



## Original Research

## Co-contaminants of microplastics in two seabird species from the Canadian Arctic



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## ABSTRACT

Through ingestion and subsequent egestion, Arctic seabirds can bioaccumulate microplastics at and around their colony breeding sites. While microplastics in Arctic seabirds have been well documented, it is not yet understood to what extent these particles can act as transport vehicles for plastic-associated contaminants, including legacy persistent organic pollutants (POPs), trace metals, and organic additives. We investigated the occurrence and pattern of organic and inorganic co-contaminants of microplastics in two seabird species from the Canadian Arctic — northern fulmar (*Fulmarus glacialis*) and black-legged kittiwake (*Rissa tridactyla*). We found that fulmars had higher levels of plastic contamination and emerging organic compounds (known to be plastic additives) than kittiwakes, whereas higher concentrations of legacy POPs were found in kittiwakes than the fulmars. Furthermore, fulmars, the species with the much larger foraging range (~200 km), had higher plastic pollution and overall contaminant burdens, indicating that birds may be acting as long-range transport vectors for plastic-associated pollution. Our results suggest a potential connection between plastic additive contamination and plastic pollution burdens in the bird stomachs, highlighting the importance of treating plastic particles and plastic-associated organic additives as co-contaminants rather than separate pollution issues.

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## 1. Introduction

In the Arctic, plastic pollution (including microplastics 1 µm–5 mm and larger macroplastics) has been found across the abiotic and biotic environment [1–8]. The risks plastic pollution may pose to Arctic wildlife and humans include physical damage (e.g., internal abrasions, blockages of the gastrointestinal tract), and toxic effects from the contaminants associated with, and derived from,

the ingested plastics [9]. Plastic particles may adsorb and/or absorb environmental contaminants [10,11], and leach chemical additives with subsequent release to the environment [12,13]. These chemical additives, such as flame retardants, UV stabilisers, phthalates, surface treatment agents (e.g., per- and poly-fluoroalkyl substances (PFAS)), and dyes, are added to plastics during the production or moulding process to achieve specific properties [14,15]. Thus, plastic particles can act as potential transport vehicles for plastic

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additives, including potential long-range transport into the Arctic [11,16].

Several seabird species in the Canadian Arctic ingest plastics, including northern fulmar (*Fulmarus glacialis*) and black-legged kittiwake (*Rissa tridactyla*) [8,17]. In addition, long term monitoring of these species [18] has revealed that both species can accumulate contaminants [18–21]. This accumulation has been reported to be tissue-specific [22,23], influenced by the breeding location, and the species-specific foraging ecology [24]. Yet, the occurrence, fate, and potential effects of plastic additives in Arctic seabirds are still poorly understood.

Plastic additives such as flame retardants have been reported in various Arctic environmental media [25]. Brominated flame retardants (BFRs), and in particular polybrominated diphenyl ethers (PBDEs), have been recorded in Arctic air, seawater, sediments, and biota [25,26]. PBDEs and hexabromocyclododecane (HBCDD) have been listed under Annex A of the Stockholm Convention on Persistent Organic Pollutants (POPs) [27]. However, the added plastic products with these BFRs are still in use or persist in the environment, including in the Arctic [26]. Moreover, the restrictions on PBDE use under the Stockholm Convention on POPs have led to the increasing use of alternative BFRs (aBFRs), which often also persist in the environment and bioaccumulate in organisms [28], but have yet to be phased out. Exposure to halogenated flame retardants has been reported to alter seabird behaviour, growth, and development, and thus may negatively influence reproductive success [29].

Another widely used replacement for restricted BFRs is organophosphate esters (OPEs). They are used as flame retardants and plasticisers with annual production volumes almost twice that of all BFRs combined [30]. Despite their high production volumes, OPE concentrations reported in biota (including birds) are generally lower than BFR concentrations [31]. However, Sührling et al. [32] reported high OPE concentrations in water samples from across the Canadian Arctic, specifically in proximity to seabird colonies [33]. To date, OPE measurements in Arctic seabirds are very limited [26].

UV stabilisers, including benzotriazole UV stabilisers (BZT-UVs), have been reported in Arctic and non-Arctic biota [34,35]. These chemicals have attracted increasing scientific and public concern because of their large production volumes, occurrence in the environment, and potential environmental risks [34,36]. Some BZT-UVs have been identified for risk management action under the Canadian federal Chemicals Management Plan [37], listed in the High Production Volume Challenge Program and Toxic Substances Control Act Inventory by the United States or classified as Substances of Very High Concern in Europe [38].

PFAS are a large group of diverse chemicals used in various applications, including as plastic additives, surfactants, and flame retardants [39]. PFAS have been reported in a wide range of Arctic biota, including zooplankton, molluscs, amphipods, fish, marine mammals, polar bears (*Ursus maritimus*), and seabirds [25,40]. PFAS can accumulate in the liver and may influence stress hormones, body condition and reproductive success in seabirds [41,42]. Perfluorooctane sulfonate (PFOS), perfluorooctanoic acid (PFOA), as well as perfluorohexanesulfonic acid (PFHxS) have been listed under Annex A of the Stockholm Convention on POPs [27]. Yet, the vast majority of the PFAS known have little environmental data and have yet to be phased out, restricted or banned [43].

Given the importance of understanding how the fate and transport of POPs and chemicals of emerging Arctic concern are associated with plastics, this study uses a range of tissue types and eggs from northern fulmars and black-legged kittiwakes from two sites in the Canadian Arctic to explore how the exposure pathways of flame retardants, UV stabilisers, phthalates, PFAS, organochlorine pesticides, and trace metals relate to tissue distribution, differences

between species, differences between sampling location, and the accumulated plastic particles. We combine new and published chemical concentration data to specifically examine multiple contaminants of concern in relation to plastic pollution.

## 2. Methods

### 2.1. Sample collection and preparation

Samples were collected from Prince Leopold Island (74° N, 90° W), Nunavut, in the Canadian High Arctic, and from the Labrador Sea, approximately 100 km from the eastern shore of Labrador, in the Canadian Sub Arctic (Fig. S1) [17,44]. Ten northern fulmars (five females, five males) and 11 black-legged kittiwakes (six females, five males) were collected from Prince Leopold Island using noose poles in early July 2013, and eggs were collected from all nests of female birds sampled. These birds were collected as a part of a long-term monitoring program under Canada's Northern Contaminants Program, and have been used for a suite of contaminant studies [17,21,34,44]. Additionally, fulmars were collected at sea by Nunatsiavut Inuit hunters from the Labrador Sea in mid-July 2015 [44], and a subset of ten females was further used in this study. Fulmars and kittiwakes were selected for this work in the region over others as these are the two species which regularly ingest and accumulate plastic particles [8,17].

Details on bird collection, storage and processing have been previously described [17,44]. A brief description is provided in the supporting information (S1). Details on the samples are presented in Table S1.

### 2.2. Tissue contaminant analysis

Nine plastic additive groups, including legacy POPs and chemicals of emerging Arctic concern (CEACs) [45] known to be used as plastic additives, were analysed in different bird tissue types to examine the relationship between plastic ingestion and chemical contaminants. A full list of the target analytes is provided in the supporting information (Table S2). The contaminants analysed in bird tissue and eggs for this study were the legacy POPs PBDEs, HBCDD, PFOS, PFOA, and tris(2-chloroethyl) phosphate (TCEP). The analysed CEACs included several alternative brominated flame retardants (aBFRs), Dechlorane Plus isomers (*syn*-DDC-CO and *anti*-DDC-CO), PFAS, OPEs, and phthalates. Additionally, essential elements and trace metals (TMs) and organochlorine pesticides (OCPs) were analysed as contaminants that could be adsorbed onto plastic particles. All analytical methods used for this study have been previously published. The method for the analysis of PBDEs, alternative brominated flame retardants, HBCDD, and the Dechlorane Plus isomers was published by McKinney et al. [46]. The OPE analysis method was published by Chu and Letcher [47], the PFAS analysis was described in Chu et al. [48], the phthalate method was developed by Provencher et al. [49], the TM method was published by Braune et al. [50], and the OCP method was previously published by Gebbink et al. [51]. Plastic particle ingestion data and methods were previously published in Avery-Gomm et al. [44] and Poon et al. [17] for Prince Leopold Island and the Labrador Sea, respectively. Data on UV-stabilisers (UV) and substituted diphenylamine antioxidants (SDPAs) were previously published by Lu et al. [52]. Additional statistical data analysis was conducted here to investigate the relationship between plastic ingestion and additive or metal contamination levels.

An overview of which analytes were measured in (a) several tissue types, (b) both locations, (c) both species, and (d) in combination with ingested plastics is provided in Table 1.

Details on the sampling locations, individual analytes and analytical methods are presented in the supporting information (Fig. S1, Table S2).

**Table 1**

Overview of compound groups that have been analysed in (a) several tissue types, (b) both locations, and (c) both species. Plastic particles data for all birds were available from Avery-Gomm et al. [44] and Poon et al. [17].

		Tissue	PBDEs	HBCDD	aBFRs	DDC-COs	OPEs	UVs <sup>a</sup>	SDPAs <sup>a</sup>	Phthalates	PFAS	TM	OCP
Northern Fulmar	Labrador Sea	Fat	x	x	x	x	x						
		Brain	x	x	x	x	x						
		Liver										x	
	Prince Leopold Island	Muscle	x	x	x	x		x					
		Liver	x	x	x	x			x	x	x	x	x
		Muscle	x	x	x	x		x					
Black-legged Kittiwakes	Prince Leopold Island	Eggs	x	x	x	x		x	x	x		x	
		Liver	x	x	x	x		x	x	x		x	
		Eggs	x	x	x	x		x	x	x		x	

<sup>a</sup> Data by Lu et al. [52].

### 2.3. Quality assurance

Average recoveries for isotope-labelled standards ranged from  $29 \pm 20\%$  for <sup>13</sup>C-perfluorononanoic acid (MPFNA) to  $205 \pm 106\%$  for <sup>13</sup>C-tetrachlorodiphenylethane (<sup>13</sup>C-*p,p'*-DDD) with a median recovery of 91% (Table S3a). Spike recoveries using pig liver ranged from 21% for tris(3-bromo-4-methylphenyl) phosphate (T3B4MP) to  $350.3 \pm 19.6\%$  for Ethanol,2-butoxy-,1,1',1''-phosphate (TBOEP) with a median of 99% (Table S3b). At least one procedural blank was included with every batch (tissue type) and target analyte group. Blanks ranged from non-detectable to  $0.32 \text{ ng g}^{-1}$  wet weight (ww) for BDE-47. All concentrations were blank and recovery corrected. The limit of detection (LOD) was calculated based on the average blank  $\pm 3x$  standard deviation of the blank or a signal to noise ratio of 3 (in the absence of blanks). The LOD ranged from  $0.50 \text{ pg g}^{-1}$  ww for dibutyl-diphenylamine (C4C4) to  $19 \text{ } \mu\text{g g}^{-1}$  ww for di (2-ethylhexyl) phthalate (DEHP). The median LOD for the target analytes was  $0.085 \text{ ng g}^{-1}$  ww (Table S4). Details on the quality assurance protocols and results are presented in the supporting information.

### 2.4. Data analysis

Statistical analysis was performed using R (version 4.0.5) and RStudio (version 1.1.456). Data distribution was investigated using summary statistics and histograms. Pearson correlations and Student's *t*-tests were performed for datasets that approximated normal distributions. Mann-Whitney-U tests were performed for non-normally distributed data. Contaminants were analysed in the nine groupings, with similar congeners and related classes of contaminants grouped. Each grouping of contaminants (e.g., PBDEs, HBCDD, DDC-CO isomers, aBFRs, OPEs, PFAS, OPEs, UV-stabilisers, SDPAs, phthalates, TMs, and OCPs) was examined in relation to tissue distribution, location, species and the mass of plastic pollution depending on which comparisons were feasible based on the analysed samples (Table 1). Species- and location-based comparisons were conducted for contaminants analysed in the same tissue types, while tissue distribution was investigated for contaminants analysed in at least two different tissue types of the same bird (Table 1). All concentrations were correlated with the ingested plastics found in the bird's GIT. Sum concentrations were calculated based on compounds in each compound class that were consistently analysed across the different matrices. In particular, sum PBDE concentrations were based on 12 congeners (Table S6) and  $\alpha$ -, $\beta$ -tetrabromoethylcyclohexane (TBECHE) were excluded from the sum aBFR concentrations (Table S9). Values below the limit of detection (LOD) were set to "0" for summary statistic calculations unless specifically stated as "based on detectable concentrations" in which case only detected concentrations were used in the calculation (used for figures). For all tests, results were considered statistically significant if  $p \leq 0.05$ .

## 3. Results and discussion

### 3.1. Ingested plastic particles

Fulmars had considerably higher plastic pollution levels (both particle count and mass) than kittiwakes, with 90% of fulmars and 9% of kittiwakes containing plastic debris (Table S5). Ingested plastic pollution in Labrador Sea fulmars was significantly higher than that of either fulmars or kittiwakes from Prince Leopold Island (Fig. S2; Mann-Whitney-U test,  $p < 0.05$ ). In both fulmars and kittiwakes, plastic pollution was dominated by user plastic fragments (Table S5). These results were consistent with recent studies elsewhere in the Canadian Arctic that suggested that fulmars are highly susceptible to plastic ingestion [8,17]. Plastic particle loads in seabirds increase towards southern latitudes in Western North Atlantic, Eastern North Atlantic, and Eastern North Pacific [44]. The species-specific differences in plastic ingestion may be explained by differences in foraging strategies or trophic niches of fulmar and kittiwake [8,17]. In general, fulmars travel long distances from the breeding colony site (~200 km) to forage and prey on crustaceans and cephalopods, whereas kittiwakes typically forage closer to the colony site (~50 km) and are primarily piscivorous [53,54]. The larger foraging range might take the northern fulmar closer to ocean plastics accumulation zones (e.g., east and south of sea-ice-choked waters of the Canadian Arctic during the breeding season), and therefore plastic pollution, than the more short-range black-legged kittiwakes. Overwintering locations may also influence the ingested plastic pollution levels as the retention time for ingested items in these species is thought to be on the order of months [55]. Overall, location and species appeared to determine the overall plastic particle loads in the sampled birds, whereas the sex of the birds did not seem to impact the ingested plastic particle loads (Mann-Whitney-U test,  $p > 0.05$ ).

### 3.2. Plastic additives

The detection frequencies of the analysed contaminant groups ranged from 13 to 100% (Table 2) and differed considerably between locations and species for some contaminants. Overall detectable concentrations ranged from a median of  $0.028 \text{ ng g}^{-1}$  ww for  $\sum$ SDPAs in eggs of black-legged kittiwakes from Prince Leopold Island to  $120 \text{ ng g}^{-1}$  ww for the phthalate, di-*n*-octyl phthalate (DnOP), in the liver of northern fulmar from Prince Leopold Island (Table 2). However, DnOP could only be detected in two eggs from Prince Leopold Island with  $350 \text{ ng g}^{-1}$  ww and  $9600 \text{ ng g}^{-1}$  ww, as well as five liver samples from fulmars collected at Prince Leopold Island with an average concentration of  $80 \pm 87 \text{ ng g}^{-1}$  ww, making it one of the compounds with the lowest overall detection frequency (19%).

**Table 2**

Median plastic additive concentrations (minimum–maximum) and detection frequency in different tissues of northern fulmar (*Fulmarus glacialis*) from the Labrador Sea, northern fulmar from Prince Leopold Island, and black-legged kittiwakes (*Rissa tridactyla*) from Prince Leopold Island. N/A indicates where tissues were not analysed, <LOD indicates measurements below the limits of detection.

	Northern Fulmar					Black-legged Kittiwakes				Detection Frequency (%)
	Labrador Sea		Prince Leopold Island			Prince Leopold Island				
	Liver (n = 10)	Brain (n = 10)	Fat (n = 6)	Muscle (n = 6)	Egg (n = 5)	Liver (n = 10)	Muscle (n = 10)	Egg (n = 6)	Liver (n = 11)	
ΣPBDEs	N/A	<LOD (<LOD–0.22)	19 (2.8–32)	0.11 (<LOD–0.65)	0.15 (0.10–0.32)	0.085 (<LOD–0.38)	0.39 (0.099–0.72)	2.2 (1.3–4.4)	3.1 (1.9–11)	74
ΣHBCDD	N/A	<LOD	<LOD	<LOD (<LOD–2.2)	3.1 (0.88–3.7)	<LOD	<LOD	1.2 (0.88–7.8)	1.3 (0.69–11)	45
ΣaBFR	N/A	<LOD	<LOD	0.089 (<LOD–2.2)	<LOD	<LOD (<LOD–1.5)	0.076 (<LOD–0.14)	<LOD	<LOD	24
DDC-COs	N/A	<LOD	0.70 (<LOD–1.7)	<LOD (<LOD–0.27)	<LOD (<LOD–14)	<LOD	<LOD	<LOD	<LOD	13
Cl-OPEs	N/A	0.47 (0.14–0.98)	1.7 (0.62–6.3)	0.23 (0.12–0.80)	N/A	N/A	0.17 (0.099–0.31)	N/A	N/A	100
Non-Cl-OPEs	N/A	0.049 (<LOD–2.7)	3.3 (1.6–34)	<LOD (<LOD–257)	N/A	N/A	<LOD (<LOD–1.2)	N/A	N/A	56
ΣUVs <sup>a</sup>	N/A	N/A	N/A	N/A	0.28 (<LOD–3.0)	<LOD (<LOD–3.8)	N/A	<LOD (<LOD–0.43)	<LOD	24 24
ΣSDPAs <sup>a</sup>	N/A	N/A	N/A	N/A	0.028 (0.015–0.064)	<LOD (<LOD–0.74)	N/A	0.028 (0.011–0.052)	0.31 (<LOD–0.63)	100
DnOP	N/A	N/A	N/A	N/A	<LOD	120 (<LOD–210)	N/A	<LOD (<LOD–9600)	N/A	19
ΣPFAS	103 (40–206)	N/A	N/A	N/A	N/A	23 (9.0–32)	N/A	N/A	N/A	100

<sup>a</sup> Data previously published by Lu et al. [34].

### 3.2.1. Polybrominated diphenyl ethers (PBDEs)

**3.2.1.1. Tissue distributions.** The highest ΣPBDE concentrations were detected in abdominal fat tissue of northern fulmars from the Labrador Sea with a median concentration of 19 ng g<sup>-1</sup> ww (Table 2). Other tissue types had ΣPBDE concentrations in the low ng g<sup>-1</sup> ww range (liver and eggs of black-legged kittiwakes from Prince Leopold Island) or pg g<sup>-1</sup> ww (fulmar tissues other than fat) (Table 2). These results are consistent with previous studies that have reported that PBDE concentrations increase with lipid content in different tissue types [56,57].

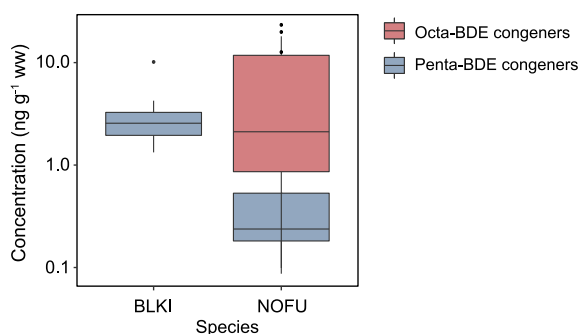
**3.2.1.2. Differences between species.** Concentrations and detection frequencies of ΣPBDEs in liver and eggs were significantly higher in kittiwakes compared to fulmars at Prince Leopold Island (*t*-test, *p* < 0.05) (Table 2). This is contrary to what has been observed in contaminant patterns in other tissues for these species [18,58]. Similar to ingested plastic pollution, this may be explained by differences in foraging strategies and diets between the two species [8,17]. The comparatively high PBDE concentrations in black-legged kittiwakes could indicate exposure through the food web. As is discussed later, several BDE congeners (e.g., BDE-47, -99, -100, -154, and -183) biomagnify in the food web [57], which will lead to higher exposures. Given that black-legged kittiwakes and northern fulmars both prey upon zooplankton and fish [54,59], these differences in concentrations may be attributed to differences in the proportions of different prey in their diet, or specific prey exposure to these contaminants. For example, at Prince Leopold Island, kittiwakes consistently occupy a higher trophic niche than fulmars early in the breeding season under all ice conditions [60], suggesting perhaps a higher proportion of fish in their diet. However, little is known about the diet of kittiwakes in

the Canadian High Arctic, so potentially these birds are foraging on different prey species with high PBDE concentrations. Other potential uptake pathways include inhalation of dust and aerosols and dermal uptake through the legs while swimming [61]. However, due to the hydrophobicity of PBDEs and the remote location of the birds, uptake from water or dust can be considered minimal compared to uptake through ingestion.

BDE-47 had the highest concentrations among the BDE congeners in kittiwakes from Prince Leopold Island (liver and eggs) followed in descending order by BDE-99 > BDE-100 > BDE-153 > BDE-154 > BDE-28 > BDE-49 (Table S6). These BDE congeners are associated with the technical Penta-BDE formulations (Fig. 1). BDE congener patterns in biota are usually dominated by congeners from the Penta-BDE mixture [57]. This is due to the higher bioavailability of Penta-BDE compared to Octa- and Deca-BDE congeners. Moreover, it has been reported that some species (e.g., fish) de-brominate higher brominated congeners to lower brominated congeners (with BDE-47 as one metabolic product) [57]. The detected PBDE congener patterns indicated that kittiwakes were predominantly exposed to PBDEs via their diet of fish, perhaps yielding additional support for the notion that kittiwakes may have more fish in their diet than fulmars (above).

In the eggs and liver of fulmars from Prince Leopold Island, most BDE congeners could only be detected in one sample. The only congeners detectable in more than one sample were Octa-BDE mixture congeners (BDE-153 and BDE-99) (Fig. 1), with the highest detection frequencies in the livers of males (Table S6). BDE-153 and BDE-99 have previously been reported as dominant PBDE congeners in terrestrial bird species [62], whereas seabirds (and typically other aquatic-feeding birds) usually are predominantly contaminated with BDE-47 > BDE-99 > BDE-100 > BDE-153 (the





**Fig. 1.** Concentrations of Octa-BDE congeners (red) and Penta-BDE congeners (blue) in northern fulmar (NOFU) and black-legged kittiwakes (BLKI) from the Labrador Sea and Prince Leopold Island. The black horizontal line inside each box represents the median based on detected concentrations; the boxes represent the 25th and 75th percentiles of concentrations above the LOD, the black vertical lines mark the 95% confidence interval, and the dots represent outliers based on a 95% confidence interval.

pattern we detected in kittiwakes). Jin et al. [62] attributed the observed congener pattern differences in terrestrial (urban) birds having direct exposure to active Deca- and Octa-BDE sources compared to rural birds exposed to the food web. This suggests that the fulmars in this study were exposed directly to PBDE sources (e.g., products containing PBDEs). Recently, Verreault et al. [63] reported comparably high concentrations of Deca-BDE in livers of glaucous gulls (*Larus hyperboreus*) sampled in the Canadian Arctic. They hypothesised that local landfills might contribute to the Deca-BDE exposure. The results from this study and Verreault et al. [63] suggest that seabirds that forage close to human settlements (e.g., glaucous gulls), or those species like fulmars that forage in areas where plastic waste accumulates (e.g., upwellings), may be exposed to PBDEs directly from ingested plastic. As a plastic additive, this could mean exposure through the ingestion of plastics or via leachates from plastic products.

**3.2.1.3. Differences between sampling locations and connection to ingested plastics.** The muscle tissue of fulmars from Prince Leopold Island and the Labrador Sea had similar  $\Sigma$ PBDE concentrations, contrary to our prediction that more southern individuals would have higher levels (Mann-Whitney-U test,  $p > 0.05$ ). This finding suggests that the distance of the collection location to PBDE sources in more southerly latitudes was not the determining factor for  $\Sigma$ PBDE concentrations in this species.

The  $\Sigma$ PBDE concentrations were not correlated with the ingested plastic mass for either sampling location or species (Figs. S3–S5) (Pearson  $R < 0.1$ ). However, the observed pattern of high Octa-PBDE congeners in northern fulmars compared to black-legged kittiwakes suggested a connection between the differences in foraging behaviour and exposure to PBDEs. This connection was also apparent for plastic ingestion. Previous results by Neumann et al. [64] showed that BDE-209 might be associated with ingested plastic particles in fulmars from Svalbard. While we did not observe a significant correlation for  $\Sigma$ PBDEs with the mass of ingested plastics (Pearson  $R < 0.2$ ,  $p > 0.05$ , Tables S4–S6), our results suggest that the pattern of PBDE congeners – and particularly the presence of Octa- PBDE congeners and BDE-209 (Deca-BDE) – could be a potential indicator for plastic ingestion. More research on larger sampling sites should be conducted to investigate this hypothesis.

### 3.2.2. Hexabromocyclododecane (HBCDD)

**3.2.2.1. Tissue distributions.** We found that HBCDD concentrations in northern fulmars were in the low  $\text{ng g}^{-1}$  ww range for eggs and

$\text{pg g}^{-1}$  ww range for muscle tissue, and HBCDD was not detected in brain, liver and fat tissue (Table 2). The HBCDD concentrations in eggs were of the same order of magnitude as concentrations previously reported in Arctic seabirds from Iceland, while they were lower than concentrations reported in eggs of the herring gull, Atlantic puffin (*Fratercula arctica*) and black-legged kittiwake from the Norwegian Arctic [25].

**3.2.2.2. Differences between species and sampling locations and connection to ingested plastics.** Concentrations in eggs were similar for fulmars and kittiwakes from Prince Leopold Island, whereas only fulmars had detectable HBCDD concentrations in liver samples at Prince Leopold Island (Table 2). HBCDD could not be detected in fulmar muscle tissue at Prince Leopold Island, whereas muscle tissue was the only tissue type with (low) detectable HBCDD concentrations in fulmars from the Labrador Sea (Table 2). There were insufficient data points for a statistical evaluation of the correlation between ingested plastic mass and HBCDD concentrations. The findings suggest that patterns of HBCDD in these two species, across sites, and between tissues do not necessarily follow specific patterns that were observed for other contaminants. The lack of pattern could indicate that the birds are exposed to diffuse sources of HBCDD across their habitats. However, the sample size in this study was very small. Therefore, further analysis should be performed to confirm this observation. This observation was consistent with previous research on HBCDD in kittiwake and fulmar eggs from Prince Leopold Island [65] and observations in six different seabird species from Iceland [66]. Murvoll et al. [67] reported a different trend in bird eggs collected at Kongsfjorden, Svalbard, where eggs of thick-billed murre (*Uria lomvia*) had significantly higher detection frequencies and concentrations than eggs of common eiders (*Somateria mollissima*). This was attributed to the difference in feeding behaviour between the predominantly piscivorous murre and benthivorous eiders [67]. The comparatively high concentrations in the bird eggs compared to other tissue types observed by Murvoll et al. [67] indicate the maternal transfer of HBCDD with subsequent exposure during embryonic and nestling development. HBCDD concentrations in the individual samples are presented in Table S7.

### 3.2.3. Emerging halogenated flame retardants (aBFRs, DDC-Cos)

**3.2.3.1. Tissue distribution.** Emerging halogenated flame retardants had low concentrations in all bird tissue samples. The highest median concentrations of  $0.70 \text{ ng g}^{-1}$  ww were detected for DDC-Cos in the fat of fulmars from the Labrador Sea (Table 2). Fulmar eggs also had the highest individual DDC-CO concentrations with up to  $14 \text{ ng g}^{-1}$  ww in fulmar eggs from Prince Leopold Island (Table 2). Concentrations for aBFRs and DDC-COs in other tissue types were in the  $\text{pg g}^{-1}$  ww range with higher concentrations in fat than in muscle tissue (Table 2). The anti-DDC-CO isomer had higher concentrations and detection frequencies than the syn-isomer (Table S8) which is consistent with reports of DDC-CO in eggs and livers of black guillemots and glaucous gulls from Greenland [68]. EHTBB was the most frequently detected aBFR with a median concentration of up to  $0.076 \text{ ng g}^{-1}$  ww in muscle tissue of northern fulmar from Prince Leopold Island (Table S9). The concentrations of  $\Sigma$ aBFRs detected in this study were similar to or lower than previously reported concentrations in Arctic seabirds [25]. BTBPE was the only aBFR with higher muscle tissue concentrations compared to previous reports in seabird eggs and livers from the European Arctic [25], but it was only detected in muscle tissue of two birds. Moreover, because different tissue types and species were analysed, it is impossible to conclude whether BTBPE concentrations in seabirds from the Canadian Arctic were generally higher than in seabirds from the European Arctic. Studies on DDC-

CO isomers (*syn* and *anti*), and other isomers (e.g., Dec602 and Dec603) in Arctic seabirds and other wildlife, are still limited [26]. The DDC-CO concentrations in this study were considerably higher than the concentrations reported in black guillemot (*Cepphus grylle*) eggs collected in East Greenland [68] and eggs of common eider, European shag (*Phalacrocorax aristotelis*) and herring gull collected in the Norwegian Arctic [69]. More research should be conducted to investigate the comparably higher concentrations in Canadian Arctic seabirds, and to identify whether egg DDC-CO concentrations are generally higher than muscle concentrations.

**3.2.3.2. Differences between species, sampling locations and possible connection to ingested plastics.** aBFRs and *syn*- and *anti*-DDC-CO isomers could only be detected in northern fulmar tissues, suggesting that the fulmars examined in this study were more exposed to emerging halogenated flame retardants than kittiwakes from the same location. In the Labrador Sea fulmar samples, *syn*- and *anti*-DDC-CO isomers were detected in the fat and muscle of the fulmars, whereas in fulmars from Prince Leopold Island, *syn*- and *anti*-DDC-CO isomers were not detectable in muscle tissue. Similarly, maximum concentrations of aBFRs in muscle tissue of fulmars from the Labrador Sea were approximately an order of magnitude higher than in muscle tissue of fulmars sampled at Prince Leopold Island, whereas the median concentrations were similar (Table S9). In eggs, only *syn*- and *anti*-DDC-CO isomers were detectable and only in eggs of fulmars sampled at Prince Leopold Island, suggesting that the sampling location played an important role in the contamination with *syn*- and *anti*-DDC-CO isomers. We did not find a correlation between the concentrations of *syn*- and *anti*-DDC-CO isomers or aBFRs and the mass of ingested plastic pollution (Pearson  $R < 0.2$ ,  $p > 0.05$ , Figs. S4 and S5).

### 3.2.4. Organophosphate esters (Cl-OPEs, non-Cl-OPEs)

**3.2.4.1. Tissue distributions.** The highest median  $\Sigma$ OPE concentrations of  $4.7 \text{ ng g}^{-1} \text{ ww}$  were quantified in fat tissue of fulmars sampled at the Labrador Sea, while  $\Sigma$ OPE concentrations in other tissue types were in the  $\text{pg g}^{-1} \text{ ww}$  range (Table S10). Brain concentrations of  $\Sigma$ Cl-OPEs were similar to concentrations in muscle, and  $\Sigma$ Cl-OPE concentrations in fat were around one order of magnitude higher than in muscle and brain in fulmars from the Labrador Sea (Fig. S6).

**3.2.4.2. Differences between species and sampling locations.** OPEs were only analysed in fulmar tissue samples. For  $\Sigma$ Cl-OPEs, muscle concentrations were comparable at the two sites (Fig. S6). Concentrations of  $\Sigma$ non-Cl-OPEs were significantly higher than  $\Sigma$ Cl-OPE concentrations in muscle and fat tissue of fulmars from the Labrador Sea and significantly higher than  $\Sigma$ Cl-OPE and  $\Sigma$ non-Cl-OPE concentrations in muscle tissue of fulmars from Prince Leopold Island and brain tissue from the Labrador Sea fulmars (*t*-test,  $p < 0.05$ , Table 2). The comparably higher concentrations of OPEs (particularly non-Cl-OPEs) highlight the increasing environmental burden of OPEs as replacements for PBDEs and other brominated flame retardants. The comparatively higher concentrations of OPEs in these seabirds also challenge the current hypothesis in scientific and regulatory assessments that OPEs are not bioaccumulative – at least in the case of seabirds [25,26].

**3.2.4.3. Connection to ingested plastics.** The high proportion of  $\Sigma$ non-Cl-OPEs to  $\Sigma$ OPE contamination (94% of the total OPE concentrations measured in this study) was particularly interesting, because Cl-OPEs are the predominant OPEs in water and air in the Canadian Arctic [16,70,71]. However, the observation matched high reported contributions of non-Cl-OPEs in water samples taken close to a large thick-billed murre colony in the Canadian Arctic

[16], and studies of OPEs in seabirds outside the Arctic [31]. Similar concentrations of the non-Cl-OPE, TEHP, and the Cl-OPE, TCEP, were reported in liver samples of kittiwakes sampled on Svalbard in the Norwegian Arctic [72]. At the same time, Greaves et al. [73] reported rapid metabolism of OPEs in *in vitro* studies of OPEs using liver microsomes of Great Lakes herring gulls. This raises the question of why the non-Cl-OPEs in the present seabird tissues do not appear to be effectively influenced by metabolic degradation in these birds. Alternately, it is plausible that the OPE exposure and uptake rate is higher than the rate of metabolism in these birds, which results in elevated tissue and egg levels. Another possible explanation could be that the detected non-Cl-OPEs in the analysed tissues are associated with microplastics. This hypothesis is supported by the correlation of  $\Sigma$ non-Cl-OPE (Pearson  $R = 0.33$ ,  $p < 0.05$ ) and  $\Sigma$ Cl-OPE (Pearson  $R = 0.34$ ,  $p < 0.05$ ) concentrations with ingested plastics in northern fulmars from Prince Leopold Island (Fig. S5). Being (non-covalently) associated with microplastics could shield OPEs from degradation and metabolism within the bird's body. However, this hypothesis does not explain the dominance of non-Cl-OPEs, because both non-Cl-OPEs and Cl-OPEs are used in plastic products [25]. Non-Cl-OPEs are more hydrophobic than Cl-OPEs, resulting in a higher leaching efficiency out of microplastics and subsequent metabolism for Cl-OPEs compared to non-Cl-OPEs [74].

### 3.2.5. UV stabilisers and SDPAs

As reported in Lu et al. [52],  $\Sigma$ UV stabilizer and  $\Sigma$ SDPA concentrations were in the low  $\text{ng g}^{-1} \text{ ww}$  or  $\text{pg g}^{-1} \text{ ww}$  range for all analysed tissue types (Tables S11 and S12). We examined  $\Sigma$ UV stabilizer and  $\Sigma$ SDPA concentrations in relation to the ingested mass of plastic pollution levels and found no significant correlations (Pearson  $R < 0.15$ ,  $p > 0.05$ , Fig. S3).

### 3.2.6. Phthalates (DnOP)

**3.2.6.1. Differences between species, sampling locations, and possible connection to ingested plastics.** Among the analysed phthalates, only DnOP could be detected and quantified in the liver and eggs. DnOP was found in five fulmar liver samples and two kittiwake egg samples in concentrations ranging from  $120 \text{ ng g}^{-1} \text{ ww}$  in a fulmar liver sample to  $9600 \text{ ng g}^{-1} \text{ ww}$  in a kittiwake egg from Prince Leopold Island. No phthalates were detected in fulmar eggs or kittiwake livers (Table 2, Table S13). Phthalate concentrations in the liver were in the same order of magnitude as concentrations reported for the pectoralis muscle of a variety of seabirds sampled in the Aleutian Archipelago, while the detection frequencies in our study were significantly lower than what was reported for the Aleutian Archipelago seabirds [75]. Our study and the study of Padula et al. [75] provide evidence that seabirds are exposed to (and may accumulate) phthalates in the North American Arctic. For the different sampling locations, phthalates were only analysed in birds sampled at Prince Leopold Island. There were no significant correlations between the phthalate concentrations and the mass of plastic pollution found in the birds under study (Pearson  $R = -0.07$ ,  $p > 0.05$ , Fig. S3). However, the high LOD for DnOP and phthalates, in general, could mask potential correlations with the ingested plastic.

### 3.2.7. Per- and Poly-Fluoroalkyl Substances (PFAS)

**3.2.7.1. Differences between sampling locations and possible connection to ingested plastics.** PFAS (mainly PFAAs) were only analysed in liver tissue, and only for northern fulmars. PFAS concentrations in fulmar livers sampled at Prince Leopold Island were significantly lower ( $p < 0.05$ ) than concentrations in fulmars from the Labrador Sea, with median hepatic  $\Sigma$ PFAS concentrations of  $23 \text{ ng g}^{-1} \text{ ww}$  (range:  $9.0\text{--}32 \text{ ng g}^{-1} \text{ ww}$ ) in fulmars from Prince Leopold

Island, and 103 ng g<sup>-1</sup> ww (range: 40–206 ng g<sup>-1</sup> ww) in fulmars from the Labrador Sea (Table 2). This pattern is consistent with previous contaminant profiles, with the more northern individuals having lower levels than those at southern locations. In both locations, PFOS contributed to >80% of the  $\Sigma$ PFAS concentrations. Detectable PFAS at Prince Leopold Island were PFOS > PFTrDA > PFUdA > PFNA > PFTeDA > PFDA > PFDaA > PFHxDA > PFHxA with only PFOS and PFTrDA detectable at concentrations above 1 ng g<sup>-1</sup> ww (Table S14). For the fulmar livers from the Labrador Sea, higher PFAS levels could be detected with contamination profiles of PFOS > PFUdA > PFTrDA > PFDA > PFNA > PFPeA > PFDaA > PFTeDA > PFOA > PFHxA > PFHxS > FOSA. Apart from PFOS, PFUdA, PFTrDA, and PFDA were detected in concentrations >1 ng g<sup>-1</sup> ww (Table S14). These results were also consistent with those reported by Choy et al. [76], i.e., in blood plasma collected (2016–2018) from thick-billed murres at Coats Island, a colony in northern Hudson Bay. PFUdA, PFOS, and PFTrDA were the dominant PFAAs and accounted for 77% of the  $\Sigma$ PFAAs. The pattern and concentrations were also similar to PFAS concentrations previously reported in fulmar and kittiwake eggs from Prince Leopold Island [20]. PFAS concentrations in fulmar livers collected at Prince Leopold Island in 2015 were five times lower than those reported in 2007–2008 [58]. The reduction in overall PFAS concentrations compared to the 2007/2008 levels reflects the reduction in PFOS concentrations compared to the earlier study. PFOS was restricted under Annex B (restricted use) of the Stockholm Convention in 2009 [45]. The lower concentrations in fulmar livers from Prince Leopold Island collected after the restriction (this study) compared to prior to the restriction [58] is encouraging to see, because it could indicate the restrictions are leading to a reduction in environmental concentrations and exposure for Arctic seabirds. However, further monitoring studies need to be conducted to investigate whether PFOS concentrations are declining among Arctic seabirds overall. Furthermore, it will have to be investigated whether remobilization due to climate change can reverse the declining trend in the future, as was discussed in the recent AMAP assessment on POP climate change interactions [77]. In the present study, there were no significant correlations between the PFAS concentrations and the mass of plastic pollution found in the birds under study (Pearson R < 0.15, *p* > 0.05, Figs. S4 and S5).

### 3.3. Metals

$\Sigma$ Metal concentrations were similar for both species and sample types but differed in patterns and abundance of potentially hazardous non-essential trace metals (TM) such as arsenic, cadmium, lead, and mercury different between the analysed tissue types (a full list of the analysed non-essential trace metals and essential elements are presented in Table S15).

#### 3.3.1. Tissue distributions

More individual TM could be detected in liver samples than eggs, and the TM pattern differed between the sample types.  $\Sigma$ Metal concentrations were dominated by essential elements such as potassium, sodium, calcium, magnesium, iron, zinc, and selenium (Fig. S7).

In bird eggs, the essential elements and strontium were the only metals with concentrations >1 ng g<sup>-1</sup> ww (Table S15). In liver tissue, cadmium, rubidium, mercury, and arsenic could be detected in concentrations >1 ng g<sup>-1</sup> ww (in addition to the essential elements) (Table S15). Livers had significantly higher sum concentrations of toxic TMs such as mercury and arsenic than egg samples (Mann Whitney-U test: *p* < 0.05).

#### 3.3.2. Differences between species and sampling locations and connection to ingested plastics

The TM patterns seemed to be driven by the tissue type rather than species (Mann Whitney-U test between species: *p* > 0.05), indicating that both species are exposed to similar TM sources. For sampling locations, metals were only analysed in birds sampled at Prince Leopold Island. Unlike previous studies that detected correlations between ingested plastics and trace metals [78], we found no significant correlations between the TM concentrations and the mass of plastic pollution in the birds (Pearson R < 0.1, *p* > 0.05, Figs. S3 and S5).

#### 3.4. Organochlorine pesticides (OCPs)

Legacy OCPs [27] (Table S1), were only analysed in liver tissue of fulmars sampled at Prince Leopold Island. Quantifiable OCP concentrations ranged from a median of 1.3 ng g<sup>-1</sup> ww for PeCB to a median of 87 ng g<sup>-1</sup> ww for oxychlorane (Table S16). Other measurable OCPs included *p,p'*-DDE > HCB > mirex > heptachlor epoxide > dieldrin > photomirex > octachlorostyrene > *p,p'*-DDD > PeCB. Half of the measurable OCPs had 100% detection frequencies, and the other half were measurable in three out of the four liver samples (Table S16), indicating that, despite international restrictions under the Stockholm Convention on POPs [79], northern fulmars are still exposed to several legacy OCPs. These observations and the detected concentrations were consistent with previous reports on OCP concentrations in Arctic seabirds [19]. There was not enough data to evaluate a correlation between OCPs and ingested plastic mass in the analysed birds.

#### 3.5. The chemical soup and concentration hierarchy

The contaminant patterns observed in northern fulmars and black-legged kittiwakes did not indicate an obvious common source or transport pathway. Legacy contaminants such as OCPs and PBDEs were prevalent in the birds, but so were contaminants of emerging Arctic concern, such as PFAS and OPEs (Fig. 2).

Overall, OCPs had the highest concentrations of the analysed contaminants despite years of restrictions [79]. Among the plastic additives, the phthalate DnOP had the highest individual concentrations, but low detection frequencies compared to some of the other contaminant groups, suggesting accumulation is highly

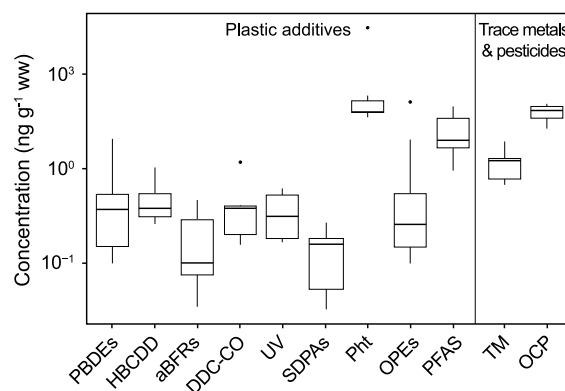
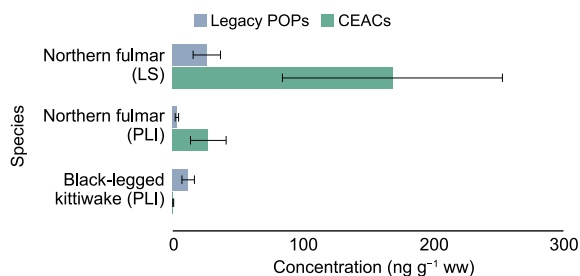


Fig. 2. Concentrations of  $\Sigma$ PBDEs,  $\Sigma$ HBCDD,  $\Sigma$ aBFRs,  $\Sigma$ DDC-COs,  $\Sigma$ UVs,  $\Sigma$ SDPAs, DnOP (PhT),  $\Sigma$ OPEs,  $\Sigma$ PFAS,  $\Sigma$ TM, and  $\Sigma$ OCP in fulmar and kittiwake tissue and eggs from the Labrador Sea and Prince Leopold Island. The black horizontal line inside each box represents the median based on detected concentrations; the boxes represent the 25th and 75th percentiles of concentrations above the LOD, the black vertical lines mark the 95% confidence interval, and the dots represent outliers based on a 95% confidence interval.





**Fig. 3.** Average concentrations of legacy POPs (blue) and CEACs (green) in black-legged fulmar and black-legged kittiwakes from the Labrador Sea (LS) and Prince Leopold Island (PLI).

variable. The flame retardant and surface treatment agents PFAS, OPEs, and PBDEs had similar concentrations and high detection frequencies, whereas aBFRs, DDC-COs, HBCDD, and UV-stabilisers generally had low concentrations and detection frequencies (Fig. 2).

An interesting observation was the different patterns of legacy POPs, namely PBDEs and CEACs such as HBCDD, DDC-COs, OPEs, aBFRs, and PFAS [25], between different bird species. In general, CEACs were only found in fulmars, and predominantly in fulmars from the Labrador Sea (Fig. 3). This suggests that fulmars are exposed to a greater range of contaminants, including some (i.e., HBCDD, DDC-COs, OPEs, and aBFRs) that are associated with plastic pollution. In contrast, and contrary to our predictions, average concentrations of legacy POPs were higher in black-legged kittiwakes than in northern fulmars from the same location (Prince Leopold Island) (Fig. 3).

Similar to the results from the Labrador Sea, northern fulmars collected at Prince Leopold Island were predominantly contaminated with CEACs even though black-legged kittiwakes from the same area were more exposed to legacy POPs. This observation supported the hypothesis drawn from the different observed BDE patterns that black-legged kittiwakes seem to be predominantly exposed to the types of plastic-associated contaminants through accumulation in the food web, whereas northern fulmars may have at least some exposure directly from active contaminant sources. Importantly, the different observed contaminant patterns were also consistent with the detection and level of ingested plastic pollution, indicating that ingested plastics could play a role in exposing northern fulmars to CEACs.

The different contamination patterns between black-legged kittiwakes and northern fulmars may be explained by the difference in the species' foraging and feeding behaviour [62]. Arctic-breeding black-legged kittiwakes that forage relatively closer to their respective colonies can be expected to represent local contamination patterns and, in the case of remote colonies, be mostly exposed through biomagnification. The larger distances that Arctic-breeding northern fulmars travel [53] mean that they can reach areas with more plastic pollution, like convergence zones, and consequently active or at least more recent pollution sources. For example, early in the breeding season fulmars from Prince Leopold Island are capable of flying the distance to open water along coastal Greenland [80] where active shipping and industrial activity are underway, whereas kittiwakes are constrained to forage within Lancaster Sound [81], usually free of shipping or industry during that time of year. Another explanation for the difference in observed contamination patterns between fulmars and kittiwakes could be the exposure through ingested plastic particles, as discussed for non-Cl-OPEs.

The role of plastic particles as transport vehicles for organic contaminants is being discussed in the scientific literature regarding to chemical transport into organisms [23,82]. However,

there is little information on the potential to facilitate long-range chemical transport through plastic particles. Zarfl and Matthies [83] estimated the potential long-range transport of POPs in water and air through microplastics compared to non-plastic based long-range transport, concluding that microplastic-enabled transport seemed to play a minor role in the overall long-range transport of POPs. However, they assumed that the POPs would partition onto the microplastics from air or seawater and did not consider plastic additives nor the bioconcentration of microplastics and their respective additives through seabirds. The concentrations of such adsorbed contaminants can be expected to be considerably lower than the concentrations of plastic additives that were deliberately added to the polymer during the production process [84]. More recently, Andrade et al. [85] estimated that between 8100 and 18,900 t of various additives are likely transported to the Arctic with buoyant plastic as the vehicle. Additionally, recent research has shown that seabirds in the Canadian Arctic act as transport vectors and concentrators of microplastics [86,87]. The higher plastic additive concentrations in northern fulmars with high plastic pollution compared to the black-legged kittiwakes from the same location could be an indication that the ingested plastic pollution can be another important source of chemical pollutants to seabirds, potentially leading to higher plastic additive exposure compared to birds that are just exposed through long-range transport of contaminants which are subsequently incorporated into marine food webs.

### 3.6. The next steps and questions about plastic-derived contaminants in Arctic seabirds

Arctic birds are exposed to a variety of plastic-derived contaminants. The results from our study highlighted the importance of analysing the contaminant mixture in seabirds rather than individual compounds if we are to understand the contaminant sources and pathways that lead to exposure.

This research is a starting point, based on small sample sizes, with the intent to explore important questions on contamination sources and pathways in Arctic seabirds. The major new question raised as a consequence of this research is the role of plastic particles as vectors for plastic additives and exposure in Arctic seabirds. Based on our findings, we conclude that experimental work needs to be conducted to evaluate the potential of the bird's gastrointestinal tract to leach out different plastic additives from plastic particles. The research priorities we identified here are the combined measurement of ingested litter and microplastics with: (a) OPEs, (b) PFAS, (c) Octa-BDE and Penta-BDE congeners, and (d) PFAS due to their high concentrations and/or detection frequencies in the birds. Future research should investigate the relationship of these contaminants to the foraging behaviour and trophic niche of the seabirds, and their potential linkage to ingested plastic load.

### Declaration of competing interest

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

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## Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.ese.2022.100189>.

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