Science of the Total Environment 718 (2020) 137327

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

First assessment of pollutant exposure in two balaenopterid whale populations sampled in the Svalbard Archipelago, Norway

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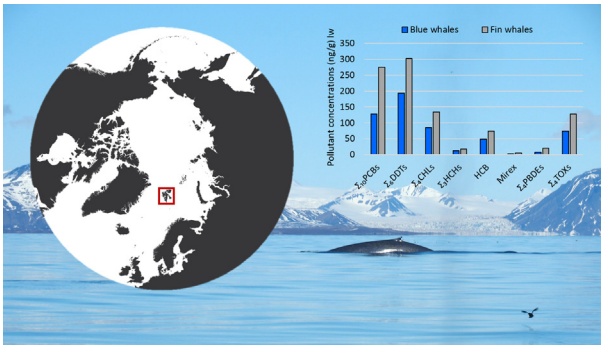
HIGHLIGHTS

- Biopsies were taken from 18 blue whales and 12 fin whales from Svalbard.
- Pollutant levels were 1.6–3 times higher in fin whales than in blue whales.
- Fin whales fed at a higher trophic level and at higher latitudes than blue whales.
- Pollutant levels were twice as high in males as females in both species.
- Pollution levels in whales vary substantially regionally.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 14 November 2019
Received in revised form 12 February 2020
Accepted 13 February 2020
Available online 14 February 2020

Editor: Yolanda Picó

Keywords:
Arctic
Svalbard
Blue whale
Fin whale
Contaminant
Stable isotope

ABSTRACT

Pollutant concentrations are poorly known for the largest animals on Earth, blue whales *Balaenoptera musculus* and fin whales *Balaenoptera physalus*. In this study, concentrations of persistent organic pollutants (POPs) were determined in blubber biopsies and stable isotope values for nitrogen (δ^{15}N) and carbon (δ^{13}C) were measured using skin biopsies for 18 blue whales and 12 fin whales sampled in waters surrounding the Svalbard Archipelago, Norway. The samples were collected in summer during the period 2014–2018. POPs were dominated by DDTs, PCBs and toxaphenes, with median concentrations in blue/fin whales being 208/341, 127/275 and 133/233 ng/g lipid weight, respectively. Linear models indicated that pollutant concentrations were 1.6–3 times higher in fin whales than in blue whales, which is likely related to the higher trophic positions of fin whales, as indicated by their higher δ^{15}N. Lower δ^{13}C in fin whales suggests that they feed at higher latitudes than blue whales; these values were not correlated with pollutant concentrations. Pollutant levels were approximately twice as high in males compared to females (intraspecifically), which indicates that females of these species offload pollutants to their offspring during gestation and lactation, similar to many other mammalian species. Pollutant concentrations in balaenopterid whales from Svalbard waters were generally much lower than in conspecific whales from the Mediterranean Sea or the Gulf of California, but higher than those in conspecifics from the Antarctic Peninsula.

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https://doi.org/10.1016/j.scitotenv.2020.137327

1. Introduction

The Arctic is warming twice as fast as anywhere else on Earth, and recent reports have documented an increasing frequency of southern species in the Arctic (Kortsch et al., 2012; Laidre et al., 2015). For instance, there has been an expansion of the distributional ranges of many invertebrate and fish species into the Arctic (Doney et al., 2012; Fossheim et al., 2015; Kortsch et al., 2012; Vihtakari et al., 2018), and the new communities of boreal species have attracted new predators such as balaenopterid whales. Along the shelf breaks west and north of Svalbard Archipelago, the sighting rate of blue whales (*Balaenoptera musculus*) and fin whales (*Balaenoptera physalus*) has increased in recent years (Storrie et al., 2018; Vacquié-Garcia et al., 2017). The increasing presence of these large balaenopterid whales coincides with an increase in the intrusion of Atlantic Water with associated Atlantic prey into these areas (Fossheim et al., 2015; Pavlov et al., 2013). Blue whales feed almost exclusively on krill, whereas fin whales have a more varied diet that includes krill, but also amphipods, copepods, shrimps, small fish and squid (Gavrilchuk et al., 2014; Nemoto, 1970). In addition to inducing distributional shifts, global warming is also causing remobilization of a large number of POPs from repositories in the Arctic, due to rising temperatures (Ma et al., 2011).

There is however a general lack of knowledge regarding pollutant levels in balaenopterid whales in High Arctic areas, although they are likely exposed to a wide variety of chemicals. Despite its remote location, the Arctic is the recipient of pollution from elsewhere on the planet via both long-range atmospheric and ocean current transport (Wania and Mackay, 1993). Persistent organic pollutants (POPs) biomagnify in marine food webs and thus apical feeding predators are exposed to high levels of POPs (Borgå et al., 2001; Hop et al., 2002; Kelly et al., 2007), which may cause adverse health effects (Dietz et al., 2019; Fossi and Panti, 2018). Due to concerns regarding the impacts of POPs on wildlife and human health, the use and production of these compounds is now either regulated or banned by the Stockholm Convention (http://www.pops.int, see Web references list for complete URL). Twelve POPs including polychlorinated biphenyls (PCBs), organochlorine pesticides (OCPs) such as chlordanes (CHLs), dichlorodiphenyltrichloroethane (DDT), hexachlorobenzene (HCB), mirex and toxaphene were initially listed as being environmentally dangerous substances in 2001. The hexacyclochlorohexanes (HCHs), α-HCH and β-HCH, and the polybrominated diphenyl ethers (PBDEs), tetra-, penta-, hexa- and hepta-BDEs, were added to the Convention's list in 2009, while deca-BDE was added in 2017. These so-called legacy POPs are, however, still present in the Arctic environment and in its biota (Rigét et al., 2019).

Balaenopterid whales do not feed at an apex position in the food web, but high levels of pollutants have been reported for some populations feeding close to industrial areas (Fossi et al., 2016, 2014; Metcalfe et al., 2004). For example, the average PCB concentration in fin whale blubber from the Mediterranean is ~13,000 ng/g lipid weight (Fossi et al., 2016), whereas slightly lower concentrations have been reported for fin whales from the Gulf of California (Fossi et al., 2014). High concentrations of pollutants are of concern for blue and fin whales, which are currently classified as *Endangered* and *Vulnerable*, respectively on the International Union for Conservation of Nature's (IUCN) Red List of Threatened Species (Cooke, 2018a, 2018b). Particularly high concentrations of POPs are generally found in male whales (Fossi et al., 2014; Metcalfe et al., 2004), since females transfer pollutants to their foetus during gestation and to their calf during lactation (Aguilar et al., 1999; Aguilar and Borrell, 1994).

The few studies reporting POP levels in fin and blue whales in different oceans of the world, suggest that there is considerable variation regionally (Fossi et al., 2016; Metcalfe et al., 2004; Muñoz-Arnanz et al., 2019; Pinzone et al., 2015). This pattern is thought to reflect pollutant concentrations in the prey of these whales regionally, because the ability of cetaceans to biotransform pollutants is limited (Boon et al., 1997; Hecker et al., 2019). Traditionally, blue and fin whales have been assumed to feed in highly productive areas at high latitudes and to migrate to breed at lower, warmer latitudes, where feeding is reduced or absent (Geijer et al., 2016). However, the consistency of long-distance migrations among baleen whale populations has been questioned recently (Geijer et al., 2016). Some studies suggest that winter feeding at lower latitudes is common among North Atlantic blue and fin whales (Silva et al., 2019). Migration patterns of these species are poorly known for the Northeast Atlantic (Moore et al., 2019). Knowledge of pollutant exposure of blue and fin whale populations may give insights into their feeding areas, as pollutant levels in the North Atlantic show high latitudinal variation, which is related to the proximity to emission sources, patterns and routes of long-range transport and ocean biogeochemistry (Breivik et al., 2002; Lohmann et al., 2007; Wagner et al., 2019). The aim of this study was to investigate levels of POPs in relation to feeding habits in a Northeast Atlantic population of blue and fin whales that reside around Svalbard during summer.

2. Material and methods

2.1. Fieldwork

Fieldwork was conducted from the end of May to September in the period 2014–2018 off the west coast of the Svalbard Archipelago (Fig. 1). Eighteen adult blue whales and 12 adult fin whales were biopsied using a custom-made biopsy dart (10 cm long, 8 mm in diameter) shot from a crossbow, targeting the upper back in the middle of the animal. Biological information for the biopsied whales is available in the supporting information (Table S1). The dart was attached to a string secured to the crossbow that enabled rapid recovery of the sample. Skin and blubber were separated, the upper part of the skin (a few millimetres) was divided vertically into two parts, one part used for stable isotope analyses and the other was used for genetic analyses. The blubber closest to skin-blubber interface (1.5 cm from the 3–4 cm thick layer) was used for POP analyses. All samples were packed in aluminium foil or pre-cleaned glass vials and were frozen at −20 °C until analyses. All sampling procedures were approved by the Norwegian Animal Care Authority and the Governor of Svalbard.

2.2. Stable isotope analyses and molecular sexing

From the skin samples, nitrogen ($\delta^{15}N$) and carbon ($\delta^{13}C$) stable isotope ratios were analysed and used as proxies of trophic level and foraging habitat, respectively (Hobson, 1999). Because $\delta^{15}N$ values increase with increasing trophic level, they reflect trophic position of individual whales (Hobson et al., 1996). In contrast, $\delta^{13}C$ varies marginally as a function of trophic level but it indicates the sources of primary production in the food consumed by the whales, for example pelagic vs benthic, inshore vs offshore (Hobson, 1999; Hobson et al., 1996).

The analyses were carried out following methods described in Marcoux et al. (2012). Briefly, skin samples were lyophilized at −48 °C at a pressure of 133×10^3 mbar for 48 h, homogenized into small pieces using a scalpel, lipid extracted using a 2:1 chloroform:methanol mixture to remove lipids (which can bias $\delta^{13}C$ measurements), and then the samples (400–600 μg) were placed into tin cups. Values of $\delta^{13}C$ and $\delta^{15}N$ were determined on a Delta V Advantage Thermoscientific Continuous Flow Mass Spectrometer (Thermo Scientific, Bremen, Germany) coupled to a 4010 Elemental Combustion System (Costech Instruments, Valencia, CA, USA). Instrument accuracy met laboratory quality assurance criteria, as values were within 0.2‰ for NIST 8547, NIST 8573 and NIST 8574 for $\delta^{15}N$, and 0.1‰ for NIST 8573, 8542, and 8544 for $\delta^{13}C$. Precision was assessed with our laboratory standards (NIST 1577c, tilapia muscle, USGS 40 and Urea ($n = 104$ for each)), run every 12 samples, and was 0.2‰. Sample reproducibility of $\delta^{13}C$ and $\delta^{15}N$ was measured in triplicate every 10 samples and was ±0.1‰.

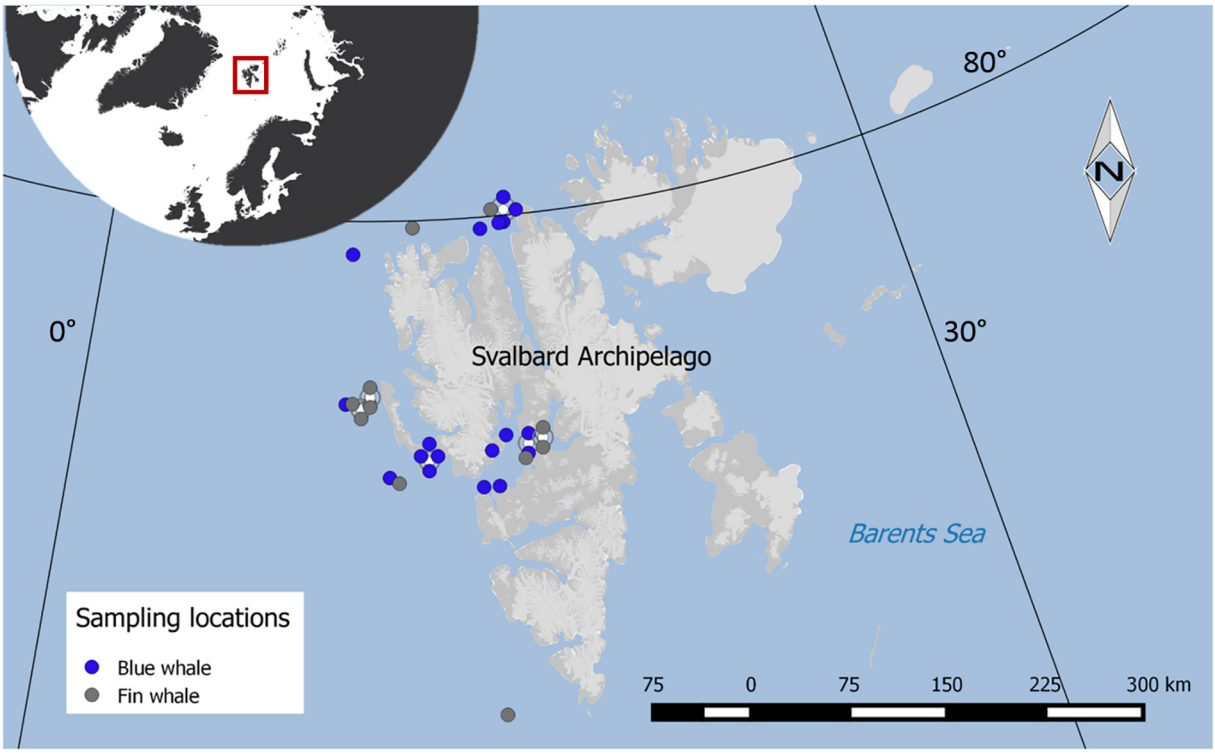

Fig. 1. Sampling locations of blue whales (*B. musculus*) and fin whales (*B. physalus*) in the coastal waters of Svalbard, Norway. Whales were biopsied between 2014 and 2018.

Sex of the whales was identified by molecular sexing using the methods described in Berube and Palsbøll (1996). Briefly, three oligonucleotides primers ZFYX0582F (5′-ATAE GTCTGCAGACTCITCrA-3′), ZFYX1204K and ZFYX0938R (5′-CITACACCTAAATGGAAGATCC-3′) were used for PCR amplification of the ZFX/ZFY sequence specific to mysticetes.

2.3. Pollutant analyses

Methods to determine lipid-normalized concentrations of organochlorine contaminants from blubber biopsies have been previously validated for balaenopterid whales (Gauthier et al., 1997a). In this study, the blubber biopsies (weight: 0.105–0.478 g) were analysed for OCPs, HCB, α-, β-, γ-HCH, oxy-, trans-, cis-CHL, trans-, cis-nonachlor, mirex; *o,p′*-DDT, *p,p′*-DDT, *o,p′*-DDD, *p,p′*-DDD, *o,p′*-DDE, *p,p′*-DDE, toxaphenes (#26, #32, #38, #40, #42, #50, #58, #62, #69), 2) PCBs (CB-28, -52, -99, -101, -105, -118, -138, -153, -180, -183, -187 and -194) and 3) PBDEs (BDE-28, -47, -99, -100, -138, -153, -183, -209). Extraction was performed using the methods described in Scotter et al. (2019). Briefly, blubber was homogenized with dried sodium sulphate and put into a freezer overnight. 20 μl of internal standard (ISTD), containing approximately 4 ng ^{13}C labelled PCBs and 4–50 ng PBDEs and OCPs (Cambridge Isotopes Laboratories Inc., Cambridge, UK; Chem Service Inc., West Chester, PA, USA), was added and contaminants were extracted with 3:1 cyclohexane:acetone. The filtered extract was evaporated, and the weight of the extractable organic material (lipids) was determined gravimetrically. Clean-up of the samples was done using Supelclean EZ-POP NP cartridges (Sigma- Aldrich Co. LLC, St. Louis, MO, USA). The evaporated eluate was cleaned-up by using solid phase extraction (SPE) cartridges packed with 1 g of Florisil (heated at 450 °C for 8 h). The eluate was evaporated, and the solvent was changed to isooctane, concentrated and spiked with a recovery standard (^{13}C CB-159).

PCBs and OCPs were measured as described in Scotter et al. (2019). Briefly, they were determined using gas chromatography coupled with mass spectrometry (GC–MS quadrupole instrument from Agilent, Santa Clara, CA, USA, GC 7890, MSD 5975C). PCBs and DDT were separated using a DB5-MS column (30 m × 250 μm, 0.25 μm df), whereas the remaining pesticides were separated using an Agilent Ultra2 column (25 m × 200 μm, 0.11 μm df connected to a 5 m × 0.32 mm ID retention gap column). Analyses of PBDEs in batch 1 and 2 were performed on a Waters Quattro Micro GC–MS (Waters Corp., MA, USA) equipped with a Restek 1614 column 15 m × 0.25 mm ID, 0.25 μm connected to a 5 m × 0.32 mm ID retention gap column. PBDEs in the third batch were measured on a Q-exactive GC (Thermo Fisher Scientific Inc., Waltham, MA, USA) in electron ionization mode and targeted SIM mode (window of 10 *m/z* units and a resolution of 30,000 FMHW @ 200 *m/z*). Sample aliquots of 2 μl were injected using a PTV (large volume injection mode) with a 40 °C start temperature, 0.1 min injection phase, then heated (at a rate of 2.5 °C/s) up to 330 °C with a hold time of 5 min. The carrier gas was helium, introduced at a constant flow of 1.5 ml/min with an oven temperature program: 80 °C for 2 min, then 30 °C/min increase to 340 °C, held for 3 min. Analyses were conducted on a Restek 1614 column 15 m × 0.25 mm ID, using a 0,1 μm stationary phase.

One method blank and standard reference material sample (Standard Reference Material [SRM] 1945 Organics in Whale Blubber, supplied by NIST, Gaithersburg, MD, USA) was run for every 10th sample; at least three blanks and one SRM were run per batch. Recoveries of the recovery standard (^{13}C CB-159) from the sample clean-up ranged from 28% to 99%. PCB, OCP and PBDE concentrations were within ± 20% of the certified values. The limit of detection (LOD) was calculated as the average peak height of the laboratory blank plus 3 × standard deviation (SD), while the limit of quantification (LOQ) was the laboratory blank average plus 10 × SD of the background ratio. Only pollutants that were above the LOD in >70% of the individuals were considered for statistical analyses. HCB and mirex were analysed as single compounds, whereas other compounds that included several congeners or metabolites were grouped as sums (Σ). LODs, LOQs and sample sizes (above the LOD limit described above) are given in Table S2. Compounds with values below LOQ were replaced by the LOQ value and values below limit of detection (LOD) were replaced by ½ LOD. If a compound was below LOD for more than one sample, random numbers ("*runif*"

Table 1
Model selection table for models explaining concentrations of POPs in blue and fin whales according to ΔAICc and AICc-weights. Predictors include $\delta^{15}N$ and $\delta^{13}C$ values in skin, species, and sex. Sign "+" means that a categorical variables is included in the model. Numbers for intercept, $\delta^{15}N$ and $\delta^{13}C$ are parameter estimates. Degrees of freedom (df), and adjusted R^2 are given.

	Intercept	Sex	Species	$\delta^{15}N$	$\delta^{13}C$	df	AICc	ΔAICc	AICc-weight	R^2
ln(Σ_{10}PCBs)	4.462	+	+			4	36.51	0.000	0.997	0.58
	4.864		+			3	48.36	11.85	0.003	0.31
	2.045	+		0.289		4	54.35	17.84	0.000	0.23
	4.944	+				3	56.11	19.60	0.000	0.10
	5.122					2	56.56	20.05	0.000	0.00
	3.101			0.206		3	56.96	20.44	0.000	0.07
	4.991	+			0.002	4	58.82	22.31	0.000	0.10
	5.479				0.019	3	59.01	22.49	0.000	0.00
ln(Σ_6DDTs)	4.611	+	+			4	49.22	0.000	0.997	0.57
	2.398	+		0.2687		4	62.26	13.04	0.001	0.33
	5.090	+				3	62.56	13.34	0.001	0.26
	5.816	+			0.038	4	65.07	15.85	0.000	0.26
	5.238		+			3	66.96	17.74	0.000	0.13
	5.447					2	68.61	19.39	0.000	0.00
	6.777				0.07	3	70.60	21.38	0.000	0.02
	4.240			0.123		3	70.63	21.41	0.000	0.02
ln(Σ_5CHLs)	4.065	+	+			4	33.92	0.000	0.987	0.49
	2.113	+		0.232		4	44.61	10.69	0.005	0.25
	4.433	+				3	45.51	11.58	0.003	0.15
	4.473		+			3	45.66	11.73	0.003	0.15
	4.622					2	47.56	13.64	0.001	0.00
	3.852	+			-0.031	4	48.04	14.12	0.001	0.16
	3.390			0.126		3	49.12	15.20	0.000	0.03
	4.248				-0.020	3	50.01	16.09	0.000	0.00
ln(Σ_3HCHs)	2.038	+	+			4	30.45	0.000	0.920	0.48
	-0.410	+		0.275		4	35.79	5.34	0.064	0.37
	2.348	+				3	39.70	9.25	0.009	0.20
	1.583	+			-0.040	4	42.01	11.56	0.003	0.26
	2.441		+			3	43.30	12.85	0.001	0.09
	2.552					2	43.53	13.08	0.001	0.00
	0.999			0.159		3	44.26	13.81	0.001	0.06
	2.012				-0.028	3	45.88	15.43	0.000	0.01
ln(HCB)										
	3.321	+	+			4	31.81	0.00	0.994	0.54
	1.153	+		0.255		4	43.63	11.81	0.003	0.30

	3.707	+				3	45.37	13.56	0.001	0.18
	3.764		+			3	46.42	14.61	0.001	0.15
	2.303	+			-0.074	4	46.91	15.10	0.001	0.21
	3.916					2	48.40	16.56	0.000	0.00
	2.567			0.138		3	49.80	17.99	0.000	0.04
	2.749				-0.061	3	50.24	18.42	0.000	0.02
ln(Mirex)	0.500	+	+			4	46.56	0.00	0.891	0.46
	0.834		+			3	50.91	4.34	0.102	0.30
	1.095					2	58.50	11.93	0.002	0.00
	-1.115			0.226		3	58.88	12.32	0.002	0.07
	-1.916	+		0.292		4	59.79	13.23	0.001	0.13
	1.009	+				3	60.42	13.86	0.001	0.02
	0.808				-0.015	3	60.99	14.42	0.001	0.00
	0.627	+			-0.020	4	63.12	16.55	0.000	0.02
ln(Σ_4BDEs)	1.397	+	+			4	58.627	0.00	0.99	0.58
	1.926		+			3	68.194	9.57	0.01	0.35
	2.328					2	78.327	19.70	0.00	0.00
	-1.417	+		0.353		4	78.697	20.07	0.00	0.15
	2.118	+				3	78.939	20.31	0.00	0.06
	-0.166			0.254		3	79.328	20.70	0.00	0.05
	1.715				-0.032	3	80.749	22.12	0.00	0.00
	1.118	+			-0.052	4	81.433	22.81	0.00	0.07
ln(Σ_2TOXs)	3.068	+	+			4	37.138	0.00	1.00	0.63
	-0.861	+		0.444		4	51.276	14.14	0.00	0.40
	3.542		+			3	52.836	15.70	0.00	0.30
	0.531			0.334		3	58.082	20.95	0.00	0.16
	3.592	+				3	59.081	21.94	0.00	0.13
	3.815					2	60.658	23.52	0.00	0.00
	2.335	+			-0.066	4	61.117	23.98	0.00	0.15
	2.975				-0.044	3	62.890	25.75	0.00	0.01

function in the software R-3.2.5) ranging between the LOD value and ½ LOD were generated. Concentrations are given in ng/g lipid weight (lw).

2.4. Statistical analyses

Statistical analyses were performed using R version 3.6.1 (R Core Team, 2019). Patterns of POPs including ΣDDTs, ΣPCBs, ΣPBDEs, ΣCHLs, ΣHCHs, HCB and mirex, were investigated using principal component analysis (PCA). Because proportions summing up to one were used, the PCA was derived from the covariance matrix of centred log ratio concentrations of pollutants (ln ($POP_x/\Sigma POP$)) in R package ade4 (Dray and Dufour, 2007; Aitchison and Greenacre, 2002). Linear models were used to investigate variability in POPs. These models included both categorical (i.e., sex and species) and continuous ($\delta^{15}N$, $\delta^{13}C$) independent variables (Table 1). Correlated predictor variables, for example species and $\delta^{15}N$, were not included in the same model. The models were ranked using an information-theoretic approach (Burnham and Anderson, 2002) based on Akaike's information criterion corrected for small sample size (AICc, R package *MuMIn* (Barton, 2016)). The number of parameters (K), the difference in AICc values between the "best" model and the model at hand (ΔAICc) and a normalized weight of evidence in favour of the specific model, relative to the whole set of candidate models, derived by $e^{(-0.5(\Delta AICc))}$ (AICc weights) were obtained. Conditional model averaging was used to make inferences from all of the models (Burnham and Anderson, 2011). This method produces averaged estimates of all predictor variables in the candidate model list, weighted using the AICc weights (Burnham and Anderson, 2002; Lukacs et al., 2009). From this, conditional parameter-averaged estimates (β) and 95% confidence intervals (CIs) for all the predictors included in the models were obtained. To determine if parameters were significantly different from 0 at the 5% level, 95% CI of the model averaged estimates were used. The 95% CIs provide information about a range in which the true value lies with a certain degree of probability, and about the direction and strength of the demonstrated effect (du Prel et al., 2009). If the confidence interval does not include the value of zero effect, it can be assumed that the result is statistically significant. Model fit was assessed using residual diagnostic plots. All POPs were ln-transformed to meet assumptions of linear models.

3. Results and discussion

3.1. Stable isotope values

Based on linear model estimates, stable isotope values for $\delta^{15}N$ were 0.92‰ units (95% CI: 0.38, 1.45) higher in fin whales than in blue whales, whereas $\delta^{13}C$ values were 1.14‰ lower (95% CI −2.24, −0.03) in fin whales than in blue whales (Table 2). Stable isotope values were similar in males and females, when species-differences were taken into account (−0.11, 95% CI: −0.62, 0.41 and −0.03, 95% CI: −1.10, 1.05 for $\delta^{15}N$ and $\delta^{13}C$, respectively) (Table 2). Lower $\delta^{15}N$ values in blue whales indicate that they feed at lower trophic level than fin whales. This finding is in accordance with a study from the Gulf of St Lawrence, where stable isotope values suggested that krill is the most important prey for both species, but that fin whales also forage on various fish species (Gavrilchuk et al., 2014). Similar conclusions were also attained by food web network analyses focusing on marine mammals in the Barents Sea (Blanchet et al., 2019). Acoustic surveys of krill and density distributions of fin whales in the Barents Sea suggest that the diet of this cetacean species was dominated by krill and capelin (Ressler et al., 2015).

The lower $\delta^{13}C$ values in fin whales compared to blue whales suggest that fin whales feed more at higher latitudes than blue whales, because pelagic ecosystems at high latitudes generally have lower $\delta^{13}C$ values than lower-latitude ecosystems due to low temperatures and high productivity (de la Vega et al., 2019; Newsome et al., 2010). Feeding at different latitudes may have occurred prior to the summer migration into the Arctic, as incorporation rates of stable isotopes in whale skin are relatively slow and would reflect feeding over three to six month period (Busquets-Vass et al., 2017; Giménez et al., 2016). The incorporation rates for $\delta^{15}N$ in blue whale and bottlenose dolphin (*Tursiops truncatus*) skin are 163 ± 91 and 180 ± 71 days, respectively, whereas for $\delta^{13}C$, estimated only for dolphins, is 104 ± 35 days (Busquets-Vass et al., 2017; Giménez et al., 2016). The results of the current study are in accordance with results from passive acoustic sampling, which also suggest that at least some fin whales are present in polar waters of the North Atlantic during winter, whereas blue whales appear to be present at those high latitudes only from June to October (Ahonen et al., 2017; Klinck et al., 2012; Moore et al., 2012). Additional acoustic and satellite tracking studies would provide further insights regarding movement patterns and feeding grounds and, thus, possible additional sources of contaminants in blue and fin whales that spend the summer in Svalbard.

3.2. Pollutants concentrations and patterns

The following compounds were detected in >70% of the individuals HCB, α-, β- and γ-HCH, *oxy*-, *trans*- and *cis*-chlordane, *trans*- and *cis*-nonachlor, mirex, *o,p′*-DDT, *p,p′*-DDT, *o,p′*-DDD, *p,p′*-DDD, *o,p′*-DDE, *p,p′*-DDE, CB-52, -99, -101, -105, -118, -138, -153, -180, -183, and -187, and, BDE-47, -99, -100 and -153 (Table 2). For toxaphenes, four congeners were quantified and detected in >70% of the individuals in Batch 1: (toxaphene-26 [B8-1413]; -40 [B8-1414]; -42 [B8-806/809]; -50 [B9-1679]: Σ_4TOXs) using Parlar and AV-code toxaphene identifiers in brackets. In the second and third batches of POP analyses, only toxaphenes-26 and -40 (Σ_2TOXs) were detected in >70% of the individuals (Table 2).

POPs determined in blubber of both whale species were dominated by DDTs, PCBs and toxaphenes (Table 2). Median concentrations (ng/g lw) and ranges of pollutants in blue whale and fin whale blubber for each species and sex are given in Table 2. For comparative purposes, mean concentrations and standard deviations for each species are given in Table S3. Σ_6DDTs were dominated by *p,p′*-DDE, PCBs by PCB-153 and -138 and toxaphenes by toxaphene-50 (Fig. 2). Σ_5CHLs, Σ_5HCHs and Σ_4PBDEs consisted mainly of *trans*-nonachlor, β-HCH and BDE-47, respectively (Fig. 2).

3.3. Factors explaining pollutant concentrations

All of the highest ranked models explaining variation in pollutant concentrations included species and sex as predictor variables (ΔAICc > 4.3); 46–58% of the total variation was accounted for in these models (Table 1). Model-averaged estimates indicated that concentrations of $\Sigma_5$chlordanes (CHLs), Σ_3HCH and HCB were approximately twice as high in fin whales as blue whales (range for 95% CI: 1.3–2.9), whereas the difference was 2.5-fold for Σ_{10}PCBs, Σ_6DDTs, mirex and Σ_2TOXs (range for 95% CI: 1.6–4.0) and almos four-fold for Σ_4PBDEs (95% CI: 2.3, 6.4). The PCA plot showing the POP patterns (Fig. 3) indicated that the proportion of Σ_4BDEs contributing to ΣPOPs was higher in fin whales compared to blue whales. The higher concentrations of pollutants in fin whales are likely the result of them feeding at a higher trophic position, which was shown in this study by their respective $\delta^{15}N$ values. Pollutants biomagnify in pelagic food webs, and higher concentrations of pollutants in fish, compared to krill, have been reported in waters surrounding Svalbard (Hallanger et al., 2011b). The model-averaged estimates in the current study showed that $\delta^{15}N$ explained some of the variation in pollutant concentrations, although it was not included in the highest ranked models (Table 1). Overall, concentrations of Σ_2TOXs increased 55% (95% CI: 19, 105) with one per mille unit increase in $\delta^{15}N$. The average increase for Σ_{10}PCBs, Σ_5CHLs, Σ_3HCH, HCB, mirex and Σ_4PBDEs was 25–36% with one per mille unit increase in $\delta^{15}N$, but the increase was significant at the 5% level only for Σ_3HCH (95% CI: 5, 64), though it was close to significance for the other compounds (range 95% CI: −12, 114; Table 3). Positive correlation between blubber

Table 2
Median (minimum, maximum) stable isotope values in skin, lipid percentage and concentrations of pollutants[a] in ng/g lipid weight in blubber of blue whales (*B. musculus*) and fin whales (*B. physalus*) sampled from Svalbard (2014-2018). Summed compounds are shown in bold.

	Blue whales		Fin whales	
	Females (n=7)	Males (n=11)	Females (n=8)	Males (n=4)
δ^{15}N	9.34 (8.47, 10.68)	9.25 (8.58, 10.29)	10.60 (9.39, 11.37)	10.61 (9.59, 10.70)[b]
δ^{13}C	-18.10 (-19.50, -16.38)	-18.93 (-21.98, -15.86)	-20.05 (-20.31, -19.25)	-19.40 (-19.92, -16.82)[b]
Lipid %	43 (25 , 55)	52 (24 , 63)	47 (26 , 58)	44 (32 , 49)
CB-52	5.7 (2.4, 14)	14.2 (9.1, 29)	19.2 (7.3, 34)	44.8 (30.0, 55)
CB-99	3.4 (2.0, 11)	10.9 (7.5, 24)	16.6 (5.3, 26)	32.6 (20.8, 41)
CB-101	5.7 (2.3, 13)	12.4 (7.5, 23)	18.9 (6.4, 32)	41.1 (29.2, 53)
CB-105	0.72 (0.37, 2.1)	2.12 (0.92, 4.1)	4.1 (1.87, 7.0)	5.7 (5.12, 8.9)
CB-118	5.1 (2.7, 15)	15.5 (10.3, 35)	22.6 (7.4, 36)	46.5 (31.0, 56)
CB-138	9.3 (6, 26)	22.9 (18, 59)	41 (14.1, 64)	82 (48.9, 94)
CB-153	16 (9.1, 41)	35 (29.6, 90)	58 (20, 97)	118 (70, 132)
CB-180	6.2 (3.2, 19)	10.9 (9.0, 34)	23.2 (8.2, 33)	34.2 (20.1, 49)
CB-183	1.3 (0.45, 3.8)	2.0 (0.58, 6.5)	4.4 (1.61, 5.6)	6.4 (4.23, 10.8)
CB-187	5.8 (3.3, 16)	11.0 (8.1, 31)	20.6 (7.0, 28)	30.0 (19.8, 41)
Σ_{10}PCBs	**86 (55, 150)**	**134 (106, 334)**	**219 (79, 341)**	**451 (279, 522)**
HCB	30 (12, 49)	58 (41, 82)	61 (35, 104)	85 (79, 139)[b]
Mirex	1.4 (0.9, 6.2)	2.5 (1.6, 6.0)	4.3 (2.5, 8.6)	4.7 (3.5, 8.4)[b]
α-HCH	1.1 (<0.3, 3.0)	2.3 (1.55, 3.4)	2.8 (1.3, 6.1)	5.2 (4.2, 7.0)[b]
β-HCH	4.5 (1.5, 9.3)	10.1 (6.6, 17.4)	10.1 (4.0, 22.7)	18.7 (16.4, 31.7)[b]
γ-HCH	0.80 (0.22, 1.3)	1.21 (0.97, 1.5)	0.99 (0.42, 1.7)	1.82 (1.33, 2.5)[b]
Σ_3HCHs	**8.4 (4.6, 13)**	**13.9 (9.6, 22)**	**12.2 (7.8, 28)**	**25.7 (21.9, 41)[b]**
Oxychlordane	5.4 (2.5, 12)	14.1 (9.7, 27)	16.7 (7.3, 32)	34.4 (23.1, 50)[b]
trans-Chlordane	0.26 (<0.08, 0.38)	0.25 (0.18, 0.78)	0.52 (<0.09, 1.02)	0.28 (0.24, 0.56)[b]
cis-Chlordane	4.3 (1.8, 8.2)	7.2 (4.7, 9.5)	7.6 (4.2, 12.8)	12.6 (12.4, 14.3)[b]
trans-Nonachlor	23 (12, 55)	57 (41, 109)	73 (25, 118)	130 (89, 191)[b]
cis-Nonachlor	7.4 (3.8, 20)	20.6 (14.7, 43)	26 (8.6, 42)	51 (34.0, 70)[b]
Σ_4CHLs	**65 (33, 96)**	**102 (71, 189)**	**122 (49, 202)**	**227 (161, 323)[b]**
o,p'-DDT	7.6 (4.5, 37)	36.2 (18.5, 87)	33 (4.1, 68)	92 (43, 114)
p,p'-DDT	7.4 (5.5, 24)	19 (13, 61)	26 (9.6, 103)	21 (19, 39)
o,p'-DDD	6.3 (2.4, 21)	17 (10, 45)	18 (6.0, 25)	42 (30, 49)
p,p'-DDD	18 (8.3, 47)	40 (15, 131)	32 (8.9, 49)	90 (75, 113)
o,p'-DDE	2.3 (0.7, 5.7)	6.4 (3.3, 10.0)	5.6 (1.7, 8.0)	10.3 (8.6, 14)
p,p'-DDE	46 (18, 133)	128 (90, 335)	165 (47, 238)	367 (231, 400)
Σ_6DDTs	**84 (40, 268)**	**244 (163, 669)**	**287 (113, 482)**	**645 (409, 681)**
Toxaphene 26	11 (6.5, 26)	28 (14, 54)	31 (17, 65)	63 (45, 100)
Toxaphene 40	8.6 (4.9, 20)	20 (<0.02, 29)	24 (17, 46)	42 (34, 65)
Toxaphene 42	21 (11, 46)[c]	53 (27, 66)[c]	37 (35, 54)[c]	76 (64, 93)[c]
Toxaphene 50	28 (13, 65)[c]	63 (30, 147)[c]	70 (61, 86)[c]	142 (132, 210)[c]
ΣTOX26+40	**23 (11, 46)**	**48 (21, 81)**	**56 (34, 110)**	**105 (79, 166)**
Σ_4TOX	**74 (35, 157)[c]**	**142 (85, 288)[c]**	**178 (138, 179)[c]**	**301 (287, 418)[c]**
BDE47	2.1 (1.1, 5.5)	6.1 (4.1, 10.0)	9.9 (2.5, 16)	22 (14, 25)
BDE99	0.92 (0.35, 2.8)	2.3 (1.3, 4.5)	3.5 (0.94, 6.5)	9.0 (5.1, 12)
BDE100	0.48 (0.21, 1.1)	0.90 (<0.09, 1.4)	1.3 (0.17, 71)	2.1 (1.6, 2.9)
BDE153	0.15 (<0.04, 0.88)	0.35 (<0.07, 0.80)	0.61 (<0.08, 1.80)	1.3 (0.77, 2.1)
ΣPBDEs	**3.7 (2.0, 9.6)**	**10 (5.9, 17)**	**16 (4.7, 94)**	**35 (22, 40)**

PCB=Polychlorinated biphenyls, HCB=Hexachlorobenzene; HCH=Hexachlorocyclohexane; DDT=(1,1,1-trichloro-2,2-bis(p-chlorophenyl)ethane; DDE=1,1-dichloro-2,2-bis(p-chlorophenyl)ethylene; DDD=1,1-dichloro-2,2-bis(p-chlorophenyl)ethane; PBDE=polybrominated diphenyl ethers.

[a] CB-28 and -194, toxaphenes #32, #38, #58, #62 and #69, and BDE-28, -138, -183 and -209 were detected above the limit of detection in <70% of the individuals

[b] δ^{15}N, δ^{13}C, HCB, HCHs, Mirex and chlordanes were analyses in 3 male fin whales.

[c] Toxaphenes 42 and 50 and Σ_4TOX were only analysed in the first batch of 17 samples (n=4/7, 3/3 for blue whales and fin whales (F/M).

pollutant concentrations and skin δ^{15}N values has also been demonstrated in other whale species (Das et al., 2017; Pinzone et al., 2015). Pollutant concentrations were not related to δ^{13}C values (Table 3). This suggests that differences in pollutant concentrations are driven by trophic position rather than carbon source. Feeding at different latitudes, as indicated by δ^{13}C values, does not seem to impact POP exposure in fin and blue whales, which suggests similar POP concentrations in their prey across latitudes. This is in accordance with similar measured concentrations of CB-153 in seawater across mid and high latitudes of the North Atlantic (Wagner et al., 2019). Another explanation could be that δ^{13}C may not be a reliable spatial signal in whales because of their movements between coastal δ^{13}C-enriched and δ^{13}C-depleted oceanic ecosystems (Newsome et al., 2010) within a specific latitudinal foraging zone, as previously observed in blue whales (Bailey et al., 2009).

Concentrations of all compounds were 1.7–2.8 times higher in males than in females (range for 95% CI: 1.1–4.2) (Table 3). This is in accordance with previous studies and trends observed in marine mammals, including fin whales and blue whales (Aguilar et al., 1999; Aguilar and Borrell, 1994; Fossi et al., 2014; Gauthier et al., 1997b; Hobbs et al., 2001; Metcalfe et al., 2004; Muñoz-Arnanz et al., 2019: Pinzone et al., 2015). This pattern suggests that maternal transfer of pollutants to their offspring takes place via lactation, and to a lesser degree to the foetus during gestation as indicated by modelling and empirical studies on whales (Hickie et al., 1999; Trumble et al., 2013).

3.4. Intra- and interspecies differences in pollutant concentrations

To provide a comprehensive overview of the levels of pollutants found in blue and fin whales in comparison to their counterparts from other regions, in addition to different whale species feeding in the same area, pollutant concentrations were explored by sex whenever the information was available. It should be noted that comparing PCB concentrations should be considered to be only indicative as most studies provide values as a sum and the number and type of congeners present in the sums often differ among studies. Geographical comparisons

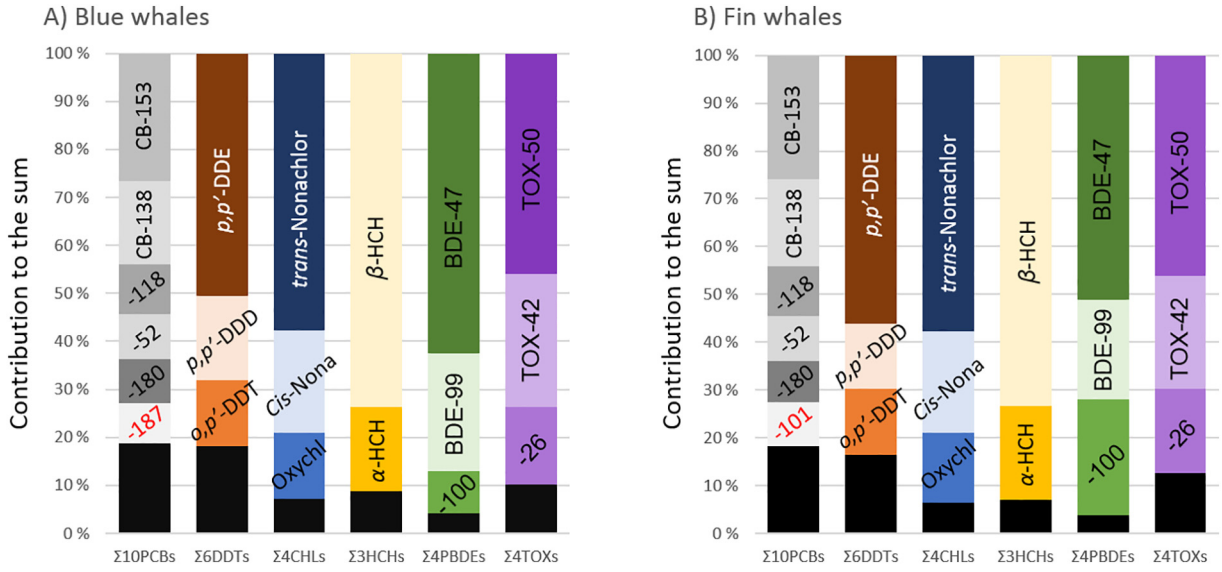

Fig. 2. Major congeners and metabolites (contribution > 80% to the sum of compounds) in A) blue whales and B) fin whales blubber samples (n = 30 for PCBs, DDTs and PBDEs, 29 for CHLs and HCHs and 17 for Toxaphenes, respectively) collected in Svalbard, Norway (2014–2018). Black areas represent minor (contribution < 20% to the sum of compounds).

may also be biased by temporal changes in pollutant levels (Rigét et al., 2019), if samples collected during different time periods are compared. To avoid bias of temporal changes in pollutant levels, the comparison included only samples collected after 2004, when the United Nations Environment Program's Stockholm Convention on restriction or elimination of POPs entered into force.

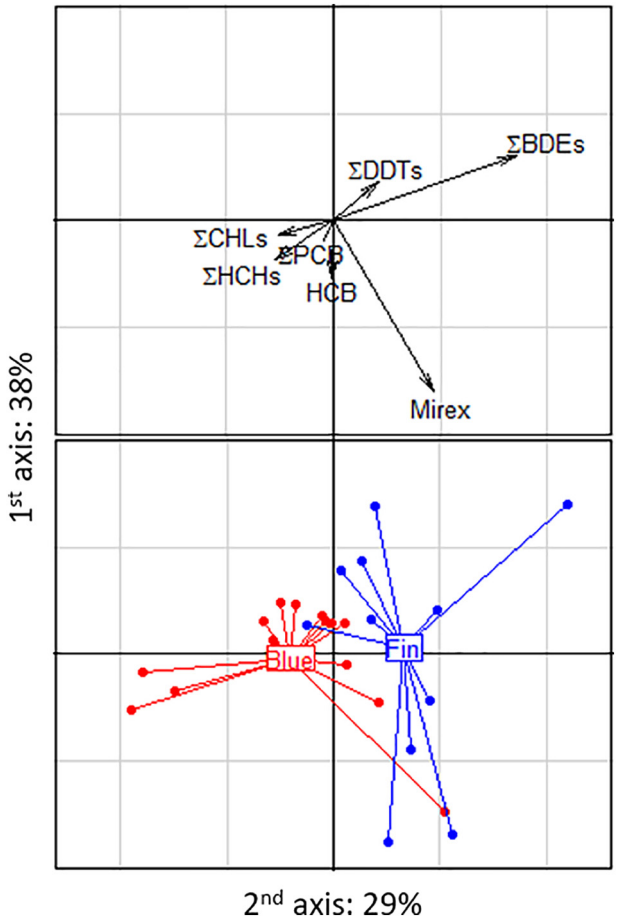

Fig. 3. Pollutant patterns in the blubber of blue and fin whales (n = 29), collected in Svalbard, Norway. These PCA ordination plots are based on the covariance matrix of log ratio concentrations of pollutants (ln ($POP_x/\Sigma POP$)). Sample scores are grouped by species. The 1st axis explains 38% of the variation and the 2nd axis explains 29% of the variation.

A limited number of studies have assessed POP levels in blue whales. In general, these studies suggest that blue whales sampled from Svalbard are less polluted than conspecifics from the west coast of Mexico and similarly or less polluted than blue whales from southern Chile (Fossi et al., 2014; Muñoz-Arnanz et al., 2019). For instance, DDT concentrations in blue whales from the Gulf of California, Mexico (sampled in 2010) were an order of magnitude higher than in Svalbard blue whales (Fossi et al., 2014). Median concentrations of HCB and Σ_6DDT in blue whales sampled from Svalbard were three to seven times higher in comparison to blue whales sampled from their feeding areas close to Isla de Chiloé, Southern Chile, between 2011 and 2013 (Muñoz-Arnanz et al., 2019). PCB and PBDE concentrations were similar between Svalbard and Chile, but congener patterns differed. Σ_{10}PCBs was dominated by CB-153 > CB-138 > CB-118 > CB-52 in blue whales from Svalbard (Fig. 2), whereas CB-95 > CB-101 > CB-153 > CB-149 were the dominating congeners, accounting for ~50% of the Σ_{20}PCBs reported, for blue whales from Chile. In whales sampled from Svalbard, PBDEs were dominated by BDE-47 > BDE-99 > BDE-100 > BDE-153 (Fig. 2), whereas BDE-47 > BDE-28 > BDE-85 > BDE-99 contributed ~85% to the Σ_{15}PBDE content reported for the blue whales sampled from Chile. Higher HCB and DDT concentrations in blue whales from Svalbard may be in part related to much lower usage of these compounds in the Southern Hemisphere than in the Northern Hemisphere and slow interhemispheric mixing of HCB (Barber et al., 2005; Stemmler and Lammel, 2009; Zhang and Lohmann, 2010). However, global distribution of POPs in marine environments does not solely reflect their application and use in neighbouring continents. The similar concentrations of PCBs in blue whales from Svalbard and Chile may be related to oceanic transport of PCBs from their main area of usage in the Northern Hemisphere to the Southern Hemisphere (Breivik et al., 2002; Wagner et al., 2019). In addition, meteorological and biogeochemical conditions such as temperature, ocean and air currents, deep water formation, sea ice, light penetration, productivity and suspended particle dynamics may all influence distribution and residence time of POPs in seawater (Barber et al., 2005; Stemmler and Lammel, 2009; Wagner et al., 2019).

Fin whales from Svalbard were generally less polluted than fin whales sampled in the Gulf of California and in the Mediterranean Sea, but more polluted than fin whales sampled in Antarctica (Fossi et al., 2016; Niño-Torres et al., 2010; Pinzone et al., 2015; Taniguchi et al., 2019). For instance, mean concentrations of ΣPCBs and Σ_6DDTs were

Table 3
Variation in pollutant concentrations (ln(ng/g lw)) according to species, sex and dietary tracers (δ^{15}N and δ^{13}C). Blue whales (*B. musculus*, n = 18) and fin whales (*B. physalus*, n = 10–11) were sampled from 2014 to 2018 off the coast of Svalbard, Norway. Conditional averaged estimates and 95% confidence intervals were derived from linear models. Values in bold are significantly different from 0 at the 5% level. Males = M, females = F.

	Intercept	Species fin vs. blue	Sex M vs. F	δ^{15}N	δ^{13}C
ln(Σ_{10}PCBs)	**4.46 (4.17, 4.76)**	**0.90 (0.56, 1.24)**	**0.66 (0.32, 0.99)**	0.27 (−0.02, 0.57)	0.01 (−0.16, 0.18)
ln(Σ_6DDTs)	**4.61 (4.19, 5.03)**	**0.90 (0.48, 0.1.32)**	**1.03 (0.62, 1.44)**	0.27 (−0.06, 0.6)	0.04 (−0.15, 0.23)
ln(Σ_5CHLs)	**4.06 (3.61, 4.50)**	**0.69 (0.34, 1.04)**	**0.67 (0.33, 1.01)**	0.22 (−0.04, 0.49)	−0.03 (−0.18, 0.12)
ln(Σ_3HCHs)	**1.88 (0.55, 3.22)**	**0.58 (0.25, 0.91)**	**0.65 (0.33, 0.97)**	**0.27 (0.05, 0.50)**	−0.04 (−0.17, 0.10)
ln(HCB)	**3.32 (2.93, 3.70)**	**0.72 (0.39, 1.06)**	**0.73 (0.40, 1.05)**	0.25 (−0.01, 0.51)	−0.07 (−0.22, 0.08)
ln(Mirex)	**0.53 (0.03, 1.03)**	**0.93 (0.47, 1.39)**	**0.55 (0.13, 0.97)**	0.25 (−0.08, 0.59)	−0.02 (−0.21, 0.18)
ln(Σ_4BDEs)	**1.40 (0.98, 1.83)**	**1.34 (0.85, 1.85)**	**0.87 (0.38, 1.35)**	0.31 (−0.13, 0.76)	−0.04 (−0.29, 0.21)
ln(Σ_2TOXs)	**3.07 (269, 3.44)**	**0.98 (0.64, 1.32)**	**0.77 (0.44, 1.11)**	**0.44 (0.17, 0.72)**	−0.06 (−0.24, 0.12)

Σ_{10}PCBs (CB-52, -99, -101, -105, -118, -138, -153, -180, -183, and -187); Σ_6DDTs (*o,p′*-DDT, *p,p′*-DDT, *o,p′*-DDD, *p,p′*-DDD, *o,p′*-DDE, *p,p′*-DDE): Σ_5CHLs (*oxy*-, *trans*-, *cis*-chlordane, *trans*-, *cis*-nonachlor); Σ_3HCHs (α-, β-, γ-HCH); Σ_4PBDEs (BDE-47, -99, -100, -153); Σ_2TOXs (TOX-26, -40).

10–45 times higher in fin whales from the Gulf of California and the Mediterranean Sea (sampled in 2006–2013) compared to fin whales from the present study (Fossi et al., 2016; Pinzone et al., 2015). In contrast, mean concentrations of Σ_3HCH were similar between the Gulf of California and Svalbard (Niño-Torres et al., 2010). Median concentrations of Σ_6DDTs in fin whales from Svalbard were 16 times higher compared to blubber concentrations reported in fin whales sampled from the Antarctic Peninsula in 2013 (Taniguchi et al., 2019). ΣPCB and HCB concentrations were only approximately twice as high in the fin whales from Svalbard compared to those spending the summer in the Antarctic Peninsula. The larger difference in DDT, compared to PCBs, between whales sampled from the Arctic and Antarctic is in accordance with differences in sea water concentrations (Gao et al., 2018; Hallanger et al., 2011a; Wagner et al., 2019). In comparison to other whale species sampled from polar waters in the North Atlantic since 2004, male humpback whales (*Megaptera novaeangliae*) from the Northwest Atlantic (Gulf of Maine, 2004) were more contaminated than the whales from the current study, with 5 to 58 times higher ΣPCB and Σ_6DDT concentrations (Elfes et al., 2010). Higher POP concentrations in humpback whales are likely the result of this species feeding at a higher trophic level than fin and blue whales (Pauly et al., 1998).

4. Conclusion

Higher POP concentrations in fin whales than in blue whales from Northeast Atlantic are likely related to the higher trophic level diet of fin whales. The comparison of pollutant levels between different areas suggests that blue and fin whales feeding in Svalbard waters during summer are exposed to lower levels of pollutants than their conspecifics sampled close to areas at lower latitudes with dense human populations (Mediterranean Sea, Gulf of California). Lower δ^{13}C values in fin whales than blue whales suggested that more of their feeding occurred at higher latitudes. Pollutant levels in females were twice those of males, which indicates that females offload considerable amounts of pollutants to their offspring. Future studies are needed to identify the movement patterns and wintering areas of these ocean giants to identify the sources of their pollutant exposure. While most POPs show decreasing trends in Arctic biota, the levels measured in marine mammals remain high in some populations (mid-latitudes) and raise concerns regarding potential health effects, which warrants further study.

CRediT authorship contribution statement

Sabrina Tartu: Formal analysis, Data curation, Writing - original draft, Writing - review & editing, Visualization. **Aaron T. Fisk:** Resources, Validation, Writing - original draft, Writing - review & editing. **Arntraut Götsch:** Investigation, Validation, Writing - original draft, Writing - review & editing. **Kit Kovacs:** Conceptualization, Investigation, Writing - review & editing, Funding acquisition. **Christian Lydersen:** Conceptualization, Investigation, Writing - review & editing. **Heli Routti:** Conceptualization, Formal analysis, Writing - original draft, Writing - review & editing, Visualization, Supervision, Project administration, Funding acquisition.

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.

Acknowledgements

The authors thank all of the people who helped with the fieldwork over the whole of the sampling program and M. Berube and P. Palsbøll at the Groningen Institute of Evolutionary Life Sciences for genetic sex identification of the animals in this study. The study was supported financially by the Norwegian Polar Institute, the Fram Centre Hazardous Substances Flagship Program (Project #602018 to H.R.) and the Norwegian Research Council ICE whales grant (Project #244488/E10 to K.M.K.).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2020.137327.

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