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Occurrence and tissue distribution of 33 legacy and novel *per*- and polyfluoroalkyl substances (PFASs) in Baikal seals (*Phoca sibirica*)

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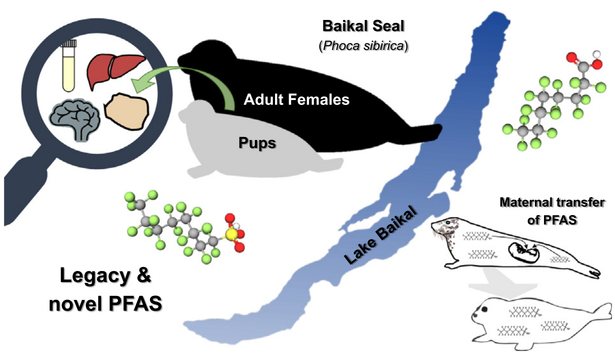
HIGHLIGHTS

- PFASs found in plasma, liver, blubber, and brain of Baikal seals.
- PFASs in suckling and pre-weaned Baikal seal pups demonstrated maternal transfer.
- PFOS and long chain PFCAs demonstrated highest concentrations in plasma and liver.
- Novel PFASs were either infrequently detected or not found in Baikal seals.
- Human consumption of Baikal seal liver and muscle can exceed current regulatory guidelines for specific PFASs.

GRAPHICAL ABSTRACT



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ABSTRACT

Per- and polyfluoroalkyl substances (PFASs) are bioaccumulative and associated with adverse effects in both wildlife and humans. The occurrence of 33 PFASs was assessed in the plasma, liver, blubber, and brain of 18 Baikal seals (*Phoca sibirica*) (16 pups and 2 adult females) from Lake Baikal, Russia (in 2011). Of the 33 congeners analysed for: perfluorooctanosulfonic acid (PFOS), 7 long chain perfluoroalkyl carboxylic acids (C_8–C_{14} PFCAs) and 1 branched PFCA (perfluoro-3,7-dimethyloctanoic acid; P37DMOA) were most frequently detected. The PFASs in plasma and liver with the highest median concentrations were legacy congeners: perfluoroundecanoic acid (PFUnA; plasma: 11.2 ng/g w.w.; liver: 7.36 ng/g w.w.), PFOS (plasma: 8.67 ng/g w.w.; liver: 9.86 ng/g w.w.), perfluorodecanoic acid (PFDA; plasma: 5.13 ng/g w.w.; liver: 6.69 ng/g w.w.), perfluorononanoic acid (PFNA; plasma: 4.65 ng/g w.w.; liver: 5.83 ng/g w.w.) and perfluorotridecanoic acid (PFTriDA; plasma: 4.29 ng/g w.w.; liver: 2.55 ng/g w.w.). PFASs were detected in the brain of Baikal seals, indicating that PFASs cross through the blood–brain barrier. In blubber, the majority of PFASs were detected in low abundance and concentrations. In contrast to legacy PFASs, novel congeners (e.g., Gen X) were either detected infrequently or not found in Baikal seals. The worldwide occurrence of PFASs in pinnipeds was compared and lower median concentrations of PFOS were observed in Baikal seals relative to other pinnipeds. Conversely, similar concentrations of long chain PFCAs were found in Baikal seals compared to other pinnipeds. Furthermore, human exposure was assessed by estimating weekly intakes (EWI) of PFASs through Baikal seal

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consumption. Although PFASs concentrations were comparatively low relative to other pinnipeds, consumption of Baikal seal could exceed current regulatory guidelines.

1. Introduction

Per- and polyfluoroalkyl substances (PFASs) are a group of organic fluorinated contaminants consisting of at least five thousand congeners produced for a multitude of industrial and commercial purposes (National Institute of Environmental Health Services, 2022). Several PFASs are persistent and known for their adverse effects in wildlife and humans. In contrast to lipophilic persistent organic pollutants (POPs), PFASs associate with proteins and phospholipids (Dassuncao et al., 2019), and similarly biomagnify in food webs. Therefore, high concentrations of PFASs are reported in top predators (Dietz et al., 2019; Houde et al., 2011), potentially causing health related effects. The immunosuppressive and endocrine disruptive effects of some PFASs are of particular concern (Dietz et al., 2019; Sunderland et al., 2019).

Lake Baikal, the world's oldest, most voluminous, and deep freshwater lake, is home to several endemic species including the Baikal seal (*Phoca sibirica*). The Baikal seal, a top predator, is the only species of seal to live exclusively in fresh water. The Baikal seal diet is consistent throughout the year regardless of season, and it mainly consists of pelagic fish from the family *Comephoride*, endemic to Lake Baikal (Ciesielski et al., 2006; Pastukhov, 1993; Yoshii et al., 1999).

In recent decades, the Baikal seal has been under increasing natural and anthropogenic stress, including from climate change (Moore et al., 2009), zoonotic diseases (Mamaev et al., 1995), hunting, and contamination from both POPs and toxic metals (Ciesielski et al., 2006; Mamontov et al., 2019). Moreover, concentrations of PFASs in Baikal seal serum and liver were previously reported for seals sampled in 1992 and 2005 (Ishibashi et al., 2008), with increasing hepatic concentrations of perfluorooctanosulfonic acid (PFOS), perfluorononanoic acid (PFNA) and perfluorodecanoic acid (PFDA), indicating ongoing sources of PFASs into Lake Baikal. Ishibashi et al. (2008a) detected higher concentrations of PFASs in pups and juveniles relative to adults, indicating maternal-fetal transfer. Therefore, Baikal seal pups can be more susceptible to health effects from PFASs than adults. Baikal seals are also consumed by local indigenous communities (e.g., Evenki and Buryats), either directly as a traditional food or indirectly through poultry feed (Mamontova et al., 2020; Nomokonova et al., 2013). Consequently, human consumption of Baikal seals can lead to adverse health effects on humans, and therefore, elevated concentrations of PFASs in Baikal seals pose a direct human consumption risk.

In the late 1980s, a morbillivirus outbreak resulted in the mass death of several thousand Baikal seals (Mamaev et al., 1995, 1996). A contributing factor to the severity of the outbreak was hypothesised to be immunosuppression from chronic exposure to environmental contaminants (De Swart et al., 1996; Nakata et al., 2002). Analysis of Baikal seal tissue demonstrated high concentrations of POPs, including polychlorinated biphenyls (PCBs), dibenzofurans (PCDFs) and other organochlorine contaminants (Iwata et al., 2004; Nakata et al., 1997). Although concentrations of PCBs are decreasing over time in most pinnipeds, concentrations of PFASs are being increasingly detected in the environment (Ross et al., 2013; Xiao, 2017). Contamination of PFASs in biota has been extensively studied in Western Europe and North America, while scarce data is available from Russian territories (Southern Siberia). With the risk of immunosuppression (Dong et al., 2009; Pachkowski et al., 2019) from PFASs combined with the increased incidence of epizootic diseases (Sanderson and Alexander, 2020), monitoring PFASs in wildlife is deemed necessary.

The present study aimed to provide a comprehensive assessment of PFASs in Baikal seal plasma, liver, blubber, and brain. The primary objectives were to: (1) investigate the occurrence of legacy and emerging (novel) PFASs in the target biological matrices; (2) assess the tissue distribution of PFASs in Baikal seals, particularly pups; and (3) establish associations amongst the concentration profiles of the target analytes. The secondary objectives were to: (1) investigate maternal transfer to pups and the association between organ concentration and body mass; and (2) uncover any toxicological implications and potential human risks. To our knowledge, this is the first study on the occurrence of 33 legacy and novel PFASs in various biological matrices (plasma, liver, blubber, and brain) from Baikal seals.

2. Materials and methods

2.1. Chemicals, reagents, and materials

Analytical standards (purity ≥98 %) were purchased for 33 PFASs from Wellington Laboratories Inc. (Guelph, ON, Canada) and included: perfluoropentanoic acid (PFPA), perfluorohexanoic acid (PFHxA), perfluoroheptanoic acid (PFHeA), perfluorooctanoic acid (PFOA), PFNA, PFDA, perfluoroundecanoic acid (PFUnA), perfluorododecanoic acid (PFDoDA), perfluorotridecanoic acid (PFTriDA), perfluoro-1-tetradecanoic acid (PFTDA), perfluoro-n-hexadecanoic acid (PFHxDA), perfluoro-3,7-dimethyloctanoic acid (P37DOMA), 7H-dodecafluoroheptanoic acid (7H-PFHpA), perfluorobutane sulfonate (PFBS), perfluorohexane sulfonate (PFHxS), perfluoroheptane sulfonate (PFHpS), PFOS, 1H,2H-perfluorohexane sulfonate 4:2 (4:2 FTS), 1H,2H-perfuorooctane sulfonate 6:2 (6:2 FTS), 1H,2H-perfluorodecan sulfonate 8:2 (8:2 FTS), 1H,2H-perfluorododecan sulfonate 10:2 (10:2 FTS), perfluoro-1-octanesulfonamidoacetic acid (FOSAA), *N*-methyl-perfluoro-1-octanesulfonamidoacetic acid (MeFOSAA), perfluorooctane sulfonamide (PFOSA), *N*-methylperfluoro-1-octanesulfonamide (MeFOSA), N-(2-hydroxyethyl)-*N*-methylperfluorooctane sulfonamide (MeFOSE), N-ethyl-N-(2-hydroxyethyl)-*N*-methylperfluorooctane sulfonamide (EtFOSE), 2,3,3,3-tetrafluoro-2-(1,1,2,2,3,3,3-heptafluoropropoxy)propanoate (GenX), sodium dodecafluoro-3H-4,8-dioxanoanoate (ADONA), N-ethylperfluoro-1-octanesulfonamide (EtFOSA), 9-chlorohexadecafluoro-3-oxanonane-1-sulfonate (9Cl-PF3ONS), 2-(N-ethylperfluorooctane-1-sulfonamido)ethyl phosphate (SAMPAP) and bis[2-(N-ethylperfluorooctane-1-sulfonamido)ethyl] phosphate (diSAMPAP). Isotopically labelled internal standards, sodium perfluoro-1-octanesulfonate (PFOS-$^{13}C_8$) and perfluorooctanoic acid (PFOA-$^{13}C_8$) were purchased from Cambridge Isotope Laboratories, Inc. (Tewksbury, MA, USA) with purity ≥99 %. Individual stock solutions were prepared in methanol (MeOH) and stored at −20 °C. A 12-port disposable liner Visiprep solid phase extraction (SPE) vacuum manifold was purchased from Supelco (Bellefonte, PA, USA). HPLC grade methanol (MeOH), HPLC grade water and ammonium acetate (≥97 %) together with 15 mL polypropylene (PP) tubes were purchased from VWR Chemicals (Trondheim, Norway). Ammonium formate (anhydrous, reagent grade 97 %), HybridSPE cartridges (Supelco, 30 mg bed weight, 1 mL) and disposable liners (PFTE) were purchased from Sigma Aldrich (Vienna, Austria).

2.2. Sample collection

A total of 69 samples (16 plasma, 18 liver, 17 blubber and 18 brain) from 18 Baikal seals were collected from the Mishikha region, Lake Baikal, Russia, in April 2011 with permission from the Angara-Baikal Directorate of the Russian Fisheries Agency (ref. 000024). The seal samples were collected in cooperation with local hunters. During the annual seal hunt, sampling and handling of the Baikal seals were performed in accordance with the Federal Law on fisheries and conservation of aquatic biological resources of the Russian Federation. The sample set consisted of two adult females (8 and 12 years old) and 16 pups. All pups were 1–2 months of age, while two of the pups born were from the two sampled females. In total, 9 pups were female and the remaining 7 were males. Blood samples were collected directly from the heart with a 20 mL heparinised syringe and transferred into Vacuette© Heparin tubes and centrifuged (3000 rpm, 10 min). Subsequently, plasma was transferred into cryovials and were immediately frozen with liquid nitrogen (−196 °C), and thereafter, transferred to −80 °C until further sample preparation and analysis. Samples of liver, brain, and blubber were also collected in the field, weighed, wrapped in aluminium foil, and immediately transferred to −20 °C until analysis. Specific details regarding the samples are provided in supplementary information (SI, Table S1).

2.3. HybridSPE extraction protocol

The HybridSPE extraction was performed according to previous work with minor modifications (Asimakopoulos and Thomaidis, 2015; Vike-Jonas et al., 2021). Each HybridSPE cartridge (30 mg, 1 mL) was preconditioned with 1 mL MeOH prior to the loading step of the sample extract.

For each plasma sample, a volume of 250 μL was transferred into a 15 mL PP tube and 20 ng internal standard mixture were added. Thereafter, 750 μL of MeOH containing 0.1 % w/v ammonium formate were added, vortex-mixed for 1 min, and centrifuged (4000 rpm, 10 min) for protein precipitation/removal. The supernatant was collected and passed through the pre-conditioned HybridSPE cartridge, and the eluent was collected and directly stored at -20 °C until analysis.

For each liver, blubber and brain sample, 150 mg (±25 mg) of tissue were transferred into a 15 mL PP tube and 20 ng internal standard mixture were added. Thereafter, 500 μL MeOH containing 0.1 % *w/v* ammonium formate were added to the tissue sample, and the derived extract was vortex-mixed, ultrasonicated (45 min), and centrifuged (3500 rpm for 5 min) for protein precipitation/removal. The supernatant was collected (250 μL in volume) and passed through the pre-conditioned HybridSPE cartridge and the eluent was collected and directly stored at −20 °C until analysis.

2.4. UHPLC-MS/MS analysis

Analysis of PFASs was conducted on an Acquity UPLC-I-Class system (Waters, Milford, CT, USA) coupled to a triple quadrupole mass analyser (QqQ; Xevo TQ-S) with a ZSpray ESI ion source (Waters, Milford, CT, USA). Separation was performed with a Phenomenex Kinetex C18 column (30 × 2.1 mm, 1.3 μm, 100 Å) connected to a Phenomenex C18 guard column (2.1 mm) maintained at 30 °C. The mobile phase consisted of solvent (A) water containing 2 mM ammonium acetate and (B) MeOH. The flow rate was 200 $\mu L\ min^{-1}$ and the injection volume was 4 μL. The gradient elution began with 10 % B, held for 0.2 min, and then increased to 100 % B over 2.8 min, held for 1 min, and decreased back to 10 % B over 0.1 min with a hold time of 0.9 min for a total run time of 5 min. Electrospray ionisation was performed in electrospray negative ionisation (ESI-) and multiple reaction monitoring (MRM) mode. Optimised source conditions were as follows: capillary voltage 2.0 kV; source temperature 150 °C; desolvation temperature 450 °C; cone gas flow 150 L/Hr; desolvation gas flow 650 L/Hr and nebulizer gas pressure 6 bar. Target analyte quantification was accomplished using internal standards and matrix-matched calibration curves prepared by spiking the target analytes and internal standards into the matrix prior to extraction (Sait et al., 2023). More details concerning ultrahigh performance liquid chromatography tandem mass spectrometry (UHPLC-MS/MS) analysis, specific mass transitions and instrumental parameters, including quality assurance and quality control data, are available in supplementary information (Tables S2–S5, Figs. S1–S2).

2.5. Data analysis and statistical treatment

All UHPLC-MS/MS data were acquired with Intellistart, MassLynx and TargetLynx software packages (Waters, Milford, USA). Excel (Microsoft, 2021) and GraphPad prism 9 (San Diego, CA, USA) were used for statistical analysis. Statistical analysis was performed on major PFASs with detection rates (DR) ≥ 70 %. Data were subjected to the Shapiro-Wilk normality test and the statistical significance was set to $p < 0.05$. As the concentration data was not normally distributed, the Kruskal-Wallis test followed by Dunn's post hoc test were applied to evaluate the differences in concentrations of PFASs between the 4 biological matrices. To evaluate differences between sex, the student *t*-test was performed on log-transformed concentrations. To assess the global distribution of PFASs in pinnipeds, data on concentrations in blood (whole blood, serum and plasma) and liver were collected following an extensive search of relevant scientific literature, while the (meta-)analysis was conducted in Jupyter notebook with Python software (version: 3.9.12); further details on methodology, and supplementary figures categorised by pinniped species and sampling year are provided in SI (Figs. S3–S6). Spearman correlations and principal component analysis (PCA) were used to assess potential correlations between major PFASs. Concentrations of PFASs detected below the method limit of detection (mLOD) were substituted with mLOD/2. Data was centred and scaled before PCA analysis in PAST 4 software (Version 4.04, https://www.nhm.uio.no/english/research/infrastructure/past/).

3. Results and discussion

3.1. Occurrence and distribution of PFASs in Baikal seals

A global comparison of PFOS concentrations (overall global median: 66.2 ng/g w.w.) and sum concentrations of C_8-C_{14} PFCAs ([Σ_7PFCAs] global median: 22.6 ng/g w.w.) in pinnipeds from different continents are visualised in Figs. 1 and 2, respectively (the data used in those figures are presented detailed in Table 1). Lower median PFOS concentrations (plasma: 8.67 ng/g w.w. and liver: 9.86 ng/g w.w.) were observed in Baikal seals than in other pinnipeds, except for Antarctica. The low prevalence of PFOS in Baikal seals can indicate low historical and current use of PFOS around Lake Baikal. The long chain PFCAs were similar in Baikal seal plasma (median [Σ_7PFCAs]: 27.6 ng/g w.w.) and liver (median [Σ_7PFCAs]: 24.2 ng/g w.w.) as the Σ_7PFCAs concentrations globally detected in pinnipeds. This indicates that Σ_7PFCAs concentrations are relatively evenly distributed in freshwater and marine ecosystems globally where seals/pinnipeds are top predators.

From the 33 PFASs analysed in Baikal seals, the most prevalent were PFOS, the 7 long chain perfluoroalkyl carboxylic acids (C_8–C_{14} PFCAs) and the 1 branched PFCA (P37DMOA) (Fig. 3; and detailed data is presented in Table S6). The distribution of PFASs in the Baikal seals was highest in plasma and decreased in the following order: liver > brain > blubber. Concentrations of PFASs in plasma and liver were significantly higher than those determined in brain (Kruskal-Wallis; $p < 0.01$) and blubber (Kruskal-Wallis; $p < 0.0001$). In plasma, the highest median concentrations were detected for PFUnA (detection rate: DR = 94 %; 11.2 ng/g w.w.), PFOS (DR = 88 %; 8.67 ng/g w.w.), PFDA (DR = 94 %; 5.13 ng/g w.w.), PFNA (DR = 88 %; 4.65 ng/g w.w.), PFTriDA (DR = 100 %; 4.29 ng/g w.w.) and PFDoDA (DR = 81 %; 1.24 ng/g w.w.). Plasma concentrations of PFASs were higher than previously determined in Baikal seal serum in 2005 (Ishibashi et al., 2008), suggesting increasing inputs of PFASs in Lake Baikal from 2005 to 2011.

In Baikal seal liver, similar concentrations and profiles were observed as in plasma for PFOS concentrations (DR = 94 %; median: 9.86 ng/g w.w.) followed by those of PFUnA (DR = 94 %; 7.36 ng/g w.w.), PFDA (DR = 94 %; 6.69 ng/g w.w.), PFNA (DR = 89 %; 5.83 ng/g w.w.) and PFTriDA (DR = 100 %; 2.55 ng/g w.w.). In contrast to plasma/serum, liver concentrations were consistent with those previously reported in Baikal seal liver from 2005 (Ishibashi et al., 2008).

In the brain of Baikal seals, PFTriDA (DR = 100 %; median: 4.05 ng/g w.w.), PFUnA (DR = 83 %; 0.92 ng/g w.w.), and PFDoDA (DR = 94 %; 0.66 ng/g w.w.) were the dominant PFASs. Other PFASs were detected sporadically in the brain, including PFDA (DR = 22 %; 1.20 ng/g w.w.), PFOS (DR = 17 %; 1.46 ng/g w.w.), PFNA (DR = 6 %; 19.2 ng/g w.w.) and

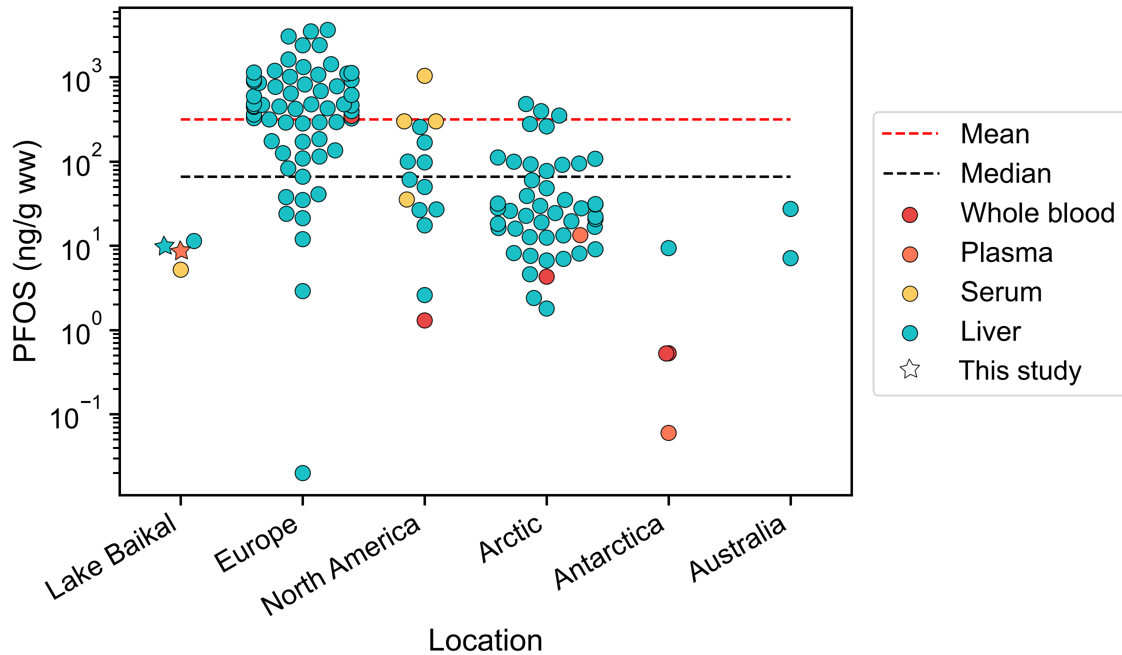

Fig. 1. PFOS concentrations (ng/g w.w.) determined in blood (whole blood, plasma, serum) and liver in pinnipeds globally.

PFOA (DR = 17 %; 1.21 ng/g w.w.). This study demonstrates that PFASs can cross the blood–brain barrier in Baikal seals, while the distribution of PFASs in the brain of pinnipeds and other aquatic mammals remains largely understudied. To our knowledge, there is only one other report on the occurrence of PFASs in the brain of pinnipeds (Ahrens et al., 2009a). Ahrens et al. (2009a, 2009b) determined PFASs in various organs and tissues in harbour seals (*Phoca vitulina*) from the German Bight (North Sea), including the brain where PFOS was the main contributor (mean: 99 ± 49 ng/g w.w.). PFASs were also detected in the brain of other marine mammals including harbour porpoises (*Phocoena phocoena*) (Van De Vijver et al., 2007) and polar bears (*Ursus maritimus*) (Eggers Pedersen et al., 2015; Greaves et al., 2013). Interestingly, brain region-specific distribution of PFASs is documented in polar bears, with the inner brain regions displaying higher concentrations of PFASs than the outer regions of the brain (Eggers Pedersen et al., 2015; Greaves et al., 2013). The authors attributed the observed accumulation patterns of PFASs in polar bear brains to the increased blood flow into the inner relative to the outer brain regions. In our study, high variation (RSD: 73.4-289 %) was evident for PFASs in the brain of Baikal seals which can further suggest brain region-specific accumulation. Although due to low detection rates herein (for most PFASs <35 %), this phenomenon should be investigated further in pinnipeds.

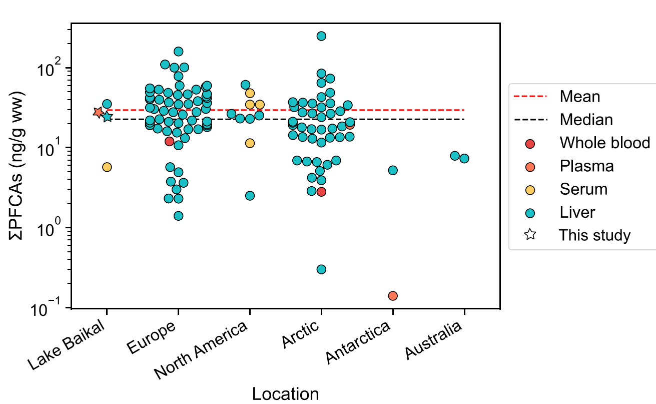

Fig. 2. Σ_7PFCAs (sum of C_8-C_{14} PFCAs) concentrations (ng/g w.w.) determined in blood (whole blood, plasma, serum) and liver in pinnipeds globally.

The majority of PFASs were detected at low concentrations and in low abundance in Baikal seal blubber (Table S6). This is in accordance with other studies on pinnipeds from remote regions, where low concentrations of PFASs were quantified in seal blubber from arctic regions (Boisvert et al., 2019; Powley et al., 2008). Furthermore, due to the polar hydrophobic nature of fluorine containing compounds, PFASs tend to not accumulate in lipid-rich tissues (Dassuncao et al., 2019; Pérez et al., 2013). In contrast, high blubber concentrations of PFOS (18.9-297 ng/g w.w.) were quantified in harbour seals from the Wadden Sea (Van de Vijver et al., 2005). However, the elevated concentrations reported in harbour seal blubber in that study are likely a consequence of denser human population impacts and industrial activities. A greater proportion of long chain PFCAs (>C9) was previously reported in pinniped blubber relative to other tissues and organs, due to their higher lipophilicity (Boisvert et al., 2019). However no clear patterns for long chain PFCAs (>C9) were observed for the Baikal seals in our study.

The following PFASs: PFOSA, PFPA, EtFOSA and 9Cl-PF3ONS were detected at low concentrations and detection frequencies in Baikal seal tissues (Table S6). Several PFASs, including novel PFASs (PFBS, PFHxS, PFHpS, PFHeA, PFHxA, PFHxDA 7H-PFHpA, 4:2 FTS, 6:2 FTS, 8:2 FTS, 10:2 FTS, FOSAA, MeFOSE, EtFOSE, MeFOSA, MeFOSAA, GenX, ADONA, SAMPAP and diSAMPAP) were not detected in Baikal seals. Low concentrations and detection frequency of PFOSA is consistent with literature and likely attributed to the efficient biotransformation of PFOSA to PFOS in pinnipeds (Bossi et al., 2005; Butt et al., 2007; Galatius et al., 2013; Ishibashi et al., 2008; Taylor et al., 2021). The absence or low abundance of novel PFASs can reflect low or no local usage of these compounds at the time of sample collection and/or lower potential for bioaccumulation in wildlife.

3.2. Associations between PFASs

In wildlife, PFOS concentrations are generally higher than those of PFCAs. However, some PFCAs (PFNA, PFDA, PFUnA) have presented similar or elevated concentrations relative to PFOS in wildlife including in Baikal seals (Ishibashi et al., 2008), bottlenose dolphins (*Tursiops truncates*), beluga (*Delphinapterus leucas*) and northern fulmars (*Fulmarus glacialis*) (Houde et al., 2005; Muir et al., 2004). Observed differences in concentrations of PFASs, especially the lower concentrations of PFOS determined herein are likely attributed to different sources of local pollution and/or dietary differences between Baikal seals and other pinnipeds. Watanabe et al.

Table 1
PFOS and C_8-C_{14} PFCAs concentrations (mean or median; ng/g w.w., unless otherwise stated) in blood media and liver of pinnipeds from different locations.

Species	Tissue/organ (n)	PFOS (C8)	PFOA (C8)	PFNA (C9)	PFDA (C10)	PFUnA (C11)	PFDoDA (C12)	PFTriDA (C13)	PFTDA (C14)	ΣPFCAs	Location (Year)	Reference
Serum/plasma/whole blood												
Baikal seal (*Phoca sibirica*)	Plasma[#] (n = 16)	8.67	0.72	4.65	5.13	11.2	1.24	4.29	0.44	27.6	Lake Baikal, Russia (2011)	This Study
	Serum (n = 24, F)	4.5	0.4	2	0.58	1.9	0.34	NA	NA	5.22	Lake Baikal, Russia (2005)	(Ishibashi et al., 2008)
	Serum (n = 20, M)	5.8	0.17*	2.5	0.85	2.3	0.36	NA	NA	6.18	Lake Baikal, Russia (2005)	(Ishibashi et al., 2008)
Bearded seal (*Eriganthus barbatus*)	Blood (n = 1)	1.3	ND	ND	ND	ND	ND	NA	NA	–	Beaufort Sea, Canada (2004)	(Powley et al., 2008)
Elephant seal (*Mirounga leonine*)	Blood[†] (n = 59)	0.53	NA	NA	NA	NA	NA	NA	NA	–	Antarctic (2004–2005)	(Tao et al., 2006)
Grey seal (*Halichoerus grypus*)	Plasma[†] (n = 26)	37	NA	NA	NA	NA	NA	NA	NA	–	Baltic Sea (1990s)	(Giesy and Kannan, 2001)
	Plasma[†] (n = 12)	28	NA	NA	NA	NA	NA	NA	NA	–	Canadian Arctic (1990s)	(Giesy and Kannan, 2001)
	Plasma[†] (n = 9)	42	NA	NA	NA	NA	NA	NA	NA	–	Baltic Sea (1996)	(Kannan et al., 2001)
	Plasma[†] (n = 10)	43.9	NA	NA	NA	NA	NA	NA	NA	–	Baltic Sea (1997)	(Kannan et al., 2001)
	Plasma[†] (n = 7)	25.5	NA	NA	NA	NA	NA	NA	NA	–	Baltic Sea (1998)	(Kannan et al., 2001)
	Plasma[†] (n = 12)	27.7	NA	NA	NA	NA	NA	NA	NA	–	Sable Island, Canada (1999)	(Kannan et al., 2001)
Harbour seal (*Phoca vitulina*)	Serum[†] (n = 6)	1044	7.83	18	6.6	8.77	6.69	NA	NA	47.9	San Francisco Bay-South Bay-Mowry (2004)	(Sedlak and Greig, 2012)
	Serum[†] (n = 34)	301	2.85	11.7	9.66	6.28	4.02	NA	NA	34.5	San Francisco Bay-Central Bay-Castro Rocks (2006-2008)	(Sedlak and Greig, 2012)
	Serum[†] (n = 21)	35.5	1.07	2.56	1.93	5.08	0.71	NA	NA	11.4	San Francisco Bay-Tomales Bay (2007-2008)	(Sedlak and Greig, 2012)
	Blood (n = 4)	349	0.62	3.93	4.38	1.71	0.47	0.76	0.08	12.0	German Bight (2007)	(Ahrens et al., 2009a)
Hooded seal (*Cystophora cristata*)	Plasma (n = 15, mothers)	13.4	0.31	2.29	2.75	9.71	1.45	5.07	NA	19.3	West Ice, east of Greenland (2008)	(Grønnestad et al., 2017)
	Plasma (n = 15, pups)	30.2	0.54	1.61	1.61	11.9	3.38	13.1	NA	32.1	West Ice, east of Greenland (2008)	(Grønnestad et al., 2017)
Northern fur seal (*Callorhinus ursinus*)	Plasma[†] (n = 44)	3.0**	NA	NA	NA	NA	NA	NA	NA	–	Coastal waters of Alaska (1995)	(Kannan et al., 2001)
Ringed seal (*Phoca hispida*)	Plasma[†] (n = 11)	29.8	0.3	7.11	3.26	8.35	0.85	5.09	1.47	26.4	Kongsfjorden, Svalbard (2012)	(The Norwegian Environmental Agency, 2013)
	Plasma (n = 11)	76	0.27**	1.9	1.3	3.6	0.56	0.89	0.04*	8.56	Kongsfjorden, Svalbard, Norway (1990)	(Routti et al., 2016)
	Plasma (n = 10)	44	0.03*	2.2	1.6	4.4	0.51	0.59	0.03*	7.76	Kongsfjorden, Svalbard, Norway (1993)	(Routti et al., 2016)
	Plasma (n = 10)	51	0.61**	3.2	2.6	5.8	0.96	1.88	0.05**	15.1	Kongsfjorden, Svalbard, Norway (1997)	(Routti et al., 2016)
	Plasma (n = 10)	48	0.8	2.8	1.8	5.3	0.34	0.17	0.025*	11.2	Billefjorden, Svalbard, Norway (2002)	(Routti et al., 2016)
	Plasma (n = 9)	94	2.05	8.5	5.6	13.8	1.66	2.34	0.165	34.1	Kongsfjorden, Svalbard, Norway (2004)	(Routti et al., 2016)
	Plasma (n = 10)	18	0.6	2.3	1.4	4	0.33	0.29	0.03*	8.95	Tempelfjorden, Svalbard, Norway (2007)	(Routti et al., 2016)
	Plasma (n = 11)	48	0.22	8	5.2	9.2	1.16	2.24	0.12**	26.1	Kongsfjorden, Svalbard, Norway (2010)	(Routti et al., 2016)
	Blood[†] (n = 5)	5.6[##]	ND	1.1[##]	0.75[##]	1.7[##]	0.2[##]	NA	NA	3.8	Beaufort Sea, Canada (2004)	(Powley et al., 2008)

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Table 1 (*continued*)

Species	Tissue/organ (n)	PFOS (C8)	PFOA (C8)	PFNA (C9)	PFDA (C10)	PFUnA (C11)	PFDoDA (C12)	PFTriDA (C13)	PFTDA (C14)	ΣPFCAs	Location (Year)	Reference
	Plasma[†] (n = 24)	6**	NA	NA	NA	NA	NA	NA	NA	–	Canadian Arctic (1990s)	(Giesy and Kannan, 2001)
	Plasma[†] (n = 18)	110	NA	NA	NA	NA	NA	NA	NA	–	Baltic Sea (1990s)	(Giesy and Kannan, 2001)
Ringed seal (*Phoca hispida*) cont'd	Plasma[†] (n = 18)	9	NA	NA	NA	NA	NA	NA	NA	–	Norwegian Arctic (1990s)	(Giesy and Kannan, 2001)
	Plasma[†] (n = 10)	133	NA	NA	NA	NA	NA	NA	NA	–	Baltic Sea (1996)	(Kannan et al., 2001)
	Plasma[†] (n = 9)	92	NA	NA	NA	NA	NA	NA	NA	–	Baltic Sea (1997)	(Kannan et al., 2001)
	Plasma[†] (n = 10)	242	NA	NA	NA	NA	NA	NA	NA	–	Baltic Sea (1998)	(Kannan et al., 2001)
	Plasma (n = 10)	8.1	NA	NA	NA	NA	NA	NA	NA	–	Spitsbergen (1996)	(Kannan et al., 2001)
	Plasma (n = 8)	10.1	NA	NA	NA	NA	NA	NA	NA	–	Spitsbergen (1998)	(Kannan et al., 2001)
Stellar sea lion (*Eumetopias jubatus*)	Plasma[†] (n = 12)	3.0*	NA	NA	NA	NA	NA	NA	NA	–	Coastal waters of Alaska (1999)	(Kannan et al., 2001)
Weddell seal[#] (*Leptonychotes weddellii*)	Plasma[†] (n = 10)	0.06	0.005*	0.005*	0.005*	0.12	0.005*	NA	NA	0.14	McMurdo Sound, Antarctica (2006)	(Routti et al., 2015)
Liver												
Antarctic fur seal (*Arctocephalus gazella*)	Liver (n = 17)	9.4	0.2*	3.3	0.6	0.9	0.2*	NA	NA	5.2	Antarctic (2004)	(Schiavone et al., 2009)
Australian fur seal (*Arctocephalus pusillus doriferus*)	Liver (n = 20)[#]	27.4	0.98	2.50	0.61	0.72	0.50	1.00	1.00	7.31	Australia (2017-2020)	(Taylor et al., 2021)
Australian sea lion (*Neophoca cinerea*)	Liver (n = 28)[#]	7.14	2.73	2.96	0.5	0.5	0.25*	0.50*	0.50*	7.94	Australia (2017-2020)	(Taylor et al., 2021)
Baikal seal (*Phoca sibirica*)	Liver (n = 18)[#]	9.86	1.3	5.8	6.7	7.36	0.46	2.55	n.d.	24.2	Lake Baikal, Russia (2011)	This Study
	Liver (n = 24, F)	13	1.5	16	8.5	8.0	0.69	NA	NA	34.7	Lake Baikal, Russia (2005)	(Ishibashi et al., 2008)
	Liver (n = 20, M)	9.7	1.6	19	7.1	7.2	0.66	NA	NA	35.6	Lake Baikal, Russia (2005)	(Ishibashi et al., 2008)
Bearded seal (*Eriganthus barbatus*)	Liver (n = 1)	2.6	ND	1.3	0.4	0.7	0.1	NA	NA	5.1	Beaufort Sea, Canada (2004)	(Powley et al., 2008)
	Liver (n = 17)	4.6	ND	3.7	1.1	1.3	0.5	NA	NA	6.6	Bering and Chukchi Seas, Alaska (2003-2007)	(Quakenbush and Citta, 2008)
California sea lion (*Zalophus californianus*)	Liver (n = 6)	26.6	NA	NA	NA	NA	NA	NA	NA	–	West Coast, United States (1994-1997)	(Kannan et al., 2001)
Grey seal (*Halichoerus grypus*)	Liver (n = 5)	328	2.1	50.5	16.7	19.3	2.6	8.5	0.9	101	Sweden (2012-2016)	(Spaan et al., 2020)
	Liver (n = 1)	172	0.7	35.9	9.2	7.5	0.9	2.8	0.41*	57.4	Gävleborgs län, Sweden (2016)	(Spaan et al., 2020)
	Liver (n = 5)	169	0.4	7.7	8.8	21.1	4.8	15.6	2.6	61	US Atlantic coast (2000-2004)	(Spaan et al., 2020)
	Liver (n = 5)	2.9*	0.15*	3.1	1.6	5.2	1.0	4.5	0.41*	16	Iceland (2009-2010)	(Spaan et al., 2020)
	Liver (n = 1)	12	ND	0.8	0.6	0.6	0.2	0.1	ND	2.30	The Baltic Sea (1969)	(Kratzer et al., 2011)
	Liver (n = 2)	115	ND	2.6	1.5	1.1	0.3	0.2	ND	5.70	The Baltic Sea (1974)	(Kratzer et al., 2011)
	Liver (n = 1)	24	ND	0.6	0.3	0.3	0.1	0.1	ND	1.40	The Baltic Sea (1975)	(Kratzer et al., 2011)
	Liver (n = 2)	35	ND	0.9	0.6	0.6	0.1	0.1	0.01	2.31	The Baltic Sea (1976)	(Kratzer et al., 2011)
	Liver (n = 2)	109	0.03	2.1	0.6	0.6	0.1	0.2	ND	3.63	The Baltic Sea (1977)	(Kratzer et al., 2011)
	Liver (n = 6)	41	0.01	2.0	0.7	0.8	0.2	0.04	0.004	3.75	The Baltic Sea (1978)	(Kratzer et al., 2011)
	Liver (n = 4)	83	0.01	2.7	0.8	1.0	0.2	0.2	0.01	4.92	The Baltic Sea (1979)	(Kratzer et al., 2011)
	Liver (n = 1)	0.02	ND	1.3	0.5	0.8	0.2	0.2	ND	3.00	The Baltic Sea (1980)	(Kratzer et al., 2011)
Grey seal (*Halichoerus*	Liver (n = 1)	126	ND	8.9	2.8	2.7	0.4	0.5	0.1	15.4	The Baltic Sea (1981)	(Kratzer et al., 2011)

Table 1 (*continued*)

Species	Tissue/organ (n)	PFOS (C8)	PFOA (C8)	PFNA (C9)	PFDA (C10)	PFUnA (C11)	PFDoDA (C12)	PFTriDA (C13)	PFTDA (C14)	ΣPFCAs	Location (Year)	Reference
grypus) cont'd	Liver (n = 1)	185	ND	13	4.2	3.9	0.6	0.7	0.2	22.6	The Baltic Sea (1983)	(Kratzer et al., 2011)
	Liver (n = 2)	136	0.06	9.4	3.2	2.9	0.5	0.7	0.2	17.0	The Baltic Sea (1984)	(Kratzer et al., 2011)
	Liver (n = 2)	295	0.02	12	3.7	4.4	0.8	0.7	0.2	21.8	The Baltic Sea (1985)	(Kratzer et al., 2011)
	Liver (n = 3)	291	0.3	12	3.3	3.8	0.6	1.0	0.03	21.0	The Baltic Sea (1986)	(Kratzer et al., 2011)
	Liver (n = 1)	362	0.8	35	11	9.7	1.2	1.5	0.2	59.4	The Baltic Sea (1987)	(Kratzer et al., 2011)
	Liver (n = 4)	284	0.1	15	5.1	5.6	0.7	1.1	0.1	27.7	The Baltic Sea (1988)	(Kratzer et al., 2011)
	Liver (n = 3)	293	2.1	19	5.5	5.8	0.8	1.5	0.01	34.7	The Baltic Sea (1989)	(Kratzer et al., 2011)
	Liver (n = 1)	326	3.3	29	6.9	5.8	0.7	0.6	0.1	46.4	The Baltic Sea (1990)	(Kratzer et al., 2011)
	Liver (n = 1)	484	0.1	31	9.7	9.3	1.1	1.7	0.2	53.1	The Baltic Sea (1993)	(Kratzer et al., 2011)
	Liver (n = 2)	620	0.2	32	8.5	9.9	1.3	3.1	0.02	55.0	The Baltic Sea (1995)	(Kratzer et al., 2011)
	Liver (n = 4)	429	0.3	26	8.0	9.4	1.0	2.0	0.02	46.7	The Baltic Sea (1996)	(Kratzer et al., 2011)
	Liver (n = 7)	457	6.0	66	12	12	1.3	2.6	0.02	99.9	The Baltic Sea (1997)	(Kratzer et al., 2011)
	Liver (n = 2)	825	11	91	23	25	3.3	5.8	0.02	159	The Baltic Sea (1998)	(Kratzer et al., 2011)
	Liver (n = 2)	447	2.8	27	7.5	8.0	0.9	2.2	0.01	48.4	The Baltic Sea (1999)	(Kratzer et al., 2011)
	Liver (n = 1)	465	0.2	15	4.1	5.9	0.8	2.8	ND	28.8	The Baltic Sea (2000)	(Kratzer et al., 2011)
	Liver (n = 3)	317	0.4	25	7	7.4	0.9	2.8	0.04	43.5	The Baltic Sea (2002)	(Kratzer et al., 2011)
	Liver (n = 3)	645	0.4	19	11	16	1.6	4.7	0.4	53.1	The Baltic Sea (2003)	(Kratzer et al., 2011)
	Liver (n = 1)	944	ND	14	12	16	1.7	5.2	0.4	49.3	The Baltic Sea (2004)	(Kratzer et al., 2011)
	Liver (n = 4)	423	0.05	21	6.8	7.8	1.1	3.4	0.3	40.5	The Baltic Sea (2005)	(Kratzer et al., 2011)
	Liver (n = 8)	479	0.04	20	7.9	8.7	1.3	3.9	0.3	42.1	The Baltic Sea (2006)	(Kratzer et al., 2011)
	Liver (n = 3)	451	0.6	74	17	13	1.4	3.5	0.04	110	The Baltic Sea (2008)	(Kratzer et al., 2011)
Harp seal (*Pagophilus groenlandicus*)	Liver (n = 5)	21.3	0.15*	6.1	4.2	11.9	1.6	7.2	0.41*	31.6	Iceland (2009–2010)	(Spaan et al., 2020)
	Liver (n = 1)	39.1	0.4	5.3	4.0	10.7	1.2	4.7	0.41*	26.7	Nuuk, West Greenland (2016)	(Spaan et al., 2020)
Harbour seal (*Phoca vitulina*)	Liver (n = 13)	689	1.8	8.7	14.8	5.1	NA	NA	NA	30.4	Wadden Sea (2002)	(Galatius et al., 2013)
	Liver (n = 59)	398	1.6	5.6	6.6	5.2	NA	NA	NA	19	Danish waters (2002)	(Dietz et al., 2012)
	Liver (n = 24)	175	ND	7.0	7.1	3.94*	ND	NA	NA	18.0	Wadden Sea (2002)	(Van de Vijver et al., 2005)
	Liver (n = 8, Adults, M)	98	0.8*	5.8	5.6	11	3.2	NA	NA	26.4	Northwest Atlantic coast (2000–2007)	(Shaw et al., 2009)
	Liver (n = 10, Adults F)	100	0.8*	5.4	4.9	9.0	2.8	NA	NA	22.9	Northwest Atlantic coast (2000–2007)	(Shaw et al., 2009)
	Liver (n = 50, pups)	258	1.9	6.6	4.4	9.9	2.4	NA	NA	25.2	Northwest Atlantic coast (2000–2007)	(Shaw et al., 2009)
	Liver (n = 5)	858	1.8	24.7	14.9	21.4	4.2	9.8	1.3	78.1	Sweden (2015)	(Spaan et al., 2020)
	Liver (n = 5)	50	1.3	4.4	2.4	5.2	1.1	7.3	1.4	23.1	US Atlantic coast (2000–2008)	(Spaan et al., 2020)
	Liver (n = 5)	37.9	0.5	6.5	4.2	19.8	2.5	10.6	1.4	45.5	Iceland (2009–2010)	(Spaan et al., 2020)
Harbour seal (*Phoca vitulina*) cont'd	Liver (n = 4)	1017	0.7	15.3	15.2	5.26	1.47	1.53	0.22	39.7	German Bight (2007)	(Ahrens et al., 2009a)
	Liver (n = 10)	66.2	0.80	4.43	3.3	6.87	0.84	2.86	0.015*	19.1	Norway (2012)	(The Norwegian Environmental Agency, 2013)

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Table 1 (*continued*)

Species	Tissue/organ (n)	PFOS (C8)	PFOA (C8)	PFNA (C9)	PFDA (C10)	PFUnA (C11)	PFDoDA (C12)	PFTriDA (C13)	PFTDA (C14)	ΣPFCAs	Location (Year)	Reference
cont'd	Liver[R] (n = 31)	1203** (7.2-2407)	4.2** (ND-8.4)	13.5** (0.3-27)	18** (0.2-36)	6** (0.5-12)	1.2** (ND-2.3)	1.8** (ND-3.6)	0.1** (ND-0.2)	44.8** (1.0-89.5)	German Bight (1988–2008)	(Ahrens et al., 2009b)
	Liver[R] (n = 19)	1838** (204-3676)	2.9** (ND-5.7)	9.5** (0.9-19)	8.0** (0.4-16)	5.5** (2.3-11)	2.45** (0.5-4.9)	3.1** (1.0-6.1)	0.3** (ND-0.6)	31.8** (5.1-63.3)	German Bight (1996–2007)	(Ahrens et al., 2009b)
Ribbon seal (*Histriophoca fasciata*)	Liver (n = 8)	6.7	ND	6.8	3.2	9.5	1.3	NA	NA	20.8	Bering and Chukchi Seas, Alaska (2003–2007)	(Quakenbush and Citta, 2008)
Ringed seal (*Phoca hispida*)	Liver (n = 5)	26##	NA	4.1##	2.7##	5.8##	0.9##	NA	NA	13.5	Beaufort Sea, Canada (2004)	(Powley et al., 2008)
	Liver (n = 6)	22.7	0.43*	1.7	1.6	5.5	1.4	2.0	0.3	12.9	Arviat, Nunavut (1992)	(Butt et al., 2007)
	Liver (n = 10)	91.6	1.13	5.0	4.9	15.6	2.8	4.0	0.49	33.9	Arviat, Nunavut (1998)	(Butt et al., 2007)
	Liver (n = 10)	35.1	1.67	5.1	4.7	17.6	2.9	4.5	0.51	37.0	Arviat, Nunavut (2003)	(Butt et al., 2007)
	Liver (n = 10)	19.6	0.98	5.1	3.9	12.0	1.9	3.3	0.36	27.5	Arviat, Nunavut (2005)	(Butt et al., 2007)
	Liver (n = 2)	1.8	1.1	0.33	0.43	0.34	0.55	0.11	0.004	2.9	Resolute Bay, Nunavut (1972)	(Butt et al., 2007)
	Liver (n = 9)	7.0	4.5	1.9	1.2	2.8	0.47	0.54	0.16	11.6	Resolute Bay, Nunavut (1993)	(Butt et al., 2007)
	Liver (n = 9)	22.1	3.9	3.7	2.8	6.4	1.2	2.1	0.39	20.5	Resolute Bay, Nunavut (2000)	(Butt et al., 2007)
	Liver (n = 9)	16.8	6.2	6.8	4.3	11.0	1.4	2.0	0.27	32.0	Resolute Bay, Nunavut (2004)	(Butt et al., 2007)
	Liver (n = 9)	8.1	0.43*	4.8	3.4	7.5	0.96	NA	0.21	17.3	Resolute Bay, Nunavut (2005)	(Butt et al., 2007)
	Liver (n = 10)	99.7	0.8	13.7	10.9	24.2	2.8	10.8	1.2	64.4	Ittoqqortoormiit, East Greenland (2012)	(Spaan et al., 2020)
	Liver (n = 5)	483	10.6	124.8	48.6	45.4	5.3	11.9	1.4	248	Northern Baltic (2015)	(Spaan et al., 2020)
	Liver (n = 5)	28.1	0.15*	9.7	5.9	18.1	2.6	10.7	1.3	48.5	Illulisat, North-West Greenland (2013)	(Spaan et al., 2020)
	Liver (n = 10)	93	0.56	11	9.4	18	2.3	1.3	0.24	42.8	Ittoqqortoormiit, East Greenland (2012 – 2013)	(Gebbink et al., 2016)
	Liver (n = 10)	12.5	0.6*	1.5	0.5	1.3	NA	NA	NA	3.9	Qeqertarsuaq, West Greenland (1982)	(Rigét et al., 2013)
	Liver (n = 8)	29.8	0.6*	0.7*	0.9	2.0	NA	NA	NA	4.2	Qeqertarsuaq, West Greenland (1994)	(Rigét et al., 2013)
	Liver (n = 10)	31.3	0.6*	1.2	1.3	3.6	NA	NA	NA	6.7	Qeqertarsuaq, West Greenland (1999)	(Rigét et al., 2013)
	Liver (n = 10)	27.9	0.6*	1.5	1.2	3.6	NA	NA	NA	6.9	Qeqertarsuaq, West Greenland (2003)	(Rigét et al., 2013)
	Liver (n = 19)	397	2.4	2.4	5.7	8.6	NA	NA	NA	19.1	Qeqertarsuaq, West Greenland (2006)	(Rigét et al., 2013)
	Liver (n = 19)	262	1.2	4.5	4.4	11	NA	NA	NA	21.1	Qeqertarsuaq, West Greenland (2008)	(Rigét et al., 2013)
	Liver (n = 19)	16.3	0.6*	2.0	2.2	8.5	NA	NA	NA	13.3	Qeqertarsuaq, West Greenland (2010)	(Rigét et al., 2013)
	Liver (n = 9)	31.0	0.6*	0.7*	2.1	3.5	NA	NA	NA	6.9	Ittoqqortoormiit, East Greenland (1986)	(Rigét et al., 2013)
	Liver (n = 6)	20.8	0.6*	0.7*	1.0	2.8	NA	NA	NA	5.1	Ittoqqortoormiit, East Greenland (1994)	(Rigét et al., 2013)
	Liver (n = 8)	77.4	1.0	5.4	3.7	7.8	NA	NA	NA	17.9	Ittoqqortoormiit, East Greenland (1999)	(Rigét et al., 2013)
	Liver (n = 9)	94.8	0.6*	4.0	3.3	8.9	NA	NA	NA	16.8	Ittoqqortoormiit, East Greenland (2003)	(Rigét et al., 2013)
	Liver (n = 14)	352	1.6	3.2	7.1	12.1	NA	NA	NA	24	Ittoqqortoormiit, East Greenland (2006)	(Rigét et al., 2013)
	Liver (n = 20)	280	2.2	8.1	4.8	16.8	NA	NA	NA	31.9	Ittoqqortoormiit, East Greenland (2008)	(Rigét et al., 2013)
	Liver (n = 16)	112	0.6*	7.6	8.4	19.9	NA	NA	NA	36.5	Ittoqqortoormiit, East Greenland (2010)	(Rigét et al., 2013)
	Liver (n = 10)	24.5	0.05	12.7	5.85	6.16	0.63	0.89	0.24	26.5	Lake Melville, Cananda (2017)	(Xiong, 2021)
	Liver	12.7	3.7	8.7	4.5	5.2	2.2	2.8	1.4	28.5	Pangnirtung,	(Muir et al., 2004)

Table 1 (*continued*)

Species	Tissue/organ (n)	PFOS (C8)	PFOA (C8)	PFNA (C9)	PFDA (C10)	PFUnA (C11)	PFDoDA (C12)	PFTriDA (C13)	PFTDA (C14)	ΣPFCAs	Location (Year)	Reference
	(n = 11)										Nunavut, Canada (2002)	
Ringed seal (*Phoca hispida*)	Liver (n = 10)	48.3	24.3	25.2	12.1	15.6	3.3	2.8	1.2	84.5	Sachs Harbour, Northern Territories, Canada (2003)	(Muir et al., 2004)
cont'd	Liver (n = 17)	8.2	3.1	9.1	1.8	2.4	0.6	NA	NA	17.0	Bering and Chukchi Sea, Alaska (2003–2007)	(Quakenbush and Citta, 2008)
	Liver (n = 9)	16	1.0*	5.9	2.1	3.3	0.44	0.57	0.25*	13.6	Ulukhaqtuuq (2001)	(Martin et al., 2004)
	Liver (n = 10)	19	1.0*	4.9	2.9	3.8	0.76	0.95	ND	14.3	Ausuittuq (1998)	(Martin et al., 2004)
	Liver (n = 16)	108	1.0	20	13	26	3.5	8.7	0.8	73	Ittoqqortoormiit, East Greenland (2012)	(Boisvert et al., 2019)
	Liver (n = 9)	31.8	0.87	–	–	–	–	–	–	35.6[a]	Arviat, Nunavut, Canada (2018)	(Facciola et al., 2022)
	Liver (n = 10)	9.1	0.09	–	–	–	–	–	–	6.1[a]	Nain, Nunatsiavut, Canada (2018)	(Facciola et al., 2022)
	Liver (n = 9)	13.3	0.16	–	–	–	–	–	–	18.4[a]	Resolute Bay, Nunavut, Canada (2018)	(Facciola et al., 2022)
	Liver (n = 9)	18.3	0.27	–	–	–	–	–	–	35.8[a]	Sachs Harbour, Canada (2018)	(Facciola et al., 2022)
Spotted seal (*Phoca largha*)	Liver (n = 9)	7.6	ND	5.7	2.1	5.3	0.6	NA	NA	13.7	Bering and Chukchi Seas, Alaska (2003–2007)	(Quakenbush and Citta, 2008)
Walrus (*Odobenus rosmarus*)	Liver (n = 5)	2.4	0.3	NA	NA	NA	NA	NA	NA	0.3	East Arctic, Kinngait, Nunavut (1998)	(Tomy et al., 2004)

Where #indicates median, *LOD/2 used as substitute value, **max concentration/2 was used as the substitute value in this meta-analysis table (e.g., because only the max concentration was reported in the corresponding study), ##midpoint used as substitute (min + max/2) in this meta-analysis table (e.g., because only the range of concentrations was reported in the corresponding study), †concentration in ng/mL, ND not detected, and NA not analysed, [a]only total ΣPFCAs provided (C_6 to C_{14} PFCAs), [R]Study using concentration ranges.

(2020) investigated the diet of Baikal seals and observed consumption of amphipods alongside other types of dietary sources (e.g., pelagic sculpins). Thus far, there is uncertainty on what proportion amphipods make up the diet of Baikal seals (Watanabe et al., 2020), but feeding at a lower trophic level could potentially result in lower dietary exposure, and consequently, lower concentrations of PFASs in their tissues and organs.

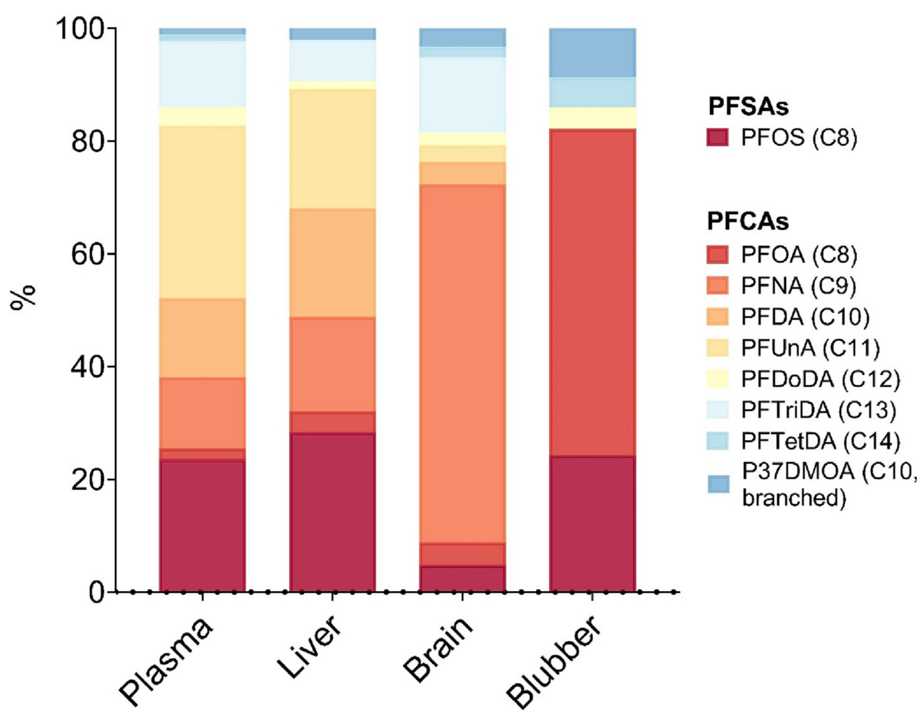

Fig. 3. Distribution of PFASs (for those with DR > 30 %) in the 4 biological matrices [plasma (n = 16), liver (n = 18), brain (n = 18) and blubber (n = 17)] of Baikal seals (n = 69). Determined from median concentrations.

Odd-carbon chain length PFCAs (e.g., C9 and C11) are often detected in higher concentrations in wildlife relative to their next shortest even-carbon homologues (e.g., C8 and C10) (Bossi et al., 2005; Butt et al., 2007; Martin et al., 2004). This odd-even pattern was similarly observed herein in Baikal seal plasma and liver, with a rank order of decreasing median concentrations of: PFTriDA (C13) > PFDoDA (C12); PFUnA (C11) > PFDA (C10); and PFNA (C9) > PFOA (C8). This odd-even pattern is hypothesised to originate either to direct release from local sources (Prevedouros et al., 2006) or indirectly through the environmental degradation of fluorotelomer based precursors (e.g., fluorotelomer alcohols (FTOHs)) (Ellis et al., 2004). Upon degradation, FTOHs yield even- and odd-chain PFCAs via oxidation in the atmosphere (Ellis et al., 2004) and in vivo (Butt et al., 2014). As an example, 8:2 FTOH is transformed to equal amounts of PFNA and PFOA, but odd-chain PFNA exceeds the concentration of the even-chain PFOA in biota due to the higher bioaccumulation potential of the former. Therefore, the long range transport of FTOHs and their subsequent degradation, combined with the increasing bioaccumulation potential of longer chain PFCAs may explain this odd-even pattern especially in wildlife from remote regions (Martin et al., 2003, 2004).

The differences between the major PFASs in Baikal seals between the matrices were assessed by principal component analysis (PCA) together with Spearman rank correlations (r_s). The PCA accounted for 83.4 % of the variance (Figs. S7–S9, Table S7). Significant correlations were observed between: PFUnA and PFDoDa ($r_s = 0.62$; $p < 0.001$); PFOS and PFDA ($r_s = 0.59$; $p < 0.001$); and between PFDoDA and PFTriDA ($r_s = 0.71$; $p < 0.001$). Positive correlations between PFOS and other PFASs, and correlations between long chain PFCAs were observed in other studies suggesting similar sources and concurrent exposure routes (Taylor et al., 2021). Both PFDoDA ($r = -0.31$; $p < 0.05$) and PFTriDA ($r = -0.32$; $p < 0.05$) were moderately inversely correlated to body mass. This can indicate that PFDoDA and PFTriDA are efficiently excreted from mother to the foetus in utero, but not easily excreted from the mother into her milk, as shown in hooded seals (*Cystophora cristata*) (Grønnestad et al., 2017). Thus, this results in a biodilution in the growing pup, and hence an inverse relationship between body mass and the concentrations of these two long chain PFCAs (Grønnestad et al., 2017).

As only two adult females were sampled, statistics were not employed to evaluate differences in PFASs concentrations between pups and adults. However, median concentrations of Σ_9PFASs (PFOS, C_8–C_{14} PFCAs, P37DMOA) were higher in plasma, liver and brain of pups than in the corresponding tissues of the adult females [pups (medians): 36.2 ng/g w.w.; 28.6 ng/g w.w.; 6.70 ng/g w.w., respectively, versus adults (medians): 22.2 ng/g w.w.; 17.2 ng/g w.w.; 1.04 ng/g w.w, respectively]. This is consistent with literature for Baikal seals (Ishibashi et al., 2008) and other pinnipeds (Grønnestad et al., 2017; Shaw et al., 2009). Higher concentrations of PFASs in seal pups demonstrated maternal transfer of PFASs, of which in-utero transfer is considered the primary pathway (Grønnestad et al., 2017). Baikal seal pups are born in snow-covered burrows on the ice, nursed for 2-2.5 months, before transitioning to an adult diet (Ozersky et al., 2017). Therefore, concentrations of PFASs in suckling/recently weaned pups reflects in utero and lactational transfer. Consequently, the ratios between pups and mother ($n = 2$) were calculated to investigate the extent of maternal pup transfer of PFASs in Baikal seals (Table S8), where a ratio of >1 represented offloading from mother to pup (i.e., higher concentration in pup than in mother). In general, ratios between mothers and pups in plasma, liver and the brain were >1 for the majority of PFASs, except for PFOS (where the ratios ranged from 0.5 to 0.9). As this study consisted of only two mother-pup pairs, it is challenging to elucidate any clear trends. Therefore, studies with larger sample sizes are needed to better assess maternal transfer of PFASs in various seal species. Concentration differences in PFASs with sex and age were also assessed. There were no significant concentration differences in PFASs between sexes, potentially due to the analysis being carried out on sexually immature Baikal seal pups.

3.3. Human exposure and consumer risk assessment

The Baikal seal is a traditional food for local communities, in particular the Evenki and Buryat people that inhabit the shores of Lake Baikal. In recent years, the population of Baikal seals has increased (Petrov et al., 2021). Currently, the liver, meat, heart, and lungs of the Baikal seal are consumed. Liver in particular is perceived as a delicacy and often eaten raw by local hunters and their families. Tolerable weekly intake (TWI) is a metric used to assess the maximum amount of a chemical (e.g., contaminants) in food that can be consumed weekly without risking adverse health effects.

Recently, the provisional TWI for the combined exposure to the sum of four PFASs (Σ_4PFASs; PFOS, PFOA, PFNA and PFHxS) from diet was reduced by the European food safety authority (EFSA) to just 4.4 ng kg^{-1} body weight (bw) week^{-1} (European Food Safety Authority et al., 2020). Therefore, the possible risks arising from consumption of Baikal seal liver and muscle (meat) were assessed at 3 concentration levels via the determination of estimated weekly intake (EWI; ng kg^{-1} bw week^{-1}; Table S9). The EWIs were calculated using the following steady-state kinetic equation (Eq. (1)):

$$EWI = \frac{[\Sigma_4\text{PFASs}] \times amount\ consumed\ per\ week}{body\ weight} \quad (1)$$

Eq. (1) was used for the determination of EWI of those PFASs, where Σ_4 PFASs is the sum concentration of PFOS, PFOA, PFNA and PFHxS in ng/g w.w. The average body weight for human adults and children was set at 70 and 30 kg, respectively. The amount consumed per week was set at 150 and 75 g for adults and children, respectively. The three different concentrations were set as the minimum (Liver: 2.52 ng/g w.w; muscle: 0.34 ng/g w.w.), median (Liver: 14.8 ng/g w.w; muscle: 1.98 ng/g w.w.), and maximum (Liver: 29.4 ng/g w.w; muscle: 3.92 ng/g w.w.) concentration of Σ_4PFASs. The reported muscle concentrations were approximated from the determined liver concentrations herein based on two previous studies on the tissue distribution of PFASs in harbour seals (Ahrens et al., 2009a; Van de Vijver et al., 2005), where the average concentration of the Σ_4PFASs in muscle tissues was 0.098 times the concentration in liver. Although, liver concentrations of PFASs were comparatively low relative to other pinnipeds, consumption of both Baikal seal liver and muscle can exceed current EFSA guidelines (Fig. 4, Table S9). The consumption of Baikal seal is however seasonal, with hunting occurring over a 1.5-month period. Therefore, TWIs may only be exceeded for a short period reflecting seasonal consumption. Nonetheless, due to the cumulative nature of PFASs and the occurrence of multiple known and unknown congeners, actual intake of PFASs is likely higher than the estimates in our study.

4. Conclusions

The occurrence of 33 PFASs was assessed in the plasma, liver, blubber, and brain of Baikal seals from Lake Baikal, Russia. The presence of PFASs in suckling and pre-weaned Baikal seal pups demonstrated maternal transfer as an important exposure pathway of pups to anthropogenic contaminants. PFOS and long chain PFCAs were the most frequently determined PFASs, with the highest concentrations determined in plasma and liver. Relative to other species of pinnipeds, Baikal seals had lower concentrations of PFOS, while concentrations of long chain PFCAs were similar (Figs. 1 and 2). In contrast to legacy PFASs, novel PFASs were either infrequently detected or not found in Baikal seals. Although, the concentrations of PFASs (especially PFOS) were comparatively low in relation to other pinnipeds, the human consumption of Baikal seal liver and muscle, could exceed current regulatory guidelines set by EFSA. This emphasises the importance of PFAS biomonitoring in wildlife and subsequent human exposure assessment. Furthermore, considering the role of pinnipeds as sentinels for monitoring the health of aquatic environments and the potential adverse effects of PFASs, continuous biomonitoring of PFASs is recommended.

CRediT authorship contribution statement

Shannen T.L. Sait: Data curation, Formal analysis, Investigation, Methodology, Resources, Software, Supervision, Validation, Visualization, Writing – original draft. **Silje F. Rinø:** Data curation, Formal analysis,

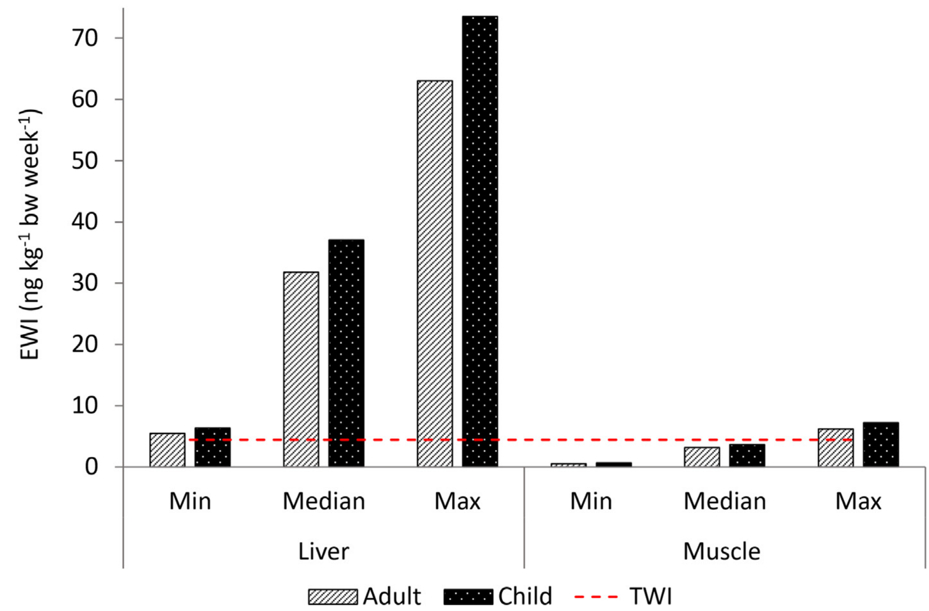

Fig. 4. Estimated weekly intake (EWI) of PFASs in Baikal seal liver and muscle (approximated from liver) at minimum, median and maximum concentrations. Red stippled line displays the provisional tolerable weekly intake (TWI) set by the EFSA (4.4 ng kg^{-1} bw week).

Investigation, Writing – review & editing. **Susana V. Gonzalez:** Resources, Writing – review & editing. **Mikhail V. Pastukhov:** Funding acquisition, Methodology, Resources, Writing – review & editing. **Vera I. Poletaeva:** Funding acquisition, Methodology, Resources, Writing – review & editing. **Julia Farkas:** Resources, Writing – review & editing. **Bjørn M. Jenssen:** Conceptualization, Funding acquisition, Investigation, Methodology, Project administration, Resources, Supervision, Writing – review & editing. **Tomasz M. Ciesielski:** Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Methodology, Resources, Supervision, Validation, Visualization, Writing – review & editing. **Alexandros G. Asimakopoulos:** Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Resources, Software, Supervision, Validation, Visualization, Writing – review & editing.

Data availability

Data will be made available on request.

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.2023.164096.

References

Ahrens, L., Siebert, U., Ebinghaus, R., 2009a. Total body burden and tissue distribution of polyfluorinated compounds in harbor seals (*Phoca vitulina*) from the German bight. Mar. Pollut. Bull. 58 (4), 520–525. https://doi.org/10.1016/J.MARPOLBUL.2008.11.030.

Ahrens, L., Siebert, U., Ebinghaus, R., 2009b. Temporal trends of polyfluoroalkyl compounds in harbor seals (*Phoca vitulina*) from the German bight, 1999-2008. Chemosphere 76 (2), 151–158. https://doi.org/10.1016/j.chemosphere.2009.03.053.

Asimakopoulos, A.G., Thomaidis, N.S., 2015. Bisphenol A, 4-t-octylphenol, and 4-nonylphenol determination in serum by hybrid solid phase extraction–precipitation technology technique tailored to liquid chromatography–tandem mass spectrometry. J. Chromatogr. B 986–987, 85–93. https://doi.org/10.1016/J.JCHROMB.2015.02.009.

Boisvert, G., Sonne, C., Rigét, F.F., Dietz, R., Letcher, R.J., 2019. Bioaccumulation and biomagnification of perfluoroalkyl acids and precursors in East Greenland polar bears and their ringed seal prey. Environ. Pollut. 252, 1335–1343. https://doi.org/10.1016/J.ENVPOL.2019.06.035.

Bossi, R., Riget, F.F., Dietz, R., 2005. Temporal and spatial trends of perfluorinated compounds in ringed seal (*Phoca hispida*) from Greenland. Environ. Sci. Technol. 39 (19), 7416–7422. https://doi.org/10.1021/ES0508469/ASSET/IMAGES/LARGE/ES0508469F00004.JPEG.

Butt, C.M., Muir, D.C.G., Stirling, I., Kwan, M., Mabury, S.A., 2007. Rapid response of arctic ringed seals to changes in perfluoroalkyl production. Environ. Sci. Technol. 41 (1), 42–49. https://doi.org/10.1021/ES061267M/SUPPL_FILE/ES061267MSI20060914_054122.PDF.

Butt, C.M., Muir, D.C.G., Mabury, S.A., 2014. Biotransformation pathways of fluorotelomer-based polyfluoroalkyl substances: a review. Environ. Toxicol. Chem. 33 (2), 243–267. https://doi.org/10.1002/ETC.2407.

Ciesielski, T., Pastukhov, M.V., Fodor, P., Bertenyi, Z., Namieśnik, J., Szefer, P., 2006. Relationships and bioaccumulation of chemical elements in the Baikal seal (*Phoca sibirica*). Environ. Pollut. 139 (2), 372–384. https://doi.org/10.1016/J.ENVPOL.2004.12.040.

Dassuncao, C., Pickard, H., Pfohl, M., Tokranov, A.K., Li, M., Mikkelsen, B., Slitt, A., Sunderland, E.M., 2019. Phospholipid levels predict the tissue distribution of poly- and perfluoroalkyl substances in a marine mammal. Environ. Sci. Technol. Lett. 6 (3), 119. https://doi.org/10.1021/ACS.ESTLETT.9B00031.

De Swart, R.L., Ross, P.S., Vos, J.G., Osterhaus, A.D., 1996. Impaired immunity in harbour seals (*Phoca vitulina*) exposed to bioaccumulated environmental contaminants: review of a long-term feeding study. Environ. Health Perspect. 104 (Suppl. 4), 823–828.

Dietz, R., Rigét, F.F., Galatius, A., Sonne, C., Teilmann, J., Bossi, R., 2012. Spatial trends of perfluorochemicals in harbor seals (*Phoca vitulina*) from Danish waters. Sci. Total Environ. 414, 732–737. https://doi.org/10.1016/j.scitotenv.2011.09.048.

Dietz, R., Letcher, R.J., Desforges, J.P., Eulaers, I., Sonne, C., Wilson, S., Andersen-Ranberg, E., Basu, N., Barst, B.D., Bustnes, J.O., Bytingsvik, J., Ciesielski, T.M., Drevnick, P.E., Gabrielsen, G.W., Haarr, A., Hylland, K., Jenssen, B.M., Levin, M., McKinney, M.A., Víkingsson, G., 2019. Current state of knowledge on biological effects from contaminants on arctic wildlife and fish. Sci. Total Environ. 696, 133792. https://doi.org/10.1016/J.SCITOTENV.2019.133792.

Dong, G.H., Zhang, Y.H., Zheng, L., Liu, W., Jin, Y.H., He, Q.C., 2009. Chronic effects of perfluorooctanesulfonate exposure on immunotoxicity in adult male C57BL/6 mice. Arch. Toxicol. 83 (9), 805–815. https://doi.org/10.1007/S00204-009-0424-0/FIGURES/7.

Eggers Pedersen, K., Basu, N., Letcher, R., Greaves, A.K., Sonne, C., Dietz, R., Styrishave, B., 2015. Brain region-specific perfluoroalkylated sulfonate (PFSA) and carboxylic acid (PFCA) accumulation and neurochemical biomarker responses in east Greenland polar bears (*Ursus maritimus*). Environ. Res. 138, 22–31. https://doi.org/10.1016/J.ENVRES.2015.01.015.

Ellis, D.A., Martin, J.W., de Silva, A.O., Mabury, S.A., Hurley, M.D., Sulbaek Andersen, M.P., Wallington, T.J., 2004. Degradation of fluorotelomer alcohols: a likely atmospheric source of perfluorinated carboxylic acids. Environ. Sci. Technol. 38 (12), 3316–3321. https://doi.org/10.1021/ES049860W/ASSET/IMAGES/LARGE/ES049860WF00003.JPEG.

European Food Safety Authority, Schrenk, D., Bignami, M., Bodin, L., Chipman, J.K., Mazo, J. del, Grasl-Kraupp, B., Hogstrand, C., Hoogenboom, L., Leblanc, J.C., Nebbia, C.S., 2020. Risk to human health related to the presence of perfluoroalkyl substances in food. EFSA J. 18, e06223.

Facciola, N., Houde, M., Muir, D.C.G., Ferguson, S.H., McKinney, M.A., 2022. Feeding and contaminant patterns of sub-arctic and arctic ringed seals: potential insight into climate change-contaminant interactions. Environ. Pollut. 313, 120108. https://doi.org/10.1016/J.ENVPOL.2022.120108.

Galatius, A., Bossi, R., Sonne, C., Rigét, F.F., Kinze, C.C., Lockyer, C., Teilmann, J., Dietz, R., 2013. PFAS profiles in three North Sea top predators: metabolic differences among species? Environ. Sci. Pollut. Res. 20 (11), 8013–8020. https://doi.org/10.1007/S11356-013-1633-X/TABLES/2.

Gebbink, W.A., Bossi, R., Rigét, F.F., Rosing-Asvid, A., Sonne, C., Dietz, R., 2016. Observation of emerging per- and polyfluoroalkyl substances (PFASs) in Greenland marine mammals. Chemosphere 144, 2384–2391. https://doi.org/10.1016/J.CHEMOSPHERE.2015.10.116.

Giesy, J.P., Kannan, K., 2001. Global distribution of perfluorooctane sulfonate in wildlife. Environ. Sci. Technol. 35 (7), 1339–1342. https://doi.org/10.1021/ES001834K.

Greaves, A.K., Letcher, R.J., Sonne, C., Dietz, R., 2013. Brain region distribution and patterns of bioaccumulative perfluoroalkyl carboxylates and sulfonates in East Greenland polar bears (*Ursus maritimus*). Environ. Toxicol. Chem. 32 (3), 713–722. https://doi.org/10.1002/ETC.2107.

Grønnestad, R., Villanger, G.D., Polder, A., Kovacs, K.M., Lydersen, C., Jenssen, B.M., Borgå, K., 2017. Maternal transfer of perfluoroalkyl substances in hooded seals. Environ. Toxicol. Chem. 36 (3), 763–770. https://doi.org/10.1002/ETC.3623.

Houde, M., Wells, R.S., Fair, P.A., Bossart, G.D., Hohn, A.A., Rowles, T.K., Sweeney, J.C., Solomon, K.R., Muir, D.C.G., 2005. Polyfluoroalkyl compounds in free-ranging bottlenose dolphins (*Tursiops truncatus*) from the Gulf of Mexico and the Atlantic Ocean. Environ. Sci. Technol. 39 (17), 6591–6598. https://doi.org/10.1021/ES0506556/SUPPL_FILE/ES0506556SI20050519_091426.PDF.

Houde, M., De Silva, A.O., Muir, D.C.G., Letcher, R.J., 2011. Monitoring of perfluorinated compounds in aquatic biota: an updated review. Environmental Science and Technology. vol. 45, Issue 19. American Chemical Society, pp. 7962–7973. https://doi.org/10.1021/es104326w.

Ishibashi, H., Iwata, H., Kim, E.Y., Tao, L., Kannan, K., Amano, M., Miyazaki, N., Tanabe, S., Batoev, V.B., Petrov, E.A., 2008. 1. Contamination and effects of perfluorochemicals in baikal seal (*Pusa sibirica*). 1. Residue level, tissue distribution, and temporal trend. Environ. Sci. Technol. 42 (7), 2295–2301. https://doi.org/10.1021/ES072054F/SUPPL_FILE/ES072054F-FILE001.PDF.

Iwata, H., Watanabe, M., Okajima, Y., Tanabe, S., Amano, M., Miyazaki, N., Petrov, E.A., 2004. Toxicokinetics of PCDD, PCDF, and coplanar PCB congeners in Baikal seals, Pusa sibirica: age-related accumulation, maternal transfer, and hepatic sequestration. Environ. Sci. Technol. 38 (13), 3505–3513.

Kannan, K., Koistinen, J., Beckmen, K., Evans, T., Gorzelany, J.F., Hansen, K.J., Jones, P.D., Helle, E., Nyman, M., Giesy, J.P., 2001. Accumulation of perfluorooctane sulfonate in marine mammals. Environ. Sci. Technol. 35 (8), 1593–1598. https://doi.org/10.1021/ES001873W.

Kratzer, J., Ahrens, L., Roos, A., Bäcklin, B.M., Ebinghaus, R., 2011. Temporal trends of polyfluoroalkyl compounds (PFCs) in liver tissue of grey seals (*Halichoerus grypus*) from the Baltic Sea, 1974–2008. Chemosphere 84 (11), 1592–1600. https://doi.org/10.1016/J.CHEMOSPHERE.2011.05.036.

Mamaev, L.V., Denikina, N.N., Belikov, S.I., Volchkov, V.E., Visser, I.K.G., Fleming, M., Kai, C., Harder, T.C., Liess, B., Osterhaus, A.D.M.E., Barrett, T., 1995. Characterisation of morbilliviruses isolated from Lake Baikal seals (*Phoca sibirica*). Vet. Microbiol. 44 (2–4), 251–259. https://doi.org/10.1016/0378-1135(95)00018-6.

Mamaev, L.V., Visser, I.K.G., Belikov, S.I., Denikina, N.N., Harder, T., Goatley, L., Rima, B., Edginton, B., Osterhaus, A., Barrett, T., 1996. Canine distemper virus in Lake Baikal seals (*Phoca sibirica*). Vet. Rec. 138 (18), 437–439.

Mamontov, A.A., Mamontova, E.A., Tarasova, E.N., 2019. Persistent organic pollutants in Baikal seal (*Pusa sibirica*) blubber. Russ. J. Gen. Chem. 89 (13), 2791–2797.

Mamontova, E.A., Tarasova, E.N., Mamontov, A.A., Mamontov, A.M., 2020. Freshwater seal as a source of direct and indirect increased human exposure to persistent organic pollutants in a background area. Sci. Total Environ. 715, 136922. https://doi.org/10.1016/J.SCITOTENV.2020.136922.

Martin, J., Mabury, S., Solomon, K., Muir, D., 2003. Bioconcentration and tissue distribution of perfluorinated acids in rainbow trout (*Oncorhynchus mykiss*). Environ. Toxicol. Chem. 22 (1), 196–204. https://doi.org/10.1002/ETC.5620220126.

Martin, J., Smithwick, M., Braune, B., Hoekstra, P., Muir, D., Mabury, S., 2004. Identification of long-chain perfluorinated acids in biota from the Canadian Arctic. Environ. Sci. Technol. 38 (2), 373–380. https://doi.org/10.1021/ES034727.

Moore, M.V., Hampton, S.E., Izmest'Eva, L.R., Silow, E.A., Peshkova, E.V., Pavlov, B.K., 2009. Climate change and the World's "Sacred Sea"—Lake Baikal, Siberia. BioScience 59 (5), 405–417. https://doi.org/10.1525/BIO.2009.59.5.8.

Muir, D., Alaee, M., Butt, C., Braune, B., Helm, P., Mabury, S., 2004. New contaminants in Arctic biota. Synopsis of Research Conducted Under the 2003–2004 Northern Contaminants Program, pp. 139–148.

Nakata, H., Tanabe, S., Tatsukawa, R., Amano, M., Miyazaki, N., Petrov, E.A., 1997. Bioaccumulation profiles of polychlorinated biphenyls including coplanar congeners and possible toxicological implications in Baikal seal (*Phoca sibirica*). Environ. Pollut. 95 (1), 57–65.

Nakata, H., Tanabe, S., Tatsukawa, R., Amano, M., Miyazaki, N., Petrov, E.A., 2002. Persistent organochlorine residues and their accumulation kinetics in Baikal seal (*Phoca sibirica*) from Lake Baikal, Russia. Environ. Sci. Technol. 29 (11), 2877–2885. https://doi.org/10.1021/ES00011A026.

National Institute of Environmental Health Services, 2022. Perfluoroalkyl and Polyfluoroalkyl Substances (PFAS). https://doi.org/10.1542/9781610022194-part04-ch36.

Nomokonova, T., Losey, R.J., Iakunaeva, V.N., Emel'ianova, I.A., Baginova, E.A., Pastukhov, M.V., 2013. People and seals at Siberia's Lake Baikal. J. Ethnobiol. 33 (2), 259–280.

Ozersky, T., Pastukhov, M.V., Poste, A.E., Deng, X.Y., Moore, M.V., 2017. Long-term and ontogenetic patterns of heavy metal contamination in Lake Baikal seals (*Pusa sibirica*). Environ. Sci. Technol. 51 (18), 10316–10325. https://doi.org/10.1021/ACS.EST.7B00995/ASSET/IMAGES/LARGE/ES-2017-00995K_0005.JPEG.

Pachkowski, B., Post, G.B., Stern, A.H., 2019. The derivation of a reference dose (RfD) for perfluorooctane sulfonate (PFOS) based on immune suppression. Environ. Res. 171, 452–469. https://doi.org/10.1016/J.ENVRES.2018.08.004.

Pastukhov, V.D., 1993. The Baikal Seal. (in Russian)VO Nauka.

Pérez, F., Nadal, M., Navarro-Ortega, A., Fàbrega, F., Domingo, J.L., Barceló, D., Farré, M., 2013. Accumulation of perfluoroalkyl substances in human tissues. Environ. Int. 59, 354–362. https://doi.org/10.1016/J.ENVINT.2013.06.004.

Petrov, E.A., Kupchinsky, A.B., Fialkov, V.A., 2021. Summer coastal rookeries and perspectives of the Baikal seal (*Pusa sibirica*) population in the conditions of the global warming. Biosyst. Divers. 29 (4), 387–392. https://doi.org/10.15421/012149.

Powley, C.R., George, S.W., Russell, M.H., Hoke, R.A., Buck, R.C., 2008. Polyfluorinated chemicals in a spatially and temporally integrated food web in the Western Arctic. Chemosphere 70 (4), 664–672. https://doi.org/10.1016/J.CHEMOSPHERE.2007.06.067.

Prevedouros, K., Cousins, I.T., Buck, R.C., Korzeniowski, S.H., 2006. Sources, fate and transport of perfluorocarboxylates. Environ. Sci. Technol. 40 (1), 32–44. https://doi.org/10.1021/ES0512475/SUPPL_FILE/ES0512475.PDF.

Quakenbush, L.T., Citta, J.J., 2008. Perfluorinated contaminants in ringed, bearded, spotted, and ribbon seals from the Alaskan Bering and Chukchi seas. Mar. Pollut. Bull. 56 (10), 1809–1814. https://doi.org/10.1016/J.MARPOLBUL.2008.06.005.

Rigét, F., Bossi, R., Sonne, C., Vorkamp, K., Dietz, R., 2013. Trends of perfluorochemicals in Greenland ringed seals and polar bears: indications of shifts to decreasing trends. Chemosphere 93 (8), 1607–1614. https://doi.org/10.1016/j.chemosphere.2013.08.015.

Ross, P.S., Noël, M., Lambourn, D., Dangerfield, N., Calambokidis, J., Jeffries, S., 2013. Declining concentrations of persistent PCBs, PBDEs, PCDEs, and PCNs in harbor seals (*Phoca vitulina*) from the Salish Sea. Prog. Oceanogr. 115, 160–170. https://doi.org/10.1016/J.POCEAN.2013.05.027.

Routti, H., Krafft, B.A., Herzke, D., Eisert, R., Oftedal, O., 2015. Perfluoroalkyl substances detected in the world's southernmost marine mammal, the Weddell seal (*Leptonychotes weddellii*). Environ. Pollut. 197, 62–67. https://doi.org/10.1016/J.ENVPOL.2014.11.026.

Routti, H., Gabrielsen, G.W., Herzke, D., Kovacs, K.M., Lydersen, C., 2016. Spatial and temporal trends in perfluoroalkyl substances (PFASs) in ringed seals (*Pusa hispida*) from Svalbard. Environ. Pollut. 214, 230–238. https://doi.org/10.1016/J.ENVPOL.2016.04.016.

Sait, S.T.L., Survilienė, V., Jüssi, M., Gonzalez, S.V., Ciesielski, T.M., Jenssen, B.M., Asimakopoulos, A.G., 2023. Determination of steroid hormones in grey seal (*Halichoerus grypus*) blood plasma using convergence chromatography tandem mass spectrometry. Talanta 254, 124109. https://doi.org/10.1016/J.TALANTA.2022.124109.

Sanderson, C.E., Alexander, K.A., 2020. Unchartered waters: climate change likely to intensify infectious disease outbreaks causing mass mortality events in marine mammals. Glob. Chang. Biol. 26 (8), 4284–4301. https://doi.org/10.1111/GCB.15163.

Schiavone, A., Corsolini, S., Kannan, K., Tao, L., Trivelpiece, W., Torres, D., Focardi, S., 2009. Perfluorinated contaminants in fur seal pups and penguin eggs from south Shetland, Antarctica. Sci. Total Environ. 407 (12), 3899–3904. https://doi.org/10.1016/J.SCITOTENV.2008.12.058.

Sedlak, M.D., Greig, D.J., 2012. Perfluoroalkyl compounds (PFCs) in wildlife from an urban estuary. J. Environ. Monit. 14 (1), 146–154. https://doi.org/10.1039/C1EM10609K.

Shaw, S., Berger, M.L., Brenner, D., Tao, L., Wu, Q., Kannan, K., 2009. Specific accumulation of perfluorochemicals in harbor seals (*Phoca vitulina concolor*) from the northwest Atlantic. Chemosphere 74 (8), 1037–1043. https://doi.org/10.1016/J.CHEMOSPHERE.2008.10.063.

Spaan, K.M., van Noordenburg, C., Plassmann, M.M., Schultes, L., Shaw, S., Berger, M., Heide-Jørgensen, M.P., Rosing-Asvid, A., Granquist, S.M., Dietz, R., Sonne, C., Rigét, F., Roos, A., Benskin, J.P., 2020. Fluorine mass balance and suspect screening in marine mammals from the northern hemisphere. Environ. Sci. Technol. 54 (7), 4046–4058. https://doi.org/10.1021/acs.est.9b06773.

Sunderland, E.M., Hu, X.C., Dassuncao, C., Tokranov, A.K., Wagner, C.C., Allen, J.G., 2019. A review of the pathways of human exposure to poly-and perfluoroalkyl substances (PFASs) and present understanding of health effects. J. Expo. Sci. Environ. Epidemiol. 29 (2), 131–147.

Tao, L., Kannan, K., Kajiwara, N., Costa, M.M., Fillmann, G., Takahashi, S., Tanabe, S., 2006. Perfluorooctanesulfonate and related fluorochemicals in albatrosses, elephant seals, penguins, and polar skuas from the southern ocean. Environ. Sci. Technol. 40 (24), 7642–7648. https://doi.org/10.1021/ES061513U/SUPPL_FILE/ES061513USI20060919_111750.PDF.

Taylor, S., Terkildsen, M., Stevenson, G., de Araujo, J., Yu, C., Yates, A., McIntosh, R.R., Gray, R., 2021. Per and polyfluoroalkyl substances (PFAS) at high concentrations in neonatal Australian pinnipeds. Sci. Total Environ. 786, 147446. https://doi.org/10.1016/J.SCITOTENV.2021.147446.

The Norwegian Environmental Agency, 2013. Perfluorinated alkylated substances, brominated flame retardants and chlorinated paraffins in the Norwegian environment - screening 2013 - Miljødirektoratet. https://www.miljodirektoratet.no/publikasjoner/2013/

oktober-2013/perfluorinated-alkylated-substances-brominated-flame-retardants-and-chlorinated-paraffins-in-the-norwegian-environment—screening-2013/.

Tomy, G.T., Budakowski, W., Halldorson, T., Helm, P.A., Stern, G.A., Friesen, K., Pepper, K., Tittlemier, S.A., Fisk, A.T., 2004. Fluorinated organic compounds in an eastern arctic marine food web. Environ. Sci. Technol. 38 (24), 6475–6481. https://doi.org/10.1021/ES049620G/SUPPL_FILE/ES049620GSI20040721_033719.PDF.

van de Vijver, K.I., Hoff, P., Das, K., Brasseur, S., van Dongen, W., Esmans, E., Reijnders, P., Blust, R., de Coen, W., 2005. Tissue distribution of perfluorinated chemicals in harbor seals (*Phoca vitulina*) from the Dutch Wadden Sea. Environ. Sci. Technol. 39 (18), 6978–6984.

Van De Vijver, K.I., Holsbeek, L., Das, K., Blust, R., Joiris, C., De Coen, W., 2007. Occurrence of perfluorooctane sulfonate and other perfluorinated alkylated substances in harbor porpoises from the Black Sea. Environ. Sci. Technol. 41 (1), 315–320. https://doi.org/10.1021/ES060827E/SUPPL_FILE/ES060827ESI20060724_033112.XLS.

Vike-Jonas, K., Gonzalez, S.V., Mortensen, Å.K., Ciesielski, T.M., Farkas, J., Venkatraman, V., Pastukhov, M.V., Jenssen, B.M., Asimakopoulos, A.G., 2021. Rapid determination of thyroid hormones in blood plasma from glaucous gulls and Baikal seals by HybridSPE®-LC-MS/MS. J. Chromatogr. B 1162, 122447. https://doi.org/10.1016/J.JCHROMB.2020.122447.

Watanabe, Y.Y., Baranov, E.A., Miyazaki, N., 2020. Ultrahigh foraging rates of Baikal seals make tiny endemic amphipods profitable in Lake Baikal. Proc. Natl. Acad. Sci. U. S. A. 117 (49), 31242–31248. https://doi.org/10.1073/PNAS.2014021117/SUPPL_FILE/PNAS.2014021117.SM03.MP4.

Xiao, F., 2017. Emerging poly- and perfluoroalkyl substances in the aquatic environment: a review of current literature. Water Res. 124, 482–495. https://doi.org/10.1016/J.WATRES.2017.07.024.

Xiong, D., 2021. Bioaccumulation and Biomagnification of Perfluoroalkyl Substances (PFAS) in a Subarctic Ringed Seal Food Web in Lake Melville, Northern Labrador, Canada. http://hdl.handle.net/1807/108611.

Yoshii, K., Melnik, N.G., Timoshkin, O.A., Bondarenko, N.A., Anoshko, P.N., Yoshioka, T., Wada, E., 1999. Stable isotope analyses of the pelagic food web in Lake Baikal. Limnol. Oceanogr. 44 (3), 502–511. https://doi.org/10.4319/LO.1999.44.3.0502.