Chemosphere 355 (2024) 141721

Contents lists available at ScienceDirect

Chemosphere

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

Can plastic related chemicals be indicators of plastic ingestion in an Arctic seabird?

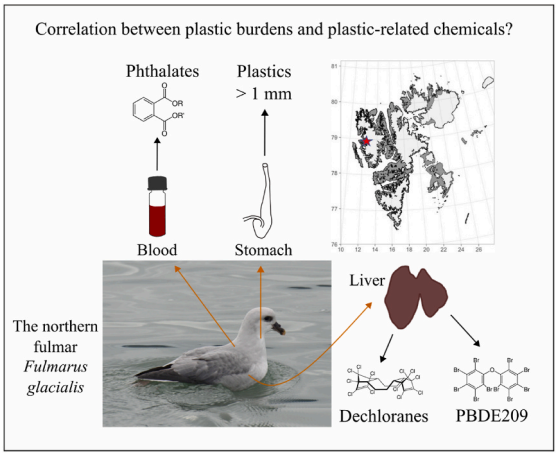
France Collard [a,b,1,*], Felix Tulatz [a,1], Mikael Harju [c], Dorte Herzke [c], Sophie Bourgeon [d], Geir W. Gabrielsen [a]

[a] Norwegian Polar Institute (NPI), Fram Centre, N-9296, Tromsø, Norway
[b] Norwegian Institute for Water Research (NIVA), Fram Centre, N-9296, Tromsø, Norway
[c] The Climate and Environmental Research Institute (NILU), Fram Centre, N-9296, Tromsø, Norway
[d] Department of Arctic and Marine Biology, The Arctic University of Norway (UiT), N-9037, Tromsø, Norway

HIGHLIGHTS

- Plastic-related chemicals were measured in the fulmar *Fulmarus glacialis*.
- PBDE209 was detected in 28% of fulmars' liver with an average of 1.36 ng/g.
- Dechloranes were detected in all livers with an average of 0.36 ng/g.
- Detectable phthalate concentrations were found in 33% of plasma samples.
- No significant positive correlation between any contaminants and plastic burdens.

GRAPHICAL ABSTRACT



ARTICLE INFO

Handling editor: Michael Bank

Keywords:
Polybrominated diphenyl ether
Fulmar
Fledgling
Phthalate
Microplastic
Svalbard

ABSTRACT

For decades, the northern fulmar (*Fulmarus glacialis*) has been found to ingest and accumulate high loads of plastic due to its feeding ecology and digestive tract morphology. Plastic ingestion can lead to both physical and toxicological effects as ingested plastics can be a pathway for hazardous chemicals into seabirds' tissues. Many of these contaminants are ubiquitous in the environment and the contribution of plastic ingestion to the uptake of those contaminants in seabirds' tissues is poorly known. In this study we aimed at quantifying several plastic-related chemicals (PRCs) -PBDE209, several dechloranes and several phthalate metabolites- and assessing their relationship with plastic burdens (both mass and number) to further investigate their potential use as proxies for plastic ingestion. Blood samples from fulmar fledglings and liver samples from both fledgling and non-fledgling fulmars were collected for PRC quantification. PBDE209 and dechloranes were quantified in 39

* Corresponding author. Norwegian Institute for Water Research (NIVA), Fram Centre, N-9296, Tromsø, Norway.
E-mail address: france.collard@niva.no (F. Collard).
[1] Both authors worked equally on this study and manuscript, and share first co-authorship.

https://doi.org/10.1016/j.chemosphere.2024.141721
Received 12 December 2023; Received in revised form 12 March 2024; Accepted 13 March 2024
Available online 22 March 2024

and 33 livers, respectively while phthalates were quantified in plasma. Plastic ingestion in these birds has been investigated previously and showed a higher prevalence in fledglings. PBDE209 was detected in 28.2 % of the liver samples. Dechlorane 602 was detected in all samples while Dechloranes 601 and 604 were not detected in any sample. Dechlorane 603 was detected in 11 individuals (33%). Phthalates were detected in one third of the analysed blood samples. Overall, no significant positive correlation was found between plastic burdens and PRC concentrations. However, a significant positive relationship between PBDE209 and plastic number was found in fledglings, although likely driven by one outlier. Our study shows the complexity of PRC exposure, the timeline of plastic ingestion and subsequent uptake of PRCs into the tissues in birds, the additional exposure of these chemicals via their prey, even in a species ingesting high loads of plastic.

1. Introduction

Northern fulmars (*Fulmarus glacialis*), hereafter called fulmars, are known to ingest and accumulate high quantities of marine plastic (Mallory, 2008; Trevail et al., 2015; e.g. Baak et al., 2020; Collard et al., 2022a; van Franeker et al., 2022; Tulatz et al., 2023). Fulmars feed opportunistically at the sea surface (Hobson and Welch, 1992) and are incapable of regurgitating food -or hard items-once they reached the gizzard (e.g. Furness, 1985), making them prone to accumulating plastics. Therefore, this species is used as a bioindicator for marine plastic pollution by the Convention for the Protection of the Marine Environment of the North-East Atlantic (OSPAR Commission, 2008). OSPAR has established guidelines for monitoring plastic burdens in fulmars and has defined a long-term Ecological Quality Objective (EcoQO) to qualify the health status of the marine ecosystem regarding plastic pollution (OSPAR Commission, 2015; van Franeker et al., 2021). The Arctic Monitoring and Assessment Programme (AMAP) placed the fulmar as priority recommendation for monitoring of litter and microplastics in the Arctic (AMAP, 2021).

Plastic contains many chemicals that can leach out and reach the organism's tissues (Teuten et al., 2009; Andrady and Rajapakse, 2016). Plastic related chemicals (PRCs) could then be used as proxies for plastic ingestion or exposure. For this purpose, tissue concentrations of plastic additives have been investigated (e.g.Tanaka et al., 2020). In fulmars, PRCs were found to be leaching from the ingested plastic pieces into different tissues (Tanaka et al., 2013, 2020; Neumann et al., 2021) and/or absorbed from their prey (Fjeld et al., 2004). It was demonstrated that stomach oil of fulmars is a potent absorbent for several PRCs, including the phthalate DEHP (Kühn et al., 2020). Due to their long lifespan and high trophic level, fulmars can accumulate a broad range of PRCs in their tissues (Burger and Gochfeld, 2004; Mallory, 2006), reported to induce negative health effects (Guigueno and Fernie, 2017; Mortensen et al., 2022). Among the flame retardants, polybrominated diphenyl ether compounds (PBDEs) have been reported to be transferred through ingested plastic to fish (Rochman et al., 2014) and procellariform seabirds (Tanaka et al., 2013, 2015), including streaked shearwaters (*Calonectris leucomelas*) (Tanaka et al., 2020) and fulmars (Neumann et al., 2021). Specifically, the congener PBDE209 demonstrating high hydrophobicity (Wania, 2003), was found in different tissues of fulmars that had ingested plastic (Herzke et al., 2016; Neumann et al., 2021; Collard et al., 2022b). Even though Herzke et al. (2016) and Collard et al. (2022b) did not report any correlation between muscle or liver PBDE209 concentrations and plastic loads, a recent study suggested that PBDE209 could be a potential indicator for plastic ingestion in fulmars (Sühring et al., 2022).

Dechloranes, another class of flame retardants associated to plastics, known to bioaccumulate and biomagnify (e.g. Chen and Hale, 2010; de Wit et al., 2010; de Wit et al., 2020), were paradoxically scarcely studied in Arctic seabirds (Sühring et al., 2022) despite their negative physiological and hormonal impacts on those organisms (Mortensen et al., 2022). Dechloranes include several isomers of organochlorides: dechlorane (or Mirex©), used as a pesticide until its ban in the United States in 1978 (Feo et al., 2012), dechlorane plus (DP, both anti- and syn-isomers used as replacements for Mirex) and dechlorane 602, 603 and 604 congeners. While Mirex has been banned, DP is currently under assessment by the European Chemicals Agency for inclusion in the list of "Persistent, bioaccumulative and Toxic (PBT)" chemicals and of "Substance of very high concern (SVHC)" (ECHA, 2023).

Diesters of phthalic acid (hereafter referred to as "phthalates") are used as additives to soften plastic, in particular PVC (Heudorf et al., 2007), the most common congener being di-(2-ethylhexyl) phthalate (DEHP). According to previous studies, plastic ingestion can be a pathway for phthalates, particularly DEHP, into marine organisms such as zooplankton (Baini et al., 2017), fin whales (*Balænoptera physalus*) (Fossi et al., 2012) and seabirds (Hardesty et al., 2015). Hardesty et al. (2015) found a correlation between plastic burden in two Procellariiformes species (*Puffinus tenuirostris* and *P. pacificus*) and DEHP concentrations in their preen oil. Based on these findings, they urged to further investigate this method as a non-lethal monitoring approach for plastic exposure. However, a study on fulmars from the Canadian Arctic failed to detect phthalates in preen oil and challenged this approach (Provencher et al., 2020a). Alternatively, phthalate quantification can be performed in blood although this, to the best of our knowledge, has never been investigated in relation to plastic ingestion by seabirds.

This study aims at quantifying the concentrations of three groups of PRCs (phthalates, PBDE209 and dechloranes) in northern fulmars previously investigated for plastic loads by Tulatz et al. (2023). Since the fledglings used in our study were shown to have a higher load of plastic following parental transfer, we predicted this age class to show higher concentrations of PRCs. In addition, the second objective was to investigate the correlation between these chemicals' concentrations and both number and mass of ingested plastics to assess their suitability as proxies for plastic ingestion in the northern fulmar.

2. Materials and methods

The fulmars studied here are the same individuals that were used in a previous study on plastic ingestion and parental transfer (Tulatz et al., 2023).

Ethical statement

Liver tissues of fulmars could only be assessed by necropsy, making it necessary to sacrifice a certain sample size of fulmars. The sampling was approved by the Governor of Svalbard (permit nr. 20/02252-2) and sampling methods were in accordance with the Norwegian animal welfare law and performed by skilled and licensed staff.

2.1. Sampling

Thirty-nine fulmars were collected at sea from a boat in Kongsfjorden (Svalbard; 78°55′N, 11°56′E), as part of a project registered in 'Research in Svalbard' (RiS-ID 11562), between the 8th and September 11, 2020. Flightless fledglings were caught using a D-shaped landing net with a telescopic rod, their blood was collected before they were sacrificed by cervical dislocation. A shotgun was used to collect older flying birds (non-fledglings). Blood was therefore only collected in fledglings, the only individuals caught alive. Between 3 and 4 ml of blood were

sampled from the brachial vein by using a sterile heparinized plastic syringe (10 mL Terumo® syringe) and a 23G needle. Blood samples were transferred into sterile glass vials with plastic screw caps. In addition, 6 field blanks were simultaneously made by using milliQ water instead of blood. Blood samples were centrifuged to isolate the plasma. Each blood sample was then transferred into two 1.5 ml Eppendorf tubes. The Eppendorf tubes were then centrifuged at 500 rotations per minute for 5 min (centrifuge VWR Galaxy 7D 5). The supernatants were pipetted into sterile glass vials, capped, and frozen.

All dead birds were frozen at −20 °C within 1–4 h after sampling until further dissection in Tromsø, Norway.

2.2. Dissection

All dissections were performed in the laboratory following a standard protocol (van Franeker, 2004; OSPAR Commission, 2015). The liver was sampled in all birds (n = 39). New scalpel blades and gloves were used for each bird and the tools were rinsed using soap, milliQ water and ethanol. Birds were weighed (±2 g) and several morphometric measurements were taken. Blood, liver, feathers, pectoral muscle and fat were collected although this study utilized only plasma and liver. Blood, and therefore plasma, was sampled from fledglings only.

2.3. Aging and sexing

The birds were confirmed as fledglings by the development state of their gonads (for males: small black testes; for females: small smooth ovaries without follicles), large bursa of Fabricius and generally thick layers of subcutaneous fat (van Franeker, 2004; OSPAR Commission, 2015).

Attempts were made to age the older fulmars by, for instance, looking at the follicle in females and the presence of marks in the surrounding tissues, and at the testes in males. Unfortunately, we could not gather enough information on the testes and were therefore not able to determine previous breeding activity. Consequently, all fulmars that were not characterized as “fledglings” fall into a single other category: “non-fledglings”.

2.4. Quantification of plastic-related compounds

Overall, 10 fledglings were analysed for all three plastic-related chemical groups.

2.4.1. Polybrominated diphenyl ether 209 (PBDE209) and dechloranes

The method used for this study is the same as the one used by Collard et al. (2022b) modified from Carlsson et al. (2014) and Herzke et al. (2016).

Liver samples (n = 39 for PBDE209 and n = 33 for dechloranes) were spiked with an internal standard (IS) including ^{13}C labelled PBDE209 and ^{13}C labelled Dechlorane Plus syn and Dechlorane 602 prior to extraction (Cambridge Isotope Laboratory; CIL, Tewksbury, MA, USA).

Two grams of liver tissue were homogenised with pre-treated sodium sulphate (600C, 8h) (Merck, Darmstadt Germany) and extracted three times with 40 ml, 30 ml, and 30 ml cyclohexane: acetone mixtures (ratio 3:1) in an ultrasonic water bath for 10 min. The extract was concentrated in a RapidVap evaporation vacuum system (labconco, MO, USA) until they were dry, and an aliquot of the extract was used for lipid determination. Samples were cleaned-up and fractionated using the EZprep 123 sample prep system (Fluid Management Systems; FMS, Billerica, MA, USA) and the prepacked 0.5 g fat removal kit with 6 g acidic silica column connected to a 4 g basic aluminium oxide column. The column set was washed with 20 ml of n-hexane and then samples were loaded with 10 ml of n-hexane and eluted with 150 ml of n-hexane. The basic aluminium oxide column was reversed and eluted with 50 ml dichloromethane, which was collected and evaporated. ^{13}C PCB159 was used as a recovery standard.

Liver samples were analysed for PBDE209 (Wellington laboratories, Ontario), Dechlorane Plus (syn and anti) and Dechloranes 602, 603, 604 (CIL, MA, USA). For analyte detection, we used gas chromatography with high-resolution accurate mass spectrometry (GC-HRAM) (TRACE 1310-Q Exactive GC Orbitrap™, Thermo Fisher Scientific, Waltham, MA, USA). The GC-HRAM was equipped with a 15 m RTx 1614 MS column (0.25 μm id and 0.1 μm film thickness, Restek Corp, Bellefonte, PA, USA). Helium was used as a carrier gas at a flow rate of 1.6 mL/min. More information regarding the method can be found in Carlsson et al. (2014) and Herzke et al. (2016). The LOD was 0.6 ng/g wet weight (ww) and the LOQ was 1.99 ng/g ww.

2.4.2. Phthalate metabolites

In total, eight different phthalate metabolites were quantified in blood plasma of 15 fulmar fledglings: Monomethyl phthalate (MMP, CAS 4376-18-5), monoethyl phthalate (MEP, CAS 2306-33-4), mono-iso-butyl phthalate (MIBP, CAS 30833-53-5), mono-*n*-butyl phthalate (MNBP, 131-70-4), monobenzyl phthalate (MBzP, CAS 2528-16-7), mono(2-ethylhexyl) phthalate (MEHP, CAS 4376-20-9), mono(2-ethyl-5-hydroxyhexyl) phthalate (MEHHP, 40321-99-1) and mono(2-ethyl-5-oxohexyl) phthalate (MEOHP, CAS 40321-98-0) (Chiron AS, Trondheim, Norway).

The method for deconjugation and extraction was based on a protocol by Jeong et al. (2011) with a few modifications.

Half a millilitre of plasma was added to a 2 ml glass screw cap vial and added 100 μl 1 M ammonium acetate (Sigma-Aldrich), 20 μl internal standard (D4-MEP, D4- MIBP, D4-MBzP and D4-MEHP) and 10 μl β-glucuronidase (BGTurbo® >200,000 units/ml) all purchased from Chiron AS (Trondheim, Norway). Samples were mixed gently and incubated at 37 °C for 2 h. Then the samples were diluted with 1 ml 1% formic acid in acetonitrile (ACN), vortex mixed and sonicated for 10 min, and later centrifuged for 10 min. A 1 ml ion exchange Solid Phase Extraction (SPE) column (OASIS MAX, Waters) was washed with 1 ml methanol and conditioned using 20 % methanol in cleaned and deionized milliQ water (Advantage A10, MilliporeSigma, MA, USA). One ml of 5% ammonium hydroxide solution and the supernatant were added on top of the SPE column and thoroughly mixed with a Pasteur pipette before elution on the 12 position SPE manifold which was connected to a membrane vacuum pump. The column was rinsed with a 1 ml 5% ammonium hydroxide solution and 1 ml methanol which were discarded. The analytes were eluted using 1 ml 2% formic acid in methanol into a 2 ml glass. Samples were evaporated under a gentle flow of N_2 to 100 μl. Twenty microliters of D4-MOP recovery standard were added. The sample was transferred and diluted with 300 μl Milli-Q water onto a Mini-UniPrep 0.45 μm filter and analysed using Liquid Chromatography with tandem mass spectrometry (LC/MSMS) (Thermo TSQ Vantage). 10 μl were injected into the LC/MSMS with a Waters Acquity BEH C18 column (100 mm × 2.1 mm ID, 1.7 μm particles) and an elution gradient of 0.1% formic acid in water and 0.1% formic acid in methanol at 0.3 ml/min. The mass spectrometry (MS) was run in the negative mode using Electro Spray Ionization (ESI) at 310 °C and 2500 V and a capillary temperature of 300 °C.

The LODs varied among metabolites and ranged between 0.20 (MEOHP, MBzP and MEHHP) and 7.18 ng/ml plasma (MMP). The LOQs varied also between 0.60 (MEOHP, MBzP and MEHHP) and 14.6 ng/ml plasma (MMP).

2.4.3. Quality assurance

All glassware was initially burnt at 450 °C for 8 h and rinsed with n-hexane and acetone. For each sample, new equipment was used to avoid cross-contamination. Laboratory tools were rinsed in acetone and cyclohexane in an ultrasonic water bath. In addition, sample preparation was carried out in a laminar flow clean cabinet (Bigneat Ltd. Waterlooville, Hampshire, UK).

For a batch of 10 liver samples, two laboratory blanks and standard reference materials (SRMs) were analysed for quality control of the

method. WMF-03 freeze-dried fish tissue (Wellington Laboratories Inc., Ontario, Canada) was used as reference material. The limit of detection (LOD) was set at average blank level + 3 x standard deviation for each congener and the limit of quantification (LOQ) was calculated as average blank level + 10 x standard deviation.

2.5. Data analysis

Statistical data analysis was performed with the statistics program R (R Core Team, 2019) As none of the data followed a normal distribution (Shapiro-Wilk tests, $p < 0.05$ in all cases), non-parametric two-sided Wilcoxon rank sum tests (Mann Whitney U tests) were used to compare concentrations of PRCs among groups. A Spearman's test was used to measure the degree of correlation between plastic burdens (mass or number) and PRC concentrations. A p-value of 0.05 was used as a significance threshold to accept or reject the null-hypothesis that groups are equal or different. Values below the limit of detection were set at "0.5*LOD" for statistic calculations.

3. Results

While the data on ingested plastics by fulmars were previously reported by Tulatz et al. (2023), our study aimed at examining the numbers and masses of ingested plastics in relation to the concentrations of plastic-related chemicals.

3.1. PBDE209 in livers

Liver PBDE209 concentrations above the LOD were observed in 11 out of 39 individuals (28.2 %) with a higher proportion in non-fledglings (38.9%) compared to fledglings (19.0%), and in females (36.4%) compared to males (17.6%) (Table 1, Fig. 1). Average concentrations did not significantly differ between non-fledglings and fledglings (2.33 ± 1.36 SE vs. 0.52 ± 0.12 SE ng/g ww; Wilcoxon rank sum test: w = 146.5; p = 0.14) nor between females and males (1.88 ± 1.11 vs. 0.682 ± 0.303 ng/g ww; w = 224; p = 0.19) across both age categories.

Within each age category, we investigated the correlation between plastic burdens and PBDE209 concentrations. Within non-fledglings, we did not find a significant relationship when using mass ($p = 0.361$) nor when using plastic numbers ($p = 0.365$). Within fledglings, a significant positive relationship was however found between PBDE209 and plastic number ($p = 0.028$, r = 0.48; Fig. 2a) but not plastic mass ($p = 0.112$). It is worth mentioning that this positive correlation is driven by one extreme value coming from the bird with the highest number of ingested plastics. When that outlier was removed, the correlation was no longer significant (p = 0.103, Fig. 2b).

While liver PBDE209 concentrations were below the LOD in both birds that did not contain plastic, the same was observed for the individual with the highest plastic mass. Also, all fledglings with PBDE209 above the LOD (n = 4) had plastic loads higher than 0.1 g, the EcoQO defined by OSPAR. The highest concentration of PBDE209 in fledglings was found in an individual with plastic burdens below average but with one PET item, which was a 7-cm long rope (no item of this type was found in any of the other stomachs).

3.2. Dechloranes in livers

Dechlorane concentrations were quantified in livers of both fledglings (n = 17) and non-fledglings (n = 16). Only Dechlorane 602 (DEC602) was detected in all samples while Dechloranes 601 and 604 were not detected in any sample. Dechlorane 603 (DEC603) was detected in 11 individuals (33%). Only one bird -a non-fledgling- did not show a concentration of the Dechlorane Plus isomers "syn" (syn-DP) above the LOD, while two birds -one fledgling and one non-fledgling- had no detected anti-DP isomer in their liver. All fledglings showed concentrations of DEC602 and syn-DP above LOD and their total concentration of all dechloranes ranged from 0.02 to 0.81 ng/g ww. While all non-fledglings also showed DEC602 concentrations above LOD, their total concentration of all dechloranes ranged from 0.12 to 3.89 ng/g ww and was significantly 4.5 times higher than in fledglings (Wilcoxon rank sum test, W = 28, $p < 0.01$, Table 2). The total concentrations of dechloranes also significantly differed between males and females (W = 62, $p = 0.007$), both age categories included, with males having higher concentrations than females.

Unexpectedly, when comparing dechlorane concentrations of each congener separately and as sums for all birds, there was a significant negative correlation between the number of plastics and DEC602, DEC603 and the total sum of all dechloranes (Spearman's test, p = 0.003, 0.012, 0.009 and rho = −0.51, −0.43, −0.45, respectively). Similarly, the plastic mass was significantly and negatively correlated with the concentrations of DEC602, DEC603 and the total sum of all dechloranes (Spearman's test, $p = 0.005$, 0.006, 0.048 and rho = −0.48, −0.46, −0.35, respectively). However, when dechlorane concentrations and both plastic mass and numbers were compared in fledglings and non-fledglings separately, no significant correlation was found for any congener (Spearman tests, $p > 0.05$).

3.3. Phthalate concentrations in blood (plasma)

We used the determination of phthalate metabolites as a proxy for the presence/exposure to phthalates. Among the eight phthalate metabolites that were measured in plasma, three were not detected, i.e., MMP, MEOHP and MEHHP, and two were only detected in one fledgling (MEP and MEHP, Table 3, Fig. 3). Five out of 15 fledglings displayed detectable concentrations of at least one phthalate in their plasma. MBzP quantifications were impaired for 12 out of 15 blood samples due to unknown technical issues and no values are given. This metabolite was however detected in the three samples that could be analysed with a maximum value of 1.45 ng/ml.

Due to a high proportion of fledglings showing non-detectable concentrations of metabolites, statistical analyses could not be run for individual congeners. We however did not report any differences in plastic mass between individuals with phthalate metabolites above or below the LOD (Wilcoxon rank sum test, w = 31, $p = 0.513$).

4. Discussion

This study provides new data on plasma and liver concentrations of three groups of PRCs in Svalbard northern fulmars, acknowledged bio-indicators of marine plastic pollution. Our sampling effort included

Table 1
Mean and maximum concentrations of PBDE209 in ng/g wet weight and standard errors in fulmar liver and corresponding mean number and mass (g) ± standard error of plastics in all birds collected. The LOD was 0.598 ng/g ww and the minimum concentration was <LOD for each category.

	N	Plastic number	Plastic mass	Detection rate (%)	Mean concentration ± SE	Maximum concentration
Fledglings	21	57.9 ± 17.0	0.34 ± 0.08	19.0 (n = 4)	0.52 ± 0.12	2.18
Non-fledglings	18	10.6 ± 3.2	0.05 ± 0.01	38.9 (n = 7)	2.33 ± 1.36	25.5
Females	22	50.5 ± 16.9	0.25 ± 0.06	36.4 (n = 8)	1.88 ± 1.11	25.5
Males	17	17.3 ± 3.6	0.15 ± 0.08	17.6 (n = 3)	0.68 ± 0.30	5.63
Total	**39**	**36.1 ± 10**	**0.21 ± 0.05**	**28.2**	**1.36 ± 0.65**	**25.5**

Table 2
Mean liver concentrations ± standard error (ng/g ww) of all detected dechlorane congeners, and mean number and mass (g) ± standard error of plastics in fulmar fledglings and non-fledglings. DEC: dechlorane, DP: Dechlorane Plus, sumDEC: all dechlorane congeners, F: fledglings, NF: non-fledglings, Fe: females, M: males.

	n	Plastic number	Plastic mass	DEC602	DEC603	Syn-DP	Anti-DP	sumDEC
F	17	58.8 ± 21.4	0.35 ± 0.10	0.05 ± 0.02	<0.001	0.027 ± 0.067	0.059 ± 0.075	0.14 ± 0.05
NF	16	11.8 ± 3.6	0.06 ± 0.02	0.57 ± 0.22	<0.001	0.014 ± 0.002	0.052 ± 0.008	0.64 ± 0.23
Fe	17	54.2 ± 21.9	0.26 ± 0.07	0.09 ± 0.03	<0.001	0.009 ± 0.001	0.036 ± 0.006	0.14 ± 0.03
M	16	17.5 ± 0.16	0.16 ± 0.09	0.53 ± 0.23	<0.001	0.034 ± 0.018	0.076 ± 0.002	0.64 ± 0.23
All	33	36.4 ± 11.7	0.21 ± 0.06	0.31 ± 0.12	<0.001	0.021 ± 0.009	0.056 ± 0.010	0.38 ± 0.12

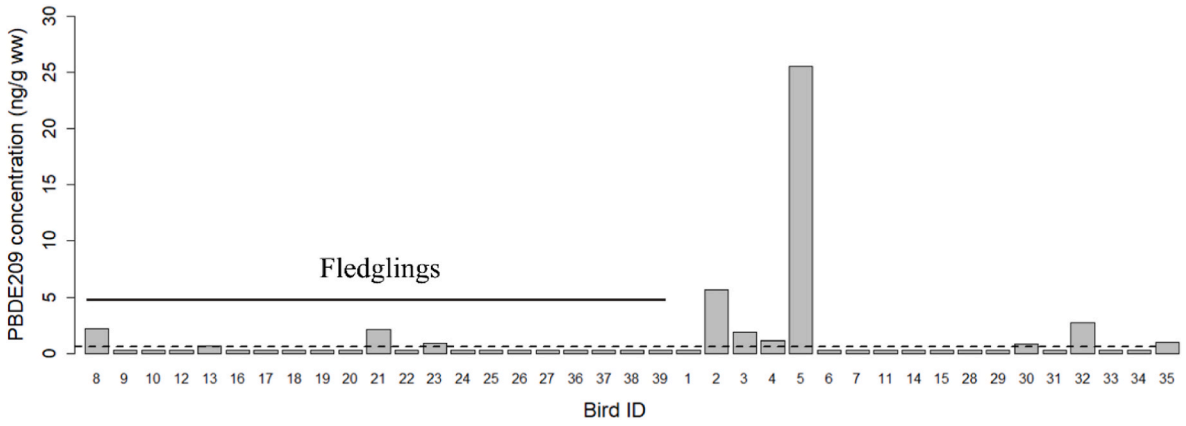


Fig. 1. PBDE209 concentrations in fulmars' (both fledglings and non-fledglings) livers. The dashed line indicates the limit of detection (LOD).

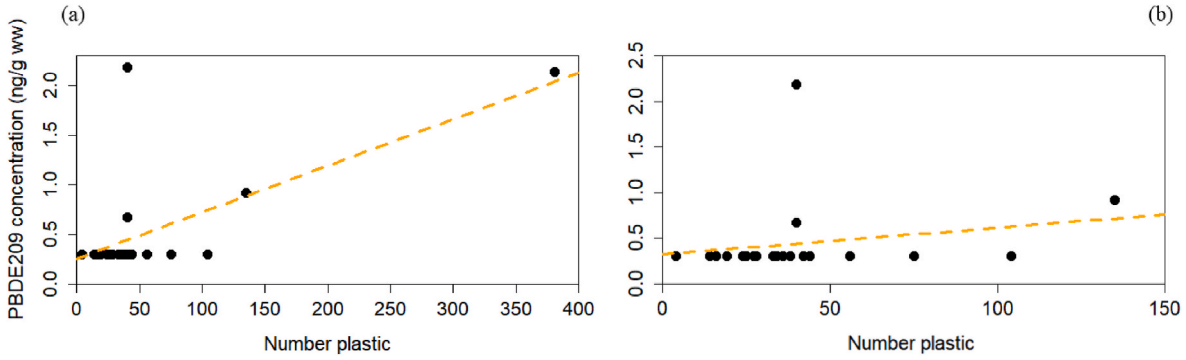


Fig. 2. Correlation between PBDE209 concentrations in fulmar fledglings' livers and their corresponding plastic numbers in the stomach, (a) all values included (Spearman test, p = 0.028, r = 0.48), (b) without the outlier on the top right of plot (a) (Spearman test, p = 0.103).

different age classes with a focus on fledglings that have been less extensively studied so far. We show that Svalbard fulmar fledglings were contaminated with dechloranes, PBDE209 and phthalate metabolites, the latter group contributing the least with one single individual measured with above LOD values for five metabolites out of the eight analysed. Previous studies have reported the concentration of these compounds in fulmars in the Arctic for monitoring purposes (Fängström et al., 2005; Jorundsdottir et al., 2013; Guzzo et al., 2014; Braune et al., 2015; Padula et al., 2020; Bianchini et al., 2022; Mortensen et al., 2022; Sühring et al., 2022) (Table 4). However, only a handful have investigated these compounds in relation to ingested plastics (Herzke et al., 2016; Provencher et al., 2020a; Neumann et al., 2021; Collard et al., 2022b).

PBDE209 was more frequent and reported at higher concentrations in non-fledglings and females (both age categories confounded). PBDEs are persistent organic pollutants that accumulate in seabird tissues and eggs (Chen and Hale, 2010) and that can biomagnify at different rates depending on the bromination of the congeners. Fulmars, like many other seabirds, are exposed to PBDE209 through their diet, via their prey and ingested plastics. As opportunistic surface-feeders, fulmars feed on different organisms such as planktonic crustaceans, polychaetas, small squids and juveniles of fish (Furness and Todd, 1984; Mehlum and Gabrielsen, 1993). During the chick-rearing season, males and females make similar foraging trips (Furness and Todd, 1984), have similar diets (Owen et al., 2013) and provide similar food items to their chicks, which explain the absence of significant difference in PBDE209 concentration between age categories. Females were found to have higher (but not significant) concentrations of PBDE209 which could explain a higher representativity of fledglings within the female category. However, this could not be verified given the low number of individuals of each sex in each age category (10 male non-fledglings vs. 7 male fledglings and 8 female non-fledglings vs. 14 female fledglings), and that only 11 birds showed PBDE209 concentrations above LOD. Therefore, we chose not to compare sexes within each age category.

Because fulmars are known to ingest plastic in high quantities, plastic was suspected to be a major source for tissue PBDE209 of fulmars in an earlier study (e.g. Neumann et al., 2021). In our study, females ingested significantly more plastics than males (Tulatz et al., 2023). Two-third of females were fledglings, likely explaining this difference between sexes. Females ingested on average 2.8 times more plastic pieces, a ratio that

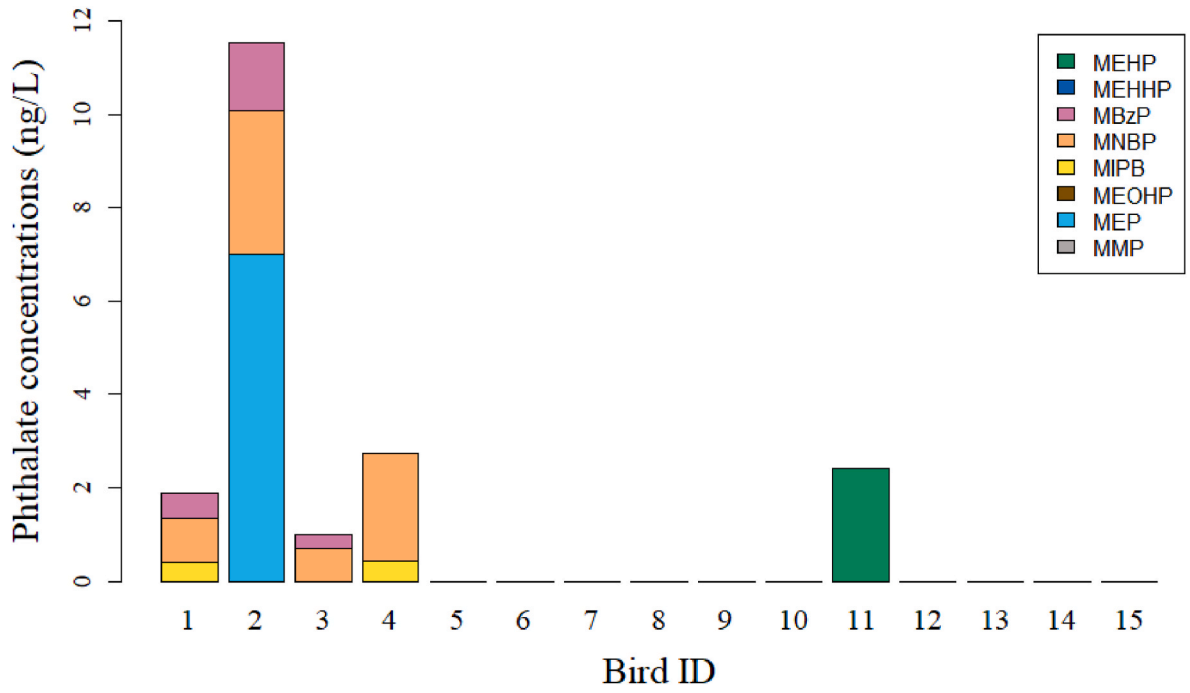


Fig. 3. Concentrations of different phthalate metabolites in fulmar fledglings' plasma (n = 15).

Table 3
Overview of the different phthalate metabolites quantified in 15 fulmar fledglings. The minimum concentration was <LOD for each metabolite.

	Number of samples > LOD (n = 15)	Maximum concentration (ng/ml)
MMP	0	<LOD
MEP	1	6.99
MEOHP	0	<LOD
MIBP	2	0.44
MNBP	5	2.31
MBzP	3	1.45
MEHHP	0	<LOD
MEHP	1	2.41

was comparable to the mean PBDE209 concentration ratio between sexes (2.9 times higher in females). The fledglings used in our study were shown to have ingested more plastics than non-fledglings, their heavier plastic load being hypothesized to result from parental transfer, at least partly (Tulatz et al., 2023). When only considering fledglings, we reported a significant and positive correlation between plastic numbers and PBDE209 concentrations although many of these fledglings showed PBDE209 concentrations below the LOD. Yet, since such a correlation was not reported in older birds (i.e., non-fledglings) and one outlier drove the correlation, this supports Neumann et al. (2021)'s assumption on PBDE209 being a good indicator of recent exposure through plastic ingestion. Fulmar fledglings have ingested plastics within the last 50–60 days and therefore show a recent exposure. Among non-fledglings, the diet likely represents a major exposure source to PBDE209 compared to ingested plastics, as already reported by Herzke et al. (2016). Altogether, these results restrict the use of PBDE209 as a potential proxy for plastic ingestion to fledglings.

Phthalates can be metabolised by several classes of organisms (Zhang et al., 2021) and subsequently excreted rapidly, depending on their chemical structure and level of exposure. Accordingly, phthalate esters that have long alkyl chains and high molecular weights show increasing hydrophobicity and potential for bioaccumulation and biomagnification along the food chain (Zhang et al., 2021). Chronic exposure to phthalates can lead to adverse health effects (reviewed in Staples et al., 1997; and in Zhang et al., 2021). In fish and amphibians, phthalates have been shown to disturb development and reproduction through disruption of the thyroid system, reduction in offspring number and decrease in hatching success, even at environmental relevant concentrations (reviewed in Oehlmann et al., 2009). Adverse effects have also been evidenced in humans (reviewed in Benjamin et al., 2017). However, both concentrations and health effects of phthalates remain largely unknown among Arctic fauna, including seabirds.

Few studies have reported the occurrence of phthalates in fulmars, but none have investigated blood plasma. Namely, Padula et al. (2020) reported the concentrations of the same phthalate congeners (DEHP, DMP, DEP, BBP, DnOP, DBP) in muscle of seabirds from the Aleutian archipelago. While Padula et al. (2020) reported the sum of concentrations to be ranging from 3.64 to 539.6 ng/g per individual, they did not report neither congeners nor species concentrations separately. Another study investigated phthalate concentrations of the same six congeners in preen oil of fulmars but reported non-detectable concentrations (Provencher et al., 2020a). Likewise, the liver and eggs of fulmars from the Canadian High Arctic were investigated recently for the occurrence of several phthalates (Sühring et al., 2022). No phthalate was detected in eggs, and only di-*n*-octyl phthalate (DnOP) was detected in some livers (five out of 10), with the same order of magnitude as reported by Padula et al. (2020), i.e., median of 120 ng/g (Sühring et al., 2022). In line with the latter studies, we found low frequencies of occurrence with phthalate being detected in one-third of the samples. However, the use of different matrices and associated units, as well as the quantification of metabolites rather than the parent compounds does not allow a comparison of concentrations across studies.

Interestingly, a similar study investigating phthalate metabolites in Arctic biota (Routti et al., 2021) reported lower frequency and lower plasma concentrations in Svalbard polar bears (*Ursus maritimus*) compared to our data on fulmars. In polar bears, only two metabolites showed concentrations above the LOD, i.e., MiBP and MnBP (Routti et al., 2021) while five fledglings from this study showed detectable concentrations of at least 2 metabolites each. In addition, while the highest metabolite concentration in polar bears was 1.2 ng/ml (MiBP), our data showed several metabolites exceeding 1.2 ng/ml, sometimes in several individual fulmars, with a maximum for MEP of 6.99 ng/ml. Both studies reported similar LODs for all metabolites, some of them

Table 4
Overview of Arctic studies investigating PBDE209, dechloranes and/or phthalates in fulmars with their corresponding results. The study of Padula et al. (2020) was not included as phthalate concentrations were not provided specifically for the fulmars. All DEC: the sum of dechlorane 601, 602, 603 and 604. "Metabolites all": the sum of MMP, MEP, MEOHP, MIBP, MNBP, MBzP, MEHHP, MEHP, DP: Dechlorane Plus, LOD: limit of detection, lw: lipid weight, ww: wet weight. Concentrations are reported as range minimum-maximum unless stated otherwise. *number of pools, [a]: n for PBDE209 analyses, [b]: n for dechlorane analyses.

Tissue	PBDE209	Dechloranes		Phthalates		Studies
		Syn + anti DP	Others	Parent compounds	Metabolites (all)	
Fat (n = 9)	<LOD	–	–	–	–	Fängström et al. (2005)
Muscle (n = 24)	<LOD – 62 ng/g lw	–	–	–	–	
Egg (n = 19)	<LOD	–	–	–	–	
Egg (n = 10)	<LOD	–	–	–	–	Jörundsdóttir et al., 2013
Egg (n = 215, 63*)	Not reported	–	–	–	–	Braune et al. (2015)
Muscle (n = 75)	<LOD – 259 ng/g ww	–	–	–	–	Herzke et al. (2016)
Liver (n = 75)	<LOD	–	–	–	–	
Ingested plastics (n = 61)	Only sum PBDE reported	–	–	–	–	
Preen oil (n = 10)	–	–	–	DEP: <LOD DEHP: <LOD DBP: <LOD	–	Provencher et al. (2020a)
Liver (n = 15)	<LOD – 2793 ng/g lw	–	–	–	–	Neumann et al. (2021)
Ingested plastics (n = 15)	<LOD – 0.340 ng/part.	–	–	–	–	
Liver (n = 18)	<LOD – 8.20 ng/g ww	<LOD – 0.76 ng/g ww	DEC602: 0.11–1.30 ng/g ww	–	–	Mortensen et al. (2022)
Liver (n = 20)	<LOD – 2.06 ng/g ww	<LOD – 0.19 ng/g ww	All DEC: 0.026–1.36 ng/g ww	–	–	Collard et al. (2022b)
Fat (n = 6)	<LOD – 4.2 ng/g ww	<LOD – 1.675 ng/g ww	–	–	–	Sühring et al. (2022)
Brain (n = 10)	<LOD	<LOD	–	–	–	
Muscle (n = 16)	<LOD – 0.65 ng/g ww	<LOD	–	–	–	
Liver (n = 20)	<LOD	<LOD	–	DEP: <LOD DEHP: <LOD DBP: <LOD	–	
Egg (n = 5)	<LOD	<LOD-13.65 ng/g ww	–	DEP: <LOD DEHP: <LOD DBP: <LOD	–	
Plasma (n = 15)	–	–	–	MEP: <LOD – 6.99 ng/ml, MEHP: <LOD – 2.41 ng/ml MNBP: <LOD – 3.10 ng/ml	< LOD – 16.4 ng/ml	*This study*
Liver (n = 39[a] & n = 33[b])	<LOD – 25.5 ng/g ww	< LOD – 0.142 ng/g ww	0.039–3.89 ng/g ww	–	–	

being quite high (e.g., 7.18 ng/ml for MMP) due to the ubiquitous presence of phthalates in indoor environments, leading to analytical contamination displayed by blank samples. The reported differences among polar bears and fulmars can be due to different sources of exposure and/or the species-specific capacity to metabolize phthalates.

Phthalates are plastic additives, and are added to the plastic matrix during manufacturing, especially polyvinyl chloride (PVC), to soften the plastic material (Heudorf et al., 2007). Phthalates are not chemically bound to the plastic matrix and therefore leach into the environment (e.g. Heudorf et al., 2007) and plastic ingestion can be a pathway for those additives to the organisms' tissues (e.g. Fossi et al., 2012; Hardesty et al., 2015). Accordingly, we expected a relationship between numbers or masses of ingested plastic and phthalate concentrations as reported by an earlier study (based on preen oil of 24 shearwaters) by Hardesty et al. (2015). However, our data support that phthalate blood metabolites cannot be considered as reliable proxies of plastic ingestion in fulmars, as already suggested by Herzke et al. (2016) and Provencher et al. (2020a). The use and mass proportion of phthalates in plastic matrices is polymer dependent. PVC did not occur in any of our samples (Tulatz et al., 2023), suggesting that phthalates came from other polymers where they are used in smaller proportions, likely explaining the absence of correlation between plastic burdens and phthalate concentrations in our study, but also the high number of values below LOD. On the other hand, the difference in phthalate concentrations between the polar bear and the fulmar could be due to the ability of metabolising parent compounds of phthalates. Living organisms are known to metabolize phthalates, with some species-specific differences in the rates of leaching or metabolisation (Vanstreels et al., 2023). Fulmars could metabolize more efficiently, or more parent compounds, than the polar bear, possibly resulting in higher metabolite concentrations in fulmars compared to polar bears. The metabolisation of phthalate is also poorly known in seabirds (Vanstreels et al., 2023) and therefore it cannot be assessed whether fulmars are more exposed to phthalates than polar bears. The high loads of plastics found in the fulmars collected for this study certainly constitute a source of phthalates, but the importance of that source, compared to the food or environment, is unknown. Further studies quantifying phthalates or phthalate metabolites in blood plasma and in ingested plastics in the same organism are needed to disentangle the influence of ingested plastics on the phthalate contamination of birds, and how fast they are metabolised and excreted.

Similarly to phthalates, several studies have investigated dechlorane contamination in seabirds but few of them focused on fulmars despite their role as bioindicators for plastic pollution (Collard et al., 2022b; Mortensen et al., 2022; Sühring et al., 2022). Dechlorane concentrations in the Arctic environment are well documented, with many abiotic compartments contaminated (Möller et al., 2010; Ma et al., 2015; Vorkamp et al., 2015; Carlsson et al., 2018). The range of hepatic dechlorane concentrations in our study is similar to those previously reported

in fulmars (Herzke et al., 2019; Collard et al., 2022b; Mortensen et al., 2022) or other European Arctic biota (Vorkamp et al., 2015) but higher than data reported on fulmars from the Canadian Arctic (Sühring et al., 2022). Unexpectedly, we reported a negative correlation between some dechlorane congeners and plastic loads. There could be several hypotheses explaining this trend. Plastic may not be the only source of dechloranes, invalidating their use as proxies for plastic pollution in seabirds. Our sample set included both fledglings and older birds, some of them likely adults, being exposed differently to dechloranes. Fledglings were indeed found to have ingested more plastics than older birds (Tulatz et al., 2023), likely driving the negative correlation between plastic burdens and dechlorane concentrations. The dechlorane concentrations in young birds such as fledglings are likely of maternal origin as dechloranes were found in Arctic or Nordic seabird eggs (Schlabach et al., 2011; Vorkamp et al., 2015), as well as in eggs of gulls (*Larus michahellis* and *L. audouinii*) from lower latitudes (e.g. Munoz-Arnanz et al., 2012) with DP concentrations similar to those we reported in the fulmars' liver. Other sources such as air, sediment and seawater can lead to exposure of dechloranes as they are subject to both long-range transport (Vorkamp et al., 2015; AMAP, 2017) and local sources (Carlsson et al., 2018) making the understanding of sources challenging.

Our results show the absence of both DEC601 and DEC604 from our samples while DEC602 was detected in all of them. Previous studies reported similar observations where both DEC601 and DEC604 were not detected at all in several Arctic marine organisms and different tissues (Schlabach et al., 2017), and in Arctic air (Skogeng et al., 2023), and where DEC604 was not detected at all while both DEC603 and DEC602 were detected in most of the fish samples quantified (Von Eyken et al., 2016). DEC602 is likely more bioaccumulative and more bioavailable than DP isomers and has the highest biota-sediment accumulation factor than DP and the other dechlorane congeners (Sverko et al., 2011), potentially explaining its high occurrence in marine biota. All those congeners are used as flame retardants but for different applications: DEC602 is for example used in Fiberglass-Reinforced Nylon and DEC604 is used in grease for lubrication of metal-to-metal and metal-to-plastic substrates (Chanda and Roy, 2007); cited in Sverko et al., 2011). The manufacturing details for each of those congeners are largely unknown while information on DP manufacturing is less scarce (Sverko et al., 2011; AMAP, 2017; Skogeng et al., 2023). DEC602, 603 and 604 are mostly manufactured in Asia but one manufacturer of DEC602 and DEC603 is based on the European continent (ChemicalBook, 2023), but the numbers related to the production are unknown. The absence of DEC601 and DEC604 in Arctic samples could reflect an absence of local sources and/or a very low volume of production and small release in the environment. DEC604 is the least volatile congener (AMAP, 2017) and is therefore less prone to long-range atmospheric transport and is less likely to reach areas lacking local sources of DEC604 (AMAP, 2017).

As for PBDE209, correlation within each age category was not performed as they were too few females and males in each age category to perform reliable statistical tests (6 male fledglings vs. 10 male non-fledglings, 11 female fledglings vs. 6 female non-fledglings).

Plastic related chemicals in an organism do not necessarily come from plastic exposure, but diet and environmental exposure can also be a source of chemicals. This study shows that none of the chemicals investigated here can be used as a proxy, probably because of the role played by uptake through diet, and the polymer-dependency of the phthalate concentrations in the ingested plastics. The fulmar is opportunistic and feeds on many different prey (e.g. Mehlum and Gabrielsen, 1993), as mentioned earlier. Fulmars also have large foraging areas, varying between years and seasons (Phillips et al., 1999; Weimerskirch et al., 2001). These temporal and spatial variabilities make estimations of chemicals exposure through plastic ingestion almost impossible. Working under controlled conditions, such as in the study of Tanaka et al. (2020) where they fed streaked shearwaters chicks in semi-field conditions, could be a way to further understand the dynamic of PRC after ingestion of contaminated plastics. However, there is no control on the environmental exposure, through air for example, and diet before and after the experiment, as well as between controlled feeding events. These types of studies could however provide more insights on the importance of ingested plastic in the exposure to plastic-related chemicals.

Understanding which contaminants are transferred to wildlife through plastic ingestion is an important issue when considering the ecological impacts of plastic pollution (Provencher et al., 2020b). Similarly, exploring the relationships between plastic ingestion and tissue concentrations of contaminants is of utmost importance to (1) understand the relative importance of plastic ingestion as a pathway for hazardous chemicals, (2) improve our knowledge on leaching of chemicals linked to plastic materials once ingested, (3) highlight proxies to be used or tested from blood samples for a non-lethal assessment of plastic ingestion and (4) understand the adverse health effects caused by plastic ingestion.

CRediT authorship contribution statement

France Collard: Writing – review & editing, Writing – original draft, Supervision, Resources, Project administration, Methodology, Funding acquisition, Conceptualization. **Felix Tulatz:** Writing – review & editing, Writing – original draft, Visualization, Investigation, Data curation. **Mikael Harju:** Writing – review & editing, Investigation. **Dorte Herzke:** Writing – review & editing, Supervision. **Sophie Bourgeon:** Writing – review & editing, Supervision, Investigation. **Geir W. Gabrielsen:** Writing – review & editing, Funding acquisition.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

France Collard reports financial support was provided by the Fram Centre and the Research Council of Norway. Dorte Herzke reports financial support was provided by the Research Council of Norway.

Data availability

Data will be made available on request.

Acknowledgements

The authors thank Rupert Krapp, Svenja Neumann, Anna Lippold, Magdalene Langset and the staff at the Sverdrup Station in Ny-Ålesund, Svalbard for their help during this study. We also thank the Fram Centre research program 'Plastic in the Arctic' that funded the 'PlastFul' project (PA072018) which includes this study. The fieldwork was funded by the Research Council of Norway through the 'FulPlast' project (#317386) and the PlastPoll project (#275172).

References

AMAP, 2017. AMAP Assessment 2016: Chemicals of Emerging Arctic Concern. Oslo, Norway, xvi+353pp-xvi+353ppp.

AMAP, 2021. Litter and microplastics monitoring plan. Tromsø 1–23p.

Andrady, A.L., Rajapakse, N., 2016. Additives and Chemicals in Plastics.

Baini, M., Martellini, T., Cincinelli, A., Campani, T., Minutoli, R., Panti, C., Finoia, M.G., Fossi, M.C., 2017. First detection of seven phthalate esters (PAEs) as plastic tracers in superficial neustonic/planktonic samples and cetacean blubber. Anal. Methods 9, 1512–1520.

Benjamin, S., Masai, E., Kamimura, N., Takahashi, K., Anderson, R.C., Faisal, P.A., 2017. Phthalates impact human health: epidemiological evidences and plausible mechanism of action. J. Hazard Mater. 340, 360–383. https://doi.org/10.1016/j.jhazmat.2017.06.036.

Bianchini, K., Mallory, M.L., Braune, B.M., Muir, D.C.G., Provencher, J.F., 2022. Why do we monitor? Using seabird eggs to track trends in Arctic environmental contamination. Environ. Rev. 30, 245–267. https://doi.org/10.1139/er-2021-0078.

Braune, B.M., Letcher, R.J., Gaston, A.J., Mallory, M.L., 2015. Trends of polybrominated diphenyl ethers and hexabromocyclododecane in eggs of Canadian Arctic seabirds

reflect changing use patterns. Environ. Res. 142, 651–661. https://doi.org/10.1016/j.envres.2015.08.010.
Burger, J., Gochfeld, M., 2004. Marine birds as sentinels of environmental pollution. EcoHealth 1. https://doi.org/10.1007/s10393-004-0096-4.
Baak, J.E., Provencher, J.F., Mallory, M.L., 2020. Plastic ingestion by four seabird species in the Canadian Arctic: comparisons across species and time. Mar. Pollut. Bull. 158 https://doi.org/10.1016/j.marpolbul.2020.111386, 111386-111386.
Carlsson, P., Herzke, D., Kallenborn, R., 2014. Polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs) and perfluorinated alkylated substances (PFASs) in traditional seafood items from western Greenland. Environ. Sci. Pollut. Control Ser. 21, 4741–4750. https://doi.org/10.1007/s11356-013-2435-x.
Carlsson, P., Vrana, B., Sobotka, J., Borgå, K., Bohlin Nizzetto, P., Varpe, Ø., 2018. New brominated flame retardants and dechlorane plus in the Arctic: local sources and bioaccumulation potential in marine benthos. Chemosphere 211, 1193–1202. https://doi.org/10.1016/j.chemosphere.2018.07.158.
Chanda, M., Roy, S.K., 2007. Plastics Technology Handbook, 4th,. CRC Press, Boca Raton, FL.
Chen, D., Hale, R.C., 2010. A global review of polybrominated diphenyl ether flame retardant contamination in birds. Environ. Int. 36, 800–811. https://doi.org/10.1016/j.envint.2010.05.013.
ChemicalBook, 2023. https://www.chemicalbook.com/ProductList_En.aspx?kwd=13560-92-4. Accessed *15.02.24*.
Collard, F., Bangjord, G., Herzke, D., Gabrielsen, G.W., 2022a. Plastic burdens in northern fulmars from Svalbard: looking back 25 years. Mar. Pollut. Bull. 185 https://doi.org/10.1016/j.marpolbul.2022.114333, 114333-114333.
Collard, F., Leconte, S., Danielsen, J., Halsband, C., Herzke, D., Harju, M., Tulatz, F., Gabrielsen, G.W., Tarroux, A., 2022b. Plastic ingestion and associated additives in Faroe Islands chicks of the Northern Fulmar *Fulmarus glacialis*. Water Biol. Syst. 1, 100079 https://doi.org/10.1016/j.watbs.2022.100079.
de Wit, C.A., Bossi, R., Dietz, R., Dreyer, A., Faxneld, S., Garbus, S.E., Hellström, P., Koschorreck, J., Lohmann, N., Roos, A., Sellström, U., Sonne, C., Treu, G., Vorkamp, K., Yuan, B., Eulaers, I., 2020. Organohalogen compounds of emerging concern in Baltic Sea biota: levels, biomagnification potential and comparisons with legacy contaminants. Environ. Int. 144 https://doi.org/10.1016/j.envint.2020.106037, 106037-106037.
de Wit, C.A., Herzke, D., Vorkamp, K., 2010. Brominated flame retardants in the Arctic environment–trends and new candidates. Sci. Total Environ. 408, 2885–2918. https://doi.org/10.1016/j.scitotenv.2009.08.037.
ECHA, 2023. Substance infocard-dechlorane plus. https://echa.europa.eu/substance-information/-/substanceinfo/100.033.575.
Feo, M.L., Barón, E., Eljarrat, E., Barceló, D., 2012. Dechlorane Plus and related compounds in aquatic and terrestrial biota: a review. Anal. Bioanal. Chem. 404, 2625–2637. https://doi.org/10.1007/s00216-012-6161-x.
Fjeld, E., Schlabach, M., Berge, J.A., Eggen, T., Snilsberg, P., Källberg, G., Rognerud, S., Enge, E.K., Borgen, A., Gundersen, H., 2004. Kartlegging av utvalgte nye organiske miljøgifter -bromerte flammehemmere, klorerte parafiner, bisfenol A og triclosan. Oslo 1–115p (in Norwegian).
Fossi, M.C., Panti, C., Guerranti, C., Coppola, D., Giannetti, M., Marsili, L., Minutoli, R., 2012. Are baleen whales exposed to the threat of microplastics? A case study of the Mediterranean fin whale (Balaenoptera physalus). Mar. Pollut. Bull. 64, 2374–2379. https://doi.org/10.1016/j.marpolbul.2012.08.013.
Furness, R.W., 1985. Plastic particle pollution: accumulation by procellariiform seabirds at Scottish Colonies. Mar. Pollut. Bull. 16, 103–106. https://doi.org/10.1016/0025-326X(85)90531-4.
Furness, R.W., Todd, C.M., 1984. Diets and feeding of Fulmars *Fulmarus glacialis* during the breeding season:a comparison between St Kilda and Shetland colonies. Ibis 126, 379–387. https://doi.org/10.1111/j.1474-919X.1984.tb00259.x.
Fängström, B., Athanasiadou, M., Athanassiadis, I., Bignert, A., Grandjean, P., Weihe, P., Bergman, Å., 2005. Polybrominated diphenyl ethers and traditional organochlorine pollutants in fulmars (*Fulmarus glacialis*) from the Faroe Islands. Chemosphere 60, 836–843. https://doi.org/10.1016/j.chemosphere.2005.01.065.
Guigueno, M.F., Fernie, K.J., 2017. Birds and flame retardants: a review of the toxic effects on birds of historical and novel flame retardants. Environ. Res. 154, 398–424. https://doi.org/10.1016/j.envres.2016.12.033.
Guzzo, M.M., Eckbo, N.H., Gabrielsen, G.W., Fisk, A.T., Hylland, K., Borga, K., 2014. Persistent organic pollutant concentrations in fledglings of two arctic seabird species. Environ. Pollut. 184, 414–418. https://doi.org/10.1016/j.envpol.2013.09.007.
Hardesty, B.D., Holdsworth, D., Revill, A.T., Wilcox, C., 2015. A biochemical approach for identifying plastics exposure in live wildlife. Methods Ecol. Evol. 6, 92–98. https://doi.org/10.1111/2041-210X.12277.
Herzke, D., Anker-Nilssen, T., Nøst, T.H., Götsch, A., Christensen-Dalsgaard, S., Langset, M., Fangel, K., Koelmans, A.A., 2016. Negligible impact of ingested microplastics on tissue concentrations of persistent organic pollutants in northern fulmars off coastal Norway. Environ. Sci. Technol. 50, 1924–1933. https://doi.org/10.1021/acs.est.5b04663.
Herzke, D., Rostkowski, P., Harju, M., Borgen, A., Christensen-Dalsgaard, S., 2019. Assessment of Additives Used in Plastic in Seabirds. Norwegian Institute for Air Research, Tromsø, p. 16p.
Heudorf, U., Mersch-Sundermann, V., Angerer, J., 2007. Phthalates: toxicology and exposure. Int. J. Hyg Environ. Health 210, 623–634. https://doi.org/10.1016/j.ijheh.2007.07.011.
Hobson, K.A., Welch, H.E., 1992. Observations of foraging northern fulmars (*Fulmarus glacialis*) in the Canadian high arctic. Arctic 45, 150–153.
Jeong, J.Y., Lee, J.H., Kim, E.Y., Kim, P.G., Kho, Y.L., 2011. Determination of phthalate metabolites in human serum and urine as biomarkers for phthalate exposure using column-switching LC-MS/MS. Safety and Health at Work 2, 57–64. https://doi.org/10.5491/SHAW.2011.2.1.57.
Jorundsdottir, H., Lofstrand, K., Svavarsson, J., Bignert, A., Bergman, A., 2013. Polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecane (HBCD) in seven different marine bird species from Iceland. Chemosphere 93, 1526–1532. https://doi.org/10.1016/j.chemosphere.2013.07.061.
Kühn, S., Booth, A.M., Sørensen, L., van Oyen, A., van Franeker, J.A., 2020. Transfer of additive chemicals from marine plastic debris to the stomach oil of northern fulmars. Front. Environ. Sci. 8 https://doi.org/10.3389/fenvs.2020.00138.
Ma, Y., Xie, Z., Halsall, C., Möller, A., Yang, H., Zhong, G., Cai, M., Ebinghaus, R., 2015. The spatial distribution of organochlorine pesticides and halogenated flame retardants in the surface sediments of an Arctic fjord: the influence of ocean currents vs. glacial runoff. Chemosphere 119, 953–960. https://doi.org/10.1016/j.chemosphere.2014.09.012.
Mallory, M.L., 2006. The Northern Fulmar (*Fulmarus glacialis*) in Arctic Canada: ecology, threats, and what it tells us about marine environmental conditions. Environ. Rev. 14, 187–216. https://doi.org/10.1139/a06-003.
Mallory, M.L., 2008. Marine plastic debris in northern fulmars from the Canadian high Arctic. Mar. Pollut. Bull. 56, 1501–1504. https://doi.org/10.1016/j.marpolbul.2008.04.017.
Mehlum, F., Gabrielsen, G.W., 1993. The diet of High-Arctic seabirds in coastal and ice-covered, pelagic areas near the Svalbard archipelago. Polar Res. 12, 1–20. https://doi.org/10.3402/polar.v12i1.6698.
Mortensen, Å.-K., Verreault, J., François, A., Houde, M., Giraudo, M., Dam, M., Jenssen, B.M., 2022. Flame retardants and their associations with thyroid hormone-related variables in northern fulmars from the Faroe Islands. Sci. Total Environ. 806 https://doi.org/10.1016/j.scitotenv.2021.150506, 150506-150506.
Munoz-Arnanz, J., Roscales, J.L., Vicente, A., Aguirre, J.I., Jimenez, B., 2012. Dechlorane Plus in eggs of two gull species (*Larus michahellis* and *Larus audouinii*) from the southwestern Mediterranean Sea. Anal. Bioanal. Chem. 404, 2765–2773. https://doi.org/10.1007/s00216-012-6326-7.
Möller, A., Xie, Z., Sturm, R., Ebinghaus, R., 2010. Large-scale distribution of dechlorane plus in air and seawater from the arctic to Antarctica. Environ. Sci. Technol. 44, 8977–8982. https://doi.org/10.1021/es103047n.
Neumann, S., Harju, M., Herzke, D., Anker-Nilssen, T., Christensen-Dalsgaard, S., Langset, M., Gabrielsen, G.W., 2021. Ingested plastics in northern fulmars (*Fulmarus glacialis*): a pathway for polybrominated diphenyl ether (PBDE) exposure? Sci. Total Environ. https://doi.org/10.1016/j.scitotenv.2021.146313, 146313-146313.
Oehlmann, J., Schulte-Oehlmann, U., Kloas, W., Jagnytsch, O., Lutz, I., Kusk, K.O., Wollenberger, L., Santos, E.M., Paull, G.C., Van Look, K.J.W., Tyler, C.R., 2009. A critical analysis of the biological impacts of plasticizers on wildlife. Philos. Trans. R. Soc. Lond. Ser. B Biol. Sci. 364, 2047–2062. https://doi.org/10.1098/rstb.2008.0242.
OSPAR Commission, 2008. Background Document for the EcoQO on Plastic Particles in Stomachs of Seabirds, 13-13p.
OSPAR Commission, 2015. Coordinated Environmental Monitoring Programme (CEMP) Guidelines for Monitoring and Assessment of Plastic Particles in Stomachs of Fulmars in the North Sea Area, 33-33p.
Owen, E., Daunt, F., Moffat, C., Elston, D.A., Wanless, S., Thompson, P., 2013. Analysis of fatty acids and fatty alcohols reveals seasonal and sex-specific changes in the diets of seabirds. Mar. Biol. 160, 987–999. https://doi.org/10.1007/s00227-012-2152-x.
Padula, V., Beaudreau, A.H., Hagedorn, B., Causey, D., 2020. Plastic-derived contaminants in Aleutian Archipelago seabirds with varied foraging strategies. Mar. Pollut. Bull. 158 https://doi.org/10.1016/j.marpolbul.2020.111435, 111435-111435.
Phillips, R.A., Petersen, M.K., Lilliendahl, K., Solmundsson, J., Hamer, K.C., Camphuysen, C.J., Zonfrillo, B., 1999. Diet of the northern fulmar *Fulmarus glacialis*: reliance on commercial fisheries? Mar. Biol. 135, 159–170. https://doi.org/10.1007/s002270050613.
Provencher, J.F., Avery-Gomm, S., Braune, B.M., Letcher, R.J., Dey, C.J., Mallory, M.L., 2020a. Are phthalate ester contaminants in northern fulmar preen oil higher in birds that have ingested more plastic? Mar. Pollut. Bull. 150 https://doi.org/10.1016/j.marpolbul.2019.110679, 110679-110679.
Provencher, J.F., Liboiron, M., Borrelle, S.B., Bond, A.L., Rochman, C., Lavers, J.L., Avery-Gomm, S., Yamashita, R., Ryan, P.G., Lusher, A.L., Hammer, S., Bradshaw, H., Khan, J., Mallory, M.L., 2020b. A Horizon Scan of research priorities to inform policies aimed at reducing the harm of plastic pollution to biota. Sci. Total Environ. 733 https://doi.org/10.1016/j.scitotenv.2020.139381, 139381-139381.
R Core Team, 2019. R: A Language and Environment for Statistical Computing. R Foundation for Statistical Computing, Vienna, Austria.
Rochman, C.M., Lewison, R.L., Eriksen, M., Allen, H., Cook, A.M., Teh, S.J., 2014. Polybrominated diphenyl ethers (PBDEs) in fish tissue may be an indicator of plastic contamination in marine habitats. Sci. Total Environ. 476–477, 622–633.
Routti, H., Harju, M., Lühmann, K., Aars, J., Ask, A., Goksøyr, A., Kovacs, K.M., Lydersen, C., 2021. Concentrations and endocrine disruptive potential of phthalates in marine mammals from the Norwegian Arctic. Environ. Int. 152 https://doi.org/10.1016/j.envint.2021.106458, 106458-106458.
Schlabach, M., Gabrielsen, G.W., Herzke, D., Hanssen, L., Routti, H., Borgen, A., 2017. Screening of PFAS and Dechlorane Compounds in Selected Arctic Top Predators. Norwegian Institute for Air Research (NILU), Kjeller, Norway, p. 34p.
Schlabach, M., Remberger, M., Brorström-Lunden, E., Norström, K., Kaj, L., Andersson, H., Herzke, D., Borgen, A., Harju, M., 2011. Brominated Flame Retardants (BFR) in the Nordic Environment. Copenhagen, 86-86p.
Skogeng, L.P., Lunder Halvorsen, H., Breivik, K., Eckhardt, S., Herzke, D., Moeckel, C., Krogseth, I.S., 2023. Spatial distribution of Dechlorane Plus and dechlorane related

compounds in European background air. Front. Environ. Sci. 10 https://doi.org/10.3389/fenvs.2022.1083011.
Staples, C.A., Peterson, D.R., Parkerton, T.F., Adams, W.J., 1997. The environmental fate of phthalate esters: a literature review. Chemosphere 35, 667–749.
Sverko, E., Tomy, G.T., Reiner, E.J., Li, Y.-F., McCarry, B.E., Arnot, J.A., Law, R.J., Hites, R.A., 2011. Dechlorane plus and related compounds in the environment: a review. Environ. Sci. Technol. 45, 5088–5098. https://doi.org/10.1021/es2003028.
Sühring, R., Baak, J.E., Letcher, R.J., Braune, B.M., de Silva, A., Dey, C., Fernie, K., Lu, Z., Mallory, M.L., Avery-Gomm, S., Provencher, J.F., 2022. Co-contaminants of microplastics in two seabird species from the Canadian Arctic. Environmental Science and Ecotechnology 12. https://doi.org/10.1016/j.ese.2022.100189, 100189-100189.
Tanaka, K., Takada, H., Yamashita, R., Mizukawa, K., Fukuwaka, M.-a., Watanuki, Y., 2013. Accumulation of plastic-derived chemicals in tissues of seabirds ingesting marine plastics. Mar. Pollut. Bull. 69, 219–222. https://doi.org/10.1016/j.marpolbul.2012.12.010.
Tanaka, K., Takada, H., Yamashita, R., Mizukawa, K., Fukuwaka, M.-a., Watanuki, Y., 2015. Facilitated leaching of additive-derived PBDEs from plastic by seabirds' stomach oil and accumulation in tissues. Environ. Sci. Technol. 49, 11799–11807. https://doi.org/10.1021/acs.est.5b01376.
Tanaka, K., Watanuki, Y., Takada, H., Ishizuka, M., Yamashita, R., Kazama, M., Hiki, N., Kashiwada, F., Mizukawa, K., Mizukawa, H., Hyrenbach, D., Hester, M., Ikenaka, Y., Nakayama, S.M.M., 2020. In vivo accumulation of plastic-derived chemicals into seabird tissues. Curr. Biol. 30, 723–728.e723. https://doi.org/10.1016/j.cub.2019.12.037.
Teuten, E.L., Saquing, J.M., Knappe, D.R.U., Barlaz, M.A., Jonsson, S., Bjorn, A., Rowland, S.J., Thompson, R.C., Galloway, T.S., Yamashita, R., Ochi, D., Watanuki, Y., Moore, C., Viet, P.H., Tana, T.S., Prudente, M., Boonyatumanond, R., Zakaria, M.P., Akkhavong, K., Ogata, Y., Hirai, H., Iwasa, S., Mizukawa, K., Hagino, Y., Imamura, A., Saha, M., Takada, H., Björn, A., Rowland, S.J., Thompson, R.C., Galloway, T.S., Yamashita, R., Ochi, D., Watanuki, Y., Moore, C., Viet, P.H., Tana, T.S., Prudente, M., Boonyatumanond, R., Zakaria, M.P., Akkhavong, K., Ogata, Y., Hirai, H., Iwasa, S., Mizukawa, K., Hagino, Y., Imamura, A., Saha, M., Takada, H., 2009. Transport and release of chemicals from plastics to the environment and to wildlife. Philos. Trans. R. Soc. Lond. Ser. B Biol. Sci. 364, 2027–2045. https://doi.org/10.1098/rstb.2008.0284.
Trevail, A.M., Gabrielsen, G.W., Kühn, S., Van Franeker, J.A., 2015. Elevated levels of ingested plastic in a high Arctic seabird, the northern fulmar (*Fulmarus glacialis*). Polar Biol. 38, 975–981. https://doi.org/10.1007/s00300-015-1657-4.
Tulatz, F., Gabrielsen, G.W., Bourgeon, S., Herzke, D., Krapp, R., Langset, M., Neumann, S., Lippold, A., Collard, F., 2023. Implications of regurgitative feeding on plastic loads in northern fulmars (*Fulmarus glacialis*): a study from svalbard. Environ. Sci. Technol. https://doi.org/10.1021/acs.est.2c05617.
van Franeker, J.A., 2004. Save the North Sea Fulmar-Litter-EcoQO Manual Part 1: Collection and Dissection Procedures. Wageningen, 38-38p.
van Franeker, J.A., Jensen, J.-K., Simonsen, P.J., Bravo Rebolledo, E.L., Kühn, S., 2022. Plastics in stomachs of northern fulmars *Fulmarus glacialis* collected at sea off east Greenland: latitude, age, sex and season. Mar. Biol. 169 https://doi.org/10.1007/s00227-022-04029-8, 45-45.
van Franeker, J.A., Kühn, S., Anker-Nilssen, T., Edwards, E.W.J., Gallien, F., Guse, N., Kakkonen, J.E., Mallory, M.L., Miles, W., Olsen, K.O., Pedersen, J., Provencher, J., Roos, M., Stienen, E., Turner, D.M., van Loon, W.M.G.M., 2021. New tools to evaluate plastic ingestion by northern fulmars applied to North Sea monitoring data 2002–2018. Mar. Pollut. Bull. 166 https://doi.org/10.1016/j.marpolbul.2021.112246, 112246-112246.
Vanstreels, R.E.T., Piccinin, I.N.L., Maraschin, M., Gallo, L., Serafini, P.P., Pereira, A., Santos, A.P., Egert, L., Uhart, M.M., 2023. Phthalate esters (plasticizers) in the uropygial gland and their relationship to plastics ingestion in seabirds along the coast of espirito santo, eastern Brazil. J. Zoo Wildl. Med. 53, 733–743. https://doi.org/10.1638/2022-0053.
Von Eyken, A., Pijuan, L., Marti, R., Blanco, M.J., Diaz-Ferrero, J., 2016. Determination of Dechlorane Plus and related compounds (dechlorane 602, 603 and 604) in fish and vegetable oils. Chemosphere 144, 1256–1263. https://doi.org/10.1016/j.chemosphere.2015.10.001.
Vorkamp, K., Bossi, R., Rigét, F.F., Skov, H., Sonne, C., Dietz, R., 2015. Novel brominated flame retardants and dechlorane plus in Greenland air and biota. Environ. Pollut. 196, 284–291. https://doi.org/10.1016/j.envpol.2014.10.007.
Wania, F., 2003. Assessing the potential of persistent organic chemicals for long-range transport and accumulation in polar regions. Environ. Sci. Technol. 37, 1344–1351. https://doi.org/10.1021/es026019e.
Weimerskirch, H., Chastel, O., Cherel, Y., Henden, J.A., Tveraa, T., 2001. Nest attendance and foraging movements of northern fulmars rearing chicks at Bjørnøya Barents Sea. Polar Biol. 24, 83–88. https://doi.org/10.1007/s003000000175.
Zhang, Y., Jiao, Y., Li, Z., Tao, Y., Yang, Y., 2021. Hazards of phthalates (PAEs) exposure: a review of aquatic animal toxicology studies. Sci. Total Environ. 771 https://doi.org/10.1016/j.scitotenv.2021.145418, 145418-145418.