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Maternal transfer and occurrence of siloxanes, chlorinated paraffins, metals, PFAS and legacy POPs in herring gulls (*Larus argentatus*) of different urban influence

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ABSTRACT

Urban herring gulls (*Larus argentatus*) are exposed to contaminants from aquatic, terrestrial and anthropogenic sources. We aim to assess if differences in urbanisation affect ecological niche and contaminant concentrations in female herring gulls. Furthermore, we investigated maternal transfer from mothers to eggs for all the target compounds, including chlorinated paraffins (CPs) and cyclic volatile methyl siloxane (cVMSs), which to our knowledge have not been assessed in herring gulls previously. We compare concentrations of legacy and emerging contaminants and metals in blood and eggs between two herring gull colonies located 51 km apart, in the urban influenced Norwegian Oslofjord. While both colonies are within an urbanised area, the inner fjord is more so, as it is surrounded by Oslo, the capital and largest city in Norway Stable isotopes of carbon and nitrogen indicated a more marine ecological niche in the outer than the inner fjord colony, although with overlap. Persistent organic pollutant (POP) concentrations were similar in the inner and outer fjord colonies, while the short-chained chlorinated paraffins (SCCP), which are recently added to the Stockholm convention and contaminants of emerging concern (CECs) varied, with higher concentrations of SCCP and the cVMS decamethylcyclopentasiloxane (D5) in females and eggs of the inner fjord colony. Per- and polyfluorinated substances (PFAS) concentrations were higher in the outer fjord colony, likely linked to releases from a point-source (airport and waste management facility with open access to food waste). In blood, chlorinated paraffins contributed most the total lipophilic contaminants (inner: 78%, outer: 56%), while polychlorinated biphenyls (PCBs) were the most abundant lipophilic contaminants in eggs (inner: 62%, outer: 46%). Dechloranes and brominated flame retardants (BFRs) were detected in few samples. Maternal transfer, assessed by egg to blood ratios, of cVMSs were similar to the POPs with mean log ratio 0.39 (D5), while it was lower for SCCPs, with log ratios-0.77. Our results indicate comparable POP exposure of the herring gulls in the inner and outer Oslofjord, likely due to overlap in ecological niches between the colonies and wide distribution of POPs. The differences between the colonies in concentrations of PFAS, cVMS and CPs shows that point source exposures and urban influence may be more important than ecological niche for these compounds.

1. Introduction

Human activities have led to the release of a wide range of contaminants to the environment, both human-made chemicals and naturally occurring elements. One prominent example is persistent organic pollutants (POPs), which can lead to adverse effects in wildlife (Brogan et al., 2017). Legacy POPs are regulated and well investigated, and present-day contamination is a legacy of previous releases. Contaminants of emerging concern (CEC) are either newly introduced compounds and their metabolites, or compounds with previously

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unrecognized adverse effects on the ecosystem, and are currently not regulated. Chlorinated paraffins (CPs) and cyclic volatile methyl siloxanes (cVMSs) are compounds currently receiving increased research attention. CP groups include complex mixtures of polychlorinated n-alkanes of differing length used in industry, commonly as plasticisers and flame retardants. Although the short chained chlorinated paraffins (SCCPs) have been listed under Annex A of the Stockholm Convention, they are still being produced and used around the world. In addition, other CP groups, medium chained and long chained chlorinated paraffins (MCCP and LCCP) are currently unregulated and in use worldwide, and can be considered CECs. cVMS concentrations have been linked to human settlements and urban influence (McGoldrick et al., 2014; Warner et al., 2010), as a result of present use in e.g. personal care products.

As a result of biomagnification, aquatic top predators like seabirds often have high concentrations of legacy and emerging contaminants (Elliott et al., 2009; Hebert et al., 1999; Huber et al., 2015). Toxicokinetics and body distribution of contaminants is dependent on their physicochemical properties, with lipophilic contaminants accumulating mainly in lipid rich tissues and organisms, and protein-associated contaminants accumulating to a higher degree in protein rich tissues (Jones et al., 2003). Lipophilic contaminants include the cVMSs and CPs, and legacy POPs such as polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs) and hexachlorobenzene (HCB). Protein-associated contaminants include the organic per- and polyfluorinated substances (PFASs), in addition to several metals. Metals are elements and thus non-biodegradable, and can be found throughout the environment. Several metals such as cadmium (Cd), lead (Pb) and mercury (Hg) are bioaccumulative with high concentrations in birds (Borgå et al., 2006; Campbell et al., 2005). Both organic compounds and inorganic trace-elements can cause adverse effects even at low levels of exposure (Brogan et al., 2017; Vizuete et al., 2019).

The herring gull *(Larus argentatus)* is a widely distributed aquatic bird species, and has been included in monitoring programs such as the Canadian Great Lakes Monitoring Programme and the Norwegian Environmental Contaminants in an Urban Fjord programme (Urbanfjord), to reflect the aquatic food web contaminant status (Hebert et al., 1999; Ruus et al., 2019). As a result of a long tradition of using herring gull as a monitoring species, there exists a large amount of data and knowledge on ecology and contaminants in the species (Hebert et al., 1999; Hebert et al., 2009). The herring gull is a generalist species with a wide ecological niche. In coastal areas, they are often observed to feed from terrestrial sources such as human waste in addition to marine and fresh-water organisms (Hebert et al., 2009). In the Norwegian Oslofjord, a weaker marine dietary signal was identified in herring gulls compared to other marine species in the urbanised inner fjord area (Thorstensen et al., 2020). This suggests that the herring gull searches for food in the terrestrial ecosystems surrounding the fjord, and that the inner Oslofjord herring gulls are not representative indicator species of the marine ecosystem of the fjord (Thorstensen et al., 2020). Shifting from a marine to a more terrestrial and anthropogenic influenced diet could result in differences in contaminant exposure. For example, concentrations of POPs and some metals, such as Hg, have been linked to trophic position and marine diet in birds (Borgå et al., 2005; Campbell et al., 2005; Elliott et al., 2009; Ruus et al., 2002), while elevated concentrations of CECs often are associated with urban areas and feeding at e.g. waste management facilities (Gewurtz et al., 2016; McGoldrick et al., 2014; Sorais et al., 2020; van Mourik et al., 2016).

Blood and eggs of birds are commonly used matrices to assess contaminants (Hebert et al., 1999; Verreault et al., 2006). Blood can contain recently acquired nutrients and contaminants, and reflects recent contaminant exposure (Henriksen et al., 1998). As a result of transfer of contaminants from mothers to eggs during reproduction, eggs can reflect female contaminant concentrations around or before egg production (Morrissey et al., 2010; Verreault et al., 2006). Maternal transfer of legacy POPs is well known and studied (e.g. (Verreault et al., 2006). However, maternal transfer of CPs and cVMSs is scarcely studied in wild birds. CPs have been observed in egg yolk of laying hens after exposure through the feed (Ueberschär et al., 2007). However, in wildlife studies of cold-blooded organisms, the maternal transfer potential has been found to be lower for CPs compared to other legacy POPs in water snake and fish (Guan et al., 2020, Choo et al., 2020). Comparing different CP congener groups. maternal transfer of CPs assessed by egg to liver ratios in frogs was higher for congener groups with longer carbon chains and higher logK_{ow} (Du et al., 2019). For cVMSs, octamethylcyclotetrasiloxane (D4), D5 and dodecamethylcyclohexasiloxane (D6) was found in herring gull eggs, with highest concentrations of D5, and further investigation was recommended (Lu et al., 2017). Based on this, we expect maternal transfer in herring gull of CPs to be lower than for the legacy POPs, whereas cVMS transfer is comparable.

We compare emerging and legacy contaminant concentrations, and metals, in blood and eggs of herring gulls from two breeding colonies in the urban Oslofjord in south eastern Norway, impacted by different degrees of urbanisation. By comparing colonies in the inner and outer Oslofjord, we address whether blood and eggs of female herring gulls from the inner fjord colony is more contaminated due to its proximity to higher density of urbanisation, or if the outer fjord colony shows higher contaminant concentrations due to a higher degree of feeding from the marine food web. Furthermore, we investigate differences in maternal transfer between the emerging and legacy contaminants, and metals.

2. Materials and methods

2.1. Sampling and colony

The area around the Oslofjord, with a human population of approximately 1.6 million living near the fjord, is affected by the high population density and activity of the capital Oslo, with pollution impacts from e.g. industry, traffic, agriculture, sewage and marine traffic. The Oslofjord is separated into the inner and outer fjord by a sill, making the inner fjord a fairly enclosed body of water with consequent restrictions of the water circulation (Gade 1968). The inner Oslofjord is located within the most densely populated area of Norway, with a population density of 3700 per km^2. The inner Oslofjord ecosystem has been shown to contain a range of legacy and emerging contaminants (Ruus et al., 2019). The outer Oslofjord is also located in a populated area and is affected by pollution, but the degree of urbanisation is lower, with a population density of 385,7 per km^2 in Moss municipality. As it is connected to the Skagerrak, the ocean area between Norway, Sweden and Denmark, there is a greater circulation of water masses in the outer compared to the inner Oslofjord (Gade, 1968).

Sampling of herring gull blood and eggs took place during the incubation period in May 2017, approximately three weeks after egg laying, in two island colonies in the Oslofjord, Søndre Skjælholmen in the inner fjord (59°51′14″N 010°43′48″E) and Store Revlingen (59°23′50″N 010°38′06″E) in the outer fjord. The distance between the colonies is approximately 51 km.

Herring gulls incubating on the nest were caught using a walk-in trap. Both male and female herring gulls incubate the eggs, and both forage during the incubation period. Sex of the birds was determined biometrically based on head length, and only females were sampled. As the difference in the head-length criteria between sexes varies between populations, our comparison was based on the local gulls in the Oslofjord. Adult, breeding individuals with head length <121 mm were considered female, while individuals with head length >123 mm were considered males. To ensure only sampling of females, only birds with head length <120 mm were sampled. A blood sample of 5 mL was taken from the brachial wing vein using a syringe flushed with heparin, and one egg was collected from the nest of the sampled female, ensuring that the egg belonged to the female. From each colony 15 blood samples and eggs were collected. All birds were biometrically measured (body mass, wing length, bill and head length). In addition to the sampling of blood

and eggs, all the captured birds, male and female, were ringed and registered in a database. The total number of sampled birds were 39 at both locations. The permit for sampling blood and eggs was granted by the Norwegian Food Safety Authority (FOTS id 12394), and sampling was performed following the Norwegian Animal Welfare Act.

2.2. Sample preparation and contaminant analyses

Details of sample preparation, analyses and quality assurance can be found in the supporting information. In brief, individual whole blood samples and whole egg homogenate were used as matrices for all analyses. Eggs were visually classified according to their level of development. Lipid content was determined gravimetrically using cyclohexane/acetone solvent. In total, 131 different chemical compounds were analysed for, including metals, PCBs, HCB, brominated flame retardants (BFRs; PBDEs and other emerging flame retardants), cVMSs and CPs at the Norwegian Institute for Air Research (NILU), and PFAS at the Norwegian Institute for Water Research (NIVA) (Table S1, supporting information). The methods are adapted from previously described analyses (Gao et al., 2016; Moeckel et al., 2020; Nøst et al., 2018; Warner et al., 2010).

The lipophilic contaminants were extracted using two silica columns and cleaned by sulfuric acid (H_2SO_4) rinse. PCBs, HCB and BFRs were quantified by gas chromatography – high resolution mass spectrometry (GC-HRMS) (Waters Autospec). Short-Chain CP (SCCP; C_{10}-C_{13} congeners), Medium-Chain CP (MCCP; C_{14}-C_{17} congeners) and dechloranes were quantified using gas chromatography quadrupole time-of-flight (GC/Q-ToF) mass spectrometry (Aligent 7200B) in electron capture negative ionization (ECNI) mode. CPs were quantified against a C_{10}-C_{13} (63% Cl) and a C_{14}-C_{17} (57% Cl) calibration standard mix, using an internal standard method according to Gao et al. (2016), Tomy et al. (1997), and care was taken to avoid contamination from the indoor lab environment. For cVMS analysis, samples were extracted using solid–liquid extraction with a biphasic solvent system of acetonitrile and hexane, and analysed using concurrent solvent recondensation large volume injection gas chromatography mass spectrometry (CSR-LVI-GC–MS). The samples were treated in a clean room, and care was taken to avoid background contamination. Metals were extracted using supra pure nitric acid and digested at high pressure and temperature in a microwave-based digestion unit (UltraClave, Milestone, Italy), and concentrations were determined using an Agilent 7700x inductively coupled plasma mass spectrometer (ICP-MS). PFASs were extracted with acetonitrile, cleaned using graphitized carbon and acetic acid, diluted with ammonium acetate buffer, and analysed by acquity ultra-performance liquid chromatography coupled to a Xevo G2-S Q-ToF-HRMS instrument (UPLC; Waters Corporation, Milford, MA, USA). Analyses were performed according to accredited methods, using recovery standards, control samples, blanks and limits of detection (LODs). Internal standards were added to all samples, standards and blanks prior to analyses. NILU is accredited for the analysis of PCBs (ISO/IEC 17025). For the other compounds, the same quality assurance procedures (as for the accredited compounds) were applied. NIVA's laboratory is accredited by Norwegian Accreditation ISO/IEC 17025. NIVA is not accredited for PFAS compounds, but the same quality assurance procedures, including regular interlaboratory studies are performed. For all compounds, the LOD was defined as 3 standard deviations of the mean blank response.

2.3. Dietary descriptors and body condition

Stable isotope analysis of $\delta^{13}C$ and $\delta^{15}N$ was performed at the Institute for Energy Technology (IFE), Norway. Samples were combusted in a Eurovector EA3028 elemental analyser. N_2 and carbon dioxide (CO_2) were separated on a 2 m Poraplot Q gas chromatography column. Isotopic ratios of $^{13}C/^{12}C$ and $^{15}N/^{14}N$ were determined in a horizon isotope ratio mass spectrometer (IRMS) from Nu-instruments. Lipids were not extracted prior to stable isotope analysis. The ^{13}C fraction is depleted in carbon found in lipids, resulting in more negative $\delta^{13}C$ values in lipid-rich tissue (DeNiro and Epstein 1977), especially in matrices with C:N ratio > 3.5 (Post et al., 2007). For our samples, the mean C:N ratios were 3.7 in blood and 7.0 in eggs, and did not differ between colonies. We corrected arithmetically for the matrix effect of lipids on the $\delta^{13}C$ value, using the non-linear equation $\delta^{13}C$ lipid-extracted = $\delta^{13}C$ non-extracted + 1:47–2:72*Log_{10}(C:N ratio), as suggestion by (Elliott et al., 2014). Ecological niche of the colonies was visualised based on $\delta^{13}C$ and $\delta^{15}N$ using the R package Stable Isotope Bayesian Ellipses (SIBER) (Jackson et al., 2011). Niche width was calculated based on the total area of the convex hull encompassing the data points (TA), and the Standard Ellipse Area (SEA). TA is drawn around the most extreme points and gives an indication of the niche width, while SEA contains 40% of the data (Jackson et al., 2011).

A body condition index (BCI) was calculated for each of the adult herring gulls using a multiple linear regression model. The dependent variable in the model was body mass. The independent variable was selected based on Pearson's and Kendall's correlation tests (Fox et al., 2007). The most correlated independent variables were wing length and head length, and these were used in the multiple linear regression model, although they were not strongly correlated ($R^2 = 0.32$, $p = 0.0007$ and $R^2 = 0.17$, $p = 0.01$, respectively). The BCI was defined by the residuals from the regression line (Jakob et al., 1996).

2.4. Data analyses

Prior to statistical analysis, contaminants with <75% of the values above the detection limit were removed from the dataset. Based on this, 45 of the 131 compounds analysed for were included in the statistical analyses (Table S2, supporting information). Nondetects in remaining contaminants were assigned random values between 0 and LOD based on left-censored distribution-based multiple imputation, assuming a β distribution ($\alpha = 5$, $\beta = 1$) (Helsel, 2010). The emerging BFRs and the cVMS D4 had detection frequencies < 75% and were not included in the statistical analysis, but are mentioned in the text. Nondetects were replaced using the imputation method described above before calculating means for these compounds.

Principle component analysis (PCA) and redundancy analysis (RDA) were performed using the vegan package to identify structures in the contaminant data, and to investigate significance of and relationships to explanatory variables (Oksanen et al., 2018). Absolute wet weight $log_{10}(x + 1)$ contaminant concentrations were used in the PCAs and RDAs. PCB congeners were grouped in homologue groups according to chlorination degree. As lipid content was not a significant explanatory variable, and thus did not affect the models, lipid was not treated as conditioned variable in either of the PCAs. The full set of explanatory variables were location, $\delta^{13}C$, $\delta^{15}N$, lipid content and BCI. For eggs, the variables embryonic age and egg weight were also included. Significance of explanatory variables were identified using forward manual model selection by Monte-Carlo permutation tests on the RDAs. Based on inspection of PCA plots, associations between selected individual contaminants and the stable isotopes were investigated using linear regression. In addition, to investigate the interactions of the explanatory variables $\delta^{13}C$, $\delta^{15}N$ and location on selected individual contaminants, analysis of variance (ANOVA) was used.

Maternal transfer factors (MTF) were calculated as logarithmic ratios of mean concentrations of individual contaminants between eggs and whole blood, and used as a measure of transfer efficiency of the individual contaminants, with a MTF of 0 indicating a 1:1 relationship in the contaminant concentrations between female and egg (Grønnestad et al., 2017). The MTFs were based on mean concentrations in ng/g lipid weight (lw) for lipophilic POPs, ng/g wet weight (ww) for PFASs and ng/g dry weight (dw) for metals.

3. Results and discussion

3.1. Ecological niche

$\delta^{13}C$ and $\delta^{15}N$ provide a proxy for ecological niche, with terrestrial originating organic material being depleted in $\delta^{13}C$ compared to organic material of marine origin, and $\delta^{15}N$ increasing with trophic position (Peterson and Fry, 1987). The outer Oslofjord females had higher $\delta^{13}C$ and $\delta^{15}N$ values than the inner fjord females, indicating more marine influence and higher trophic status in the outer fjord gulls, assuming that the food web baseline is the same in the inner and outer fjord (Fig. 1; $\delta^{13}C$: Welch two-sample *t*-test, t = -3.6614, df = 23.303, p-value = 0.001279, $\delta^{15}N$: Welch two-sample ttest, t = −4.4878, df = 23.705, p = 0.0001) (Peterson and Fry, 1987). In accordance with the female stable isotope ratios, eggs also had higher $\delta^{13}C$ and $\delta^{15}N$ levels in the outer than the inner fjord colony (Fig. 1; $\delta^{13}C$: Welch two-sample *t*-test, t = −2.2781, df = 26.498, p-value = 0.03102, $\delta^{15}N$: Welch two-sample *t*-test, t = −4.4878, df = 23.705, p = 0.0001). The stable isotope values in the eggs represent maternal diet before or during egg laying, whereas stable isotope values in the mother's blood represent diet up to two weeks before sampling (Bearhop et al., 2002, Hobson, 1995). Thus, the higher $\delta^{13}C$ and $\delta^{15}N$ values in both blood and eggs in the outer compared to the inner colony indicate that the foraging behaviour was consistent in both colonies before and after egg laying. The lipid correction of the $\delta^{13}C$ values in eggs resulted in less negative $\delta^{13}C$ values compared to the non-corrected egg values, and more similar values between the eggs and blood (Fig. 1, Fig. S1, supporting information). The $\delta^{13}C$ and $\delta^{15}N$ values were positively correlated between females and eggs, suggesting that the isotopic ratio of the egg reflects that of the female (Fig. S2, supporting information; $\delta^{13}C$: Estimate = 0.8902, SE = 0.2144, $R^2 = 0.3619$, $F_{1, 28} = 15.88$, p = 0.0004. $\delta^{15}N$: Estimate = 1.0362, SE = 0.1407, $R^2 = 0.6595$, $F_{1, 28} = 54.23$, p < 0.00001).

The ecological niche estimation from the stable isotope ratios shows some overlap between the locations for both matrices, reflecting the similarity of the colonies; both are in proximity to and influenced by urban areas, but to a larger degree in the inner Oslofjord gulls. The ecological niche overlap was larger for eggs than for females, suggesting a more similar diet of the females of the two colonies before than after egg laying. There is also more variation in the stable isotope values in the outer Oslofjord gulls (Fig. 1), indicating larger intra-colony variation in diet (Hebert et al., 2009). This can be explained by the niche width flexibility of gull species, as even within the same colony, diet and contaminant exposure might vary between individuals. In addition, human-made, processed food has unique stable isotope signatures, likely resulting in variations in stable isotope values in gulls with varying consumption of human refuse (Caron-Beaudoin et al., 2013, Jahren and Kraft, 2008).

3.2. Differences in contaminant concentrations between females of the inner and outer Oslofjord colonies

3.2.1. Lipophilic contaminants of emerging concern

In blood, CPs were the dominating of the lipophilic contaminants, with ΣCP making up 78.1% (inner fjord) and 56.0% (outer fjord) of Σ lipophilic contaminants (i.e. PCBs, PBDEs, HCB, CPs and cVMS; Fig. S3, supporting information). The proportion of cVMS in blood was small, with 1.67% in the inner fjord and 1.79% in the outer fjord. In eggs, ΣPCB was the dominating group, making up 62.3% (inner fjord) and 45.6% (outer fjord) of Σ lipophilic contaminants. Relative contributions of ΣCPs (inner fjord: 15.6%, outer fjord: 25.0%) and ΣcVMS (inner fjord: 16.5%, outer fjord: 27%) in eggs was similar.

The PCA of lipophilic contaminants in females shows large individual variance among birds of both colonies, indicating that location alone does not explain the degree of contaminant exposure of individual female herring gulls in the Oslofjord (Fig. 2). SCCP, MCCP and decamethylcyclopentasiloxane (D5) were negatively associated to the dietary markers, but not significantly, as indicated by the RDA ($\delta^{13}C$: Pseudo-F = 0.5752, p = 0.5839, $\delta^{15}N$: Pseudo-F = 1.0316, p = 0.3158). The first three components in the PCA of lipophilic contaminants explained 35.9%, 31.6% and 20.0% of variation in the female lipophilic contaminant data each. Investigating PC3 revealed that the variation on this axis is related to MCCP. Removing MCCP from the PCA resulted in a larger separation of the locations along PC1 and PC2 (Fig. 2B). Also in this model, $\delta^{15}N$ and $\delta^{13}C$ were not significant explanatory variables ($\delta^{13}C$: Pseudo-F = 0.5007, p = 0.5715, $\delta^{15}N$: Pseudo-F = 1.3237, p = 0.2606). With MCCP removed, we observed higher concentrations of SCCP and D5 in the inner fjord females, and higher concentrations of D6 in the outer fjord females (Fig. 2B). This was also confirmed when investigating the compounds separately, and no interaction effects between location and $\delta^{15}N$ or $\delta^{13}C$ were observed (Table S3, supporting information).

The large variation in MCCP is related to two outer fjord females with concentrations approximately five times higher than the mean MCCP

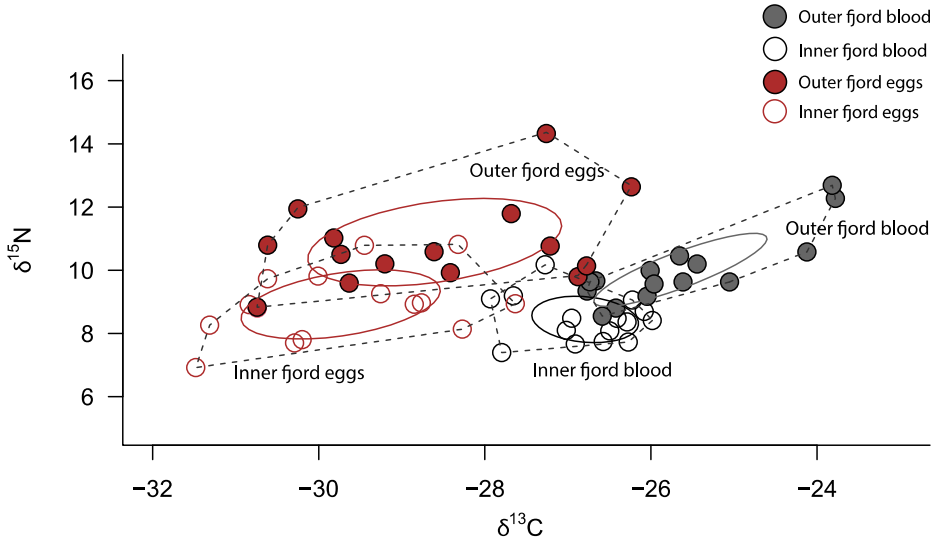

Fig. 1. Relationship between stable isotope values of carbon ($\delta^{13}C$) on the x-axis and nitrogen ($\delta^{15}N$) on the y-axis, and ecological niche estimations in herring gull females and eggs from the inner and outer Oslofjord. The $\delta^{13}C$ values were lipid-corrected arithmetically according to Elliott et al. (2014). The ecological niche of the four clusters are represented by solid lines for the standard ellipse area corrected for sample size (SEAc), and dashed lines for the total convex hull area (TA).

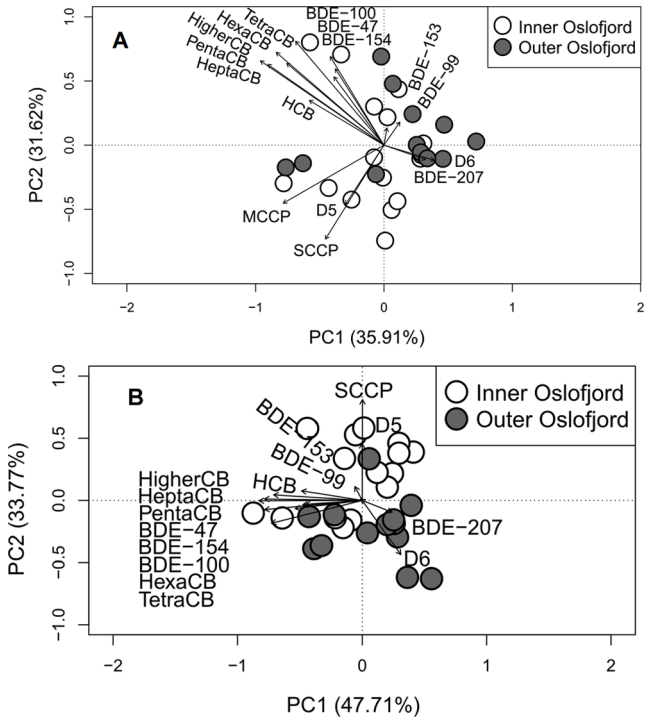

Fig. 2. Principal component analysis biplot of levels of all the included lipophilic POPs (A) and with MCCP removed (B) in herring gull whole blood from females (inner fjord n = 15, outer fjord n = 13). Contaminant concentrations are log-transformed ng/g wet weight. Individual herring gulls are represented as points and coloured according to the explanatory variable location. Response variables (contaminants) are represented as black arrows. The factor variable Location was the only significant explanatory variable, and therefore no explanatory variables were fitted in the ordinations. As lipid content was not a significant variable, and did not affect the models, lipid was not treated as a covariable. The amount of variance explained by each principal component is shown on the axis.

concentration for outer fjord females, which were also reflected in their eggs. The two individuals did not have high concentrations of other contaminants, or deviant $\delta^{15}N$ and $\delta\ ^{13}C$ values. In females from the inner fjord colony, SCCP accounted for 64% of ΣCP, and in the outer fjord females, SCCP constituted 44% of ΣCP. CP concentrations were variable between the individuals, which is also reported in biota previously (van Mourik et al., 2016). Our results indicate that individual specialisation and local sources of contaminants in the foraging area of individual gulls are more important than between-colony feeding habits. There are known analytical challenges in the quantification of CPs, relating to structural complexity, availability of suitable standards, inferences from other OC compounds, and contamination of samples from field and lab materials and environment (Sverko et al., 2012). The methods used at the NILU lab are in accordance with established methods (Gao et al., 2016). In addition, an investigation of the method has been performed within the NILU laboratory (Nipen 2017).

cVMS concentrations have been linked to human settlements and urban influence (McGoldrick et al., 2014; Warner et al., 2010), as a result of present use in e.g. personal care products. The concentrations of cVMSs in our study (Table 1) were higher than concentrations reported from remote herring gull colonies (Huber et al., 2015), and in fish from coastal monitoring stations along the Norwegian coast (Green et al., 2018), indicating urban influence. D5 was the dominating cVMS in our study (Table 1), in accordance with other studies, both in urban and remote locations, possibly explained by higher use and emissions of D5 compared to the other cVMSs (Huber et al., 2015; McGoldrick et al., 2014; Warner et al., 2010). As for the CPs, there was high variability in cVMS concentrations between individuals in our study (Table S4, supporting information). In most individuals, the variability was consistent between D5 and D6. High variability in cVMS concentrations in herring gulls was also observed in previous Oslofjord surveys (Ruus et al., 2019). D4 is a cVMS which has not been shown to biomagnify, unlike D5 and D6 (Borgå et al., 2013). In the current study, D4 was not included in the statistical analysis because of low detection frequency, and was only detected in the Inner Oslofjord, supporting higher concentrations of CECs close to the most urbanised area. In the inner fjord colony, D4 was detected in 73% of samples in females, and in 54% of samples in eggs. In the inner fjord colony females, mean concentration ± SD of D4 was 0.572 ± 0.477 ng/g wet weight, which is similar to the inner colony female concentrations of D6, while the D5 concentrations are approximately twice as high.

Table 1
Organic contaminant levels (ng/g wet weight), quantified in herring gull blood from females and in their eggs from the inner and outer Oslofjord. Contaminant levels are reported as mean ± standard deviation and range (min – max). The means include imputed values.

Mean ± SD Min – Max	Inner Oslofjord		Outer Oslofjord	
	Females[a] (n = 15)	Egg[b] (n = 15)	Females[a] (n = 15)	Egg[b] (n = 15)
SCCP	50.3 ± 32.4	35.0 ± 24.9	30.3 ± 49.0	42.0 ± 46.3
	14.0–108	13.0–91	5.00–200	18.0–178
MCCP	28.3 ± 23.8	29.1 ± 19.8	38.9 ± 64.6	69.6 ± 160
	8.00–76.0	6.00–68.0	6.00–200	3.00–630
HCB	0.423 ± 0.313	3.66 ± 2.61	0.794 ± 2.13	3.03 ± 1.67
	< LOD–1.27	0.720–9.35	< LOD–8.49	0.848–6.66
∑TetraCB	1.54 ± 1.64	15.8 ± 19.6	5.77 ± 17.9	10.0 ± 10.1
	0.220–6.18	0.610–65.4	0.206–70.4	1.37–36.2
∑PentaCB	3.71 ± 2.88	53.0 ± 53.9	11.2 ± 34.5	37.8 ± 32.5
	0.658–10.2	3.60–213	0.579–136	7.04–111
∑HexaCB	10.4 ± 9.49	132 ± 101	22.2 ± 57.4	115 ± 86.6
	2.69–40.8	16.6–385	1.74–229	25.7–317
∑HeptaCB	2.93 ± 2.08	49.0 ± 30.5	9.83 ± 29.1	37.0 ± 25.6
	0.984–9.27	8.94–120	0.561–1145	9.57–95.3
∑HigherCB	0.359 ± 0.326	6.07 ± 4.29	1.00 ± 3.03	3.47 ± 1.98
	0.104–1.30	1.35–16.1	0.0531–11.9	0.950–8.04
∑PBDE	0.892 ± 1.01	19.0 ± 34.3	1.10 ± 2.27	6.77 ± 3.54
	0.287–4.07	1.59–129	0.146–9.24	1.66–12.6
D5	1.09 ± 0.765	56.1 ± 69.2	0.493 ± 0.322	111 ± 167
	< LOD–2.42	< LOD–206	< LOD–1.21	13.0–695
D6	0.594 ± 0.558	11.9 ± 17.9	1.72 ± 0.713	8.96 ± 4.52
	< LOD–2.01	< LOD–65.5	0.706–3.56	3.89–19.7
∑PFCA	2.64 ± 1.41	3.49 ± 2.78	5.23 ± 2.25	5.11 ± 4.67
	1.46–6.42	1.26–12.3	1.89–10.8	1.15–20.7
PFSA	11.2 ± 7.14	25.6 ± 42.3	18.7 ± 11.5	38.5 ± 32.2
	2.72–6.43	4.17–172	6.45–51.4	4.39–126

[a] Whole blood.
[b] Whole egg homogenate.

3.2.2. Legacy POPs

Concentrations of PCBs, HCB and some PBDEs (ww; lipid was not a significant explanatory variable) did not differ between females of the two colonies (Fig. 2). POPs are present in the food webs of both the inner and outer colonies, but as the outer fjord colony females have a stronger marine signal in their diet, the concentrations of POPs were expected be higher in the outer fjord colony females. Our results however indicate that the marine influence on diet of the outer fjord females is not large enough to illustrate accumulation in marine food chains compared to the inner fjord colony females, supported by the overlapping ecological niches from the stable isotope ecological niche estimation (Fig. 1). Although we found higher $\delta^{15}N$ and $\delta^{13}C$ in the outer fjord colony females compared to the inner fjord females, also the outer fjord females could be foraging from anthropogenic and terrestrial sources in addition to the marine food web. The outer fjord colony at Store Revlingen is located approximately 1 km from land, and the gulls could be feeding from sources e.g. around the town Moss, approximately 3 km away, and at Stegen waste management facility in Askim, approximately 36 km

away. This is a waste management facility with open access to food waste, and 28 of the 35 herring gulls (males and females) which have been sighted and registered after capture at Store Revlingen have been observed at Stegen. Despite the ban on POPs such as PCBs and PBDEs, considerable amounts of these contaminants may still discharge from urban sources such as landfills and old building materials, leading to increased exposure in some urban areas (Brogan et al., 2017; Elliott et al., 2009; Sorais et al., 2020). In the Oslo area, higher concentrations of PCBs have been observed in terrestrial sparrow hawk eggs than in the herring gull eggs in this study (Heimstad et al., 2018). With the overlapping ecological niches in the present study, the extensive distribution of POPs could be dominating over the differences in diet between the inner and outer colony females.

In the females, the PCBs, HCB and the majority of the PBDEs were positively correlated to each other and were uncorrelated to the dietary markers $\delta^{13}C$ and $\delta^{15}N$, which were not significant explanatory variables in the multivariate model, as indicated by the RDA reported above. This contradicts previous studies linking increasing concentrations of POPs in birds to increasing levels of $\delta^{15}N$ (Elliott et al., 2009), maybe due to the low variation in diet between the colonies and inter-colony individual diet specialisation in this study (Hebert et al., 2009). The confounding effects of the unique $\delta^{13}C$ and $\delta^{15}N$ signatures in processed food and anthropogenic waste could also contribute to this lack of correlation (Jahren and Kraft, 2008, Caron-Beaudoin et al., 2013, Ramos et al., 2009). Diet and trophic position alone does not always explain contaminant concentrations in birds, especially within one species (Brogan et al., 2017), as in our study. Other important factors affecting avian contaminant concentrations include migration and breeding strategy (Baert et al., 2013; Borgå et al., 2005; Hitchcock et al., 2019). In the present study, different turnover rates between contaminant concentrations and dietary markers could be of importance for the results, as $\delta^{13}C$ and $\delta^{15}N$ in blood represent a short dietary accumulation of approximately two weeks, which in this case was during the incubation period (Bearhop et al., 2002). Blood contaminant concentrations reflect a more extended timeframe and could be influenced by diet over a longer time period than the stable isotopes, resulting in a mismatch between the contaminant concentrations and stable isotope values measured in our study (Baert et al., 2013; Borgå et al., 2005). Contradicting the majority of the lipophilic POPs, PBDE-99 PBDE-153 and PBDE-207 had higher concentrations in the outer colony females (Fig. 2). Investigated separately, regression analyses found no correlation between these BDE congeners and $\delta^{13}C$ or $\delta^{15}N$ in females (Table S 5, supporting information). Previous studies have suggested that terrestrial and urban organisms are more exposed to higher-chlorinated BDE congeners than aquatic organisms (Law et al., 2006, Park et al., 2009). In the outer Oslofjord herring gulls, negative correlation between PBDE-209, and to some degree PBDE-99 and $\delta^{13}C$ or $\delta^{15}N$ has been observed previously, suggesting that exposure to this higher-chlorinated BDE congener can be associated with land-derived sources. (Sørmo et al., 2011). Thus, our results contradict our expectations of a more marine-based diet in the outer fjord gulls, and indicates exposure to terrestrial-influenced contaminants in the outer fjord gulls. However, also differences in ability to metabolise the congeners could contribute to explain the results (Elliott et al., 2009).

BFRs and dechloranes were detected in < 75% of the samples overall, and were not included in the statistical analysis. The analysed BFRs included bis(2-ethylhexyl)-2,3,4,5-tetrabromophthalate (TBPH), 1.2-bis (2,4,6- tribromophenoxy)ethane (BTBPE), decabromodiphenyl ethane (DBDPE), pentabromoethylbenzene (PBEB) and 1,2-dibromo-4-(1,2-dibromoethyl)cyclohexane (TBECH; α, β and g/d isomers) and seven dechlorane related compounds. Of the dechloranes analysed, four had < 10% detected values. The most frequently detected dechloranes, plus syn and plus anti, were both detected in < 20% of the female samples, and in 93.3% of samples in eggs, in both colonies (Table S6, supporting information). The Dechlorane plus (DP) concentrations in eggs were variable between the individuals (Range: 13.18 ng/g – 1245 ng/g for DP syn, 44,45 ng/g – 3619 ng/g for DP anti). There were no differences in mean concentrations of DPs in eggs between the colonies (ANOVA: $p > 0.05$ for both). The other flame retardants all had low detection frequencies, with all contaminants detected in < 12% of the samples, and with similar frequencies across all locations and matrices.

3.2.3. PFASs

Female concentrations of PFASs were higher in the outer Oslofjord (Fig. 3A). PFASs have previously been linked to urban areas (Gewurtz et al., 2016; Herzke et al., 2009). In the present study, the higher PFAS concentrations in the outer fjord can be related to Rygge airport as a point source, as it is located near the Revlingen colony, and was previously used as a fire drill area. Although the exact mixture is unknown, fire-fighting foams containing PFAS have presumably been used at Rygge since the 1970s, and PFASs has been detected in lake Vansjø close to Rygge, which drains to the Oslofjord close to the outer fjord colony (Amundsen et al., 2016; Fjeld et al., 2017). Thus, PFAS are probably not suitable for indicating contamination differences between the inner and outer Oslofjord due to urbanization or marine influence, but rather reflects local point sources.

3.2.4. Metals

The metals were expected to show higher or lower concentrations in the gulls from the inner and outer fjord based on their use and properties. Hg, especially in its methylated form, is known to accumulate in marine food webs, and we expected higher concentrations to be associated with a more marine diet (Ruus et al., 2015, Borgå et al., 2006). Arsenic has been reported to accumulate in marine food webs, however trends are variable between species (Anderson et al., 2010), and other studies have found no potential for accumulation of As in seabirds (Borgå et al., 2006, Vizuete et al., 2019). Also for Cd there have been contrasting results, with some studies finding that the element bioaccumulates in marine food webs, while others have found no potential for bioaccumulation (Borgå et al., 2006, Sun et al., 2020, Anderson et al., 2010). Pb is a common contaminant in industrial and urban areas, related to activities such as industry, gasoline and agriculture, and previous studies support our results, with higher concentrations of Pb in the house sparrow (*Passer domesticus*) and the common blackbird (*Turdus merula*) with increasing urbanisation (Bichet et al., 2013; Meillere et al., 2016). In our results, Hg and arsenic (As) had comparable concentrations in females the inner and outer fjord colonies (Fig. 3B). For Hg, this was contrary to expectations when considering the more marine diet in the outer fjord females. As for the POPs, the similarity between colonies could be due to the overlap in ecological niche. Furthermore, the association between Hg and $\delta^{15}N$ was weak (Linear regression: Estimate = 0.10798, SE = 0.02512, $R^2 = 0.2416$, $p < 0.00001$)… Cd concentrations were higher in the outer fjord females compared to the inner fjord females (Fig. 3B). For Pb, concentrations were higher in the inner fjord than in the outer fjord females (Fig. 3B), which was expected with the more urbanised environment of the inner fjord.

3.3. Maternal transfer of contaminants to egg

In both colonies, the lipophilic contaminants with the exception of the CPs, were transferred from female to egg to a higher degree than PFASs (Fig. 4). As the lipophilic contaminants are not mainly sequestered in blood, our MTFs might not reflect the true rates of maternal transfer, but give an indication of the ratio of contaminants in eggs and blood. Lipids constitute around 20–30% of the total egg mass, and protein is a limiting resource during egg production, possibly explaining this difference in MTFs between the lipophilic and more protein-associated compounds (Hitchcock et al., 2019). Some PFASs, especially PFDoA and PFUdA, had lower MTFs in the outer than the inner colony. This is likely related to the higher concentrations in females of the outer colony, as high blood concentrations and a low degree of

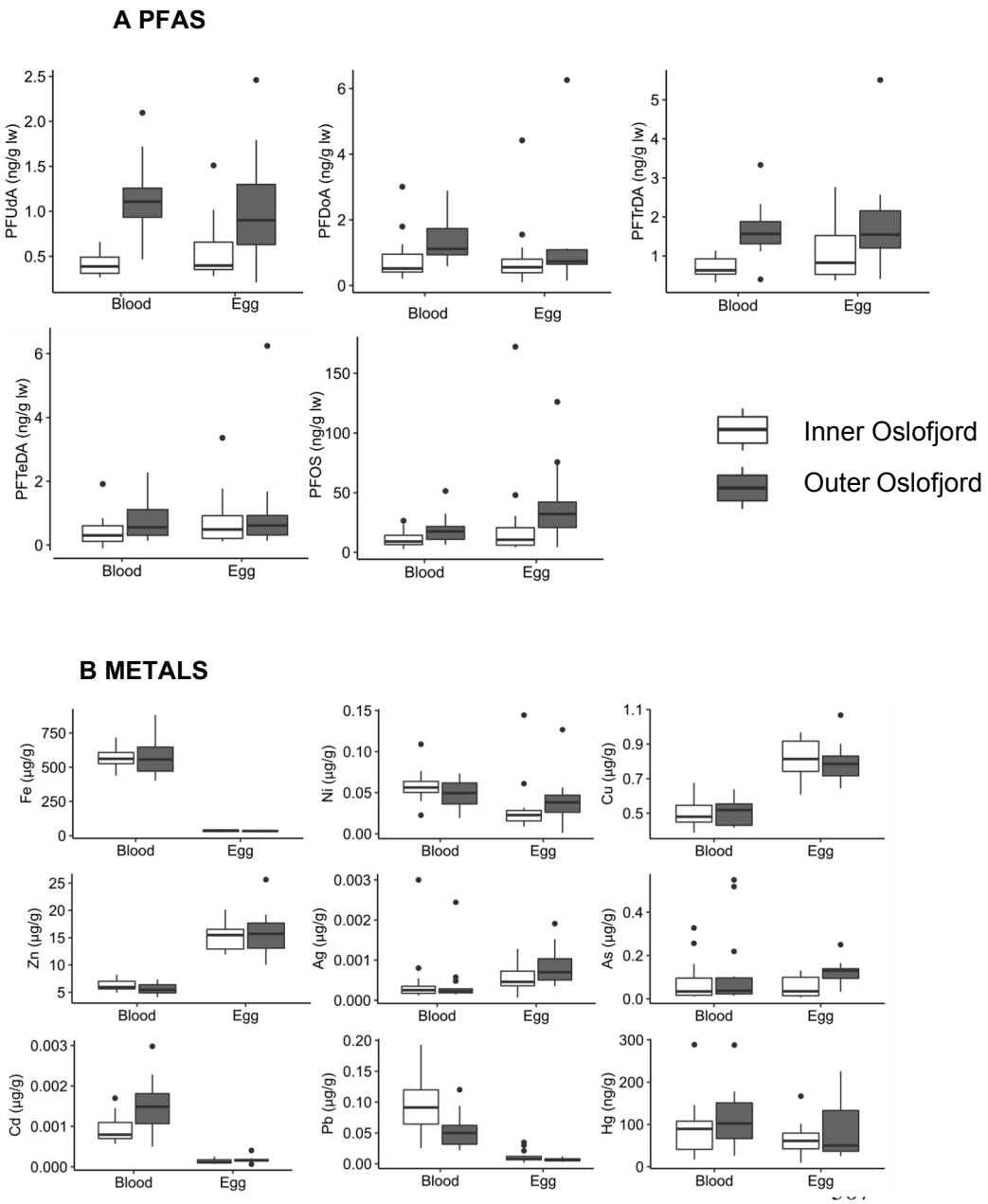

Fig. 3. Boxplot of levels of PFASs (ng/g wet weight) (A) and metals (µg/g and ng/g dry weight) (B) in herring gull females (n = 15) and eggs (n = 15) from the inner (white boxes) and outer (grey boxes) Oslofjord. The whiskers represent the standard deviation and points are the extreme values. Note the different scales on the axes.

transfer of protein-associated contaminants would lead to lower MTFs. SCCPs and MCCPs had MTFs < 0, indicating that for CPs, egg production is not an important route of elimination in herring gulls, compared to the other lipophilic contaminants. To our knowledge, only few studies are available on CP maternal transfers in birds. However, it has been suggested that in other wildlife, e.g. amphibians and fish, CPs have lower maternal transfer potential compared to many POPs, which is in accordance with our results (Choo et al., 2020; Guan et al., 2020). The cVMS D5 and D6 in the inner fjord colony and D6 in the outer fjord colony had MTFs similar to the lipophilic legacy contaminants. (Fig. 4; Fig. S4, supporting information). D5 had a higher MTF in the outer colony, resulting from one egg with a high D5 concentration of 695 ng/g ww, approximately 6.2 times higher than the mean concentration of D5 in outer fjord eggs (Table S4, supporting information). The egg did not have deviating $\delta^{15}N$ and $\delta^{13}C$ values, and the corresponding female did not have an elevated D5 concentration.

When sampling approximately three weeks after egg laying, many embryos are well developed, and the stage of incubation likely varied between the eggs, as the eggs are not laid on the same day. Although it is recommended to use freshly laid eggs in monitoring programmes, our sampling campaign was carried out later to minimise risk of nest abandonment and increased negative impact on the colony (Burger, 1981). Developmental stage of each egg was later classified by visual inspection (Table S6, supporting information), and the effect of this variable on contaminant concentrations was tested in the RDA, and was identified as not significant (RDA: Pseudo-F = 1.2555, p = 0.2829). Further, blood and eggs represent contaminant status at different times, and the MTFs could be biased as a result. In the Oslofjord, many individuals overwinter locally, while others migrate south to areas around the North Sea in winter (Systad et al., 2007). However, also the migrating gulls arrive early, and blood and egg concentrations are expected to reflect similar exposure. Furthermore, lipophilic contaminant

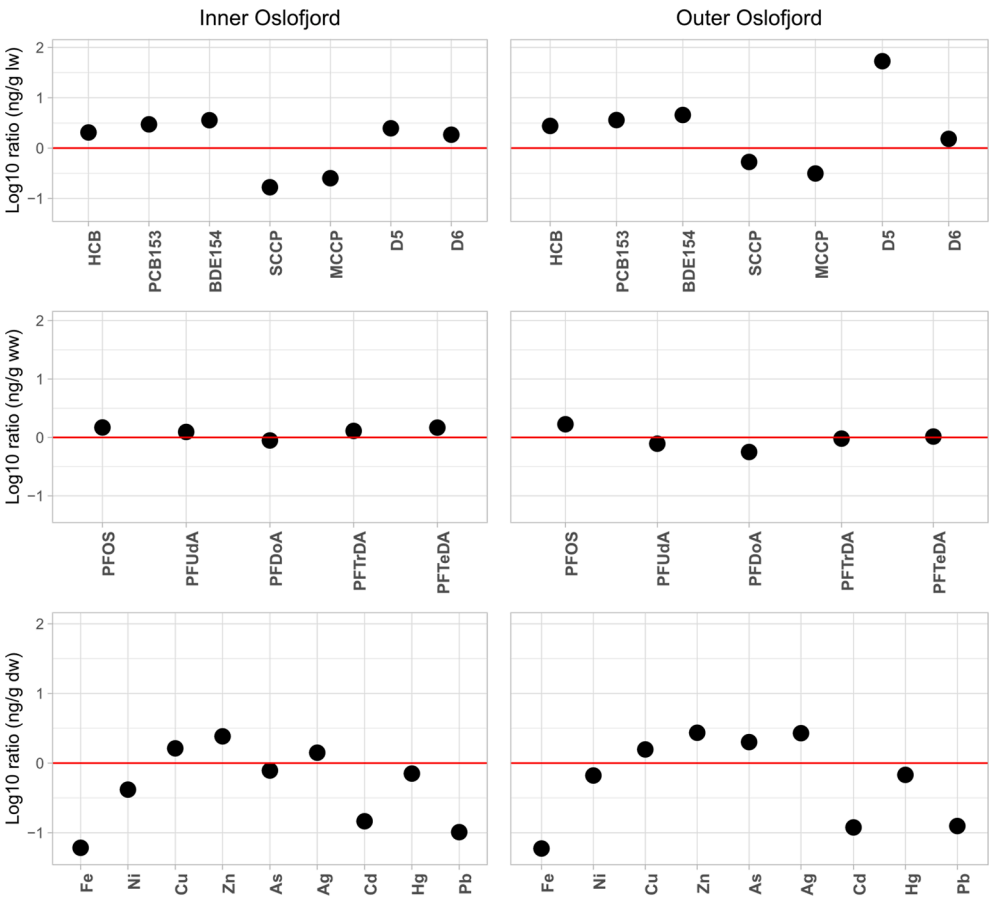

Fig. 4. Maternal transfer factors (log_{10} ratios) of contaminant levels in herring gull eggs and females from the inner (n = 15) and outer (n = 15) Oslofjord colony for the lipophilic contaminants, PFASs and metals. The lipophilic contaminants are ordered by log-K_{ow} and group (lipophilic POPs, CPs, cVMSs). The PFASs are ordered by carbon chain length (C8 – C14), and metals are ordered by atomic weight. Ratios are calculated as log_{10} (mean concentration in eggs: mean concentration in blood) in ng/g lipid weight for lipophilic contaminants, ng/g wet weight for PFASs, μg/g dry weight for the metals except Hg, and ng/g dry weight for Hg. The horizontal line (log ratio = 0) indicates a 1:1 relationship between female and egg. HCB, PCB-153 and BDE-154 are included as examples of the lipophilic POPs. For ratios of all PCB and PBDE congeners, see Fig. S4, supporting information.

concentrations have been shown to increase with laying order in herring gulls, posing another possible bias, as we did not control for laying order in our sampling. In legacy organochlorines, this effect has been shown to be negligible, with larger differences between mothers than within a clutch (Verreault et al., 2006). For CECs however, this effect has not been sufficiently studied, providing uncertainty in our results, and illustrating the need for further research on maternal transfer mechanisms on emerging contaminants.

3.3.1. Contaminant concentrations in the Oslofjord herring gulls compared to other areas

In gulls in the North American Great Lakes and urban Canada, concentrations of PCBs (De Solla et al., 2016), PBDEs (Su et al., 2017), PFAS (Su et al., 2017) and Hg (Mallory et al., 2018) were higher than in the current study of the Oslofjord herring gulls. For the cVMSs however, concentrations in the Oslofjord and in urban Canada were similar to or lower than in the Oslofjord. (Lu et al., 2017). In other contaminated European seas, studies have found similar or higher concentrations in gulls and other seabirds compared to the current study (Carravieri et al., 2020, Leat et al., 2013, Reindl et al., 2019, Morales et al., 2012, de Wit et al., 2020, Lopez-Antia et al., 2021)

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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Author statement

The authors declare no conflicts of interests. We declare that this manuscript is original, has not been published before, and is not currently being considered for publication elsewhere.

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The data used in the current manuscript is available in the report Environmental Contaminants in an Urban Fjord 2017, from the Norwegian Environment Agency (https://www.miljodirektoratet.no/globalassets/publikasjoner/m1131/m1131.pdf). The data analysis and interpretations were however done for this manuscript alone. All co-authors have contributed to data collection and/or analysis and drafting of the manuscript, and are aware and accept responsibility and submission of the manuscript.

Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2021.106478.

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