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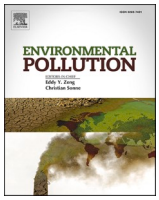

Emerging and legacy contaminants in common minke whale from the Barents sea☆

Clare Andvik [a], Tore Haug [b], Jan L. Lyche [c], Katrine Borgå [a,*]

[a] Department of Biosciences, University of Oslo, NO-0316, Oslo, Norway
[b] Institute of Marine Research, Fram Centre, PO Box 6606, Stakkevollan, NO-9296, Tromsø, Norway
[c] Department of Food Safety and Infection Biology, Norwegian University of Life Sciences, P.O. Box 5003, NO-1432, Ås, Norway

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ABSTRACT

Persistent organic pollutants (POPs), including brominated flame retardants (BFRs), perfluoroalkyl substances (PFAS) and metals, can accumulate in marine mammals and be transferred to offspring. In this study, we analyzed 64 lipophilic POPs, including four emerging BFRs, in the blubber, liver and muscle of 17 adult common minke whales (*Balaenoptera acutorostrata*) from the Barents Sea to investigate occurrence and tissue partitioning. In addition, the placental transfer concentration ratios of 14 PFAS and 17 metals were quantified in the muscle of nine female-fetus pairs to investigate placental transfer. Legacy lipophilic POPs were the dominating compound group in every tissue, and we observed generally lower levels compared to previous studies from 1992 to 2001. We detected the emerging BFRs hexabromobenzene (HBB) and pentabromotoluene (PBT), but in low levels compared to the legacy POPs. We detected nine PFAS, and levels of perfluorooctane sulfonate (PFOS) were higher than detected from the same population in 2011, whilst levels of Hg were comparable to 2011. Levels of lipophilic contaminants were higher in blubber compared to muscle and liver on both a wet weight and lipid adjusted basis, but tissue partitioning of the emerging BFRs could not be determined due to the high number of samples below the limit of detection. The highest muscle ΣPFAS levels were quantified in fetuses (23 ± 8.7 ng/g ww), followed by adult males (7.2 ± 2.0 ng/gg ww) and adult females (4.5 ± 1.1 ng/g ww), showing substantial placental transfer from mother to fetus. In contrast, Hg levels in the fetus were lower than the mother. Levels were under thresholds for risk of health effects in the whales. This study is the first to report occurrence and placental transfer of emerging contaminants in common minke whales from the Barents Sea, contributing valuable new data on pollutant levels in Arctic wildlife.

1. Introduction

Arctic marine mammals are exposed to and accumulate high levels of legacy persistent organic pollutants (POPs) and metals (Braune et al., 2015; Dietz et al., 2019; Letcher et al., 2010). These POPs and metals reach the Arctic through long range transport, with few local sources (Burkow and Kallenborn, 2000; Vorkamp et al., 2019), and may biomagnify to concentrations exceeding the threshold of negative effects in certain species (Andvik et al., 2020; Dietz et al., 2019). Many legacy POPs, such as polychlorinated biphenyls (PCBs) and brominated flame retardants (BFRs) are lipophilic, preferentially partitioning in fatty tissues, whereas the non-regulated ("emerging") contaminant group perfluoroalkyl substances (PFAS) bind to proteins (Andvik et al., 2021; Jones et al., 2003; Yordy et al., 2010). A number of emerging contaminants of concern have been identified in the Arctic (AMAP, 2017), and for several of these there is little knowledge on levels and tissue distribution in marine mammals.

Marine mammals such as whales are often considered sentinel species for marine ecosystem health due to their long lives, thick blubber layer and mid-to-high trophic position (Bossart, 2011; Ross, 2000). The common minke whale (*Balaenoptera acutorostrata*) is one of the smallest baleen whales, which in the Northeast Atlantic annually migrates from lower latitudes to Arctic waters in the spring and early summer to feed opportunistically on a variety of fish such as capelin (*Mallotus vilosus*), herring (*Clupea harengus*), mackerel (*Scomber scombrus*), cod (*Gadus morhua*), and crustaceans such as krill (Haug et al., 2002; Jonsgård,

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* Corresponding author.
E-mail address: katrine.borga@ibv.uio.no (K. Borgå).

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1966; Olsen & Holst, 2001). Both lipophilic pollutants such as PCBs, and protein-binding pollutants such as PFAS and mercury (Hg), have, over the last few decades, been found in common minke whales from the Barents Sea (Julshamn et al., 2012; Kleivane and Skåre, 1998; Maage et al., 2017). Lipophilic POP levels are often higher in males than females (Kleivane and Skåre, 1998) and depend on prey availability and migratory routes (Hobbs et al., 2003). Due to the human consumption of minke whale muscle in Norway, most previous studies have focused on contaminant levels in muscle (Frydenlund and Øvrevoll, 2002; Julshamn et al., 2012; Maage et al., 2017). The last study to quantify legacy POPs in blubber of common minke whales from the Barents Sea was in 2001 (Skåre et al., 2001), and besides a small screening of PFAS in muscle by the Norwegian Food Safety Authority (Mattilsynet) in 2011 (Julshamn et al., 2012), there have been little to no quantification of emerging contaminants in these whales.

The common minke whale has a breeding cycle of approximately 14 months, including 10 months of gestation (Horwood, 1990), and approximately 95% of all sexually mature females of the Northeast Atlantic stock of common minke whales are pregnant annually (Christensen, 1981). Whilst lipophilic POPs are efficiently transferred via milk in marine mammals (*e.g.* Bacon et al., 1992; Hickie et al., 1999), studies post-gestation suggest that protein-associated Hg and PFAS are more efficiently maternally transferred through the placenta than milk (Frodello et al., 2002; Grønnestad et al., 2017; Habran et al., 2011; Hitchcock et al., 2017; Houde et al., 2006a). Hepatic PFAS has previously been detected in a single common minke whale fetus harvested in Greenland (Spaan et al., 2020), and a killer whale (*Orcinus orca)* mother-fetus pair harvested in Greenland (Gebbink et al., 2016). Hg has been found in pilot whale (*Globicephala melas)* fetuses harvested in the Faroe Islands (Julshamn et al., 1987), but there have been no studies to date on placental transfer during gestation of metals and PFAS in common minke whales.

The objectives of the present study were to 1) investigate the presence and tissue distribution of legacy and emerging contaminants in male and female Norwegian common minke whales in the context of differences in sex and diet 2) investigate placental transfer of protein associated metals and PFAS to common minke whale fetuses during gestation and 3) compare levels to thresholds for health effects for the whales. Results are relevant for national, regional and global monitoring bodies to understand pollution status of the common minke whale population, which also reflects the status of the Arctic Barents Sea marine environment.

2. Materials and methods

2.1. Sampling

Common minke whale blubber, muscle, liver and skin samples were collected from 7 mature males and 10 pregnant females onboard a Norwegian whaling vessel between August 5 and August 22, 2019 by the Norwegian Institute of Marine Research in the northern Barents Sea (74°23′N–77°21′N and 11°28′E−24°32′E) (Fig. 1; Table 1). Muscle samples were obtained from nine of the fetuses of the pregnant females (Table 1). Samples were taken from the same spot on each whale, and each sample was wrapped in aluminum foil, placed in labelled plastic bags, and stored at −20 °C during all transport and storage. Total body length of the whale was measured in a straight line from the tip of the upper jaw to the apex of the tail fluke notch, and fetus lengths were measured in a similar way. Sex was determined by physical observation, and mature males defined as longer than 670 cm and mature females longer than 730 cm, as per Christensen (1981).

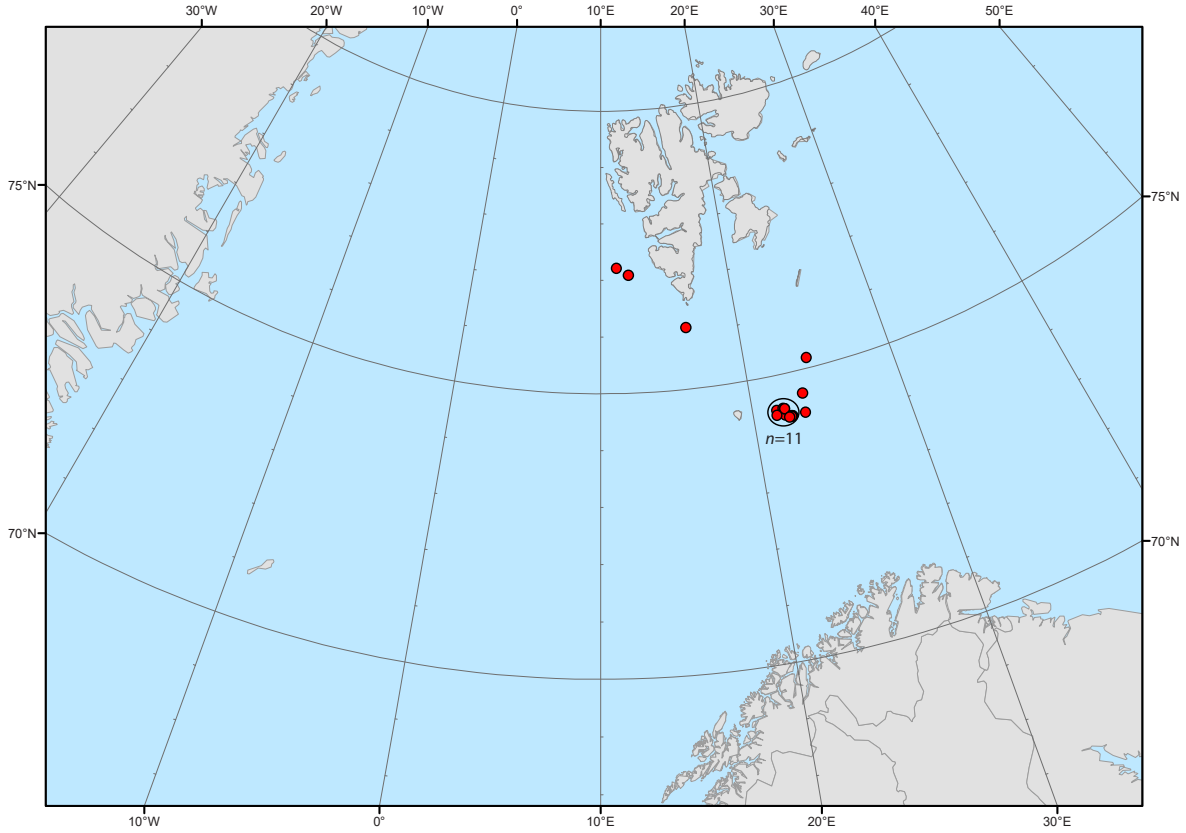

Fig. 1. Map of the Barents Sea and Svalbard showing where each of the 17 common minke whale (*Balaenoptera acutorostrata)* were sampled in August 2019 and indicating the overlapping points for 11 of the whales.

Table 1
The number of individuals (n), life stage, sex, length (mean and range) and tissue types collected from each common minke whale (*Balaenoptera acutorostrata*) harvested from the Barents Sea in August 2019. Analyses completed for each tissue type is indicated by a shaded green box. POPs = Persistent organic pollutants (lipophilic), PFAS = perfluoroalkyl substances and SIA = stable isotope analysis.

n	Life stage	Sex	Length (cm)	Tissues	POPs	PFAS	Metals	SIA
7	Adult	Male	815 (740 – 910)	Blubber				
				Liver				
				Muscle				
				Skin				
10	Adult	Female	781 (680 – 840)	Blubber				
				Liver				
				Muscle				
				Skin				
9	Fetus	NA	116 (88 – 181)	Muscle				

2.2. Laboratory analyses

Lipophilic POPs were analyzed in blubber, muscle and liver samples of all adult whales at the Laboratory of Environmental Toxicology at the Norwegian University of Life Sciences (MT-laboratory NMBU), Oslo, Norway (Table 1). Fetuses were not included due to limited sample material. Lipophilic POPs included 34 PCB congeners, 16 organochlorinated pesticides (including dichlorodiphenyltrichloroethane (DDTs), chlordanes (CHLs) and hexachlorobenzene (HCB)), 14 legacy BFRs (13 polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecane (HBCDD)), and four emerging BFRs: pentabromotoluene (PBT), hexabromobenzene (HBB), pentabromoethylbenzene (PBEB) and 3-dibromopropyl-2,4,6-tribromophenyl ether (DPTE). We used a multicomponent method on approximately 0.5 g of tissue utilizing extraction by cyclohexane and acetone, and sulfuric acid to remove fat and lipid residues, first described in Brevik (1978), and since modified for a range of compounds and biological matrices (Andersen et al., 2001; Bernhoft and Skaare, 1994; Polder et al., 2014, 2008). The method is fully described for killer whale biopsies (Andvik et al., 2020) and stranded killer whales (Andvik et al., 2021). The MT-laboratory is accredited by the Norwegian Accreditation for the determination of lipophilic POPs in biological matrices of animal origin, according to the requirements of NS-EN/IEC 17025:2005 (Test 137). Certified reference materials CRM350, CRM598 and CRM2525 are analyzed on a quarterly basis and had satisfactory reproducibility. In addition, each run included three blank samples, one blind (corn oil and internal standards), two recovery samples (corn oil, internal standards and spiked analytes of interest) and one in-house reference material (seal blubber with known concentrations). The limit of detection was determined as three times the average noise in the chromatograms. A full list of analyzed compounds can be found in Supplementary Table S1, along with limits of detection and internal standard recoveries.

We analyzed 14 PFAS in the muscle and liver of adults, and muscle of fetuses, at the Laboratory of Environmental Toxicology at NMBU: perfluorobutanoate (PFBA); perfluorohexanoate (PFHxA); perfluorohexane sulfonic acid (PFHxS); perfluoroheptanoate (PFHpA); perfluorooctanoate (PFOA); perfluorooctane sulfonate (PFOS); perfluorooctane sulfonamide (PFOSA); perfluorobutane sulfonate (PFBS); perfluorononanoic acid (PFNA); perfluorodecanoic acid (PFDA); perfluoroundecanoic acid (PFUnDA); perfluorododecanoic acid (PFDoDA); perfluorotridecanoic acid (PFTrDA) and perfluorotetradecanoic acid (PFTeD). PFASs were extracted by precipitation of proteins using methanol, clean-up using Graphitized Non-Porous Carbon (ENVI-CarbTM) and detection by liquid chromatography coupled to a tandem mass spectrometry (LC-MS/MS), as described previously for marine mammals in Grønnestad et al. (2017) and killer whales in Andvik et al. (2021). Each run of samples included three blank samples, one blind (trout liver with internal standards) and two recovery samples (trout liver with internal standards and PFAS analytes). The MT-laboratory is not accredited for PFAS, however the method for determining the analytes was performed and validated following the same principles as the accredited standard (NS- EN ISO/IEC 17025). The limit of detection was determined as three times the average noise in the chromatograms. Limits of detection for each PFAS and tissue type, and internal standard recoveries, are listed in Supplementary Table S1.

We analyzed 17 metals in the muscle, liver and skin of adult whales and the muscle of fetuses at the Faculty of Environmental Sciences and Natural Resource Management (MINA) at NMBU: Lithium (Li), Magnesium (Mg), Aluminum (Al), Vanadium (V), Chromium (Cr), Iron (Fe), Cobalt (Co), Nickel (Ni), Copper (Cu), Zinc (Zn), Arsenic (As), Selenium (Se), Molybdenum (Mo), Silver (Ag), Cadmium (Cd), Total mercury (Hg) and Lead (Pb). Samples were freeze dried, homogenized and then approximately 100 mg digested using Ultrapure nitric acid (HNO3) in an UltraClave (Milestone, Leutkirch, Germany) for 2 h, with 20 min at the top temperature of 260 °C. The samples were then diluted with Milli-Q water® by a factor of 10 prior to quantification using an 8800 Triple Quadrupole ICP-MS (Agilent Technologies, Tokyo, Japan). Sample blanks and certified reference material (DORM-4, fish protein; DOLT-5, dogfish liver, National Research Council, Ottawa, Canada and 1577 b, bovine liver, National Institute of Standards and Technology, USA) were analyzed in parallel, and the average recoveries of the certified reference materials were within 10% of the reported values (Supplementary Table S2). The limit of detection (LOD) and limit of quantification (LOQ) were calculated from 3 times and 10 times the standard deviation of the blank samples (n = 5) (Table S2). Water content was recorded before and after freeze drying and used to convert each individual result from dry weight (dw) to wet weight (ww). We did not analyze methyl mercury (MeHg) in addition to Total mercury as Maage et al. (2017) found 100% of the total mercury in common minke whale muscle was in MeHg form, and similarly high proportions have been found in a range of other marine mammal tissues (Wagemann et al., 1998). The 17 metals were chosen due to their increased presence in the environment due to anthropogenic activity, and to enable comparisons to previous studies/areas.

Stable isotopes of bulk nitrogen ($\delta^{15}N$) and carbon ($\delta^{13}C$) were analyzed in freeze-dried and homogenized skin and muscle of the adult whales, and conducted at the CLIPT Stable Isotope Laboratory at the University of Oslo. $\delta^{15}N$ values were determined from non-lipid extracted samples and $\delta^{13}C$ values from lipid-extracted samples due to the unpredictable changes in $\delta^{15}N$ values in cetacean skin following lipid

extraction, which is done to control for the low $\delta^{13}C$ values in the lipid fraction of an organism (Tarroux et al., 2010; Yurkowski et al., 2015). The full method and quality assurance is described in Jourdain et al. (2020) on killer whale biopsies, and Andvik et al. (2021) on stranded killer whales, and the internal references and calibrations in this study were within acceptable ranges, with standard deviations for $\delta^{15}N$ 0.003‰ and $\delta^{13}C$ 0.003‰. Summary results (mean and standard deviation) from the lipid-extracted $\delta^{15}N$ and $\delta^{13}C$ values in skin from the same whales are published in MacKenzie et al. (2022).

2.3. Data treatment

Data treatment and statistical analyses were conducted using R (v. 4.2.1) (R Core Team, 2022). Significant explanatory variables were determined by a p-value <0.05 in addition to the confidence interval of the point estimate excluding 0.

We used multiple imputation to replace values under the LOD with a random number between 0 and the LOD, assuming a beta distribution ($\alpha = 5$, $\beta = 1$) to retain the pattern of the dataset (Baccarelli et al., 2005). Values under the LOQ (applicable only to metals) were replaced with a random value between the LOD and LOQ, following the same distribution. When reporting contaminant levels in tables and graphs, we included all analyzed contaminants, including imputed values, to allow for direct comparison. When conducting statistical analyses, we included only those contaminants found in over 70% of the samples of the respective objective to ensure that the dataset did not include a high proportion of substituted numbers. For lipophilic POPs, this excluded PCB-31, PCB-56, BDE-28, BDE-183, BDE-196, BDE-202, BDE-206, BDE-207, BDE-208, BDE-209, PBT, HBB, PBEB and DPTE and the final dataset included 103 imputed values representing 3.7% of the dataset. For PFAS, this excluded PFBA, PFHxA, PFHpA, PFOA, PFTeDA, PFBS and PFHxS and the final dataset included 12 imputed values, 4% of the dataset. For metals, this excluded V. Co was detected in 33% of muscle samples from the mother, but in all but one of the fetuses, so was included for analysis with imputed values which made up 9% of the final dataset.

In adults, we used multivariate analysis to compare and visualize the differences in the levels of lipophilic POPs, and test the significance of the following explanatory variables: Tissue (Blubber, Muscle, Liver), Sex (Female, Male), $\delta^{15}N$ (in muscle) and $\delta^{13}C$ (in muscle). Principle Component Analysis (PCA) was used to visualize the main structure of the data, and a Redundancy Analysis (RDA) based on forward model selection followed by a Monte Carlo forward permutation test (1000 unrestricted permutations) was used to determine significant associations between response and explanatory variables. We log-10 transformed contaminant levels to reduce heteroscodacity and deviance from normal distribution, and the presence of any influential outliers were checked by the Cook's distance test. Rather than lipid normalizing the data, which can often lead to misleading conclusions in inferential statistics (Hebert and Keenleyside, 1995), lipid content (%) was used as a covariate in the PCA after verifying its significance (RDA, $F = 96.5$, $p < 0.001$, 66.3% of constrained variation). As each individual whale had three datapoints in the multivariate analysis (blubber, muscle and liver from each whale), we checked for pesudeoreplication by testing the effect of whale ID. We found the same explanatory variables were significant in the RDA with or without whale ID as a covariate, and it was not included in the analyses due to non-significance (RDA, $F = 0.652$, $p = 0.851$).

For PFAS and Hg, linear mixed effect models were used to assess the combined effect on the log-10 transformed response variables in the adult whales, using whale ID as a random effect and the following fixed effect explanatory variables: Tissue (Muscle, Liver); Sex (Female, Male); $\delta^{15}N$ (in muscle) and $\delta^{13}C$ (in muscle). The best model was selected using backward model selection based on the lowest corrected Akaike Information Criterion (AICc) values. The same explanatory variables were significant when tested on an ordinary linear model without whale ID as a random effect, but, for each, the AICc score was lower, and hence better, for the mixed effect model.

We did not test the significance of Body Length on contaminant levels in the present study, as all whales chosen for the analysis in the present study were sexually mature, and length cannot be used as a proxy for age in common minke whales. We also chose to use $\delta^{15}N$ and $\delta^{13}C$ levels in muscle as explanatory variables, and not in skin in addition, as the two metrics are strongly correlated but stable isotope levels in muscle are known to have a longer turnover rate than other tissues and thus be representative of a longer time period (Boecklen et al., 2011).

To assess placental transfer, concentration ratios between pregnant female and fetus pairs were calculated based on wet weight concentrations, and the mean of these concentration ratios then calculated for each contaminant. The placental concentration ratios are presented as both absolute ratios and $\log_{10}$ ratios to ease in comparison of transfer efficiency of each contaminant. Compounds with absolute ratios of 1 or $\log_{10}$ ratios of 0 indicate a 1:1 relationship, absolute ratios more than 1 or positive $\log_{10}$ ratios indicate higher levels in fetus than mother, and absolute ratios less than 1 or negative $\log_{10}$ ratio indicate a lower level in fetus than mother.

3. Results and discussion

3.1. Occurrence

Legacy lipophilic POPs were the dominating compound group in every quantified tissue of the adult whales (blubber, muscle and liver). The legacy POPs ΣPCBs and ΣDDTs together contributed 73% (blubber), 66% (muscle) and 67% (liver) to the total lipophilic POP load. Lipophilic POPs have previously been quantified in the blubber of common minke whales from the Barents Sea in 1992 (Kleivane and Skåre, 1998), 1998 (Hobbs et al., 2003), 2000 (Skåre et al., 2002) with the most recent study by Norway's veterinary institute in 2001 (Skåre et al., 2001), and we observed generally lower levels of lipophilic POPs than previous studies (Table 2; Supplementary Table S3). Blubber levels of ΣPCBs, ΣDDTs, ΣCHLs, ΣHCH and HCB in the present study of minke whales from 2019 were approximately half the levels in adult whales in 1992 (Kleivane and Skåre, 1998) and 1998 (Hobbs et al., 2003), for each sex respectively, and comparable to the levels in mixed sexes from 2000 (Skåre et al., 2002). Levels in females were similar to the levels of five female adult whales from 2001 (Skåre et al., 2001) (Table 2; Table S3). PCB levels in the muscle of common minke whales from the Barents Sea were last measured in 2000 (Skåre et al., 2002), and levels in the present study are comparable (Table 2; Table S3). This indicates that despite being regulated over four decades ago, levels of lipophilic POPs in these whales have only slowly decreased and/or stabilized over the last three decades. This is in accordance with the general decreasing trends in legacy POPs such as PCBs, DDTs, HCHs, HCB and Mirex that have been observed in a number of Arctic marine mammal species since 1980 (AMAP, 2016; Rigét et al., 2019, 2010; UNEP, 2021). Levels of lipophilic POPs in minke whale blubber in the present study were approximately ten times lower than previously measured in killer whales from Norway (Andvik et al., 2021, 2020), which is likely a reflection of feeding at different trophic levels.

The BFRs contributed the least to the total lipophilic POP load, with approximately 1.8% for all tissue types (Table 2). The dominating PBDE congeners were PBDE-47 followed by PBDE-99 in all tissue types. PBDEs were measured in common minke whales from the Barents Sea in both muscle in 2011 (Julshamn et al., 2012), and blubber in 1993 and 1999 (Rotander et al., 2012). Whilst muscle PBDE levels from the present study were approximately 10 fold higher than 2011, blubber levels in males were approximately three times lower than 1993 and 1999 (Table 2; Table S3). Samples from the previous studies are, however, of pooled individuals of either unknown age and/or sex, which confound the comparisons. Temporal trends of PBDE levels in other Arctic marine

Table 2
Levels of legacy and emerging lipophilic Persistent Organic Pollutants (POPs; ng/g) in blubber, muscle and liver of male (M) and female (F) adult common minke whales (*Balaenoptera acutorostrata*) harvested in the Barents Sea in August 2019. Mean ± standard deviation and range in parentheses. LOD = Limit of detection (values given in Supplementary Table S1).

			Legacy contaminants																Emerging contaminants					
			Organochlorines												Brominated flame retardants									
	Tissue	Lipid %	ΣPCBs[a] (ng/g)		ΣDDTs[b] (ng/g)		ΣCHLs[c] (ng/g)		HCB (ng/g)		ΣHCHs[d] (ng/g)		Mirex (ng/g)		ΣPBDEs[e] (ng/g)		HBCDD (ng/g)		PBT (ng/g)		HBB (ng/g)		ΣOCs+ BFRs[f] (ng/g)	
			ww	lw	ww	lw	ww	lw	ww	lw	ww	lw	ww	lw	ww	lw	ww	lw	ww	lw	ww	lw	ww	lw
Male (n=7)	Blubber	75 ± 4.9 (65-80)	1800 ± 1500 (60-4600)	2500 ± 2000 (620-6000)	1300 ± 1100 (336-3400)	1800 ± 1400 (460-4600)	870 ± 630 (250-2000)	1100 ± 840 (340-2700)	140 ± 78 (65-300)	180 ± 95 (100-380)	26 ± 14 (9.3-49)	34 ± 19 (14-66)	13 ± 11 (4.0-34)	18 ± 14 (5.5-46)	35 ± 19 (13-61)	47 ± 25 (18-83)	22 ± 10 (11-36)	29 ± 13 (17-46)	<LOD	<LOD	0.17 ± 0.12 (<LOD-0.35)	0.22 ± 0.15 (<LOD-0.44)	4300 ± 3300 (1200 - 10000)	5700 ± 4400 (1600 - 14000)
	Muscle	5.9 ± 4.4 (1.4- 13)	48 ± 41 (16-130)	1300 ± 1300 (230-3800)	35 ± 27 (12-89)	881 ± 820 (190-2600)	21± 14 (8.8-51)	540 ± 460 (130-1500)	7.2 ± 3.8 (2.5-13)	150 ± 71 (98-300)	1.2 ± 0.53 (0.33 - 1.7)	25 ± 13 (12-48)	0.30 ± 0.31 (0.11-0.98)	8.4 ± 9,4 (1.1-28)	1.3 ± 0.72 (0.47 - 2.2)	29 ± 20 (7.8-61)	0.30 ± 0.16 (0.085-0.48)	6.9 ± 4.7 (2.6-14)	<LOD	<LOD	0.076 ± 0.041 (0.048-0.16)	0.16 ± 1.7 (0.48 - 4.7)	120 ± 84 (42-280)	2900 ± 2600 (680-8200)
	Liver	6.0 ± 1.8 (3.8-8.8)	49 ± 42 (14-110)	880 ± 770 (200-2200)	33 ± 24 (12 – 72)	600 ± 460 (160 – 1500)	20 ± 13 (6.5-39)	360 ± 250 (100-790)	10 ± 4.5 (4.2 – 18)	160 ± 82 (110 -340)	1.8 ± 0.58 (0.84 - 2.6)	31 ± 13 (17-48)	0.4 ± 0.31 (0.095-0.97)	7.1 ± 6.4 (1.5-20)	1.0 ± 0.64 (0.52 - 2.2)	19 ± 11 (7.0-33)	0.16 ± 0.10 (<LOD-0.30)	3.1 ± 2.4 (<LOD-7.2)	<LOD	<LOD	0.0085 ± (<LOD-0.027)	0.14 ± 0.10 (<LOD-0.35)	110 ± 81 (40-230)	2100 ± 1500 (600-4700)
Female (n=10)	Blubber	75 ± 6.2 (63- 81)	760 ± 1000 (150-3000)	1000 ± 1500 (200-4300)	300 ± 400 (63-1400)	410 ± 590 (100-4300)	160 ± 140 (53-520)	220 ± 210 (76-750)	50 ± 32 (18-130)	68 ± 48 (29-190)	8.3 ± 6.5 (4.7-26)	11 ± 9.7 (5.8-38)	5.2 ± 2.7 (1.8-10)	7.0 ± 3.8 (2.9-15)	53 ± 130 (6.2-430)	76 ± 190 (7.7-620)	18 ± 24 (4.2-85)	75 ± 36 (5.2-620)	<LOD	<LOD	0.14 ± 0.14 (<LOD-0.45)	0.19 ± 0.19 (<LOD-0.65)	1400 ± 1700 (310-5500)	1900 ± 2400 (480-8000)
	Muscle	17.2± 7.5 (4.7- 30)	87 ± 140 (8.4-450)	700 ± 1500 (120-4900)	46 ± 58 (4.5-190)	320 ± 620 (50-2000)	24 ± 21 (3.2-64)	180 ± 200 (45-700)	12 ± 6.8 (1.6-23)	71 ± 48 (31-190)	1.5 ± 0.79 (0.49 - 2.9)	10 ± 7.9 (4.9-32)	0.61 ± 0.57 (<LOD-1.6)	3.9 ± 5.0 (<LOD-17)	8.2 ± 20 (0.62 - 66)	81 ± 220 (4.6-720)	2.1 ± 2.3 (0.17 - 7.9)	15 ± 25 (1.6-86)	0.0062 ± 0.0039 (<LOD-0.017)	0.063 ± 0.11 (<LOD-0.36)	0.054 ± 0.052 (<LOD-0.18)	0.38 ± 0.56 (<LOD-1.9)	180 ± 240 (19-800)	1400 ± 2600 (280-8800)
	Liver	6.6 ± 1.4 (4.8-9.6)	36 ± 73 (6.2-240)	560 ± 1200 (90-3900)	16 ± 28 (3.7-96)	250 ± 450 (52-1500)	7.7 ± 9.5 (1.9-33)	120 ± 150 (6.2-520)	4.9 ± 3.8 (2.1-13)	74 ± 53 (33-200)	1.1 ± 0.80 (0.40 -2.2)	17 ± 13 (33-42)	0.25 ± 0.24 (0.081-0.80)	3.7 ± 3.5 (1.2-13)	3.8 ± 10 (0.24 -33)	60 ± 170 (3.7-530)	0.37 ± 0.78 (0.0091-2.5)	5.4 ± 12 (0.14 -39)	0.013± 0.019 (<LOD-0.065)	0.22 ± 0.39 (<LOD-1.3)	0.0098 ± 0.015 (<LOD-0.052)	0.16 ± 0.24 (<LOD-0.84)	70 ± 120 (15-420)	1100 ± 2000 (230-6700)

mammal species are variable: ranging from significant increasing trends from 1980 to 2015, to decreases post 2005, to no trend throughout the whole time period (AMAP, 2016; Rigét et al., 2019; UNEP, 2021). Penta- and -octa BDEs were listed as POPs by the Stockholm Convention in 2009, and deca-BDEs in 2017, after years of regional regulations including inclusion on the Norwegian Priority List of chemicals that should be phased out since 1997.

The present study is the first to screen for four emerging BFRs in common minke whales from the Barents Sea. They composed 0.002% of the total lipophilic POP load, and none were found above the LOD in all individuals and tissues (Table 2). PBT was found in the muscle and liver of three female whales, and HBB was found in three liver, 15 muscle, and 10 blubber samples (Table 2). HBB blubber levels in whales from the present study were about half that measured in common minke whales from the St Lawrence Estuary in Canada, sampled 2015–2017 (Simond et al., 2019, Table 2; Table S3). The St Lawrence Estuary is known as a highly industrialized area with several point sources of pollution, including pulp and paper mills (Carignan et al., 2011). PBEB and DPTE were < LOD in all samples and individuals in the present study, and were similarly not found in stranded killer whales from Norway sampled 2015–2017 (Andvik et al., 2021), despite killer whales consuming higher trophic prey than minke whales (Jourdain et al., 2019). This indicates that PBEB and DPTE are not ubiquitous in the Norwegian Arctic.

We found nine PFAS in the present study, in muscle and liver (Supplementary Table S4), whereas only four (PFOS, PFNA, PFUdA and PFPeA) were previously found in muscle of minke whales sampled from the Barents Sea in 2011 by the Norwegian Food Safety Authority (Mattilsynet) (Julshamn et al., 2012). The present study is thus the first to report the presence of PFOA, PFDA, PFDoDA, PFTrDA, PFHxS and PFOSA in minke whales from the Barents Sea. The ΣPFAS levels were dominated by PFOS in all tissue types, whilst PFOA was found in only 25% of the samples (primarily liver) (Table S4). PFOS levels in males from the present study were double the levels from 2011, whereas PFOS levels in females were similar (Julshamn et al., 2012; Table S3, Table S4). PFUnDA and PFNA levels in the present study of minke whales from 2019 were similar to the 2011 whales (Julshamn et al., 2012). This differs to ringed seal (*Phoca hispida*) and polar bear (*Ursus maritimus*) from Greenland, in which PFOS and PFUnDA levels increased until the mid-2000s, followed by a decrease to reach the 1980-levels by 2015 (Rigét et al., 2019, 2013). Our data resemble pilot whales from the North Atlantic, that saw an increasing PFOS trend over the whole time series to 2013, despite deceasing PFOSA levels after a peak in 2006 (Dassuncao et al., 2017). This indicates that temporal trends of PFAS are dependent on species and/or study area, as well as PFAS type. PFOS was internationally regulated through the Stockholm Convention in 2009 and PFUnDA is still not regulated, even though it is on the Norwegian Priority list for chemicals that should be phased out.

Levels of Hg, Cd, Pb, As and Se in common minke whale muscle from the present study were comparable to those from individuals sampled in 2011 (Maage et al., 2017), and Hg levels similar to minke whale muscle from 1998 (Frydenlund and Øvrevoll, 2002) and 2002 (Kleivane and Børsum, 2003) (Supplementary Table S5; Table S3). Hg is globally regulated via the Minamata Convention on Mercury which was adopted in 2013 and entered into force in 2017 (UNEP, 2017). Whilst polar bear teeth from Svalbard showed a decrease in Hg concentrations between 1964 and 2003 (Aubail et al., 2012), other Arctic marine mammal populations have increasing Hg trends, such as Greenland beluga whale (*Delphinapterus leucas;* Desforges et al., 2022) and Greenland ringed seals (Aubail et al., 2010). A review of temporal trends for Arctic marine mammal species found a general pattern of significantly increasing trends of Hg in liver over the last 20 years (Morris et al., 2022). These increases may be due to remobilization of Hg from permafrost and other Arctic soils by climate warming and increased outputs of terrestrial Hg to the coastal regions, as well as a shift in diet to higher trophic prey (Desforges et al., 2022; Dietz et al., 2009). The local effects of climate change on temporal trends have also been recognized for other POPs (Vorkamp et al., 2022).

3.2. Effects of tissue, sex and diet on measured contaminant levels

We investigated the effect of tissue type, sex, and diet (using $\delta^{15}N$ and $\delta^{13}C$ levels in muscle as proxies) on the levels of lipophilic POPs, ΣPFAS and Hg.

3.2.1. Model summaries

For the lipophilic POPs, lipid content explained 66.3% of the total variation in levels (RDA, $F = 96.5$, $p < 0.001$), and lipid content was highest in blubber, followed by muscle and then liver (Table 2). When conditioning lipid as a co-variable in the redundancy analysis, the best model explained 16.4% of the total variation, and included the following explanatory variables, in order of decreasing explanatory power: $\delta^{13}C$ in muscle (55%, F = 25, p = 0.001), Sex (39%, F = 18, p = 0.001), Tissue Type (15%, F = 3, p = 0.015) (Fig. 2). For ΣPFAS, the explanatory variables in the best model were: Tissue, Sex and $\delta^{15}N$ levels in muscle (see Supplementary Table S6 for model summary). For Hg, the best model for explaining variation in levels included just Tissue (see Supplementary Table S7 for model summary).

3.2.2. Tissue distribution

Even after controlling for the effect of lipid content, we found that tissue type significantly explained the levels of lipophilic POPs in the RDA, with similar levels in blubber and muscle, and lowest levels in liver (Fig. 2). Only legacy POPs were included for statistical analysis due to the high number of emerging BFR samples < LOD, and thus only simple comparisons can be made between tissue partitioning of legacy and emerging contaminants in the present study. Levels were generally highest in blubber and lowest in liver on both a wet weight and lipid adjusted basis for almost all the legacy lipophilic POPs (Table 2). Whilst HBB levels in male whales followed the same distribution as the legacy POPs of highest in blubber both before and after lipid adjustment, HBB lw levels in female whales were twice as high in muscle than blubber, and PBT was < LOD in all blubber samples. PBT levels were three times higher in liver than muscle (ww and lw), whereas levels of legacy POPs, and of HBB, were lowest in liver. These conflicting results can be as a result of high variations in lipid content within and between tissues (Table 2) and a small sample size, so conclusions should not be prematurely drawn about the tissue partitioning of these emerging contaminants. Investigation of the compounds' physical-chemical properties would also be necessary for any further analysis of lipid partitioning.

ΣPFAS levels ww were 14 times higher in liver than muscle (Table S4; Fig. 3A). PFAS bind to proteins (Jones et al., 2003), and our results are in accordance to previous studies indicating a preferential partitioning in liver rather than muscle in marine mammals (Houde et al., 2006b; López-Berenguer et al., 2020). For Hg, levels were highest in liver, followed by muscle and then skin (Table S5; Fig. 3B), similar to previous studies in marine mammals (André et al., 1990; Endo et al., 2006; Frodello et al., 2000).

3.2.3. Sex

We found higher levels of lipophilic POPs in males than females, illustrated by the clear separation of the sex centroids on the PCA (Fig. 2). Female marine mammals are known to transfer contaminants to their offspring through both the placenta and the lipid-rich milk (Haraguchi et al., 2009; Pedro et al., 2017), and previous studies on common minke whales in the Barents Sea found two to three times higher levels of PCBs in the blubber of males than females (Hobbs et al., 2003; Kleivane and Skåre, 1998). Other studies on marine mammals have also shown an increase in POP levels in males with age, but not with females, due to maternal transfer (Binnington and Wania, 2014; Krahn et al., 2009).

We found twice as high PFAS levels in males than females, but no effect of sex on Hg levels. This is most likely explained by the high placental transfer of PFAS that we found in the present study, and low placental transfer of Hg (see section *Placental transfer of PFAS and metals*).

3.2.4. Diet

Higher $\delta^{15}N$ values are indicative of feeding at a higher trophic level (DeNiro and Epstein, 1981), whilst higher $\delta^{13}C$ values indicate a more benthic and/or inshore diet (Hobson et al., 1994; Hobson and Clark, 1992). Minke whales eat opportunistically across multiple trophic levels, and from both pelagic and benthopelagic sources, and have a wide ecological niche, especially in the $\delta^{15}N$ direction (Haug et al., 2002; MacKenzie et al., 2022). Contaminant levels in common minke whales from the Barents Sea have previously been shown to vary with regional differences in availability of prey along differing migration routes (Hobbs et al., 2003). In the present study, we found higher $\delta^{15}N$ values in muscle were correlated with higher PFAS levels, indicating that whales feeding at a higher trophic level accumulated more PFAS.

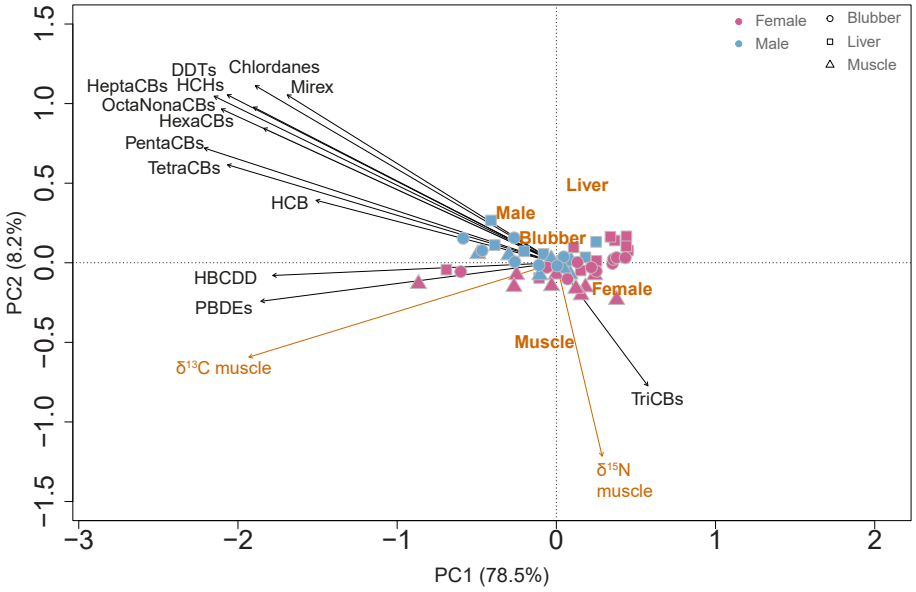

Fig. 2. Principal component analysis (PCA) biplot based on lipophilic POP levels in blubber, muscle and liver of adult minke whales (*Balaenoptera acutorostrata*) harvested from the Barents Sea in August 2019, with lipid as a co-variable. Contaminant loadings are represented as black arrows, and passive explanatory variables are added as orange arrows ($\delta^{13}C$ muscle, $\delta^{15}N$ muscle) or as text representing the centroids of each categorical variable (Male and Female for Sex, Blubber, Muscle and Liver for Tissue). The percentage of the total variation explained by PC1 and PC2 are given in brackets on each axis. TriCBs are the sum of PCB-28 and -31; TetraCBs the sum of PCB-47, -52, −66 and −74; PentaCBs the sum of PCB-87, -99, −101, −105, −110, −114 and −118; HexaCBs the sum of PCB-128, -137, −138, −141, −149, −151, −153, −156 and −157; Hepta-CBs the sum of PCB-170, -180, −183, −187 and −189; and OctaNonaCBs the sum of PCB-194, -196, −199, −206 and −209. DDTs is the sum of *p,p'*-DDE, *o,p'*-DDD, *p,p'*-DDD, *o,p'*-DDT and *p,p'*-DDT. Chlordanes is the sum of heotachlor, oxychlordane, *trans*-chlordane, *cis*-chlordane, *trans*-nonachlor and *cis*-nonachlor. HCHs is the sum of α-HCH, β-HCH and γ-HCH. PBDEs is the sum of BDE-47, -99, −100, −153 and −154.

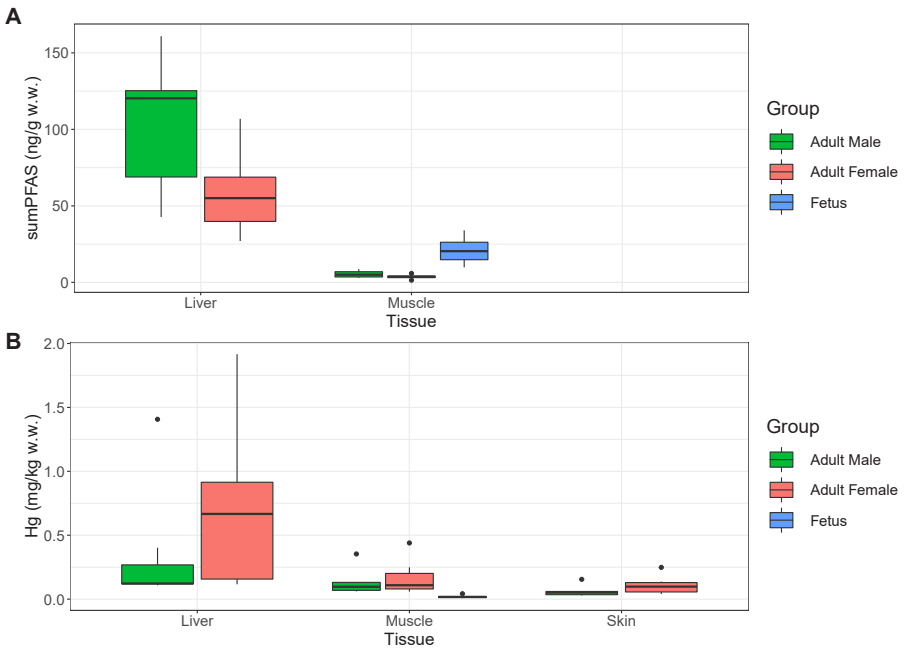

Fig. 3. A) ΣPFAS levels (ng/g ww) in liver and muscle and B) Total mercury (Hg) levels (mg/kg ww) in liver, muscle and skin of common minke whales (*Balaenoptera acutorostrata*) harvested in the Barents Sea in August 2019. Adult males are the green boxes (n =7), adult females red (n =10) and fetuses blue (n =9). Note the different units and scales on the y axis.

However, we unexpectedly found no correlation between $\delta^{15}N$ values and levels of lipophilic POPs or Hg, despite knowing that these contaminants biomagnify in marine food webs (AMAP, 2011, 2005; Borgå et al., 2001; Fujii et al., 2007; Haukås et al., 2007; Ruus et al., 2015). Rather, we found higher levels of $\delta^{13}C$ in muscle, which indicate feeding on benthic and/or coastal prey, were associated with higher levels of lipophilic POPs in the whales, but not higher PFAS or Hg levels. These contrasting results for the different contaminants can reflect contaminant sources in prey, *e.g.* higher levels of lipophilic POPs in benthic, coastal prey with higher $\delta^{13}C$ values, perhaps due to bioavailability of contaminants in the sediment, but no difference in PFAS or Hg levels. The range of $\delta^{13}C$ values in the whales can reflect different proportions of availability of benthic prey for individuals, and because there was no clear threshold or marked difference between our individuals, we suggest a generalist feeding strategy rather than of specialists seeking out specific prey, in accordance with previous assumptions of the species (*e.g.* Haug et al., 2002). Future studies should focus on quantification of compounds of prey items to better understand the effect of diet.

3.3. Placental transfer of PFAS and metals

This study is the first to analyze levels of PFAS and metals in common minke whale mother-fetus pairs, and to calculate concentration ratios for this species as an indication of placental transfer.

Mean absolute concentration ratios for PFAS between fetus and mother pairs ranged from 0.55 to 8.0, with all PFAS except PFOSA present in higher levels in the fetus than the mother (Fig. 4A; Fig. 4B). Ratios for PFOA and PFHxS were excluded from this analysis due to quantification < LOD in the pregnant females, but they were quantified > LOD in 44% and 33% of the fetus samples, respectively, which indicate a transfer from mother to fetus.

The highest mean absolute concentration ratios were in PFNA (8.0), PFOS (7.5) and PFDA (7.4), and the concentration ratios of these same PFAS were also highest in the livers of a killer whale mother-fetus pair from Greenland (Gebbink et al., 2016). This indicates a high placental transfer of these PFAS in both species, and in both muscle and liver. Increased placental transfer appears correlated with higher protein-water partition ratios ($\log K_{PW}$: from Bischel et al., 2011) (Fig. 4A; Fig. 4B), which is likely due to the partitioning of PFAS to proteins and thus being transferred to the fetus through the placenta. $\log K_{PW}$ values were not available in the literature for PFOSA and PFTrDA, however it has been demonstrated that PFOSA has a lower protein binding affinity than PFOS (Weiss et al., 2009), which can explain the low concentration ratio.

Higher levels of PFAS in offspring than adults is found in a range of marine mammals including melon-headed whales (*Peponocephala electra;* Hart et al., 2008), pinnipeds (Ahrens et al., 2009; Grønnestad et al., 2017; Ishibashi et al., 2008), killer whales (Andvik et al., 2021), and bottlenose dolphins (*Tursiops truncatus;* Houde, Balmer, et al., 2006). Although PFAS is found in the milk of marine mammals, such as bottlenose dolphins (Houde et al., 2006a) and hooded seal (*Cystophora cristata;* Grønnestad et al., 2017), the PFAS transfer in hooded seal mother-pup pairs was determined to be predominantly placental rather than from lactation (Grønnestad et al., 2017). This was determined from the low PFAS levels in milk and higher plasma PFAS concentrations in pup than mother, and is in line with the protein-associated rather than lipophilic nature of PFAS (Jones et al., 2003). In humans, the breastfeeding transfer efficiencies of PFAS were 1–2 times lower than transplacental transfer efficiencies, and yet analysis of total body burden of offspring found the postnatal exposure to PFAS via breastfeeding were approximately 10 times higher than prenatal exposure in utero (Zheng et al., 2022). The absolute concentration ratios of all PFAS in the present study (0.55–8.0) are higher than those measured for contaminants known to have greater affinity to lipids, such as chlorinated paraffins in a common minke whale mother-fetus pair from Denmark (0.22–0.29; Yuan et al., 2021), and ΣPCBs in two killer whale mother-fetus pairs (0.25; Pedro et al., 2017). This indicates that PFAS could be the dominating POP placentally transferred in cetaceans, and should be acknowledged when considering movement of PFAS in the ecosystem.

ΣPFAS levels in the male whales were almost double the levels in the females and the levels in fetuses three times that in males (Table S4; Fig. 3A), further indicating that female transfer of PFAS to their offspring is an important elimination route.

Mean absolute concentration ratios between fetus and mother pairs for metals ranged from 0.039 (As) to 5.8 (Ni) (Fig. 4C). All metals were present in the fetus in higher levels than in the mother, except for Fe, As,

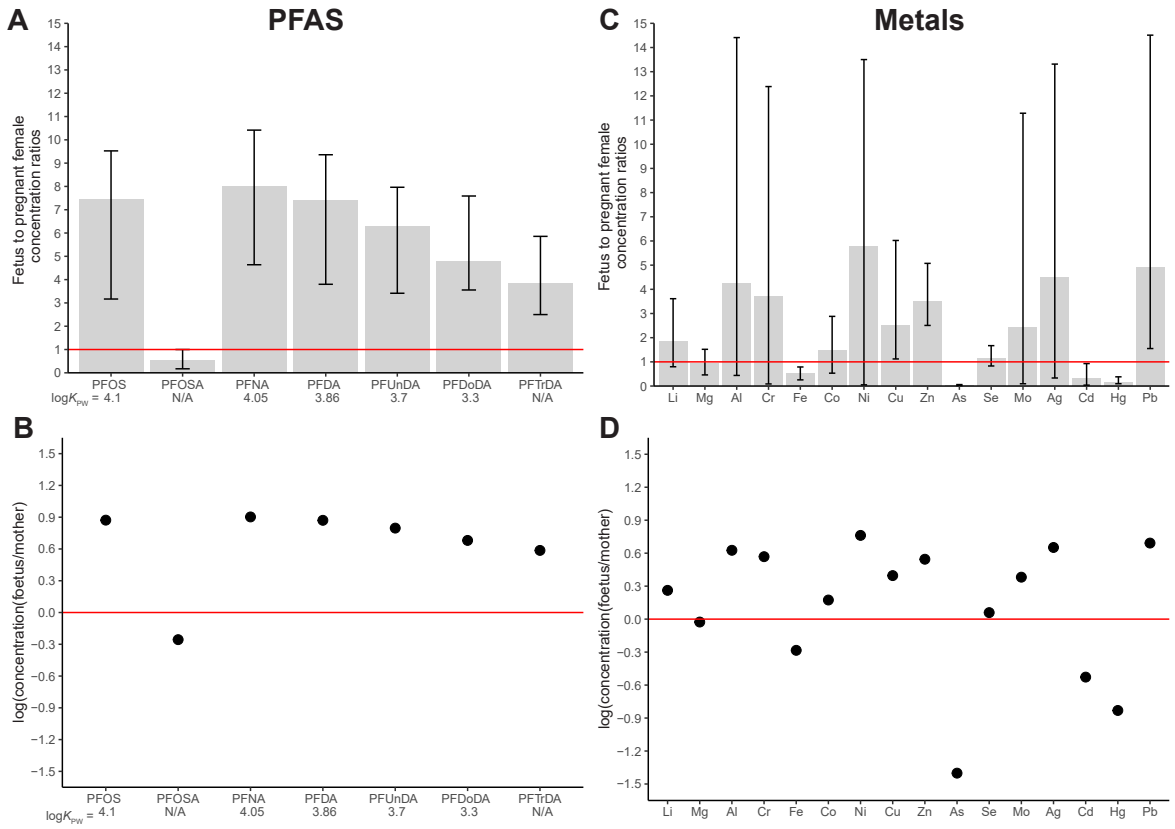

Fig. 4. Concentration ratios between the muscle in common minke whale (*Balaenoptera acutorostrata*) fetus to pregnant female harvested in the Barents Sea in August 2019 (n = 9 pairs). A) Absolute concentration ratios of perfluoroalkyl substances (PFAS) mean with range (minimum and maximum); B) Log_{10} concentration ratios of PFAS; C) Absolute concentration ratios of metals mean with range (minimum and maximum) and D) Log_{10}concentration ratios of metals. An absolute ratio = 1 and a log_{10} ratio = 0 indicates equilibrium in transfer of contaminants from mother to fetus and is indicated by a red line. The PFASs are ordered by carbon chain length (C8 – C13) and the logK_{PW}, from Bischel et al. (2011), are indicated where available (N/A = Not available). The metals are ordered by atomic weight. PFOS = perfluorooctane sulfonate; PFOSA = perfluorooctane sulfonamide; PFNA = perfluorononanoic acid; PFDA = perfluorodecanoic acid; PFUnDA = perfluoroundecanoic acid; PFDoDA = perfluorododecanoic acid; PFTrDA = perfluorotridecanoic acid.

Cd and Hg which were present in higher levels in the mother than the fetus, and Mg which was approximately equal between mother/fetus (Fig. 4C; D). Two mother-calf killer whales from Japan had significantly higher Cu levels in the muscle of calves than mature whales, and Fe and Cd levels lower in calves than mature whales (Endo et al., 2006), something that is mirrored in our results of direct mother-fetus relationship. Placental metallothionein (MT) acts as a barrier for Cd transport over the placenta, thereby reducing transfer of Cd to the fetus (Fukase et al., 2014).

The low placental concentration ratio of Hg from mother to fetus in the present study (0.14) is supported by studies in other marine mammals indicating little maternal transfer to offspring, including killer whales (Andvik et al., 2021; Endo et al., 2006), blue whales (Trumble et al., 2013) and pinnipeds (Hitchcock et al., 2017). The reason for the low placental transfer of Hg observed is likely due to the efficiency of the placental barrier, as shown in rodents (Cambier et al., 2018).

3.4. Levels below thresholds for risk of toxic effects for the whales

ΣPCB levels in the blubber of all the adult individuals were at least four times lower than the most widely used proposed threshold for toxicological effects in marine mammals (9 μg/g lw), for which adverse physiological effects can be expected (Kannan et al., 2000; adapted by Jepson et al., 2016). ΣPBDE levels above 1.5 μg/g lw have been associated with thyroid hormone disruption in grey seals pups (*Halichoerus grypus*; Hall et al., 2003), and all whales in the present study were at least 20 times below this level. Hg levels in the liver were approximately 25 times lower than the threshold for a low risk of health effects in marine mammals (16 μg/g ww) (Dietz et al., 2019) based on toxic hepatitis, uremia and renal failure observed in harp seals (*Pagophilus groenlandicus*) (Ronald et al., 1977). Thus, these whales are not at risk of PCB, PBDE or Hg-mediated health effects.

As thresholds for the possible health effects from Hg are usually based on liver levels, we assessed the relationship between Hg levels in liver and skin, as such relations are useful when assessing risk for negative effects based on common minke whales biopsy sampling, when only skin is available. The Hg levels (ww) in skin and liver correlated strongly (adj. $R^2 = 0.87$), described by the following formula: log_{10}Hg_Liver = 1.3049 + 1.5651 (log_{10}Hg_Skin) (Supplementary Fig. S1).

4. Conclusions

Emerging POPs are being increasingly used in response to global and regional bans of legacy POPs, and yet more data is needed on their presence and behavior in Arctic biota. In the present study, we recorded the first presence of emerging BFRs HBB and PBT, and of PFOA, PFDA, PFDoDA, PFTrDA, PFHxS and PFOSA in common minke whales from the Barents Sea. Whilst levels of legacy POPs appear to have decreased since 1992, in accordance with many other marine mammal populations, levels of PFOS, PFUnDA and PFNA levels were higher or comparable to levels in 2011. Whilst legacy contaminants were higher in the blubber of the whales on both a wet weight and lipid adjusted basis, there was no clear pattern for the emerging BFRs. The present study presents the first concentration ratios of PFAS and metals between mother and fetus,

indicating high placental transfer of nearly all PFAS, and low placental transfer of Hg. Contaminant levels were below thresholds for health effects for the whales. Our results are relevant for continued biomonitoring of the Arctic marine ecosystem, providing valuable data to aid regional and global monitoring bodies.

Credit author statement

Clare Andvik: Conceptualization, Formal analysis, Investigation, Methodology, Visualization, Writing – original draft. Tore Haug: Resources, Writing – review & editing. Jan Ludvig Lyche: Resources, Writing – review & editing. Katrine Borgå: Conceptualization, Funding acquisition, Project administration, Resources, Supervision, Validation, Writing – review & editing.

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.

Data availability

All data is available in the main text or in the Supplementary Data excel sheet available online.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2023.121001.

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