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Within and between breeding-season changes in contaminant occurrence and body condition in the Antarctic breeding south polar skua☆

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ABSTRACT

The Antarctic ecosystem represents a remote region far from point sources of pollution. Still, Antarctic marine predators, such as seabirds, are exposed to organohalogen contaminants (OHCs) which may induce adverse health effects. With increasing restrictions and regulations on OHCs, the levels and exposure are expected to decrease over time. We studied south polar skua (*Catharacta maccormiciki*), a top predator seabird, to compare OHC concentrations measured in whole blood from 2001/2002 and 2013/2014 in Dronning Maud Land. As a previous study found increasing organochlorine concentrations with sampling day during the 2001/2002 breeding season, suggesting dietary changes, we investigated if this increase was repeated in the 2013/2014 breeding season. In addition to organochlorines, we analyzed hydroxy-metabolites, brominated contaminants and per- and polyfluoroalkyl substances (PFAS) in 2013/2014, as well as dietary descriptors of stable isotopes of carbon and nitrogen, to assess potential changes in diet during breeding. Lipid normalized concentrations of individual OHCs were 63%, 87% and 105% higher for hexachlorobenzene (HCB), 1,1-dichloro-2,2-bis (p-chlorophenyl)ethylene (p,p'-DDE), and ∑Polychlorinated biphenyls (PCBs), respectively, in 2013/2014 compared to 2001/2002. South polar skuas males in 2013/2014 were in poorer body condition than in 2001/2002, and with higher pollutant levels. Poorer body condition may cause the remobilization of contaminants from stored body reserves, and continued exposure to legacy contaminants at overwintering areas may explain the unexpected higher OHC concentrations in 2013/2014 than 2001/2002. Concentrations of protein-associated PFAS increased with sampling day during the 2013/2014 breeding season, whereas the lipid-soluble chlorinated pesticides, PCBs and polybrominated diphenyl ether (PBDEs) showed no change. OHC occurrence was not correlated with stable isotopes. The PFAS biomagnification through the local food web at the colony should be investigated further.

1. Introduction

Antarctica is located far from point sources of pollution making it one of the least pollutant-affected and most pristine continents on earth (Corsolini, 2009). However, with growing human population and industrial development in the Southern hemisphere countries the Antarctic ecosystems are increasingly subjected to multiple environmental threats, including environmental pollution from organohalogen contaminants (OHCs) (Aronson et al., 2011; Bennett et al., 2015). OHCs include various chlorinated, brominated and fluorinated chemicals produced for industries, agriculture, disease control or consumer products (Arctic Monitoring Assessment Program, 2004). Many OHCs and their by-products are persistent to degradation, can accumulate in the environment, biomagnify in biota (Borgå et al., 2004), and may be toxic to humans and wildlife (Dietz et al., 2019; Letcher et al., 2010). OHCs reach Antarctic ecosystems primarily via long range atmospheric transport (Kallenborn et al., 2013; Wania and Mackay, 1993), as well as transport by migratory species and melting glaciers (Brink et al., 2009;

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Geisz et al., 2008; Roosens et al., 2007).

OHCs were recorded in Antarctica for the first time in the 1960s, when the chlorinated pesticide dichlorodiphenyltrichloroethane (DDT) was found in Adélie penguins (*Pygoscelis adeliae*), Weddell seals (*Leptonychotes weddellii*) and south polar skuas (*Catharacta maccormicki*) (George and Frear, 1966). Today, OHCs are found in all compartments of the Antarctic ecosystems including atmosphere, water, sea ice, seawater, sediments, ice, snow and biota (Corsolini, 2009). Though Antarctic contaminant concentrations are generally low compared to other regions of the world, some OHCs are present in high concentrations in organisms occupying high trophic positions in the Antarctic food web (Fuoco et al., 2009). As a top predator seabird, the south polar skua has among the highest contaminant concentrations recorded in Antarctic species (Bustnes et al., 2006, 2007). Moreover, concentrations of the chlorinated pesticides HCB and Mirex increased in south polar skuas during their breeding season at Svarthamaren, Dronning Maud Land (Bustnes et al., 2006). The increase in OHC concentrations suggests increasing exposure from the diet during the breeding season (Bustnes et al., 2006, 2007).

Global conventions, such as the Convention on Long-range Transboundary Air Pollution and the Stockholm Convention on Persistent Organic Pollutants, came into force during 2003 and 2004 (UNECE, 1979; United Nations Environment Programme, 2001). These restrictions have contributed to a general and global decrease of many OHCs in the environment over time (Hung et al., 2016; Rigét et al., 2019). In particular, OHC levels have decreased in both in Antarctica (Corsolini, 2009; Weber and Goerke, 2003), and in the Arctic (Rigét et al., 2019). However, a proper assessment of temporal trends of OHC concentrations in the Antarctic is challenging as studies focus on different species, tissues, analytical methods, contaminant summations, geographic locations, and period of the year (Corsolini, 2009).

In the present study, we aimed to compare OHCs and body condition in an Antarctic breeding avian top predator, the south polar skua, sampled a decade apart in 2001/2002 and in 2013/2014 during the breeding season at Svarthamaren, Dronning Maud Land. We compare organochlorine data between breeding the seasons 2001/2002 (Bustnes et al., 2006), and data measured in the present study sampled from the same colony in 2013/2014. To understand which factors contribute to OHC occurrence within the 2013/2014 breeding season, we also expand on the measured OHCs to include brominated flame retardants (BFRs), per- and polyfluoroalkyl substances (PFAS), hydroxy (OH)-metabolites of polychlorinated bihenyls (PCB) and polybrominated diphenyl ethers (PBDE), as well as dietary descriptors of carbon sources and relative trophic position, measured by stable isotopes of carbon $\delta^{13}C$ and nitrogen $\delta^{15}N$, respectively.

Due to increased global restrictions and regulations on OHCs during the same decade, the concentrations were expected to be lower in 2013/2014 than in 2001/2002. To ensure comparability, the same matrix (whole blood) was sampled and contaminant analyses were conducted in the same laboratory. Eight individuals from the 2001/2002 study were re-sampled in the 2013/2014 breeding season. To our knowledge, this is the first study of perfluoroalkyl substances (PFASs) from wildlife in Dronning Maud Land.

2. Materials and methods

2.1. Study area and sampling

Whole-blood was sampled from south polar skuas (n = 30) breeding at the Svarthamaren seabird colony (71°53′ S, 05°10′ E), Dronning Maud Land, Antarctica. Svarthamaren is an ice-free mountain approximately 200 km from the Antarctic coast (Brooke et al., 1999). In addition to approximately 120 breeding pairs of south polar skuas, the colony consists of 100,000–250,000 pairs of Antarctic petrels (*Thalassoica antarctica*) and approximately 2000 pairs of snow petrels (*Pagodroma nivea*) (Descamps et al., 2016, and unpubl. results). During the breeding season at Svarthamaren, the south polar skua stay near the colony and feed on eggs and chicks of Antarctic petrels, while petrels go on foraging trips to the coastal waters (Brooke et al., 1999). The egg laying period is very spread for skuas at Svarthamaren, and the first lay date in the 2013/2014 breeding season was 11 December 2013 and last lay date 20 January 2014. The birds were sampled between 7 December 2013 and 31 January 2014, and the nest status varied from eggs present, chicks or failed reproduction, with no relationship between sampling date and estimated hatching date (Table S1).

Skuas were caught and sampled as described previously (Helberg et al., 2005). In brief, birds were caught mostly by a snare (nest) trap, in addition to some sampled with a net gun (air-propelled net) or a large hand net. Whole blood (5 mL) was sampled from the brachial vein using a heparinized syringe, quickly frozen in the field and later stored at −20 °C for subsequent contaminant and isotopic analyses. Bill length (±0.1 mm) and height (±0.1 mm) were measured using a caliper, wing chord length (±0.5 mm) using a steel ruler, and body mass (±5 g) using a 2500 g Pesola® spring scale.

Work on the south polar skuas in Antarctica was approved by the Norwegian Animal Research Authority (NARA/FDU) under permits #3714, 5746 and 7935, following regulations and ethics for experiments with animals in the field (Norwegian Animal Welfare Act and Norwegian Regulation on Animal Experimentation, Food Safety Authority).

2.2. Sex determination

Sex was determined from blood samples in the laboratory using the chromo-helicase-DNA-binding (CHD) gene for sex identification (Griffiths et al., 1998). DNeasy Blood and Tissue kit (Qiagen, Valencia, CA, USA) was used for DNA extraction, and the primers 2550F and 2718R (Invitrogen, Thermo Fisher Scientific Inc, Waltham, MA, USA) were used for amplification of the CHD-gene (Fridolfsson and Ellegren, 1999).

2.3. Chemical analysis of pollutants

A total of 87 individual OHCs were analyzed in whole blood sampled in 2013/2014 at the Laboratory of Environmental Toxicology at the Norwegian University of Life Science. The chemical method used in the present study is accredited by the Norwegian Accreditation for analyzing OHCs in biological samples according to the requirements of the NS-EN SO/IEC 17025 (TEST 137). Analysis of organochlorine contaminants (OCs), brominated flame retardants (BFRs) and hydroxy-metabolites of polychlorinated biphenyls (OH-PCBs) and polybrominated diphenyl ethers (PBDEs) were performed according to Gabrielsen et al. (2011) and Polder et al. (2014), while analysis of perfluoroalkyl substances (PFASs) was performed as described in Grønnestad et al. (2017). In particular, OCs, BFRs and PBDEs were analyzed following lipid extraction from whole blood, whereas OH-PCBs and PFASs were analyzed from whole blood. For an overview of analyzed contaminants, see Supporting Information (Supporting Information, Table S1). The following procedural internal standards were added to whole-blood prior to extraction: PCB-29, -112, −207 (Ultra Scientific, N. Kingstown, RI, USA), BDE-77, -119, −181, ^{13}C-BDE-209 (Cambridge Isotope Laboratories, Inc., Tewksbury, MA, USA), DE-TOX 409 (LGC Standards GmbH, Wesel, Germany), 4-OH-$^{13}C_{12}$-PCB 187, 4′-OH-$^{13}C_{12}$-PCB159, ^{13}C-labeled PFAS equivalents (Wellington Laboratories, Ontario, Canada). Individual OHC concentrations were calculated from six-point linear calibration curves of internal standards and chromatographic data was analyzed with MSD ChemStation (Version E.02.01 and B.04.03, Agilent Technologies) and Mass Hunter (Agilent Technologies).

According to accreditation, each analytical series included one non-spiked sample (cattle blood), two spiked samples for recovery (cattle blood), three laboratory blanks (solvent), one harp seal (*Pagophilus groenlandicus*) blubber as internal reference material (control for organochlorine pesticides (OCPs), PCBs and BFRs) and one minke whale

(*Balaenoptera acutorostrata*) blubber (control for toxaphenes). In addition, the analytical quality was approved by satisfactory participation in several international ring tests.

The limits of detection (LOD) were defined as three times signal-to-noise ratio. For most of the contaminants, the relative recovery rates were within the acceptable range of 70–130%. Some compounds were outside this range (*p,p'*-DDT, *c*-heptachlor epoxide, oxychlordane, Mirex, 4-OH-PCB187 and PFUnA), and are reported uncorrected for recovery. For details on LOD and relative recovery rates of individual OHCs, see Table S3a. Unless otherwise specified, all OHC concentrations are expressed as ng/g wet weight (ww).

2.4. Stable isotope analyses

Stable isotopes of carbon ($^{13}C/^{12}C$) and nitrogen ($^{15}N/^{14}N$) were analyzed at the Institute for Energy Technology, Kjeller, Norway. Dried whole-blood (~2 mg) was packed into tin cups and combusted at 1700 °C to carbon dioxide (CO_2) and nitrogen gas (NO_x) with a ThermoQuest NCS 2500 elemental analyzer (EA; Thermo instrument systems Inc., Waltham, USA). CO_2 and NO_x were separated and detected on the basis of mass with a Micromass Optima Isotope Ratio Mass Spectrometer (IRMS) (Micromass, Manchester, UK). The internal standard used was trout (*Salmo trutta*). The proportion of stable isotopes in the sample (R_{sample}) relative to international standards ($R_{standard}$), Vienna Peedee belemnite (PDE) carbonate for $\delta^{13}C$ and atmospheric nitrogen (AIR) for $\delta^{15}N$, respectively, were expressed as δ values in parts per thousand (‰), (Equation (1)).

$$\delta X = (R_{sample}/R_{standard} - 1) \times 1000 \quad (1)$$

2.5. Body condition index

Body condition index (BCI) was calculated based on the scaled mass index method (Peig and Green, 2009), where a predicted body mass at a fixed body size (wing chord length) in the sample population was calculated for each individual bird. As females are larger than males (Bustnes et al., 2007), BCI was calculated separately between the two sexes. As the mean sex-specific body mass did not differ between the seasons, BCI was not calculated separately for each breeding season (see Supporting Information).

2.6. Data treatment and statistical analysis

Statistical analyses were carried out using the program R version 3.5.1 (R Core Team, 2013). Organochlorine results from the 2013 breeding season (n = 30) were combined with the published results from the 2001/2002 season (hereafter referred to as 2001) (n = 142) (Bustnes et al., 2006). As the 2001/2002 data included mainly nesting pairs, one individual from each breeding pair was randomly selected and included in the final dataset, thus reducing sample size to 71. Of the 30 south polar skuas sampled in 2013/2014, most were from different nests, except 5 pairs. In addition eight individuals were recaptured individuals from 2001/2001 (Bustnes et al., 2006).

Individual contaminants quantified below LOD in more than 25% of the sample population were excluded from further statistical analysis (for details on excluded contaminants, see Table S2 and S4 of the SI). For the remaining contaminants, only five values (from 2001/2002) were reported as non-detects (0.3% of the dataset). Contaminant concentrations in these samples were replaced with a random value between 0 and LOD, using the most conservative LOD (i.e. highest) between the breeding seasons (Table S3b of the SI). For contaminants quantified only in 2013/2014, 32 non-detects were replaced by random values (2.4% of the entire 2013 dataset). Contaminant concentrations were $\log_{10}$-transformed prior to data analysis to reduce skewness and heteroscedastic variance.

Multivariate statistics were conducted using the package "vegan" in R (Oksanen et al., 2007). Principal component analysis (PCA) was performed separately on total OHC concentration and patterns during 2013/2014 (the relative contribution in % of individual contaminants to ∑OHCs) to identify highly correlated compounds groups and to explore differences in contaminant accumulation between sexes and breeding seasons (2001/2002 vs. 2013/2014). As lipid content significantly affected the overall structure in the OHC concentration data, lipid content treated as a covariate to analyze the interannual differences in contaminant status beyond variations in lipid content. Redundancy analysis (RDA) was conducted on both concentration and pattern data to test the explanatory power of relevant biological variables (sex, breeding season, BCI, $\delta^{15}N$, $\delta^{13}C$ and sampling date) that might account for contaminant variation among individuals. Where applicable, explanatory variables were added stepwise in a forward selection process.

For the analyses of contaminant concentrations within 2013/2014 breeding season, linear regression was used to compare the relationship between PC1 axis (the axis containing the most variation) and BCI (the only variable identified significant with RDA). As PFAS did not vary along the PC1 axis (see Results and Discussion), linear regression models were used to assess the relationship between $\log_{10}$-transformed PFAS concentration and relevant biological variables (sex, breeding season, BCI, sampling date etc.).

3. Results and discussion

3.1. Occurrence of organohalogen contaminants in 2013/2014-sampled south polar skuas

Of the 87 individual OHCs analyzed in the south polar skua whole blood from 2013/2014, 41 were quantified above the LOD in more than 75% of the samples (Table S4). Organochlorine pesticides (OCPs) dominated the OHC pattern, representing 62% of ∑OHCs, followed by ∑PCBs (19%) then ∑PFAS (16%) (Fig. 1a). Mirex was the most abundant OCP and contributed to 50% of ∑OCPs. The concentrations of the individual dominating compounds, Mirex (21.2 ng/g ww), HCB (9.1 ng/g ww), *p,p'*-DDE (8.8 ng/g ww), PFOS (5.1 ng/g ww) were higher than or similar to other south polar skua colonies in the Antarctic (Tables 1 and 2). BFR levels were low (∑BFRs 0.07 ng/g ww), and were similar to other colonies, whereas all PCB congeners (∑PCBs 13.3 ng/g ww) with the exception of PCB-180, were lower in the 2013 Svarthamaren south polar skuas than in other Antarctic colonies (Table 2). BFRs and OH-metabolites (OH-PCBs and OH-PBDEs) contributed only 0.1% and 1% to ∑OHCs, respectively. In previous studies, HCB, *p,p'*-DDE and PCBs were the dominating contaminants in Antarctic biota, including permanent Antarctic residents such as Adelie penguins (*Pygoscelis adeliae*) (Cipro et al., 2013; Colabuono et al., 2015; Corsolini, 2009), and migrating species such as southern giant petrel (*Macronectes giganteus*) and wandering albatrosses (*Diomedea exulans*) (Carravieri et al., 2014; Colabuono et al., 2016).

3.2. Drivers of organohalogen contaminant concentrations in 2013/2014-sampled south polar skuas

The multivariate ordination of contaminant concentrations separated lipid-soluble compounds (OCPs, PCBs and BFRs) from protein-associated compounds (PFASs) along the PC1 and PC2 axes, respectively (Fig. 2). The predominantly protein-associated OH-metabolites (OH-PCBs and OH-PBDE) were typically loading onto the PC1 axis, along with their lipophilic parent compounds, rather than onto PC2 along with the protein-associated PFAS. As 62% of the total variation was captured by PC1, and 14% by PC2, there was a greater variability among the skuas in lipophilic contaminant concentrations and their metabolites than in protein-associated contaminants, even with lipid content as a covariable in the analyses. When removing the effect of lipid content by treating it as a covariable, overall OHC concentrations

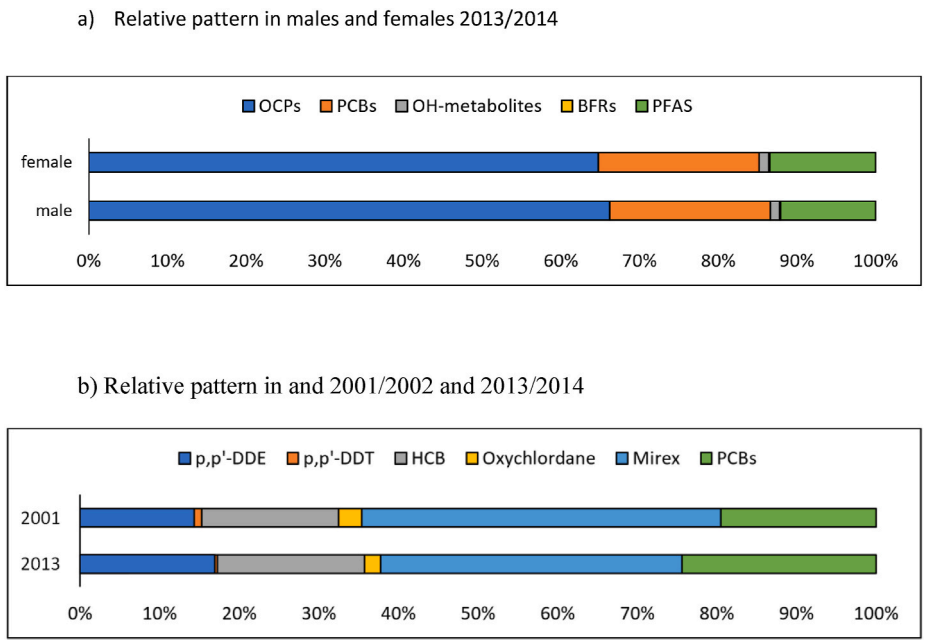

Fig. 1. Mean relative pattern of organohalogen contaminants in south polar skuas (Catharacta maccormicki) breeding in Svarthamaren, Antarctica in a) female (n = 13) and male (n = 17) from 2013/2014; and b) comparing November–January 2001/2002 (n = 71) and December–January 2013/2014 (n = 30). Values are calculated and presented as the relative contribution of each individual contaminant (%) to its respective contaminant group.

Table 1
Body Condition Index (BCI), lipid content (%), stable isotopes of carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$), and organohalogen contaminant concentrations (ng/g ww, whole blood) in female (♀) and male (♂) south polar skuas (Catharacta maccormicki) breeding at Svarthamaren, Antarctica from November–January 2001/2002 (♀ = 36, ♂ = 35, Bustnes et al., 2006) and December–January 2013/2014 (♀ = 13, ♂ = 17, present study). Note that $\delta^{13}C$, $\delta^{15}N$, BFR OH-metabolites and PFAS were not analysed (-) in 2001/2002 individuals.

		South polar skua 2001/2002 (*n* = 71)			South polar skua 2013/2014 (*n* = 30)	
		mean ± SD	min - max		mean ± SD	min - max
BCI	♀	1357 ± 60	1119–1457	♀	1344 ± 68	1221–1430
	♂	1338 ± 66	1170–1483	♂	1285 ± 85	1144–1417
LIPID (%)	♀	0.44 ± 0.32	0.01–1.74	♀	0.30 ± 0.08	0.11–0.40
	♂	0.40 ± 0.20	0.03–0.85	♂	0.25 ± 0.08	0.10–0.36
$\delta^{13}C$ (‰)	♀	–	–	♀	−24.3 ± 0.1	−24.4 to −24.1
	♂	–	–	♂	−24.3 ± 0.3	−25.3 to −23.8
$\delta^{15}N$ (‰)	♀	–	–	♀	12.0 ± 0.4	11.6–12.9
	♂	–	–	♂	12.0 ± 0.2	11.7–12.3
MIREX	♀	18.0 ± 10.9	1.0–53.9	♀	16.9 ± 12.4	4.8–47.2
	♂	23.7 ± 14.6	1.8–63.6	♂	24.4 ± 16.5	9.7–71.7
HCB	♀	6.8 ± 3.3	0.6–16.6	♀	8.1 ± 3.5	2.0–14.6
	♂	7.7 ± 4.0	1.0–21.2	♂	9.8 ± 4.9	5.1–22.0
P,P'-DDE	♀	6.1 ± 4.5	0.6–22.6	♀	7.5 ± 4.5	2.1–16.2
	♂	7.4 ± 7.0	0.6–40.9	♂	9.7 ± 4.7	4.2–19.7
OXYCHLORDANE	♀	1.2 ± 0.9	0.2–4.3	♀	0.8 ± 0.4	0.4–1.8
	♂	1.4 ± 0.7	0.2–3.7	♂	1.2 ± 0.7	0.5–3.0
∑OCPS[a]	♀	32.52 ± 18.76	2.34–91.60	♀	33.55 ± 19.25	9.22–68.66
	♂	40.47 ± 23.73	3.82–114.91	♂	45.37 ± 25.84	20.96–116.87
PCB-153	♀	2.0 ± 1.7	0.3–9.1	♀	2.9 ± 1.6	1.3–6.6
	♂	2.2 ± 1.8	0.3–9.8	♂	4.0 ± 2.1	1.6–9.4
∑PCBS[b]	♀	8.13 ± 6.38	0.99–32.97	♀	10.77 ± 6.68	4.45–27.92
	♂	9.35 ± 6.78	1.53–35.22	♂	14.38 ± 7.92	5.59–35.46
∑BFRS[c]				♀	0.06 ± 0.04	0.02–0.16
				♂	0.08 ± 0.04	0.03–0.17
∑OH-PCB/PBDES[d]				♀	0.73 ± 0.19	0.36–1.01
				♂	0.62 ± 0.21	0.27–0.96
∑PFAS[e]				♀	6.25 ± 1.35	3.62–8.32
				♂	7.38 ± 1.72	4.31–11.16

[a] ∑OCP: sum of p,p'-DDE, p,p'-DDT, HCB, oxychlordane and Mirex.
[b] ∑PCBs: sum of PCB-99, -105, −118, −118, −137, −138, −153, −156, −170, −180, −187, −197, −196 and −206.
[c] ∑BFRs: sum of BDE-153, BDE-154 and PBT.
[d] ∑OH-metabolites: sum of OH-PCB-146, OH-PCB-187 and OH-BDE-47.
[e] ∑PFAS: sum of PFNA, PFDA, PFUdA, PFDoDA, PFTrDA, PFHxS and PFOS.

Table 2
Reported or estimated mean or range concentrations of organohalogen compounds (OHCs) in various matrices of high trophic level seabirds in Antarctica. Unless otherwise specified, concentrations are given in ng/g and lipid weight (lw) basis. Wet weight is denoted as ww and dry weight dw.

Species	Sampling year	Locality	Matrix	*p,p'*-DDE	HCB	Mirex	PCB-153	PCB-180	∑PCBs	∑PBDEs	PFOS	Reference
South polar skua *Catharacta maccormicki*	20013/2014	Svarthamaren	Whole blood	3542	3554	8467	1441	1507	5374[a]	12.3	5.08 (ww)	Present study
–	2016/2017	Adelaide Island	Muscle	~10000	~1000		~1000	~1000	2000–3000[b]	~10–15		Krasnobaev et al. (2018)
–	2003	Adélie Land	Plasma						1604–128089[c]			Tartu et al. (2015)
–	2011/2012	–	Plasma (chick)	400 (dw)	900 (dw)							Carravieri et al. (2017)
–	2013	–	Plasma								~6 (ww)	Munoz et al. (2017)
–	1995–1999	Edmonson Point	Whole blood								0.88 (ng/mL)	Tao et al. (2006)
–	2010–2012	King George Island	Egg	1311	133			477	1670[d]	66.3		Mello et al. (2016)
Brown skua *Catharacta antarcticus*	2011/2012	South Shetland Islands	Egg	1230	2389	5336			4354[e]	2.9		Colabuono et al. (2015)
Northern giant petrel *Macronectes halli*	2009–2011	Marion Island	Plasma	20.1 (ww)	4.75 (ww)		9.81 (ww)		30.7 (ww)	0.0794 (ww)		Roscales et al. (2016)
Southern giant petrel *Macronectes giganteus*	2009–2011	Livingston Island	Plasma	0.4 (ww)	2.66 (ww)		1.89 (ww)		6.1 (ww)	0.0203 (ww)		–
–	2009–2010	–	Plasma								<3 (ww)	Roscales et al. (2019)
–	2012	Elephant Island	Whole blood		4.19 (ww)	8.84 (ww)			5.96 (ww)	<0.003 (ww)		Colabuono et al. (2016)
Laysan Albatrosses *Phoebastria immutabilis*	1992–1996	Southern Ocean	Plasma								3.2 (ng/ml)	Tao et al. (2006)
Wandering Albatrosses *Diomedea exulans*	2007/2008	Crozet Islands	Plasma	1138	357	234	715	217	4705[f]	97		Carravieri et al. (2014)

[a] Sum of PCB-99, -105, −118, −118, −137, −138, −153, −156, −170, −180, −187, −197, −196 and −206.
[b] Sum of PCB-28, -52, −101, −105, −114, −118, −123, −138, −153, −156, −157, −167, −180, and −189. Data estimated from figure (log_{10}-scale).
[c] Sum of PCB-28, -52, −101, −118, −138, −153 and −180.
[d] Sum of PCB-28, -52, 95, −101, −105, −114, −118, −123, −138, −153, −156, −157, −180 and −189.
[e] Sum of PCB-8, -18, −8, −18, −28, −31, −33, −44, −49, −52, −56, −60, −66, −70, −74, −77, −81, −87, −95, −97, −99, −101, −105, −110, −114, −118, −123, −126, −128, −132, −138, −141, −149, −151, −153, −156, −157, −158, −167, −169, −170, −174, −177, −180, −183, −187, −189, −194, −195, −201, −203, −206 and −209.
[f] Sum of PCB-50/28, −52, −101, −118, −153, −138, −180.

increased with decreasing body condition index, accounting for 17% of the constrained variation in pollutant concentrations (RDA: $F_{1,27} = 5.50$, $P < 0.01$; Figs. 2 and 3a). However, relative trophic position ($\delta^{15}N$), carbon source ($\delta^{13}C$), sex or sampling day did not contribute to explain the contaminant concentration variation among the individuals (sex: RDA variation = 8.1%, $F_{1,27} = 2.38$, $P = 0.09$; $\delta^{13}C$: RDA variation = 6.8%, $F_{1,27} = 1.96$, $P = 0.13$; Sampling day: RDA variation = 4.8%, $F_{1,27} = 1.36$, $P = 0.22$; $\delta^{15}N$: RDA variation = 1.0%, $F_{1,27} = 0.30$, $P = 0.89$). Both $\delta^{13}C$ and $\delta^{15}N$ varied little among the skuas ($\delta^{13}C$: females = −24.3 ± 0.1, males: 24.3 ± 0.3; $\delta^{15}N$: females = 12.0 ± 0.4; males = 12.0 ± 0.2; Table 1), as Svarthamaren south polar skuas remain in the colony throughout the breeding season, feeding exclusively on Antarctic petrel eggs and chicks (Brooke et al., 1999; Buskedier et al., 2020).

To our knowledge, this is the first study to report PFAS concentrations in wildlife from Dronning Maud Land. Similar to other Antarctic wildlife (Giesy and Kannan, 2001; Tao et al., 2006), mean ∑PFAS concentration was low (11 ng/g ww; Table 2). PFAS concentrations in the skuas increased during the breeding season (effect of sampling date on [PFAS]: $R^2 = 0.13$, $P = 0.049$; Fig. 3b). Body condition index was lower in males than in females in 2013/2014 ($t_{69} = -2.05$, $P = 0.05$), however, body condition did not affect the PFAS concentrations, and there was also no interaction between body condition and sampling day together on PFAS concentration (body condition: $t_{26} = -1.13$, $P = 0.27$; sampling day: $t_{26} = -0.38$, $P = 0.71$; body condition*time: $t_{26} = 0.49$, $P = 0.63$). Sexes were evenly distributed across the breeding season ($t_{28} = 0.68$, $P = 0.50$), suggesting sex was unrelated to sampling date. The higher PFAS concentrations in skuas sampled later in the breeding season imply either changes in local food sources (i.e. in Antarctic petrels) or mobilization from body reserves along with decreasing body condition during the breeding period. However, the egg laying dates ranged from first egg laying on 1 December 2013 to last 20 January 2014, and there was no relationship between blood sampling date of the adults and the estimated hatching date of the specific nest ($R^2 = 0.0001$, $F_{1,26} = 0.004$, $P = 0.95$) (Table S1). If increased remobilization of contaminants from utilized energy reserves affected the measured contaminants in blood, we expect that this would lead to a greater increase in lipid soluble contaminants as lipids is a more important energy source than proteins (and hence protein associated contaminants such as

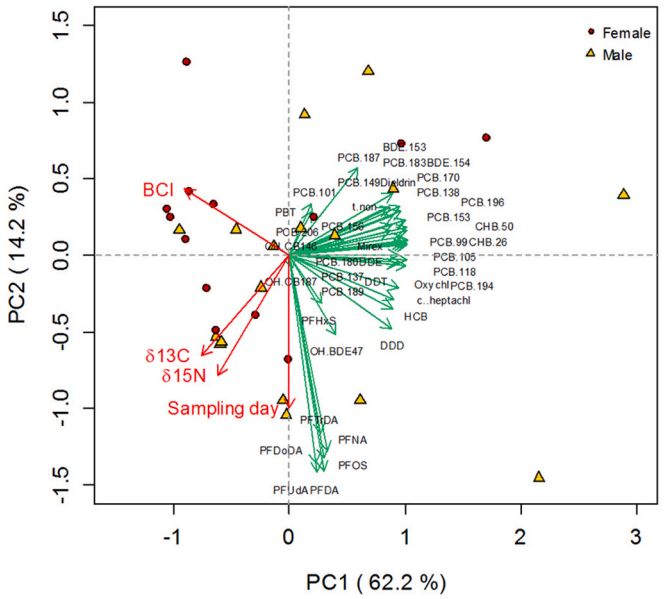

Fig. 2. Principal component analyses diagram of log_{10}-transformed organohalogen compound concentrations (ng/g ww) in female and male south polar skuas (Catharacta maccormicki) breeding at Svarthamaren (Antarctica) in December–January 2013/2014 (♀ = 13, ♂ = 17). Lipid content was included as covariable. Response variables (contaminants) are represented with green arrows, explanatory variables have been passively projected with red arrows. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

PFAS). Furthermore, there was no relationship between body condition and PFAS concentration ($R^2 = 0.09$, $P = 0.09$). There are no known local sources of PFAS exposure at Svarthamaren other than diet. However, no data are available on PFAS in Antarctic petrels from the area.

Skua males had higher PFAS concentrations than females, except for PFHxS (Table 1). Higher PFAS concentrations in males than females is in agreement with other studies on south polar skua colonies in other colonies (Cipro et al., 2013), and may be due to maternal elimination of PFAS via egg laying, or differences in feeding ecology between the sexes (Bustnes et al., 2008a, 2017). However, this later explanation is not supported by previous observations showing that females and males in Svarthamaren both feed almost exclusively on Antarctic petrels from the same colony during the entire breeding season (Brooke et al., 1999). This is also supported by similar stable isotope values ($\delta^{13}C$ and $\delta^{15}N$) between the sexes. Previous studies have shown that PFAS is typically higher in male than female seabirds, but with lower differences than for lipophilic contaminants (Hitchcock et al., 2019). Our study, however, did not find sex-specific differences for lipophilic contaminants.

3.3. Drivers of organohalogen contaminants pattern in 2013/2014-sampled south polar skuas

The multivariate ordination of the total OHC pattern showed that birds caught later in the breeding period had higher relative proportion of PFAS (Fig. 2), but a lower relative proportion of PCBs and PBDEs (RDA variation = 11.7%, $F_{1,28} = 3.69$, $P < 0.01$; Fig. S4). A reduction in relative proportion might reflect a faster elimination of these lipid soluble contaminants, and/or that the relative proportion in the diet and accumulation was low, compared to other compounds such as OCPs or PFAS. Changes in the relative proportion of OHCs over time may also reflect a shift from distant food sources to local ones, e.g. Antarctic petrels. Similar decrease of relative proportion of PCBs during the breeding period was also found in lesser black-backed gulls (*Larus fuscus*) from the coast of Norway (Bustnes et al., 2008b). However, little is currently known of different migration routes and wintering sites between females

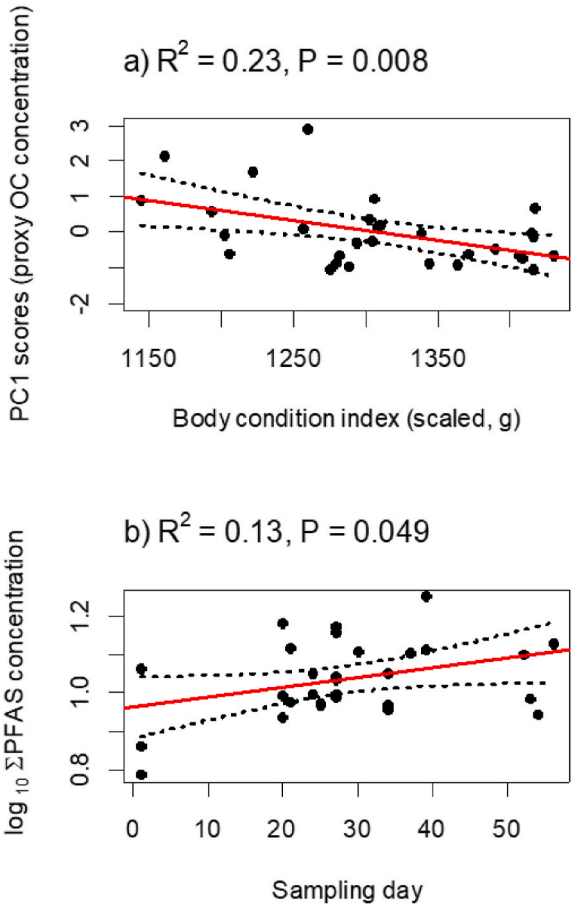

Fig. 3. Relationship between a) Principal component x-axis 1 scores as a proxy for organochlorine (OC) concentration and body condition index; and b) ΣPFAS (ng/g ww) and sampling day in December–January 2013/2014 south polar skuas (Catharacta maccormicki) breeding at Svarthamaren, Antarctica. Line of fit and confidence intervals have been indicated including summary statistics.

and males. To assess the sources of OHCs in south polar skuas from breeding and wintering grounds, future studies should address in the local food source (Antarctic petrels), contaminant and stable isotope analysis of primary feathers grown during the winter, as well as use of Global Location Sensor (GLS) loggers to confirm wintering habitats of recovered birds (Leat et al., 2019; Magnusdottir et al., 2012).

For all contaminants, except OH-metabolites, there was a slight difference in pattern (<2% for each contaminant) between males and females (Fig. 1a and S3). For OH-metabolites, females had a higher proportion of OH-PCB146 and OH-PCB187 than males (1.2% and 10% higher, respectively), whereas OH-BDE47 was higher in males (Fig. S3). While differences were small, previous studies have shown that contaminant deposition through egg laying is typically greater for lipid-soluble contaminants than protein-associated ones, leading to skewed concentration ratios between sexes (i.e. higher in male). (Hitchcock et al., 2019).

3.4. Higher organochlorine levels in 2013/2014 than in 2001/2002

When comparing pollutant concentrations between the breeding seasons 2001/2002 and 2013/2014 individuals, breeding season, body condition and sex combined explained 29% of the total variation in contaminant concentrations (Sex + Breeding season + BCI: $F_{3,96} = 12.9$, $P < 0.001$). Variation in lipid adjusted contaminant concentrations between 2001/2002 and 2013/2014 was likely driven by an increase in Mirex (39%), HCB (63%), *p,p'*-DDE (87%), ΣPCBs (105%) and a decrease in *p,p'*-DDT (53%). Note however that the recovery of Mirex

and *p,p'*-DDT in 2013/2014 was 157% and 146%, respectively, and thus outside of the 70–130% range. With the exception of PCB- 105 (decrease by 10%), all PCBs increased with percentages ranging from 9% (PCB-206) to 174% (PCB-187). When the effect of each explanatory variable was tested alone with other variables included as covariables, breeding season alone explained 16%, BCI 8% and sex 4% of the total variation in contaminant concentrations (Breeding season: $F_{1,96} = 18.92$, $P < 0.001$; BCI: $F_{1,96} = 8.0$, $P < 0.003$; Sex: $F_{1,96} = 3.95$, $P = 0.03$). In brief, when controlling for other variables, the contaminant concentrations were generally higher in 2013/2014 than in 2001/2002, and concentrations increased with decreasing body condition index, where males had overall higher concentrations of contaminants than females (Fig. 4). Male body condition was poorer in 2013 than in 2001 ($t_{50} = -2.48$, $P = 0.02$), whereas the females had a similar BCI during the two breeding seasons ($t_{47} = 0.68$, $P = 0.50$). Males also had a lower body condition than females in 2013 ($t_{69} = -2.05$, $P = 0.05$), but not in 2001 ($t_{69} = -1.30$, $P = 0.20$). In Arctic breeding seabirds, contaminant concentrations increase with decreasing body condition (Bustnes et al., 2012; Helberg et al., 2005; Tartu et al., 2015).

The dominating PCB congeners increased in concentrations from 2001/2002 to 2013/2014 (Fig. S2 and S4). These included PCB-138, -153, −183, and −187, which are among the most persistent congeners with relatively low potential for long range transport compared to more volatile substances (Borgå et al., 2005), and with a high potential for biomagnification (Borgå et al., 2004). The Svarthamaren south polar skuas migrate north to overwinter in three main areas in the Indian Ocean: the Mozambique Channel, Seychelles area, the Bay of Bengal/-coastal Sri Lanka (Weimerskirch et al., 2015). In recent years, increasing PCB concentrations in the abiotic environment have been found in coastal zones and metropolitan cities in both Africa and India (Chakraborty et al., 2016; Gioia et al., 2014). Elevated PCB concentrations in these areas may result from disposal, recycling and leaching from PCB-containing waste, such as shipwrecks and electronic equipment (Chakraborty et al., 2016). Higher PCB concentrations in 2013/2014

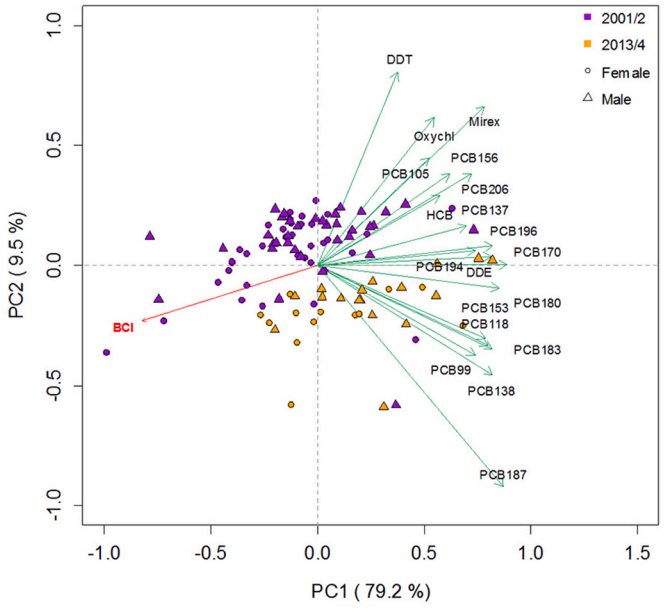

Fig. 4. Principal component analyses diagram of $\log_{10}$-transformed organochlorine contaminant concentrations (ng/g ww in female and male south polar skuas (Catharacta maccormicki) from November–January 2001/2002 (♀ = 36, ♂ = 35, Bustnes et al., 2006) and December–January 2013/2014 (♀ = 13, ♂ = 17, present study)breeding at Svarthamaren, Antarctica. For concentration, lipid content was included as covariable. Response variables (contaminants) are represented with green arrows, explanatory variables have been passively projected with red arrows. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

than 2001/2002 may thus be due to dietary exposure while skuas are the wintering grounds. However, PCB levels were low in fish from the Tanzanian coast (Mwakalapa et al., 2018), and chlorinated, brominated and fluorinated organohalogen levels were low in seabird eggs from western Indian Ocean islands (Bouwman et al., 2012; van der Schyff et al., 2020, 2021, 2021), suggesting background levels. Thus, if winter grounds are the source of contamination in the skuas, they are likely to feed in areas more affected by organohalogen-containing waste.

The skuas from 2001/2002 contained a higher relative proportion of *p,p'*-DDT and Mirex, whereas 2013/2014 skuas contained a higher relative proportion of *p,p'*-DDE (Fig. 1b). For DDT, differences between breeding seasons are most likely due to both the restricted usage of DDT and the metabolic degradation of *p,p'*-DDT to *p,p'*-DDE in the environment and biota, with diet in wintering areas representing a major contaminant source. In both breeding seasons, the ratio *p,p'*-DDE/*p,p'*-DDT was greater than one, indicating that *p,p'*-DDT in south polar skua most likely originate from old weathered DDT sources (Kallenborn et al., 2013; Zhang et al., 2015). Recovery adjustment of the p,p'-DDT concentrations would enforce these findings.

Seven of the eight individual birds sampled in both 2001/2002 and 2013/2014 had higher concentrations in 2013/2014 than in 2001/2002 (Fig S6). In 2013/2014, the eight recaptured birds were representative of the contaminant concentrations and patterns in the remainder of the 2013/2014 sampled population. Although we do not know the specific age of the birds, this finding suggests that the OHC occurrence is similar across the ages of adult south polar skua. This is in line with results from other species, although few studies have recaptured and sampled the same individuals across multiple seasons.

3.5. South polar skuas compared with other Antarctic avian species

Comparisons of OHC concentrations across Antarctic regions and species are limited due to varying matrices, geographic locations, sample sizes and contaminants of concern. Moreover, interspecies comparisons can be confounded by differences in biotransformation, which may depend on phylogeny (Borgå et al., 2005). We compared the south polar skua from Svarthamaren to other Antarctic and sub-Antarctic breeding seabirds at similar ecological niches including the south polar skua from Dumont d'Urville (Munoz et al., 2017), southern giant petrel (*Macronectes giganteus*) (Colabuono et al., 2016; Roscales et al., 2016, 2019), northern giant petrel (*Macronectes halli*) (Roscales et al., 2016), Laysean albatross (*Phoebastria immutabilis*) (Tao et al., 2006), and wandering albatrosses (*Diomedea exulans*) (Colabuono et al., 2016) (Table 2). Overall, concentrations of HCB, Mirex and PFOS were higher in south polar skua at Svarthamaren (present study), whereas *p,p'*-DDE, PCB-153 and ∑PCBs were lower, and ΣPBDEs were lowest. This indicates that the sources of OHCs in south polar skuas from Svarthamaren may differ compared to other Antarctic bird colonies, and are probably related to the location of overwintering areas, where primary sources and historic long range transport of contaminants are contributing factors.

Elevated HCB concentration could be due to long range atmospheric transport of the chemical from areas where there was historic usage of industrial chemicals and fungicides in South America or Africa (Department of the Environment and Heritage, 2006; Gerber et al., 2016). This is because the high vapor pressure and partitioning coefficient of HCB enable high atmospheric mobility, deposition and persistence at polar latitudes (Bailey, 2001; Weber and Goerke, 2003). Mirex was continuously used to combat ants and termites in South Africa and China until late 1990s, and until late 2000s in South America and Australia (Connell et al., 1999; Department of the Environment and Heritage, 2006). However, contrary to HCB, the atmospheric transportation of Mirex is reduced as it quickly sorbs to aerosols and particles in the lower troposphere and deposited in aquatic sediment (Scheringer et al., 2000). Hence, food web exposure in winter areas in Indian Ocean near the African continent are more likely sources for Mirex (Department of the Environment and Heritage, 2006; Weimerskirch et al.,

2015).

Studies of endemic Antarctic species show relatively low PFAS levels in the Antarctic food web (Giesy and Kannan, 2001; Tao et al., 2006). Hence, the higher concentrations of PFASs in Svarthamaren south polar skua in the present study are likely due to their migratory habits, in addition to their high trophic position. However, the finding of increasing PFAS levels during breeding season motivates future analyses of PFAS in the local breeding site diet, the Antarctic petrel.

4. Conclusion

The concentrations of several individual OHCs increased from the 2001/2002 study to the 2013/2014 breeding season, which was contrary to our expectations. OHCs which increased in concentration included PCBs (105%), HCB (63%), and p,p'-DDE (87%). The higher levels of PCBs with high persistence and low mobility in 2013/2014 than 2001/2002, suggest that exposure during winter migration to the Indian Ocean and surrounding countries during the non-breeding season is the dominating source to contaminants in this migrating species. South polar skua males with poorer condition also had higher pollutant concentrations.

Contrary to expectations, concentrations of lipid soluble OHCs did not vary during the 2013/2014 breeding season, whereas the protein soluble PFAS increased. The dietary signal of carbon source ($\delta^{13}C$) and relative trophic position ($\delta^{15}N$) showed low variance during the breeding season, reflecting that Antarctic petrel is the dominant food source during breeding. To better understand contaminant exposure in south polar skua, further studies should elucidate the effects of wintering grounds and winter diet, as well as the contaminant status in Antarctic petrel.

Author statement

Hilde Karin Midthaug: Formal analysis, Investigation, Writing - Original Draft. Daniel J. Hitchcock: Formal analysis, Writing - Review & Editing. Jan Ove Bustnes: Conceptualization, Resources, Writing - Review & Editing, Supervision, Funding acquisition. Anuschka Polder: Validation, Investigation, Writing - Review & Editing, Supervision. Sébastien Descamps: Conceptualization, Investigation, Resources, Writing - Review & Editing. Arnaud Tarroux: Conceptualization, Investigation, Writing - Review & Editing. Eeva M. Soininen: Investigation, Writing - Review & Editing. Katrine Borgå: Conceptualization, Writing - Review & Editing, Supervision, Project administration.

Declaration of competing interest

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

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