sink of PFAS in the environment (Yamashita et al., 2004).

While PFAS in marine environmental media and wildlife have been reported (for comprehensive reviews see Houde et al., 2011; Jian et al., 2018; Sunderland et al., 2019; Wang et al., 2019), current understanding of how environmental PFAS accumulate in tissues across species occupying different marine environments and niches remains limited (Dassuncao et al., 2019; De Silva et al., 2021). PFAS tend to bioaccumulate in phospholipids and protein-rich tissues, with phospholipids being significant predictors of tissue distribution for long-chain PFASs — those with perfluorocarbon chains longer than nine carbons — such as perfluorodecanesulfonate (PFDS), perfluorododecanoate (PFDoA), perfluorotridecanoate (PFTrA), and perfluorotetradecanoate (PFTA) in marine mammals (Dassuncao et al., 2019). Factors which may influence PFAS bioaccumulation include the carbon chain length of the molecule (long-chain PFAS tend to bioaccumulate more than short-chain counterparts), feeding ecology (predatory species exhibit higher PFAS levels due to biomagnification) and metabolic capacity (Khan et al., 2023). For example, organisms with limited ability to excrete PFAS may transform these molecules to more persistent and sometimes toxic forms (Miranda et al., 2022).

For marine mammals, it remains largely unknown how contaminant burdens correlate with their habitat or trophic position. The occurrence of PFAAs and their precursors can vary significantly in their chemical profile with water column depth and distance from input sources. Many of these compounds have high water solubility and low lipophilicity, resulting in unique long-range transport and cycling processes in the marine environment. Studies have shown that in the Mediterranean Sea and the Sea of Japan, several differences were found in the profiles of PFAS in the water column (horizontal profiling) (Yamazaki et al., 2019). Further studies collectively indicate that PFAS are found at higher concentrations in shallower coastal waters, primarily due to inputs from atmospheric deposition, riverine inflows, and coastal activities (Yamashita et al., 2008; Ahrens et al., 2010; Zhao et al., 2015).

Pelagic and bathypelagic odontocetes (toothed whales, dolphins and porpoises) feed at different depths within the water column in addition to at different trophic levels (Stockin et al., 2022; Peters et al., 2022). Accordingly, it is unclear if and/or how chemical profile differences in the marine environment may affect species feeding within different foraging ecology. This is especially pertinent within Oceanian waters, an area of high cetacean biodiversity, yet for which little knowledge on PFAS distribution is available. For example, limited data exist on the presence of PFAS in the New Zealand environment, in part because no industrial production of PFAS has occurred in the area. While a recent New Zealand study indicates that the concentrations of three PFAAs — perfluorooctane sulfonate (PFOS), perfluorohexane sulfonate (PFHxS), and perfluorooctanoic acid (PFOA) — in urban coastal waters were below the local guideline values for drinking water and recreational water (Lenka et al., 2022), no data on PFAS in the New Zealand marine environment, and especially in the context of water column depth and proximity to input sources, currently exists.

Here, we characterise PFAS tissue residue concentrations across multiple odontocete species which inhabit distinct marine habitats. New Zealand hosts more than 50% of the world's cetacean species, including 35 odontocetes (Stephenson et al., 2020), which, as apex- and mesopredators, play a crucial part in maintaining ecosystem health (Roman et al., 2014). Due to New Zealand's latitudinal expanse and isolated geographical location, odontocete species occupy a range of different habitats, ranging from shallow coastal and shelf areas, over deep ocean trenches, to subantarctic waters (Stevens et al., 2019). Specifically, we explore potential interactions between contaminants and the marine seascape (neritic, mesopelagic, bathypelagic; see S1 Supplementary Materials) in which odontocetes feed. We hypothesize that the ecological zone in which Delphinidae (dolphin) and Ziphiidae (beaked whale) species forage influences PFAS accumulation patterns. To test this hypothesis, we developed generalized linear mixed models (GLMMs) to determine the main predictors (i.e., habitat zone, sex, age) influencing the PFAS bioaccumulation patterns across species.

2. Materials and methods

2.1. Sample collection and storage

We sampled 127 animals across 16 species within Aotearoa, New Zealand, comprising four families (Delphinidae, Ziphiidae, Phocoenidae, and Kogiidae) which inhabit four different marine habitats (neritic, mesopelagic, bathypelagic and polar waters, as previously defined by Peters et al. (2022; S1 Supplementary Material)). Sampled animals originated from beach cast carcasses between 2009 and 2022. Only carcasses in good or moderate body condition and fresh to mild decomposition were included, as defined in Stockin et al. (2009). Of the 16 species examined, majority belonged to Delphinidae (n = 9) and Ziphiidae (n = 4). Following Stockin et al. (2021), we recorded sex, total body length, nutritive condition and decomposition for each specimen and sampled liver tissues from each carcass using a stainless-steel knife. Each sample was wrapped in aluminium foil prior to storage at −20 °C until subsequent contaminant analyses.

2.2. Habitat stratification

PFAS concentrations and profiles were characterised across species (n = 16) and habitat (n = 4), derived from the best-known habitat and distribution of each species within New Zealand waters (Stephenson et al., 2020; Peters et al., 2022). Species were assigned to habitat zones as follows: *neritic* (bottlenose dolphin *Tursiops truncatus*, Hector's dolphin *Cephalorhynchus hectori*); *mesopelagic* (common dolphin *Delphinus delphis*, dusky dolphin *Aethalodelphis obscurus*, killer whale *Orcinus orca*, long-finned pilot whale *Globicephala melas edwardii*, pygmy killer whale *Feresa attenuata*, pygmy sperm whale *Kogia breviceps*, striped dolphin *Stenella coeruleoalba*); *bathypelagic* (Gray's beaked whale *Mesoplodon grayi*, Cuvier's beaked whale *Ziphius cavirostris*, Arnoux's beaked whale *Berardius arnouxii* and straptoothed beaked whale *M. layardii*). As spectacled porpoise *Phocoena dioptrica* (n = 5) and hourglass dolphin *C. cruciger* (n = 1) are both vagrant species, we added these to a separate *polar* category.

2.3. Per- and polyfluoroalkyl substances (PFAS) analysis

2.3.1. Sample extraction and quality control

Hepatic tissue samples were extracted and analysed for PFAS by

AsureQuality Laboratories, Wellington, NZ, using validated in-house methods and quality control procedures, as previously described in Stockin et al. (2021). In brief, frozen liver samples were thawed at 2–5 °C, then allowed to warm to room temperature. Samples were subsequently homogenised using a Waring blender. Approximately 1 g test portion was extracted from the blended material, and an internal standard mix was incorporated. Acidified acetonitrile was added, and each test portion was further homogenised using ceramic homogeniser pellets and a Merris Minimix vibrational shaker for 8 min at 50 Hz. A portion of the extract was cleaned up by dispersive solid phase extraction (SPE) using graphitised carbon, C18 adsorbent, and zirconia-coated silica. Extracts were prepared in a 50:50 methanol/water solvent mixture prior to LC-MS/MS analysis. As it was not possible to obtain blank samples suitable to determine the limit of detection (LOD), the limit of reporting (LOR) was assigned as the equivalent to the lowest calibration standard, which was 0.5 ng/g.

Quality control (QC) procedures were compliant with requirements of the US DoD/DoE QSM 5.1.1 (2018). Each analysis batch included an ongoing precision and recovery (OPR) sample and an OPR blank. The OPR sample was prepared with analytical standards at a concentration of 1–2× of the method LoR. Each analysis batch also included a reagent blank (i.e., no matrix, but all the reagents and processes used in the extraction procedure). One submitted sample per analysis batch was randomly selected by the laboratory for spiking with the target analytes at the OPR concentration before and after extraction to assess the impact of matrix effects on the recovery of target analytes. Each analysis batch also included a randomly selected duplicate test sample.

2.3.2. Analytical methods

We analysed extracted samples for PFAS by AsureQuality Laboratories, Wellington, NZ, using validated in-house ESI-LC-MS/MS methods, as reported by Stockin et al. (2021) (S2 Supplementary Materials). Results were reported independently for the linear isomers and for two groups of branched isomers (di-methyl and mono-methyl) for PFOS and PFHxS. Total branched PFOS (B.PFOS) or total branched PFHxS (B.PFHxS) represents the numerical sums of the two measured branched isomers for each compound.

2.4. Data analysis

To assess which predictors (i.e., habitat zone, sex, and age) best explain the variation in PFAS contaminant concentration patterns across species, we built and compared generalized linear mixed models (GLMMs). We tested seven metrics characterizing PFAS concentration (in ng/g wet weight) as response variables: (1) ∑PFAS, (2) ∑PFCAs, (3) ∑PFSAs, (4) ∑Precursors, and the percentages of (5) ∑PFCAs, (6) ∑PFSAs, and (7) ∑Precursors relative to the total PFAS burden.

For each response variable, we fitted a specific GLMM to test each relationship with the predictors sex (male and female), habitat zone (neritic, mesopelagic, bathypelagic, and polar), and total body length as a proxy for age. To ensure comparability across species, we calculated a body size index (numerical, between 0 and 1) for each animal by standardizing the total body length for each sex and species based on the specific minimum length at birth and maximum adult length (see S3; Supplementary Materials).

GLMMs are preferred over classical statistical methods because they not only accommodate multiple predictors and their interactions simultaneously but also account for non-independence in the response variable (i.e., PFAS bioaccumulation). Indeed, PFAS concentrations among individuals of the same species are likely correlated, leading to potential pseudo-replication. We calculated the intraclass correlation coefficient and design effect to confirm that >30% of the total variance in our response variables was due to species clustering, and omitting this effect would inflate our sample size by a factor greater than four. We addressed these clustering effects by denoting species as a random effect in the GLMMs. Fixed effects quantified overall effects across all species, while random effects quantified variation across species (Bolker et al., 2009).

We fitted a total of 49 GLMMs using the 'lme4' and 'glmmTMB' packages in R version 4.3.0 (Brooks et al., 2017) (see model description in Tables S1, S2). We computed Akaike's information criterion weights (corrected for small sample size: wAICc) for each model to rank them and identify the top-ranked model. We also calculated the information-theoretic evidence ratio to quantify the relative support of the highest-ranked model against the others. Four species containing only one individual were excluded from the analysis: Arnoux's beaked whale, Cuvier's beaked whale, strap-toothed beaked whale and hourglass dolphin.

3. Results

3.1. Samples

Of the 127 animals tested, more than 75% of samples (n = 95) originated from nine species of Delphinidae (comprising eight genera: *Cephalorhynchus*, *Delphinus*, *Feresa*, *Globicephala*, *Aethalodelphis*, *Orcinus*, *Tursiops* and *Stenella*; see S4, Supplementary Materials). This sampling effort reflects the taxonomic and ecological diversity within Delphinidae, which covers species across all major marine habitats, including polar and bathypelagic environments. A further 11% of samples (n = 14), including four species across three genera (*Mesoplodon*, *Ziphius* and *Berardius*) originated from the family Ziphiidae (commonly known as beaked whales). This group comprised a diverse group of cetaceans known for their bathypelagic ecology, including prolonged diving capabilities to undertake deep sea foraging (Shearer et al., 2019; Alcázar-Treviño et al., 2020). The remaining samples (n = 17) represent three species across *Phocoenidae*, *Kogiidae* and *Physeteridae*, covering polar, mesopelagic and bathypelagic environments. Across 16 odontocetes examined here, we provide the first global insights into PFAS burden for eight new species, including Hector's dolphin (n = 22), spectacled porpoise (n = 5), pygmy sperm whale (n = 11) and three species of beaked whales (n = 14).

3.2. PFAS profile by habitat group

Across all samples, minimum and maximum ∑PFAS concentrations ranged up to 146.5 ng/g. Minimum and maximum ∑PFAS concentrations ranged from 0.84 to 66.3 ng/g in bathypelagic (mean = 14.4, SD = 17.2; n = 15; all mean and SD concentrations are present in ng/g wet weight) and 3 to 146.5 ng/g in mesopelagic (mean = 37.6, SD = 18.2; n = 71) species, respectively (Fig. 1). Minimum and maximum ∑PFAS concentrations reported in neritic and polar ranged from 2.1 to 111 ng/g (mean = 30.9, SD = 26.7; n = 34) and 9.2 and 113 (mean = 32.3, SD = 41.3; n = 6), respectively (Fig. 1).

PFAS profiles varied between habitats (Fig. 2) with ∑PFSA accounting for the highest proportion of ∑PFAS across all but the bathypelagic habitat (Fig. 2). Within the bathypelagic group, mean ∑PFCA accounted for 55.9% of ∑PFAS (mean = 7.5, SD = 8.9; n = 15) compared with neritic habitat which accounted for 16.3% of ∑PFAS (mean = 5.3, SD = 3.7; n = 34). ∑PFCA accounted for 36.4% (mean = 15.1, SD = 9.0; n = 71) and 22.7% (mean = 8.5, SD = 13; n = 6) of ∑PFAS mesopelagic and polar habitats, respectively. Conversely, ∑PFSA formed the largest component of ∑PFAS in the neritic habitat (56.9%, mean = 16.9, SD = 16.5, n = 34), with least ∑PFSA reported in bathypelagic (30.8%, mean = 4.4, SD = 4.7; n = 15) species.

Detected precursors, mostly PFOSA, were most prominent in the species from polar habitat, accounting for 33.8% (mean = 7.7, SD = 1.3, n = 6; Fig. 2) of ∑PFAS. In contrasts, bathypelagic species exhibited the lowest proportion of precursors (13.4%, mean = 2.5, SD = 4.8; n = 15). Both neritic (26.7%, mean = 8.7, SD = 10.1; n = 34) and mesopelagic (22.7%, mean = 7.1, SD = 3.1; n = 71; Fig. 2) species demonstrated a similar proportion of the precursors with their ∑PFAS profile.

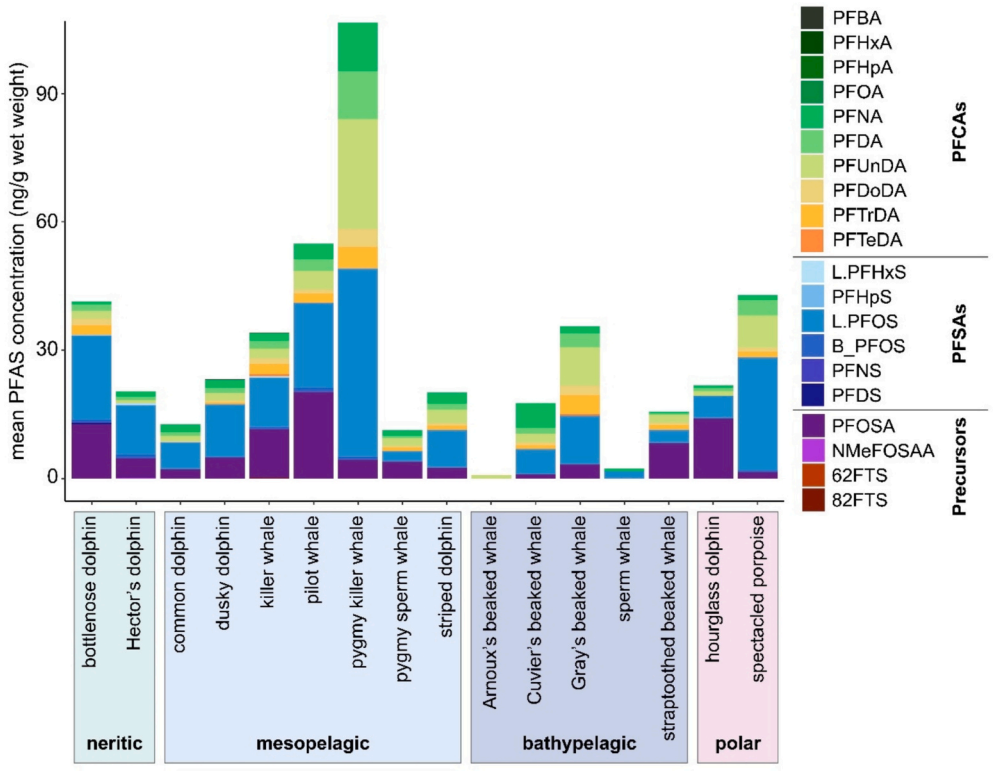

Fig. 1. Mean PFAS concentration (ng/g wet weight) by species and habitat for 16 odontocetes (n = 126) stranded between 2009 and 2022 in Aotearoa, New Zealand.

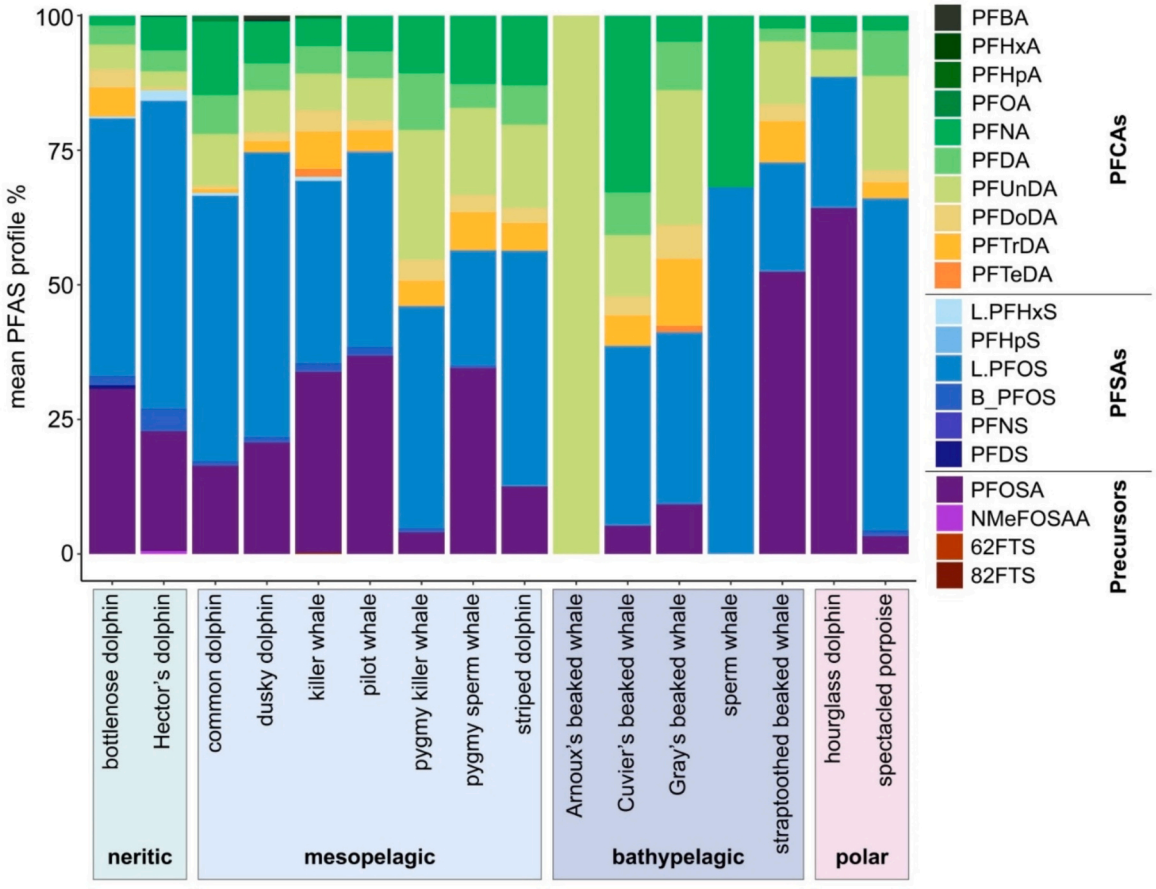

Fig. 2. Mean PFAS profile (%) by species and habitat for 16 odontocetes (n = 126) stranded between 2009 and 2022 in Aotearoa, New Zealand.

3.3. PFAS profile by species

A total of 127 tissue samples across 16 species were examined for ∑PFAS concentration and profile (Fig. 1). One outlier (Hector's dolphin H296: a 75.5 cm male with ∑PFAS = 420 ng/g) was removed prior to analysis owing to its burden exceeding four-fold that of all other samples measured. Statistical analysis was conducted using the remaining 126 samples.

3.3.1. Delphinidae

While the maximum ∑PFAS concentration of 111 ng/g within Delphinidae was detected in a Hector's dolphin (excluding outlier H296), mean ∑PFAS concentrations for *Cephalorhynchus* genus ranked only seven out of nine when considering mean ∑PFAS across the wider Delphinidae family (mean = 20.4, SD = 25.1; n = 22). Instead, pygmy killer whale (mean = 106.7, SD = 52.0; n = 3), pilot whale (mean = 54.9, SD = 19.4; n = 18), bottlenose dolphin (mean = 41.4, SD = 28.4; n = 12) and killer whale (mean = 34.1, SD = 15.4; n = 6) ranked highest for mean ∑PFAS concentrations, while common dolphin (mean = 12.7, SD = 15.9; n = 12) reported lowest mean ∑PFAS concentrations within Delphinidae (Fig. 1).

Total PFSA concentrations (∑PFSAs) accounted for the largest component of ∑PFAS in all but two genera of the Delphinidae family (Fig. 2), exceptions being pygmy killer whale (∑PFAS; 54.0%, PFCA; n = 3) and hourglass dolphin (∑Precursors 64.3%; n = 1).

∑PFCAs concentrations ranged from 0 to 78.2 ng/g. Lowest concentrations of mean ∑PFCAs were recorded in hourglass dolphin (mean = 2.5; n = 1), Hector's dolphin (mean = 2.8, SD = 3.7; n = 22) and common dolphin (mean = 4.2, SD = 7.6, n = 12; Fig. 1). Meanwhile highest concentrations of ∑PFCA were observed in pygmy killer whale (mean = 57.6, SD = 28.0, n = 3), long finned pilot whale (mean = 13.9, SD = 8.1; n = 18) and killer whale (mean = 10.2, SD = 7.4; n = 6; Fig. 1).

Individuals across Delphinidae also varied in ∑PFSA, with Hector's dolphins demonstrating both the lowest and highest concentrations, ranging from 0 to 78.0 ng/g (mean = 12.9, SD = 18.2; n = 22; Fig. 1). Second and third highest reported ∑PFSA concentrations were observed in pilot whale, ranging from 4.4 to 47.0 (mean = 20.8, SD = 13.7; n = 18) and bottlenose dolphin ranging from 4.7 to 44.3 ng/g (mean = 20.9, SD = 14.7; n = 12; Fig. 1).

Precursor compounds within Delphinidae ranged from 0 to 58.0 ng/g, with highest mean total precursors recorded in pilot whale (mean = 20.2, SD = 5.1, n = 18), killer whale (mean = 11.5, SD = 8.0; n = 6) and bottlenose dolphin (mean = 12.7, SD = 15.0; n = 12; Fig. 1). Lowest mean total precursors were recorded in common dolphin (mean = 2.1, SD = 0.8; n = 12) and striped dolphin (mean = 2.5, SD = 2.3, n = 9; Fig. 1).

3.3.2. Ziphiidae

The maximum ∑PFAS concentrations of 66.3 and 32.0 ng/g within Ziphiidae were reported in Gray's (mean = 35.6, SD = 22.1; n = 8) and strap-toothed whales (mean = 15.6, SD = 12.4; n = 4), respectively.

In contrast to Delphinidae, ∑PFCAs accounted for the largest proportion of ∑PFAS in all but one member of Family Ziphiidae (Fig. 2), the strap-toothed beaked whale (n = 4) where precursor compounds, mostly PFOSA composed 52.4% of ∑PFAS. N-MeFOSAA and 8:2FTS were each detected in only one animal (Fig. 2, Table 1).

Beaked whales ranged in ∑PFCA from 0.8 to 41.2 ng/g for Arnoux's (n = 1) and Gray's beaked whales (mean = 20.9, SD = 14.4; n = 8), respectively. Mean ∑PFCA were not possible to explore for either Arnoux's or Cuvier's beaked whales given only one sample originated from each species. However, Gray's beaked whales demonstrated higher mean ∑PFCA (mean = 20.9, SD = 14.4; n = 8) compared to strap-toothed whales (mean = 4.3, SD = 3.4; n = 4).

Individuals across the species of Ziphiidae ranged in ∑PFSA, with Gray's beaked demonstrating the highest concentrations of 22.6 ng/g (mean = 11.4, SD = 7.7; n = 8), while no PFSA were detected in the Arnoux's beaked whale (Table 1). Detected precursor compounds comprised less than 10% of ∑PFAS in all but one examined beaked whale, the exception being strap-toothed whale which composed a surprising 52.4% of ∑PFAS concentration, ranging from 2.1 to 20.0 ng/g (mean = 8.2, SD = 8.1; n = 4; Fig. 1).

3.3.3. Phocoenidae, Kogiidae and Physeteridae

∑PFAS concentrations in pygmy sperm (Kogiidae) and sperm whales (Physeteridae) ranged from 3.5 to 24.5 ng/g (mean = 11.3, SD = 6.6, n = 11) and 2.4 ng/g (n = 1), respectively, aligning most with beaked whale and other bathypelagic species (Fig. 1). Conversely, spectacled porpoise (Phocoenidae) ∑PFAS concentrations ranged from 9.2 to 113.1 ng/g (mean = 42.9, SD = 41.3, n = 5), aligning most with mesopelagic delphinids (Fig. 1). However, as the only true porpoise within this study, the spectacled porpoise provides a useful comparison to other members of Phocoenidae, especially the harbour porpoise (*Phocoena phocoena*) whose PFAS burdens have been assessed internationally (Trimmel et al., 2021).

In the case of spectacled porpoise and sperm whale, PFSAs were the dominant group of PFAS accounting for 62.7% (mean = 26.9, SD = 28.4, n = 5) and 68.1% (1.6 ng/g, n = 1) of ∑PFAS, respectively (Fig. 2). However, this trend was reversed for pygmy sperm whales, whose profile was dominated by PFCA (43.6%, mean = 4.9, SD = 3.8, n = 11), with precursor compounds also making a significant contribution (34.5%; mean = 3.9, SD = 2.4; n = 11) to the ∑PFAS profile (Table 1, Fig. 2).

3.4. Model results

The top-ranked generalized linear mixed models for (i) ∑PFAS, (ii) ∑PFCA, (iii) ∑PFSA, and (iv) ∑Precursors included sex (male or female), body size index (proxy for age), and species (Table 1). Each highest-ranked model shows a difference between males and females in all PFAS concentrations except for ∑PFCA of mean PFAS profile. The positive coefficient for sex indicates that males have higher PFAS concentrations than females. Conversely, the models show a negative relationship between age class (derived from body size index) and PFAS concentration in all cases except for precursors, indicating non-precursor PFAS decreases with age class (Fig. 3). Models including habitat (represented as group) provide much lower support to explain the PFAS concentration relative to the top-ranked models (i.e., ER including "group" > 2 across models, refer to Supplementary Materials).

4. Discussion

PFAS levels in New Zealand odontocetes were not influenced by habitat. This finding was unexpected given that PFAS exposure and accumulation often varies between inshore and offshore marine species due to differences in habitat, diet, and proximity to pollution sources (Houde et al., 2006a; Gewurtz et al., 2013; Sunderland et al., 2019). For example, inshore harbour seals (*Phoca vitulina*) in the North Atlantic demonstrate high PFAS burdens compared to their offshore counterparts due to greater exposure to contaminated runoff and sediments (Shaw et al., 2008). These factors combined with restricted mobility can also lead to elevated risks of PFAS accumulation in neritic species (Yamashita et al., 2005; Rumsby et al., 2009). Conversely, dynamic water movements that are characteristic of offshore habitats are likely to dilute PFAS concentrations lowering accumulation risks that may be more pronounced in static inshore environments (Yamashita et al., 2005; Loi et al., 2011; Munoz et al., 2017).

Odontocetes live and feed in nutritionally complex, three-dimensional marine environments. The vertical distribution of PFAS within the water column is complex and affected by an interplay of chemical (e.g., hydrophilicity, persistency and stability) and physical conditions (water mixing and stratification) that are characteristic of aquatic ecosystems (Giesy and Kannan, 2001; Lau et al., 2007;

Table 1
Performance values for the top-ranked generalized linear mixed models. These top-ranked generalized linear mixed models contain Sex (male or female), Group (neritic, mesopelagic, bathypelagic, and polar), Index (body size index used as a proxy for age), and Species to describe for each individual the concentration in (*i*) ∑PFAS, (*ii*) ∑PFCA, (*iii*) ∑PFSA and (*iv*) ∑Precursors. Shown are the summary statistics for the top-ranked generalized linear mixed: coefficients estimated for each predictor variable of the top-ranked generalized linear mixed, standard error (SE), and the probability (Pr) that the observed effect would occur by chance. Also shown are the number of parameters (*k*), minimized negative log-likelihood (LogLik), weight scaled to a sum of 1 ($wAIC_c$) and the Akaike's information criterion corrected for small sample sizes (AIC_c).

Model	Var.	Coefficients			k	AIC_c	$wAIC_c$	LogLik
		Value	SE	Pr				
Burden								
∑PFAS ~ Sex + Index + (Sex | Species)	Int.	3.66	0.26	<0.001	7	992.11	0.95	−488.56
	Sex	0.33	0.26	0.21				
	Index	−0.91	0.24	<0.001				
∑PFCA ~ Sex + Index + (1 | Species)	Int.	3.02	0.38	<0.001	5	743.92	0.63	−366.63
	Sex	0.42	0.20	0.03				
	Index	−1.74	0.43	<0.001				
∑PFSA ~ Sex + Index + (1 | Species)	Int.	3.23	0.30	<0.001	5	833.97	0.87	−411.72
	Sex	0.52	0.15	<0.001				
	Index	−1.72	0.31	<0.001				
∑Prec ~ Sex + Index + (1 | Species)	Int.	1.17	0.33	<0.001	5	683.93	0.27	−336.71
	Sex	0.29	0.15	0.056				
	Index	0.51	0.33	0.12				
Profile								
∑PFCA ~ Index + (1 | Species)	Int.	3.85	0.17	<0.001	5	956.31	0.43	−472.90
	Index	−0.68	0.17	<0.001				
∑PFSA ~ Sex + Index + (1 | Species)	Int.	3.96	0.12	<0.001	6	965.99	0.67	−476.63
	Sex	0.14	0.05	0.003				
	Index	−0.58	0.10	<0.001				
∑Prec ~ Sex + Index + (1 | Species)	Int.	2.01	0.28	<0.001	6	972.58	0.57	−479.92
	Sex	−0.18	0.08	0.03				
	Index	1.61	0.19	<0.001				

Yamashita et al., 2008; Ahrens et al., 2010; Ahrens, 2011).

Environmental conditions including temperature, salinity and pH can also influence PFAS distribution (McMurdo et al., 2008; Thompson et al., 2011; Washington et al., 2020). However, our models showed that habitat did not predict PFAS burdens (Table 1), suggesting that different marine environments have little influence on PFAS levels within our sampled odontocetes. Instead, the variance observed between PFAS burdens and profiles appears to be driven by sex and age class (Table 1), indicating physiological and life history parameters as key drivers of PFAS accumulation in New Zealand odontocetes. This is consistent with previous findings that demonstrated the influence of age-related accumulation, developmental stage and reproductive history in contaminant burdens expressed in odontocetes (Bossi et al., 2005; Houde et al., 2006b; Stockin et al., 2023). While older animals are known for exhibiting higher PFAS concentrations due to prolonged exposure and bioaccumulation than younger individuals (Houde et al., 2006b), differences in metabolism, reproductive biology, feeding habits, and exposure routes determine the ability of an organism to assimilate, transform and potentially eliminate the contaminants.

Sex and age trends in relation to PFAS and stress biomarker relationships have been identified in bottlenose dolphins (Bennett et al., 2024), which may imply a sex-specific physiological phenomenon (i.e., offloading during birth or nursing), including maternal transfer via gestation and lactation, reducing the body burden of PFAS in breeding females. This also aligns with juveniles who often demonstrate higher PFAS levels due to both maternal transfer and their developing physiology (Lee et al., 2023). Distinct physiological stages can lead to different metabolic rates and storage capacities compared to adults (Houde et al., 2006a). Age and sex influences have been highlighted as important in the PFAS levels reported in other cetacean species. For example, PFAS concentrations in adult female harbour porpoises (Galatius et al., 2013; Trimmel et al., 2021) were lower than males, likely related to maternal offloading. Sexually dimorphic foraging strategies can also lead to differential exposure to PFAS resulting in different burdens (Ross et al., 2004).

PFAS concentrations may vary among species due to differences in diet. Peters et al. (2022) investigated the isotopic niche overlap among 21 odontocete species in New Zealand (including many of the individuals examined here) and reported clear isotopic niche separation between deep-water species like Gray's beaked whales and sperm whales. However, most of the Delphinidae examined, which notably span both neritic and mesopelagic habitats, revealed high isotopic niche overlap, suggesting potential interspecific competition. Accordingly, such overlap between most of the species examined here may explain, at least in part, why we found no habitat differences in PFAS burden. However, based on the same hypothesis, we still may have expected some signal of differentiation for the bathypelagic and polar species, as was indicated by Peters et al. (2022). Although such habitat differentiation was not detected in PFAS burdens, sample sizes for both bathypelagic and polar species were considerably lower than for the other habitats owing to stranding frequency.

We acknowledge sample size may limit full interpretation of the results presented here. For example, subtle variability of PFAS between bathypelagic species may have gone undetected due to lower statistical power. Furthermore, temporal stratification of the samples may also have confounded our results. However, this is less likely given the temporal scale of this study (13 years) sits comfortably within the life span of all species examined. Likewise, as all samples originated from stranding events within New Zealand, our data represent a relatively refined spatial scale when compared to similar studies which have explored wider coastal seas and ocean basins (Ahrens et al., 2009; Ahrens and Bundschuh, 2014). Of course, migration, or at least movement patterns between habitats, may also have limited identification of trends and patterns in our results. While the 16 odontocete species included in our study are not typically considered migratory, the movement patterns of beaked whales and blackfish (pilot whales, false killer whales, killer whales) in particular, remain largely unknown in New Zealand. Accordingly, it is not possible to rule out the influence of movements between environments, though our conscious decision to exclude migratory baleen whales from this study goes some way to limit this potential bias.

One final consideration for PFAS homogeneity between habitats may

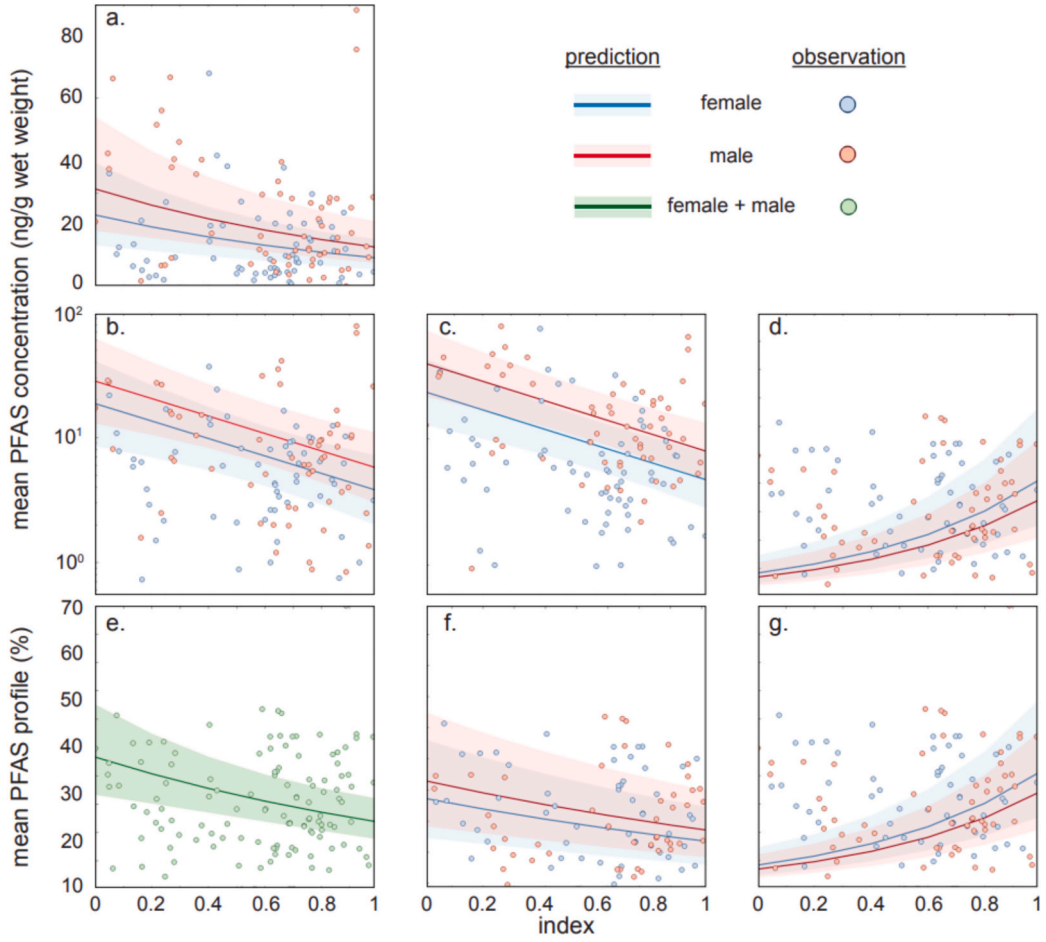

Fig. 3. Performance values for the top-ranked generalized linear mixed models for burden (a–d) and profile (e–g). ∑PFAS (a), ∑PFCA (b), ∑PFSA (c) ∑Precursors concentration (ng/g wwt) (d) and ∑PFCA (e), ∑PFSA (f) ∑Precursors profile(g) as a function of body index and sex across 16 odontocete species (n = 126) sampled between 2009 and 2022 in Aotearoa New Zealand. Details on each model's structure are provided in Table 1.

indicate a uniform pollution source or exposure pathway, e.g., through similar trophic transfer. For example, if atmospheric inputs remain the dominant source of PFAS within the seascape of Aotearoa, New Zealand, then the lack of differences in PFAS burden between habitats would make sense. Indeed, PFAS within the wider Southern Ocean was once restricted to PFOS caused by atmospheric transport of its precursors (Ahrens et al., 2010). However, climate change alongside increased PFAS entry into marine environments would surely evoke considerable change in PFAS type and concentrations within wider Oceania waters (Muir and Miaz, 2021). Further studies are therefore warranted in the wider Southern Ocean, especially since the ubiquitous nature of PFAS may have greater implications for cetaceans than previously considered.

5. Conclusions

PFAS exposure of odontocetes inhabiting different habitats along the coast of New Zealand demonstrated that PFAS variance is more likely driven by biological parameters as opposed to habitat differences. While much remains unclear about the transport and distribution of PFAS in the surrounding waters of New Zealand and the wider Southern Ocean, all cetaceans, including offshore and bathypelagic predators, face similar risk to PFAS exposure. The adverse outcomes to individuals and populations from PFAS exposure require further investigation.

CRediT authorship contribution statement

Karen A. Stockin: Conceptualization, Writing – review & editing, Writing – original draft, Resources, Project administration, Investigation, Funding acquisition, Data curation. **Katharina J. Peters:** Writing – review & editing, Writing – original draft, Visualization, Methodology, Formal analysis. **Frédérik Saltré:** Writing – review & editing, Writing – original draft, Visualization, Validation, Software, Methodology, Formal analysis. **Gabriel E. Machovsky-Capuska:** Writing – review & editing, Writing – original draft, Data curation. **Emma L. Betty:** Writing – review & editing, Writing – original draft, Methodology, Data curation. **Louis A. Tremblay:** Writing – review & editing, Writing – original draft, Investigation. **Shan Yi:** Conceptualization, Writing – review & editing, Writing – original draft, Visualization, Investigation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2025.180701.

Data availability

Data and code that support our findings are openly available at github.com/FredSaltre/PFAS_NZ and github.com/BEAM-NguraNadamari/PFAS_NZ. Data will be made available on request.

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