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Common Eider and Herring Gull as Contaminant Indicators of Different Ecological Niches of an Urban Fjord System

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ABSTRACT

Seabirds like gulls are common indicators in contaminant monitoring. The herring gull (*Larus argentatus*) is a generalist with a broad range of dietary sources, possibly introducing a weakness in its representativeness of aquatic contamination. To investigate the herring gull as an indicator of contamination in an urban-influenced fjord, the Norwegian Oslofjord, we compared concentrations of a range of lipophilic and protein-associated organohalogen contaminants (OHCs), Hg, and dietary markers in blood ($n = 15$), and eggs ($n = 15$) between the herring gull and the strict marine-feeding common eider *(Somateria mollissima)* in the breeding period of May 2017. Dietary markers showed that the herring gull was less representative of the marine food web than the common eider. We found higher concentrations of lipophilic OHCs (wet weight and lipid weight) and Hg (dry weight) in the blood of common eider (mean ± SE ∑PCB = 210 ± 126 ng/g ww, 60 600 ± 28 300 ng/g lw; mean Hg = 4.94 ± 0.438 ng/g dw) than of the herring gull (mean ± SE ∑PCB = 19.0 ± 15.6 ng/g ww, 1210 ± 1510 ng/g lw; mean Hg = 4.26 ± 0.438 ng/g dw). Eggs gave opposite results; higher wet weight and lipid weight OHC concentrations in the herring gull (mean ± SE ∑PCB = 257 ± 203 ng/g ww, 3240 ± 2610 ng/g lw) than the common eider (mean ± SE ∑PCB = 18.2 ± 20.8 ng/g ww, 101 ± 121 ng/g lw), resulting in higher OHC maternal transfer ratios in gulls than eiders. We suggest that the matrix differences are due to fasting during incubation in the common eider. We suggest that in urban areas, herring gull might not be representative as an indicator of marine contamination but rather urban contaminant exposure. The common eider is a better indicator of marine pollution in the Oslofjord. The results are influenced by the matrix choice, as breeding strategy affects lipid dynamics regarding the transfer of lipids and contaminants to eggs and remobilization of contaminants from lipids to blood during incubation, when blood is drawn from the mother. Our results illustrate the benefit of a multispecies approach for a thorough picture of contaminant status in urban marine ecosystems. *Integr Environ Assess Manag* 2021;17:422–433. © 2020 The Authors. *Integrated Environmental Assessment and Management* published by Wiley Periodicals LLC on behalf of Society of Environmental Toxicology & Chemistry (SETAC)

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INTRODUCTION

Human activities have introduced contaminants to ecosystems worldwide, resulting in the need to monitor for contamination status as part of environmental assessment and management programs (Furness and Camphuysen 1997; Elliott and Elliott 2013). In coastal marine ecosystems, seabirds have been used as indicators of contamination high in the food web due to biomagnification of recalcitrant contaminants with increasing trophic position (Fisk et al. 2001; Tomy et al. 2004). It is important that species selected for this role represent the monitored ecosystem (Baert et al. 2013; Elliott et al. 2015). In addition to diet, interspecies differences in physiology and life history traits affecting energy allocation, remobilization of contaminants, and elimination processes affect contaminant concentrations (Borgå et al. 2004; Hitchcock et al. 2019a).

The herring gull (*Larus argentatus*) has been commonly used as an indicator species of contaminants in aquatic ecosystems due to its high trophic position and wide distribution (Hebert et al. 1999; De Solla et al. 2016; Gewurtz et al. 2016). The gull has been used in the Great Lakes monitoring program, where more than 40 years

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of contaminant monitoring has provided important information about spatial and temporal contaminant trends, in addition to insights in the use of aquatic birds as monitoring species (Norstrom et al. 1978; Hebert et al. 1999; De Solla et al. 2016). The herring gull has also been included in European contaminant monitoring, such as studies of the German Environmental Specimen Bank, and the Norwegian Environmental Contaminants in an Urban Fjord program (Koschorreck et al. 2015; De Solla et al. 2016; Ruus et al. 2019).

The herring gull is a widely distributed species found in inland and coastal areas of north eastern North America, western Europe, and central Asia (Morris et al. 2003; Olsen 2010), making comparisons of studies across a wide selection of habitats and areas possible. It is an opportunistic species, and it is found in a range of environments, from remote to coastal and terrestrial areas. The herring gull can adapt to feeding in urban environments and is often known to individually specialize on specific diets (Morris et al. 2003; Hebert et al. 2009; Coulson 2015). The herring gull may feed from aquatic, terrestrial, and anthropogenic sources, and dietary items can include fish and aquatic invertebrates, small mammals and birds, plant material and terrestrial invertebrates, and anthropogenic leftovers and waste (Ewins et al. 1994; Morris et al. 2003). Therefore, in urban marine ecosystems, the contaminant status of the herring gull might reflect sources other than the marine food web (Davis et al. 2015).

The common eider (*Somateria mollissima*) is a marine benthic-feeding diver, and it is considered a midtrophic predator, preying mainly on mollusks and other invertebrates (Larsen and Guillemette 2000; Houle et al. 2017). Because of its marine diet, the common eider is expected to serve as a good representative of contaminant exposure in the marine food web (Fenstad et al. 2016), although this is as a benthic midtrophic level feeder, not of the same part of the food web as the mainly fish-eating herring gulls. In Europe, the species has been used as a contaminant indicator for example in the Baltic Sea and the Norwegian coast (Huber et al. 2015; Fenstad et al. 2016). In North America, the common eider and other sea ducks have been long used as contaminant indicators, particularly focusing on metals such as Hg (Vermeer and Peakall 1979; Elliott and Martin 1998; Mallory et al. 2004; Wayland et al. 2005). Although common eiders have low concentrations of POPs compared to other marine bird species (Mallory et al. 2004), it is an accurate indicator of metals, and the importance of studying contaminant levels in the species has been documented (Mallory et al. 2004, 2017; Wayland et al. 2005). The common eider has a circumpolar distribution, with subspecies breeding in areas in the Arctic and boreal zones, although it is not as widely distributed as the herring gull (Cramp 1977; Canadian Wildlife Service 1986). Its habitat ranges from remote sites to urban areas. In several areas, common eider populations decline. In heavily polluted sites, contaminants have been identified as potential contributors to population declines (Franson et al. 2002; Wayland et al. 2003; Matson et al. 2004).

In addition to the choice of species, the choice of matrix is important when designing monitoring studies. Blood and eggs are commonly used as matrices in contaminant monitoring, as their sampling is relatively noninvasive to the adult population, and it is therefore preferred over other destructive sampling methods such as that of liver or brain tissue (Henriksen et al. 1998; De Solla et al. 2016). Blood as a matrix reflects recent exposure to contaminants, and will therefore vary with time since the last feeding and remobilization from other tissues such as stored lipids (Henriksen et al. 1998). Eggs of seabirds have been widely used and studied in contaminant monitoring, and they are recognized as an efficient monitoring matrix (Mineau et al. 1984; Focardi et al. 1988; Morrissey et al. 2010). Egg production reflects the maternal transfer of contaminants from the reproducing mothers to the eggs, and the eggs can reflect female contaminant levels at the time of, or before, egg production (Verreault et al. 2006). As a result, contaminants in eggs can originate from contamination at the breeding site and at the overwintering site (Hitchcock et al. 2019a). Avian maternal transfer of contaminants vary with several factors, including contaminant properties and life history factors such as migration and lipid dynamics (Verreault et al. 2006).

The contaminants' physicochemical properties affect the toxicokinetics and body distribution, resulting in different partitioning of contaminant groups in the tissues and organs. Matrix composition thus influences contaminant occurrence and the effectiveness of monitoring. For example, lipophilic contaminants accumulate in lipid-rich tissues, such as egg yolk, whereas protein-associated contaminants accumulate in protein-rich tissues such as blood, muscle, and liver (Braune and Norstrom 1989; Jones et al. 2003). Thus, the relative composition of contaminants found in blood, and transferred to eggs, is dependent on a compound's affinity for different matrices (Verreault et al. 2006). Furthermore, species differences in tissue distribution and dynamics of lipids can lead to interspecies differences in contaminant occurrence in the same matrix. For example, differing maternal investment and lipid content in the eggs of 2 species can result in different transfers of contaminants from mother to egg (Hitchcock et al. 2019a).

The Oslofjord is a fjord in south eastern Norway surrounded by areas with high population densities, including the capital Oslo in the inner fjord. The inner Oslofjord ecosystem has since 2013 been monitored through the program Environmental Contaminants in an Urban Fjord (Urbanfjord) (Ruus et al. 2019). Water, sediments, and selected organisms representing the marine food web are sampled yearly and analyzed for a range of legacy and emerging environmental contaminants. In addition to marine fish and invertebrates, the sampled biota includes herring gull blood and eggs. The measured contaminants include legacy lipophilic organohalogen contaminants (OHCs) such as the polychlorinated biphenyls (PCBs) and polybrominated

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biphenyl ethers (PBDEs), and emerging nonlipophilic OHCs such as the perflourinated and polyfluorinated substances (PFASs), in addition to Hg.

The aim of the present study is to examine the suitability of the herring gull and common eider as indicator species of the contamination status of the marine food web in the urban inner Oslofjord, and how these species might best be used in environmental monitoring in northern areas. This was addressed by comparing concentrations of organohalogen contaminants and Hg in birds sampled from the inner Oslofjord during the breeding season in May 2017, in relation to trophic niche and position in the marine food web as reflected by stable isotope ratios of C and N; $\delta^{13}C$ and $\delta^{15}N$. To evaluate the birds' representativeness of the marine food web, stable isotopes were also analyzed in a range of marine species representing the inner Oslofjord ecosystem. A matrix comparison of contaminant concentrations in blood and eggs of both bird species was done by evaluating the differences in maternal transfers and considering reproductive physiology. We expected the 2 bird species to belong to different food webs, with a more marine dietary signal in the common eider, and a more terrestrially influenced signal in the herring gull, due to opportunistic feeding. Because of the accumulation of organohalogen contaminants in marine food webs, we expected to find higher concentrations in the common eider. However, we also expected contaminant findings to be influenced by matrix nutrient content and reproductive physiology.

MATERIALS AND METHODS

Study areas and field procedures

The Norwegian capital Oslo is located in the inner Oslofjord, in south eastern Norway at 59°54′N, 10°44′E (see Supplemental Data Figure S1). The Oslofjord is an urban, populated, and polluted area, with a human population of approximately 1.6 million living near the fjord. Decades of emissions from industry and other human impacts such as runoff from agriculture, traffic, marine activities, and sewage have resulted in a marine ecosystem affected by both organic and inorganic contaminants (Ruus et al. 2019). The Oslofjord is affected both by emerging contaminants originating from current urban activities and by legacy contamination from diffuse sources, for example mobilized from the sediments and transported from the terrestrial environment with stormwater (Skarbøvik et al. 2014). The inner fjord is connected to the coastal area Skagerrak by a narrow sound at Drøbak, where a sill depth of 20 m limits the water exchange (Staalstrøm and Røed 2016).

Along with competition for habitats and resources in marine areas, gull species including the herring gull are known to feed in cities and agricultural lands. The common eider has also moved from more coastal habitats into fjords (Systad et al. 2007). The inner Oslofjord supports several seabird colonies in close association with human activities (Bergan and Andersen 2017). The colonies include the herring gull and common eider as important species. The herring gull population was increasing until 2001, and has since seen a decreasing trend (Bergan and Andersen 2017). While some individuals migrate south to areas around the North Sea in winter, others overwinter in Norway (Systad et al. 2007). The common eider population has been increasing since the 1990s (Bergan and Andersen 2017), and it is known to overwinter in Norway (Systad et al. 2007).

Blood and eggs of herring gull ($n = 15$) and common eider ($n = 15$) were collected from colonies in the Inner Oslofjord during the incubation period in May 2017, approximately 3 weeks after egg laying. The herring gulls were caught using walk-in traps placed over the nests. As the gull entered the trap the door closed, leaving the bird trapped on its nest, ensuring that the sampled egg belonged to the sampled female. Both male and female herring gulls incubate the eggs, but blood samples were only collected from females. Common eider females were caught using hand nets on the nests. For both species, measurements of head length, wing length, and body weight were registered. The sex of the birds was determined based on plumage (eiders) or head length (herring gulls). A blood sample (5 mL) was drawn from the branchial vein of females using a syringe flushed with heparin, and eggs were collected from the female's nest. The eggs were frozen as quickly as possible after collection. The permit for bird sampling was granted by the Norwegian Food Safety Authority (FOTS id 12394), and sampling was performed following the Norwegian Animal Welfare Act. In addition, marine organisms representing the Inner Oslofjord food web were sampled: polychaetes, krill (Euphausiacea), prawn (*Pandalus borealis*), blue mussel (*Mytilus edulis*), herring (*Clupea harengus*), and cod (*Gadus morhua*). Collections of the marine food web species were done from the R/V Trygve Braarud, using a van Veen grab for polychaetes, a fry trawl for krill, benthic trawl for prawns, hand picking, rake or snorkeling for mussels, and trawl for herring and cod. Further details on this sampling are described in the report Environmental Contaminants in an Urban Fjord 2017 (Ruus et al. 2019). The polychaetes constituted several species (see Supplemental Data Table S1). The muscle of 15 cod was sampled for individual analysis, and for all of the other marine organisms, material for 3 pooled samples was collected (see Supplemental Data Table S2).

Sample preparation and contaminant analyses

Details of all the analyses and quality assurance can be found in the Supplemental Data. Briefly, eggs were visually classified according to their level of development and homogenized at the Norwegian Institute for Water Research (NIVA), Oslo, Norway. Homogenized eggs and whole blood samples were individually analyzed for concentrations of lipophilic OHCs and Hg at the Norwegian Institute of Air Research (NILU), and PFAS at NIVA. In total, 97 compounds were targeted in the chemical analysis. The lipophilic OHCs were extracted using cyclohexane and acetone solvents and 2 silica columns and cleaned using a sulfuric acid rinse. For

instrumental analysis, a gas chromatography high resolution mass spectrometer (Waters Autospec) was used. The Hg was extracted and cleaned using supra pure nitric acid and digested at high pressure and temperature in a microwave-based digestion unit (UltraClave, Milestone, Italy). An Agilent 7700x inductively coupled plasma mass spectrometer (ICP-MS) was used for instrumental analysis. The PFASs were extracted using acetonitrile before dilution with an ammonium acetate buffer. Instrumental analysis was performed using acquity ultra-performance liquid chromatography (UPLC) connected with a mass spectrometer (Waters, XEVO G2-S QTOF). The lipid content of the samples was determined gravimetrically at NILU using cyclohexane and acetone (3:1) solvent. Analyses were performed according to accredited methods, using recovery standards, blanks, and limits of detection (LODs). The NILU lab is accredited for the analysis of PCBs by Norwegian Accreditation (ISO/IEC 17025). For the PBDEs, the same quality assurance procedures as for the accredited compounds were applied. The NIVA's laboratory is accredited by Norwegian Accreditation ISO/IEC 17025. The NIVA lab is not accredited for these particular compounds, but documentation, preparation, analysis, and calculations are performed in accordance with accredited methods.

Included contaminants

The LOD was defined as 3 standard deviations of the mean blank response. Prior to statistical analyses, contaminants with fewer than 75% (PCBs and PBDEs) or 70% (PFAS) of the values above the detection limit were removed, and nondetects in remaining contaminants were replaced by β distribution multiple imputation ($\alpha = 5$ and $\beta = 1$) (Helsel 2010). For lipophilic OHCs, 28 congeners were included (out of 67), and for PFAS, 6 of 29 analyzed contaminants were included (see Supplemental Data Figure S2). Data treatment was performed using R v.3.4.2 (R Core Team 2017), and the Vegan package v.2.5-6 (Oksanen et al. 2018) for multivariate analyses.

Dietary descriptors and body condition

All of the samples (whole blood and eggs of both bird species, and all marine food web organisms) were analyzed for stable isotopes of C ($\delta^{13}C$) and N ($\delta^{15}N$) at the Institute for Energy Technology (IFE). Samples were combusted in a Eurovector EA3080 elemental analyzer and separated on a 2 m Poraplot Q gas chromatography column. The N_2 and CO_2 were transferred directly to a Horizon Isotope Ratio Mass Spectrometer (IRMS) from Nu-instruments for determination of the isotopic ratios of $^{13}C/^{12}C$ and $^{15}N/^{14}N$. The lipid was not extracted before the stable isotope analysis.

The ^{13}C fraction is depleted in the C found in the lipids, resulting in more negative $\delta^{13}C$ values in the lipid-rich tissue (DeNiro and Epstein 1977). This is in particular an issue in matrices where the C-to-N ratio is greater than 3.5 (Post et al. 2007). The mean C-to-N ratio was 7.21 for herring gull eggs, and 9.93 for common eider eggs. To investigate the impact of the differences in lipid content between the eggs of the herring gull and the eggs of the common eider on the species comparison, the $\delta^{13}C$ egg data was corrected for lipid content using the C-to-N ratio, according to the model of Elliott et al. (2014) for whole egg homogenate. The lipid correction slightly decreased our $\delta^{13}C$ in both species, but it did not alter the interpretation of the results (see Supplemental Data Figure S3). Because of this, and that the algebraic lipid correction of the $\delta^{13}C$ values is not always consistent (Elliott et al. 2014), the uncorrected values were used in the final evaluation of the ecological niche. Because of the low C-to-N ratio (Table 1), the lipid correction was not investigated regarding $\delta^{13}C$ in the blood.

As a measure of the individual bird's body condition, and to control for variation in body size among individuals, a body condition index (BCI) was calculated using a residual index method (Jakob et al. 1996). The BCI was expressed as standardized residuals of the points in a linear regression of head length (herring gull) and wing length (common eider) against mass. Head length and wing length were chosen as parameters based on Pearson correlation tests, and standardized residuals were used to ensure comparability between the species even though different body structures were used. Full details on the BCI calculations can be found in the Supplemental Data.

Data analyses

Differences between the concentrations of contaminant groups in the blood and eggs of herring gull and common eider were assessed using Welch's *t*-test. All of the lipophilic OHCs were tested together as a group, on both wet weight and lipid weight bases, and the PFASs together as another group on a wet weight basis. Multivariate analysis by ordination was used to identify the interrelationships among response variables, contaminant concentrations, and associations to explanatory variables. The PCB congeners were grouped in homologue groups according to chlorination degree. The full set of explanatory variables included were $\delta^{13}C$, $\delta^{15}N$, BCI, lipid content, and species for blood, in addition to egg weight and egg stage for eggs.

To identify the differences between species, multivariate analysis was performed for the matrices separately, and for lipophilic and nonlipophilic compounds separately. A biplot of a principal component analysis (PCA) was used to identify the structure in logarithmic contaminant concentrations between species. As the lipid content was a significant explanatory variable, and to control for the effect of lipid content on the lipophilic compounds, PCAs were done with and without lipid as a covariable for the lipophilic OHCs. To determine the relationships to, and explanatory power of, explanatory variables, multivariate forward model selection and variance partitioning using Monte-Carlo permutation tests in redundancy analysis (RDA) was performed. The explanatory power of a constrained component was expressed as the model inertia (RDA) divided by the total inertia of the unconstrained model (PCA). Maternal transfer factors were calculated as logarithmic ratios of the mean wet weight

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Table 1. Explanatory variables (body condition index, egg stage, egg weight, lipid content, $\delta^{13}C$, $\delta^{15}N$, and C-to-N ratio) and contaminant concentrations (wet weight (ng/g) for OHCs, dry weight (ng/g) for Hg) in blood and eggs of herring gull and common eider

	Herring gull						Common eider					
	Blood[a]			Egg[b]			Blood[a]			Egg[b]		
	Mean[c] ± SD[c]	Median[c]	Range	Mean[c] ± SD[c]	Median[c]	Range	Mean[c] ± SD[c]	Median[c]	Range	Mean[c] ± SD[c]	Median[c]	Range
BCI	−0.030 ± 1.04	0.044	−1.63–2.39	—	—	—	0.010 ± 1.02	0.017	−1.72–1.89	—	—	—
Egg stage	—	—	—	3.26 ± 1.09	4	1–5	—	—	—	2.46 ± 1.18	2	1–4
Egg weight[d]	—	—	—	78.6 ± 10.6	79.9	61.8–97.7	—	—	—	105 ± 8.17	106	91.2–119
Lipid[e]	2.28 ± 1.51	2.00	0.60–5.30	8.01 ± 1.20	7.90	5.92–9.97	0.38 ± 0.25	0.35	0.14–1.25	18.4 ± 1.20	18.1	16.9–21.0
$\delta^{13}C$[f]	−24.4 ± 0.351	−24.3	−25.1–24.0	−25.7 ± 0.771	−25.8	−26.9–24.6	−20.7 ± 1.41	−21.0	−22.5–18.1	−21.2 ± 0.951	−22.4	−23.3– −9.78
$\delta^{15}N$[f]	8.44 ± 0.725	8.41	7.39–10.1	8.91 ± 1.08	8.93	6.92–10.8	12.4 ± 0.61	12.4	11.1–13.6	12.6 ± 0.811	12.4	11.4–14.3
C-to-N ratio	4.09 ± 0.596	3.93	3.41–5.30	7.21 ± 1.54	7.41	3.97–9.10	4.44 ± 0.692	4.62	3.55–5.85	9.93 ± 0.588	9.78	9.21–11.5
HCB[g]	0.422 ± 0.314	0.300	<LOD–1.27	3.66 ± 2.61	2.52	0.72–9.35	2.92 ± 0.72	2.95	1.80–4.17	0.246 ± 0.118	0.2	0.10–0.42
∑triCB[g]	0.087 ± 0.051	0.062	<LOD–0.240	1.04 ± 1.38	0.600	0.13–5.75	1.89 ± 0.995	1.63	0.88–4.11	0.156 ± 0.112	0.12	<LOD–0.48
∑tetraCB[g]	1.55 ± 1.63	0.940	<LOD–6.18	15.8 ± 19.5	8.93	0.61–65.3	18.3 ± 12.3	13.2	7.25–49.1	2.17 ± 2.46	1.19	0.51–9.68
∑pentaCB[g]	3.75 ± 2.88	2.81	<LOD–10.1	53.4 ± 54.4	38.2	3.6–213	54.9 ± 34.1	46.3	22.1–137	4.18 ± 4.96	2.48	0.79–19.0
∑hexaCB[g]	10.3 ± 9.48	8.98	<LOD–40.8	132 ± 101	102	16.5–384	104 ± 60.6	87.5	48.8–263	8.52 ± 9.85	4.83	<LOD–37.7
∑heptaCB[g]	2.93 ± 2.07	2.27	<LOD–9.27	48.9 ± 30.4	46.9	8.94–120	28.8 ± 18.6	25.0	12.2–81.1	3.04 ± 3.41	1.75	<LOD–13.8
∑higherCB[g]	0.358 ± 0.315	0.260	<LOD–1.30	6.06 ± 4.28	4.98	1.36–16.0	1.68 ± 1.58	1.09	0.44–6.86	0.150 ± 0.192	0.080	<LOD–0.780
∑PCB[g]	19.0 ± 15.6	15.2	<LOD–65.1	257 ± 203	200	31.5–790	210 ± 126	160	92.6–543	18.2 ± 20.8	9.54	<LOD–81.8
∑BDE[g]	0.785 ± 0.890	0.363	<LOD–3.49	17.0 ± 30.3	7.00	<LOD–114	1.11 ± 0.742	0.942	0.646–3.67	0.149 ± 0.084	0.121	<LOD–0.413
∑PFAS[g]	14.1 ± 7.77	12.3	<LOD–31.3	—	—	—	20.0 ± 10.2	16.6	<LOD–45.2	—	—	—
Hg[h]	4.26 ± 0.717	4.51	2.89–5.66	3.92 ± 0.765	4.13	2.37–5.12	4.94 ± 0.438	4.93	4.30–5.79	5.15 ± 0.49	5.03	4.53–5.97

<LOD = below limit of detection; SD = standard deviation; Hyphens = values not measured or not applicable.
[a] Whole blood.
[b] Whole egg homogenate.
[c] Data including imputed values.
[d] Grams.
[e] %.
[f] ‰.
[g] ng/g (w.w.).
[h] ng/g (d.w.).

egg-to-blood ratio concentrations of individual congeners to quantify the transfer of contaminants from female to egg (Grønnestad et al. 2017). A maternal transfer factor with log-ratio greater than 0 indicates higher levels in eggs than in blood, assuming the blood concentrations are similar at the sampling time and at the egg production time.

RESULTS

Carbon source and trophic status

We observed a separation of the stable isotope signatures between the herring gulls and common eiders, with higher $\delta^{13}C$ and $\delta^{15}N$ in the eiders (Figure 1; Welch's *t*-test: $\delta^{13}C$-to-blood ratio: $t = -9.8331$, $p < 0.0001$, eggs: $t = -11.329$, $p < 0.0001$; $\delta^{15}N$: blood: $t = -16.257$, $p < 0.0001$, eggs: $t = -10.607$, $p < 0.0001$).

Contaminant concentrations in herring gull and common eider blood

We found higher concentrations of the lipophilic OHCs in the blood of common eider than in the blood of herring gull, with variation due to lipid content included and removed (Figure 2A and Supplemental Data Figure S4). We also found higher dry weight concentrations of Hg in the blood of common eider than of herring gull, although the difference between the species was smaller for Hg than for the lipophilic OHCs (Figure 2B and Table 1). There was no general species difference in the wet weight blood concentrations of the analyzed PFAS (Figure 2B). The 3 compounds, PFDA, PFUdA, and PFHxS, showed higher concentrations in the common eider, whereas PFTrDA was higher in the herring gull, and there was no difference between the species for PFOS and PFDoA (Figure 2B). Lipophilic OHCs, protein affiliated Hg, and the PFASs with higher concentrations in common eider were positively correlated with $\delta^{15}N$ and $\delta^{13}C$ in the blood (Figure 2A and B).

Maternal transfer and contaminant concentrations in herring gull and common eider eggs

In the eggs, we found higher concentrations of lipophilic OHCs in herring gull than in common eider, with and without variation due to lipid removed (Figure 3 and Supplemental Data Figure S5). For all lipophilic congeners, the maternal transfer factors were >0 in herring gull, and <0 in common eider (Figure 4). The same was observed when the lipophilic contaminants were expressed on a lipid weight basis (see Supplemental Data Figure S6). For Hg, maternal transfer factors were close to 0 in both species (Figure 4).

Effects of lipid content

Even though the lipid content was a significant explanatory variable, our study had the same outcome for lipid weight normalized and wet weight concentrations, as well as when accounting for lipid as a covariate in the PCA. Lipid content in the blood of herring gull (mean lipid content 2.28%) was higher than in the blood of common eider (mean lipid content 0.38%) (Welch's *t*-test: $t = 4.7642$, $p = 0.00026$; Table 1); however, the difference was small compared to the difference in eggs, where common eider (mean lipid content 18.4%) had more than twice as high of a lipid content than herring gull (mean lipid content 8.01%) (Welch's *t*-test $t = -23.608$, $p < 0.0001$; Table 1).

Detected contaminants

Following the concentration trends, the highest number of contaminants was detected in herring gull eggs and common eider blood, which had 46 and 43 lipophilic OHCs detected in more than 75% of the samples, respectively (see Supplemental Data Table S2). We found 22 (herring gull blood) and 17 (common eider eggs) lipophilic OHCs detected in more than 75% of the samples (see Supplemental Data Table S2). For the PFASs, there were 4 contaminants in herring gull blood and 5 in common eider blood detected in more than 70% of the samples (see Supplemental Data Table S3).

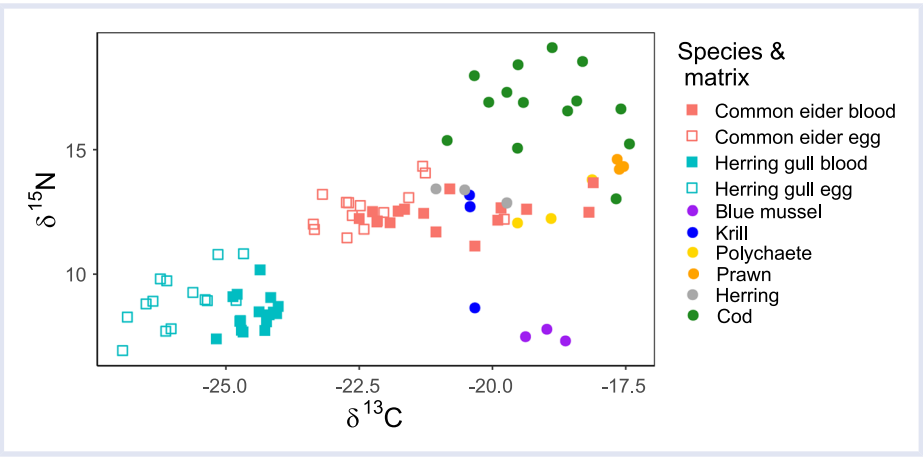

Figure 1. Scatterplot of the stable isotope values of blood and eggs of herring gull and common eider and the marine food web as sampled in the Urbanfjord program.

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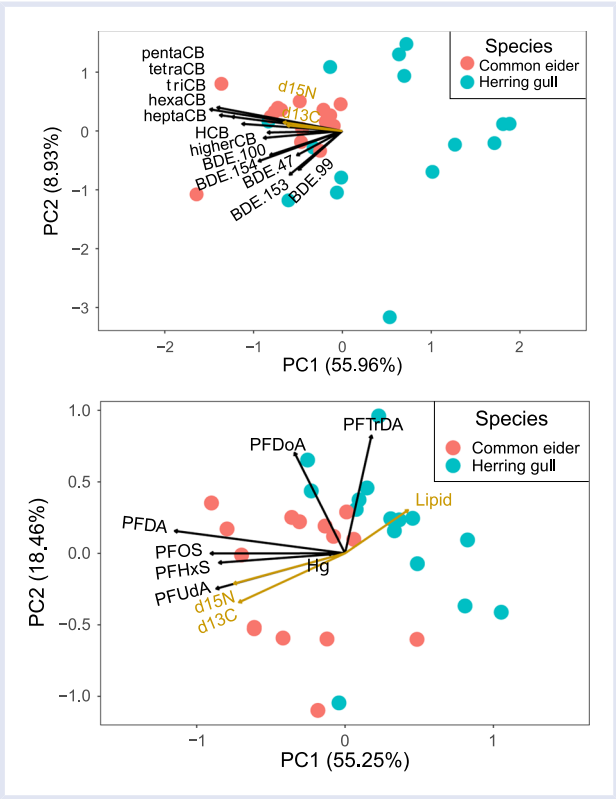

Figure 2. Principal component analysis (PCA) biplot of log concentrations (wet weight) of lipophilic organohalogen contaminants (OHCs) with lipid as the conditioned variable (**A**) and protein-associated contaminants (**B**) in the blood of herring gull and common eider. Individual birds are presented as points and colored according to the explanatory variable species. Contaminant concentrations (response variables) are presented as black vectors. Significant explanatory variables are fitted passive brown vectors. Percentage of total variation explained by each principal component is shown on the axis and expressed as the eigenvalue of the component divided by total variation in the model.

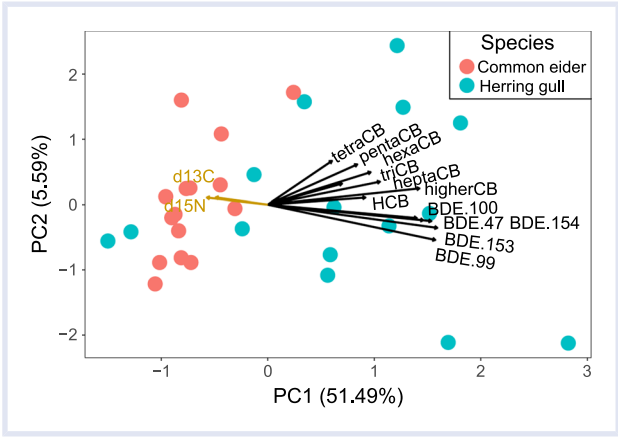

Figure 3. Principal component analysis (PCA) biplot of log concentrations (wet weight) of lipophilic contaminants in the eggs of herring gull and common eider with lipid as the conditioned variable. Individual birds are presented as points and colored according to the explanatory variable species. Contaminant concentrations (response variables) are represented as black vectors. Significant explanatory variables are fitted passive brown vectors. Percentage of total variation explained by each principal component is shown on the axis and expressed as the eigenvalue of the component/total variation in the model.

DISCUSSION

Carbon source and trophic status

Organic material of terrestrial origin is more depleted in $\delta^{13}C$ than organic material of marine origin, and $\delta^{15}N$ increases with trophic position, providing a proxy for ecological niche (Peterson and Fry 1987). The $\delta^{13}C$ signal of the common eider was similar to that of organisms of the marine food web, reflecting a marine C source of the common eider and placing it within the marine food web (Figure 1). The lower $\delta^{13}C$ in herring gull indicates a more terrestrial C source, supporting our expectation of herring gull being influenced by dietary sources other than the marine food web (Peterson and Fry 1987). Based on the $\delta^{15}N$ signal, the Oslofjord common eider fits the description as a midtrophic predator. As the species reflect different dietary sources, a direct comparison of trophic position is not possible. The low $\delta^{15}N$ values of the herring gull could indicate feeding from low trophic levels such as marine invertebrates. However, as the $\delta^{13}C$ indicates terrestrial influence on the herring gull diet, their $\delta^{15}N$ signal is likely a result of a mix of items of different trophic status and origin, resulting in different food-web baselines for the 2 species. The placement of the common eider in the marine food web is in accordance with our expectations and indicates that the common eider is a good representative of pollution in the Oslofjord midtrophic marine ecosystem.

Blood of herring gull and common eider as urban marine contaminant indicators

The higher concentrations of lipophilic organohalogen compounds and Hg in the blood of common eider than in the blood of herring gull is likely due to the common eiders feeding from the marine food web, where PCBs, PBDEs, and Hg are known to accumulate. (Fisk et al. 2001; Savinov et al. 2003; Borgå et al. 2004). The higher concentrations of lipophilic OHCs and Hg in blood of common eider compared to that of herring gull illustrate the difference between these urban birds and more marine-influenced gulls. In the Arctic, where marine feeding gulls occupy a higher trophic position compared to eiders, higher contaminant concentrations have been found in the former, due to the higher trophic position of the gulls and feeding in different parts of the food web (Savinov et al. 2003; Haarr et al. 2018). In urban areas, contaminant concentrations in gulls have been shown to increase with the proportion of marine prey in the diet (Santos et al. 2017). This supports the lower contaminant concentrations in the terrestrially influenced urban gulls in our study. Thus, for herring gull to be considered as an indicator species of contamination in routine monitoring programs, dietary parameters should also be included in the study to evaluate the ecological niche of the population to ensure that the investigated food web is the same as the one reflected by the contaminant concentrations of the bird (Davis et al. 2017). In addition to stable

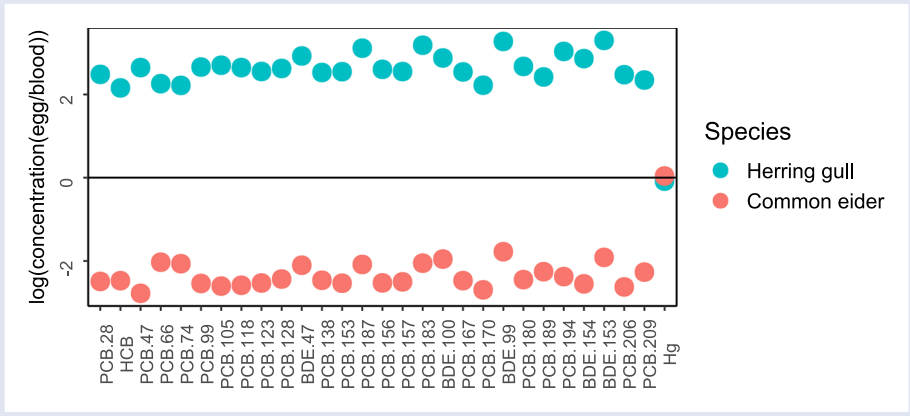

Figure 4. Maternal transfer factors of lipophilic organohalogen contaminants (OHCs) and Hg for the herring gull and common eider. The MTF is the Log ratio of (mean wet weight concentration in eggs)-to-(mean wet weight concentration in blood). Ratio = 0 indicates equilibrium in transfer of contaminants from mother to egg, ratio >0 indicates higher contaminant concentrations in eggs as compared to the blood of the mother, and ratio <0 indicates higher concentrations in the blood of the mother compared to the egg. Lipophilic congeners are sorted by K_{OW} value, increasing left to right.

isotopes, bird tracking and additional methods for studying seabird diet, such as analyzing regurgitated food, amino acid stable isotopes, fatty acid composition, and measuring DNA in the feces, will give more thorough information about the role of urban gulls as indicator species (Hebert et al. 2006; Deagle et al. 2007; Davis et al. 2015; Hebert et al. 2016; Sorais et al. 2020). We found no clear species difference in the protein associated PFAS concentrations between the herring gull and the common eider. Although some PFASs are being restricted and phased out due to environmental and toxicological effects, other compounds are still in use and are being distributed in the urban environment (Gebbink et al. 2011; Gewurtz et al. 2016). Furthermore, the phased-out PFASs have been restricted relatively recently, such as PFOS, which was implemented as a POP in the Stockholm convention in 2009. Recently restricted compounds are likely still present in consumer products currently being used and distributed in the environment, making urban areas the main source of PFOS in the environment (Gewurtz et al. 2016). All of the PFASs included in the analysis in the current study are included on the list of Priority Substances in Norway (Norwegian Environment Agency 2018), with goals of restricting or reducing use and emissions. They have, however, only been added within the last 20 years, from PFOS in 2002 to PFHxS in 2020. As a result of current emissions and recent restrictions, we also expect to find these substances in relation to terrestrial and urban feeding (Gebbink et al. 2011a, 2011b; Gewurtz et al. 2016; Heimstad et al. 2018). For example, in the Oslo area, PFAS have been found in earthworms, which can be a part of the herring gull diet (Heimstad et al. 2018). Higher levels of PFAS associated with increasing urbanization have been found in common eider eggs (Herzke et al. 2009). However, PFASs are also known to accumulate in the marine food web (Tomy et al. 2004; Houde et al. 2006), and PFASs are observed in higher levels offshore bird species as compared to coastal bird species (Miller et al. 2015). As reflected by our results, PFAS exposure in gulls cannot be directly linked to terrestrial feeding. Our contaminant findings and the dietary descriptors indicate that the herring gull might not represent the contaminant status of the marine food web in urban areas. However, by comparing the concentrations of legacy, diffuse source contaminants with emerging, point source contaminants still in use, the herring gull could be a good indicator of urban contamination, integrating the urban ecosystem from the fjord through the city to waste dumps and agriculture lands.

Blood and eggs as monitoring matrices of legacy and emerging contaminants of concern

We found higher maternal transfer ratios of lipophilic OHCs in herring gulls than in common eiders. Similar results to the ones in our study were also observed in a study of herring gull and common eider eggs on the urban Swedish west coast, south of the Oslofjord, which found higher ∑PCB and ∑PBDE in the eggs of herring gulls as compared to the eggs of common eider (Carlsson et al. 2011). This species difference could indicate that a higher amount of these contaminants is transferred to the eggs of herring gulls than to the eggs of common eider, relative to concentrations in mothers. However, the breeding strategy also contributes to the observed species differences. During incubation, female eiders rarely leave the nest, and they fast during the breeding period, losing about 25% to 40% of their body weight during incubation (Korschgen 1977; Gabrielsen et al. 1991). To cover their energetic requirements during breeding, they store large lipid reserves prior to breeding, which are then mobilized during the fast (Korschgen 1977). This mobilization of stored energy results in remobilization of contaminants from the lipid reserves to the bloodstream, and the wet weight concentration of PCB-153 in the blood of Arctic breeding common eiders has previously been found to increase 3.6-fold during the breeding period (Bustnes et al. 2010). As a result of remobilization, the contaminant concentrations in eider blood sampled during

the incubation might not reflect contaminant levels in the recent diet alone. In our study, we collected blood and eggs at the same time, late in the breeding period. This means that while the eggs represent the contaminant status at the time of egg laying, our blood samples represent the contaminant status after fasting and remobilization in the common eider. The remobilization introduces a caveat in using the common eider as an indicator species. In contrast, herring gull blood is not affected by the remobilization of lipids and contaminants to the bloodstream to the same degree, and contaminant concentrations in herring gull blood will better reflect contaminants assimilated recently.

For Hg, the maternal transfer factors in both species were close to 0, indicating equilibrium in the Hg concentration between the blood and eggs. This is also in accordance with the remobilization explanation, as Hg is not lipid-associated and thus not remobilized to the blood stream along with the lipids. Furthermore, protein is thought to be the limiting nutrient in egg production, and maternal transfer is higher for lipophilic than for protein-associated contaminants (Mallory et al. 2004; Hitchcock et al. 2019a).

Contaminant occurrence is linked to nutrient composition, and differences in matrix quality are also important to consider when choosing a monitoring matrix. Because of high lipid solubility, lipid content is of special concern for lipophilic contaminants. Our study had the same outcome for lipid weight normalized and wet weight concentrations, and when accounting for lipids as a covariate in the PCA, which indicate robust results. The difference in the lipid content of the eggs of the 2 species may reflect the more precocial breeding strategy of the eiders, which have a greater maternal investment, including investing more lipids in their clutch than the herring gull (Hitchcock et al. 2019a). The variable numbers of detected contaminants in the matrices, following the concentration trends, shows that the number of lipophilic OHCs detected does not follow the lipid content of the matrices but is likely more affected by the remobilization of contaminants to the blood stream during incubation in the common eider.

A consideration when using seabird eggs as a contaminant monitoring matrix is the origin of the egg constituents (Hitchcock et al. 2019b). Depending on the reproductive life history strategy, females can invest a greater degree of stored energy (capital breeding), or energy recently acquired (income breeding) in reproduction (Stephens et al. 2009). It is likely that the eggs of species investing more stored energy in reproduction can be more influenced by contaminants with origins outside of the breeding area, for example acquired on the overwintering grounds. The eggs of birds investing more recently acquired energy might reflect to a higher degree the contamination status of the breeding area. While placing species strictly in 1 of the 2 categories is not realistic, common eiders can be considered to be closer to a capital breeding strategy than the herring gull (Sénéchal et al. 2011). This means that the contaminant status of eider eggs can represent the contaminant status of the mother and thus the environment over a longer timescale than herring gull eggs. In the Oslofjord, the eiders arrive early in spring, in March to April, and resources required prior to breeding are thus expected to reflect local contamination. However, in areas where egg production might be more influenced by nutrients from overwintering grounds, this possible bias should be considered before using common eider eggs for monitoring.

The difference in maternal transfer factors for lipophilic OHCs between herring gull and common eider demonstrates that the blood and eggs of these species (and probably other seabirds) should only be included as monitoring matrices with careful consideration of the ecology and reproductive physiology of the species. This illustrates the importance of a robust monitoring design, as a study on only 1 matrix or species would lead to opposite conclusions on the contaminant status of the 2 species.

Implications for monitoring of contaminants

The common eider is relevant as a contaminant indicator of marine ecosystems in monitoring programs in northern urban areas as it reflects the marine food web. As it is a benthic midtrophic level feeder, the common eider does not necessarily represent the same food chain as fish-eating herring gulls, indicating that the species is not a replacement of the herring gull as an indicator species, but rather an addition. Because of relatively low POP levels, the common eider might be less efficient in measuring POPs than other seabird species, and its breeding cycle provides challenges for interpreting results. Therefore, inclusion of this species in monitoring programs requires careful data analysis. As it is a marine feeder, the common eider is not relevant for monitoring fresh-water systems. Furthermore, it is not as widely distributed as the herring gull, and in many areas where the herring gull is used as an indicator species, the common eider is not present, and therefore not relevant for monitoring. The common eider population is threatened in several areas, which limits the possibilities of using it as indicator in regular monitoring programs.

The herring gulls in the Oslo area are not representative of the marine food web of the Oslofjord, but appear to be a good indicator for urban pollution in general, providing the possibility of integrating different ecosystems into a monitoring program. This is interesting, for example, for studying the occurrence and distribution of contaminants of emerging concern (Gebbink et al. 2011b; Chen et al. 2012; Brown et al. 2019). The different roles of the herring gull and the common eider show the advantage of a multispecies approach for a thorough picture of contaminant status in urban marine systems. Our study showed that blood and eggs should be used as monitoring matrices in combination, illustrated by the opposite conclusions that would be drawn in the present study if only 1 matrix was assessed separately. Further, doing contaminant monitoring in cooperation with bird population monitoring would give useful insights, for example, as data on life history and migration would be available to consider in the evaluation of contaminant data. Ecological studies such as monitoring bird populations

should also be completed, as life history and migration data greatly help in interpreting contaminant data.

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Disclaimer—The authors declare no conflicts of interests.

Data Availability Statement—Data and associated metadata and calculation tools are available upon request by contacting corresponding author Katrine Borgå (katrine.borga@ibv.uio.no).

SUPPLEMENTAL DATA

Figure S1. Map of the Inner Oslofjord, and location within Norway. The capital Oslo is situated in the innermost part of the area, and samples were taken in the same area. Yellow areas indicate areas of high population density. The map was generated in R using data from the Norwegian Mapping Authority under CC BY 4.0 license.

Figure S2. Percent detected values in all analyzed lipophilic congeners. The vertical line illustrates the censoring level at 75% detected values.

Figure S3. Percent detected values in all analyzed PFASs. The vertical line illustrates the censoring level at 70% detected values.

Figure S4. Scatterplot of stable isotope values of blood and eggs of herring gull and common eider and the marine food web as sampled in the Urbanfjord program, with lipid corrected values of $\delta^{13}C$ for eggs of herring gull and common eider included. The $\delta^{15}N$ is shown on the y-axis, $\delta^{13}C$ on the x-axis.

Figure S5. PCA biplot of log concentrations (wet weight) of lipophilic OHCs in the blood of herring gull and common eider, with variation due to lipid included (no conditioned variable). Individual birds are presented as points and colored according to the explanatory variable species. Contaminant concentrations (response variables) are presented as black vectors. Significant explanatory variables are fitted passive brown vectors.

Figure S6. PCA biplot of log concentrations (wet weight) of lipophilic OHCs in eggs of herring gull and common eider with variation due to lipid included (no conditioned variable). Individual birds are presented as points and colored according to the explanatory variable species. Contaminant concentrations (response variables) are represented as black vectors. Significant explanatory variables are fitted passive brown vectors.

Figure S7. The log ratio of (mean concentration in eggs)-to-(mean concentration in blood) in lipophilic OHCs and Hg for herring gull and common eider. Lipophilic OHCs are in lipid weight, and Hg is in wet weight. Ratio = 0 indicates equilibrium in transfer of contaminants from mother to egg. Lipophilic congeners are sorted by K_{OW} value, increasing left to right.

Table S1. Species constituting polychaete samples (grams of each species)

Table S2. Overview of samples of marine food web organisms

Table S3. Lipophilic OHCs detected in more than 75% of samples per matrix, sorted by amount of censored values (high-low)

Table S4. PFAS compounds detected in more than 70% of samples per matrix, sorted by the amount of censored values (high-low)

Table S5. Individual concentrations (wet weight) of PCBs and HCB per sample, and the sum for each contaminant per matrix. Concentrations under the detection limit are marked with < and are not included in the sums

Table S6. Individual concentrations (wet weight for PBDEs and PFASs, dry weight for Hg) of PBDEs, PFASs, and Hg per sample, and the sum for each contaminant per matrix. Concentrations under the detection limit are marked with < and are not included in the sums.

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