Environmental Toxicology

Differences in Trophic Level, Contaminant Load, and DNA Damage in an Urban and a Remote Herring Gull (*Larus argentatus*) Breeding Colony in Coastal Norway

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Abstract: Herring gulls (*Larus argentatus*) are opportunistic feeders, resulting in contaminant exposure depending on area and habitat. We compared contaminant concentrations and dietary markers between two herring gull breeding colonies with different distances to extensive human activity and presumed contaminant exposure from the local marine diet. Furthermore, we investigated the integrity of DNA in white blood cells and sensitivity to oxidative stress. We analyzed blood from 15 herring gulls from each colony—the urban Oslofjord near the Norwegian capital Oslo in the temperate region and the remote Hornøya island in northern Norway, on the Barents Sea coast. Based on $d^{13}C$ and $d^{34}S$, the dietary sources of urban gulls differed, with some individuals having a marine and others a more terrestrial dietary signal. All remote gulls had a marine dietary signal and higher relative trophic level than the urban marine feeding gulls. Concentrations (mean ± standard deviation [SD]) of most persistent organic pollutants, such as polychlorinated biphenyl ethers (PCBs) and perfluorooctane sulfonic acid (PFOS), were higher in urban marine (PCB153 17 ± 17 ng/g wet weight, PFOS 25 ± 21 ng/g wet wt) than urban terrestrial feeders (PCB153 3.7 ± 2.4 ng/g wet wt, PFOS 6.7 ± 10 ng/g wet wt). Despite feeding at a higher trophic level ($d^{15}N$), the remote gulls (PCB153 17 ± 1221 ng/g wet wt, PFOS 19 ± 1421 ng/g wet wt) were similar to the urban marine feeders. Cyclic volatile methyl siloxanes were detected in only a few gulls, except for decamethylcyclopentasiloxane in the urban colony, which was found in 12 of 13 gulls. Only hexachlorobenzene was present in higher concentrations in the remote (2.6 ± 0.42 ng/g wet wt) compared with the urban colony (0.34 ± 0.33 ng/g wet wt). Baseline and induced DNA damage (doublestreak breaks) was higher in urban than in remote gulls for both terrestrial and marine feeders. *Environ Toxicol Chem* 2022;41:2466–2478. © 2022 The Authors. *Environmental Toxicology and Chemistry* published by Wiley Periodicals LLC on behalf of SETAC.

Keywords: POPs; PFOS; Siloxanes; Oxidative damage; Arctic; Urban gulls

INTRODUCTION

As a result of human activities, environmental contaminants are distributed worldwide with possible adverse effects on wildlife (Dietz et al., 2019). Some contaminants, including

persistent organic pollutants (POPs) are well studied and regulated through global conventions (United Nations Environment Programme, 2001). Contaminants of emerging concern (CECs), on the other hand, are not yet well studied and currently mostly not regulated, or they are of increasing concern even if they are well studied and regulated (Arctic Monitoring and Assessment Programme, 2017; de Wit et al., 2010; Muir et al., 2019). Many of these compounds are lipophilic and thus have high absorption affinity to organic matter and lipid tissues following uptake by organisms. Transfer through aquatic food chains through predator–prey relationships results in trophic magnification (Fisk et al., 2001) with high concentrations accumulating in apex predators such as seabirds

(Elliott et al., 2021; Rigét et al., 2019). Many POPs, such as the polychlorinated biphenyl ethers (PCBs), have been shown to have high concentrations in marine food webs and are not specifically linked to current anthropogenic activity, because their presence in the environment is a legacy of previous releases. Elevated concentrations of the CECs cyclic volatile methyl siloxanes (cVMSs) in biota have been linked to anthropogenic activity and emissions from populated centres in both urbanized and remote regions (McGoldrick et al., 2014; Warner et al., 2010). The long-chained perfluorooctane sulfonic acid (PFOS), an amphipatic chemical with both lipophilic and hydrophilic properties (Buck et al., 2011), has been phased out since 2000 and included in the Stockholm convention since 2009 (Buck et al., 2011).

Seabirds are commonly used as sentinels of ecosystem health and have been widely studied in relation to contaminants (Elliott & Elliott, 2013). The herring gull (*Larus argentatus*) is a large species of seagull distributed throughout north-temperate latitudes, found along the entire coast of Norway. They are surface feeders and their diet includes fish, crustaceans, and a wide variety of marine invertebrates, and in addition terrestrial items like carcasses, earthworms, insects, eggs, and plants (Ewins et al., 1994; Hebert et al., 2009; Sotillo et al., 2019). However, the herring gull is a highly opportunistic feeder, and with proximity to human settlements it will scavenge food from urban areas, such as waste dumps (Coulson, 2015; Hebert et al., 2009; Tongue et al., 2021). Because diet is the major source of contaminants in seabirds, knowing the feeding preferences of the herring gulls is essential to understand the patterns and presence of contaminants (Elliott et al., 2021; Hebert et al., 2009; Lato et al., 2021). An urban influenced colony may incorporate more food items of terrestrial and anthropogenic origins, altering their exposure to contaminants compared to colonies with a marine-based diet (Elliott et al., 2009; Knudtzon et al., 2021). In addition, urban colony sites are closer to sources of contaminants and thereby may be more exposed.

Several POPs are genotoxic, and by altering DNA structure and integrity genotoxic compounds may cause serious damage, with consequences such as cell death, mutations, or carcinogenesis (Costantini et al., 2014; Valavanidis et al., 2006; van Gent et al., 2001). In addition, reduced food availability and quality, stress in relation to reproduction, and migration will exert extra pressure on seabirds' internal repair and maintenance systems (Alonso-Alvarez et al., 2004; Costantini, 2008; Lin et al., 2021). Damage to the DNA can be identified using several methods, including the quantification of DNA strand breaks, commonly due to oxidative damage (Collins, 2014; Ericson & Larsson, 2000), a method also validated in various seabirds (Haarr et al., 2018).

In the present study, we compared an urban and a remote herring gull colony to investigate the impacts of urbanization on POP (PCBs, polybrominated diphenyl ethers [PBDEs] and PFOS) and CEC (cVMS) exposure concentrations and potential health risks associated with genotoxicity and oxidative stress between colonies located in urbanized and remote areas. We measured concentrations of POPs and CECs in the whole blood of herring gulls, and evaluated the dietary source and trophic levels using stable isotope ratios of carbon ($d^{13}C$), nitrogen ($d^{15}N$), and sulfur ($d^{34}S$). Baseline levels of DNA strand breaks and resilience to oxidative stress induced by hydrogen peroxide (H_2O_2) were quantified using the comet assay optimized for avian blood.

MATERIALS AND METHODS

Study areas and sampling

Whole blood was sampled from herring gulls during the 2016 breeding season 2–3 weeks into the egg incubation in May in a colony in the urban Oslofjord in southern Norway, and in June in a remote colony on Hornøya, off the coast of northern Norway. The urban Oslofjord colony, Søndre Skjælholmen (59°85′N, 10°73′E), is an island approximately 6 km off the city centre of the Norwegian capital Oslo. The Oslofjord area is highly populated, with a population of approximately 1.6 million living near the fjord. The fjord is exposed to legacy POPs because of previous releases and by current emissions of CECs resulting from urban activities. The remote colony Hornøya (70°23′N, 31°10′E) is located 1.5km from the small town Vardø, along the coast of Finnmark county, a remote area in the Norwegian Arctic. This is a sparsely populated area, with a human population of approximately 28 000 in the larger area Øst-Finnmark. The town Vardø, with 1800 inhabitants, is located within 1 km from the colony.

In the urban colony, herring gulls were caught on the nest with a walk-in trap and 15 females were identified biometrically in the field by measuring the head length. On the remote Hornøya, the herring gulls were caught with an automatic triggered snare trap placed around the nest and 15 individuals with a mix of males and females were sampled. Because the biometric identification of sex in the remote population was unsatisfactory, molecular determination of sex was performed at the University of Oslo using the Qiagen DNeasy kit with 2550F and 2718R primers (Fridolfsson & Ellegren, 1999). In both colonies, 5 ml of blood was drawn from the branchial vein of herring gulls using sterile syringes flushed with heparin. The samples were kept on ice for up to 8 h before further processing. In addition to blood sampling, all captured birds were biometrically measured and tagged with identification rings. Permission to handle and sample the birds at both locations was granted by the Norwegian Food Safety Authority and procedures followed the guidelines of the Norwegian Animal Welfare Act (FOTS id 12394).

Sample preparation and contaminant analyses

The samples were analyzed for PCBs and PBDEs at the Norwegian Institute for Air Research in Tromsø, Norway (Hornøya samples) and Kjeller, Norway (Oslofjord samples). Additional details of the chemical analyses can be found in the Supporting Information. A brief description follows. At the Norwegian Institute for Air Research, Tromsø, whole blood was extracted using the protocol described in Haarr et al. (2018).

Liquid–liquid extraction in a biphasic mixture of ethanol saturated with ammonium sulfate and hexane was used to extract 0.5–2.0 g of wet weight of blood. After two subsequent extractions, the hexane phases were combined and evaporated to approximately 0.2 ml to undergo further clean-up using an automated solid phase extraction (SPE) with 1 g of activated Florisil (450 °C). Extracts collected from SPE clean-up were evaporated to approximately 50 μl and ^{13}C-PCB 159 was added as a recovery standard to each sample and analyzed using an Agilent 7890 gas chromatograph equipped with a triple quadrupole mass spectrometer, Quattro Micro GC (Waters Corporation). Instrumental details have been previously described in the supporting information by Eckbo et al. (2019).

At the Norwegian Institute for Air Research, Kjeller, the samples were extracted and cleaned using two silica columns and a sulfuric acid rinse as described in Krogseth et al. (2017). The samples were dried and homogenized using sodium sulfate. The dried samples were eluted with internal standards (isotope labelled PCBs) and 150 ml of cyclohexane/acetone (3:1) and concentrated to 0.5 ml under nitrogen flow in a TurboVap Classic 500 Evaporation System before they were transferred to silica columns for extraction. Sixty millilitres of etherhexane was used as eluent. Concentrated samples (0.5 ml) were then cleaned using a 4× sulfuric acid rinse, concentrated to 0.5 ml, and eluted in a second silica column. The samples were concentrated to a volume of 100 ml using nitrogen gas, and 20 ml recovery standard was added to each sample. The prepared samples were analyzed using high-resolution gas chromatography/high-resolution electron ionization mass spectrometry (GC/EI HRMS; Waters Autospec; HP6890 GC coupled to VG AutoSpec). Further details on the analysis have been described in the supporting information of Thorstensen et al. (2020).

Perfluorooctane sulfonic acid was analyzed at the Norwegian Institute for Air Research, Tromsø for the Hornøya samples and at the Norwegian Institute for Water Research (NIVA) Oslo for the Oslofjord samples. In Tromsø, the samples were extracted using methanol and cleaned up using active carbon (Envi-Carb) as an adsorbent (Powley et al., 2005). The instrumental analysis of PFOS was conducted as described by Hanssen et al. (2013) using ultrahigh-pressure liquid chromatography triple-quadrupole mass spectrometry (UHPLC-MS/MS). At NIVA Oslo, the samples were analyzed as described in the supporting information of Thorstensen et al. (2020). Briefly, samples were extracted with acetonitrile, cleaned using graphitized carbon and acetic acid, diluted with ammonium acetate buffer, and analyzed by acquity ultra-performance liquid chromatography coupled to a Xevo G2-S Q-ToF-HRMS instrument (UPLC; Waters Corporation).

The cVMSs octamethylcyclotetrasiloxane (D4), decamethylcyclopentasiloxane (D5), and dodecamethylcyclohexasiloxane (D6) were analyzed at the Norwegian Institute for Air Research in Tromsø for both the Hornøya and the Oslofjord samples following methods specified in Krogseth et al. (2017). In brief, the blood samples (0.5–1 g wet wt) were extracted as described using a biphasic mixture of acetonitrile and hexane (1:1). Extracts were analyzed using concurrent solvent recondensation large volume injection gas chromatography mass spectrometry (CSR-LVI-GCMS) using a modified method published by Companioni-Damas et al. (2012). Instrument specifications and details were as described by Krogseth et al. (2017).

At the Norwegian Institute for Air Research, quantification of POPs is done by isotopic dilution technique using single point calibration. Siloxanes are quantified using an internal standard method with a five-point calibration curve (Krogseth et al., 2017). The laboratories at NIVA and the Norwegian Institute for Air Research are accredited by Norwegian Accreditation for ISO/IEC 17025. For all analyses, field and laboratory blanks were run with the samples. A reference material sample was run and analyzed with the samples as additional quality control. Analysis of cVMSs is the most sensitive method because there are multiple sources of background contamination of these compounds present in the ambient environment. Care was taken to avoid contamination during handling of samples. Procedural blanks were run before and after the samples to ensure the background variation did not contribute to the detection levels of the compounds. The limit of detection (LOD) was defined as the average laboratory blank response or instrumental noise value plus three times the standard deviation of the blank or instrumental noise value. For the siloxanes, the blank was subtracted from the samples because it can make up a large contribution of the signal when approaching the detection limits. After blank subtraction, 10 times the standard deviation of the blank was used to determine the limit of quantification (LOQ).

Lipid content was determined gravimetrically using n-hexane at the Norwegian Institute for Air Research, Tromsø (Hornøya samples) and the Norwegian Institute for Air Research, Kjeller (Oslofjord samples).

Stable isotope analysis

Stable isotopes of nitrogen (δ^{15}N), carbon (δ^{13}C), and sulfur (δ^{34}S) were analyzed by the Institute for Energy Technology, Kjeller. Analysis of nitrogen and carbon isotopes was done by combustion in an element analyzer. Nitrogen oxide was reduced to nitrogen gas (N_2) at 650°C in the presence of copper (Cu) before separation of N_2 and CO_2 on a GC-column and determination of δ^{13}C and δ^{15}N using an isotope ratio mass spectrometer (IRMS; Nu-instruments). Analysis of sulfur isotopes was done by combustion in an element analyzer with V_2O_5 to increase the amount of available oxygen reduction of SO_x to SO_2, separation of SO_2 from other products of combustion on a GC-column, and determination of δ^{34}S using IRMS. The stable isotope ratios were expressed as δ values relative to the internationally accepted standards: PeeDee Belemnite marine fossil limestone formation (Vienna) for δ^{13}C, atmospheric nitrogen (N_2) for δ^{15}N, and Vienna Canyon Diablo Troilite for δ^{34}S.

DNA damage: Comet assay

The comet assay procedure for detection of cellular DNA strand breaks is based on Gutzkow et al. (2013) as modified by

Haarr et al. (2018). White blood cells were obtained from the blood samples by centrifugation in a Percoll gradient (GE Healthcare). White blood cells were diluted 10, 20, and 40 times in phosphate-buffered saline ethylenediamine tetra-acetic acid (PBS-EDTA) to ensure optimal cell density for scoring. Due to logistical intractabilities and harsh field conditions, the PBS used for dilution of the first 13 Hornøya samples did not contain EDTA, and all dilution steps were done with PBS without EDTA for these samples. The last two Hornøya samples were diluted in PBS-EDTA because new shipments were received toward the end of the field campaign. To test potential effects of inclusion or exclusion of EDTA in the PBS-solution, the DNA fragmentation tail intensity was measured in human blood diluted in PBS with and without EDTA. No difference was found in tail intensity between the treatments (paired *t*-test, $p > 0.05$; Supporting Information, Figure S1a). In addition, there was no difference in tail intensity between the Hornøya gull samples treated with PBS with ($n = 2$) and without EDTA ($n = 13$; Wilcoxon test $p > 0.05$, KS-test $p > 0.05$; Supporting Information, Figure S1b). The DNA damage was therefore compared between the colonies as planned.

White blood cells were mixed with low melting point agarose (Sigma-Aldrich) and applied as 20-µl gels on Gelbond films. Deoxyribonucleic acid damage was measured on cells receiving two different treatments: baseline DNA damage and H_2O_2-induced damage. Baseline DNA damage was investigated by placing films directly into lysis buffer after the gels had set, with no further treatment. Sensitivity to a genotoxic stressor, H_2O_2, was measured by placing films in boxes containing 5 µM H_2O_2 at 4 °C in the dark for 15 min before placement in lysis buffer. Prior to the H_2O_2 treatment, the concentration of 5 µM was tested on herring gull blood to ensure the amount of induced damage was within a measurable range. The results of the concentration test proved adequate, with the resulting damage being measurable and not exceeding 80%. Human blood was used as a reference throughout the experiment, run alongside all samples, receiving all treatments. The blood belonged to the person performing the experiment and care was taken to avoid potential contamination with human pathogens.

All films were left overnight in lysis buffer at 4 °C. For unwinding and relaxation of supercoiled DNA, films were placed in electrophoresis buffer at alkaline conditions (pH 13–14) for 20 min. Electrophoresis was run for 25 min at 24 V and 4 °C. Films were then rinsed with neutralizing buffer and fixated by rinsing briefly in distilled H_2O, followed by 96% ethanol, before they were left to air dry overnight away from direct light exposure. For visualization of the DNA, the films were stained with a staining solution of TE-buffer (Tris-EDTA) SYBR Gold. A fluorescence microscope (Zeiss Axio Scope A1, ex/em 520/610) with a 20× objective coupled to a camera (Allied Vision Technologies) was used to inspect the DNA. To quantify DNA strand breaks, 50 cells were scored for each individual sample (gel) using Comet Assay IV software (Ver 4.2; Perspective Instruments). Overlapping or irregular cells were avoided, as well as cells near the edges of the gels or near any air bubbles or gel irregularities. The median of the 50 cells was calculated and used for representation of the DNA damage in each individual gull. The DNA damage is presented as tail intensity (% of the median of the total 50, log10 transformed).

Data treatment and statistical analyses

All statistics were done using R Ver 4.0.3 (R Core Team, 2017). Normal distribution of data was assessed using the Shapiro–Wilk test and homogeneity of variance was assessed using the Fligner–Killeen test. Contaminant concentrations were reported and treated in wet weight. All contaminants except cVMS were excluded from the data treatment if >20% of data values were below the LOD. The remaining nondetects in the dataset were substituted by a random value between 0.5×LOD and LOD, generated using the RAND-function in Excel. A total of 14 (of 781 data points) substitutions were made, representing 1.8% of the dataset used in the data treatment. In cVMS analysis, Warner et al. (2013) has previously shown that co-extracted matrix significantly contributes to background analytical variation at trace levels. This can result in the reporting of false positives, and to avoid this issue the use of method detection limit (MDL, matrix defined) is recommended. Due to logistical issues in obtaining a matrix which is free of siloxanes and mimics the sample matrix (whole blood) under investigation, the LOQ (average blank response+10×standard deviation in blank response, solvent defined) was used as a conservative detection limit to reduce the reporting of false positives. LOQ was found to be comparable or greater to MDL in the analysis of several environmental matrices (Krogseth et al., 2017), thus the LOQ (although not matrix defined) was considered appropriate to account for analytical variation introduced by the co-extracted matrix. Because of the low number of detected samples, no exclusion or replacement was done on the cVMS. Thus, results must be interpreted carefully and we discuss only the amount of detected samples.

Multivariate analyses were conducted using the vegan package in R (Oksanen et al., 2018). Principal component analysis (PCA) was used to analyze the structure in the variation of contaminant concentrations (wet wt and log transformed, log [$x + 1$]). The PCBs were grouped according to homologue group. Significant explanatory variables were identified by subsequent redundancy analysis (RDA). The full set of explanatory variables available included colony, trophic level, sex, baseline DNA tail intensity, lipid content, and body condition index (BCI). To assess if the baseline DNA damage could be explained by the contaminant variation, the PC1, PC2, and PC3 sample scores (individual herring gulls) were extracted from the PCA and entered as explanatory variables in a linear model with baseline DNA damage as the response.

A BCI was calculated for each herring gull using a multiple linear regression model. The body mass was the response variable, head length+wing length were predictor variables, and sex and location were included as significant factors (Fox et al., 2007). The residuals of the regression were defined as BCI (Jakob et al., 1996).

The $\delta^{15}N$ value was used to calculate the trophic level (TL) of the herring gulls from the respective colonies according to

Hobson and Welch (1992), with modifications suggested by Mizutani et al. (1991), Fisk et al. (2001), and Ruus et al. (2002).

$$TL = 3 + (\delta^{15}N_{herring\,gull} - (\delta^{15}N_{baseline} + 2.4))/3.8 \qquad (1)$$

Blue mussel (Mytilus edulis) was chosen as a representative marine species of both areas as baseline (inhabiting trophic level 2) to calculate the trophic levels of the herring gulls (trophic level) to adjust for differences in baseline $\delta^{15}N$ between the colonies. Values of $\delta^{15}N$ differ in blue mussel along the Norwegian coast (Green et al. (2016), and $\delta^{15}N$ values from a northern location (Brashavn $\delta^{15}N = 6.46$) and an urban Oslofjord location (Gressholmen $\delta^{15}N = 7.3$), relatively close to the remote and urban colony, respectively, were included from Green et al. (2016).

RESULTS AND DISCUSSION

Dietary niche

The herring gulls from the urban and remote colonies differed in stable isotope signatures beyond what would be expected based on potential differences in $\delta^{15}N$ baselines and carbon source in the food web base. A more depleted $\delta^{13}C$ indicates a relatively higher terrestrial carbon source and an enriched $\delta^{13}C$ indicates a relatively higher marine carbon source (Peterson & Fry, 1987). The $\delta^{13}C$ values in the remote Hornøya gulls were enriched with all values above −21.95‰, indicating that these birds feed primarily from the marine environment (Figure 1). The urban Oslofjord gulls were separated into two groups by their $\delta^{13}C$ values, some individuals below −23.5‰ and the rest above −22.5‰ (t-test: $t = 11.379$, $p < 0.0001$, $df = 13$; Figure 1), indicating different feeding habits within the colony with the former group having terrestrial dominated dietary input (terrestrial feeders) and the latter a more marine-derived dominated diet (marine feeders). Herring gulls are known to feed on terrestrial invertebrates, birds and mammals, plant material, and human leftovers and waste (Coulson, 2015; Ewins et al., 1994; Ingraham et al., 2020; Morris et al., 2003). Around the urban Oslofjord colony, herring gulls have been observed to forage on a range of human industrial food (personal observations). In addition, they are observed to feed in agricultural areas, and dietary items are known to include terrestrial items such as earthworms (Heimstad et al., 2018).

Enrichment in $\delta^{34}S$ has also been linked to marine-originated food, and $\delta^{34}S$ has been identified as a good tool for making detailed descriptions of diet in generalist species (Elliott et al., 2021; Eulaers et al., 2014; Moreno et al., 2010). Knoff et al. (2001) showed a higher $\delta^{34}S$ and $\delta^{13}C$ in laughing gulls (Leucophaeus atricilla) depending on more marine origin food than gulls with more estuarine prey. Lott et al. (2003) found enriched $\delta^{34}S$ in marine and coastal bird-eating raptors when compared with inland feeders. This tendency was evident in the urban Oslofjord gulls in the present study, with a strong correlation between $\delta^{13}C$ and $\delta^{34}S$. In addition to indicating the proportions of marine and terrestrial prey, depleted $\delta^{34}S$ has also been suggested to indicate association with urban areas, because sulfur emissions originating from combustion of fossil fuels typically have $\delta^{34}S$ values close to zero (Thode, 1991). Exemplifying this, Eulaers et al. (2014) found depleted $\delta^{34}S$ in white-tailed eagle (Haliaeetus albicilla) nestlings close to an urban area compared to more open coast locations, Morrissey et al. (2013) linked the depletion of $\delta^{34}S$ in Eurasian dipper (Cinclus cinclus) eggs to proximity to urban areas, and Moreno et al. (2010) found refuse to have the most depleted $\delta^{34}S$ values compared to marine and terrestrial dietary items of Spanish yellow-legged gull (Larus michahellis). In the present study, $\delta^{34}S$ values were, with the exception of one individual from the urban colony, which was similar to the remote, higher in the remote Honøya gulls than in the urban Oslofjord gulls, indicating terrestrial and urban influence in the Oslofjord. The remote Hornøya gulls $\delta^{34}S$ values ranged from 18.51‰ to 20.76‰ (Figure 1). Within the urban colony, the terrestrial individuals covered a range of 7.27‰–13.21‰, whereas a range of 13.71‰–17.36‰ was observed for the marine influenced individuals (Figure 1). This shows that the urban Oslofjord marine individual with the highest $\delta^{34}S$ was close to the lower range of the remote Hornøya gulls, whereas the three marine
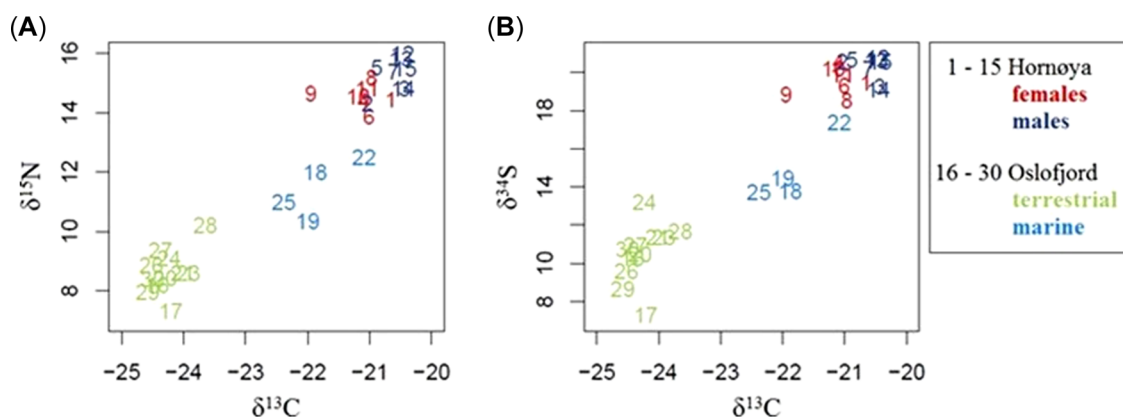

FIGURE 1: Stable isotope ratios of (**A**) $\delta^{15}N$ (‰) and $\delta^{13}C$ (‰), and (**B**) $\delta^{34}S$ (‰) and $\delta^{13}C$ (‰) in herring gulls from the urban colony Oslofjord (all females, other diet $n = 11$, marine diet $n = 4$) and the remote colony Hornøya (females $n = 7$ and males $n = 8$ m all with a marine diet), plotted by individual ID (1–30).

individuals with lower δ^{34}S were close to the range of the terrestrial gulls.

The δ^{15}N values were higher in the remote colony gulls than in the Oslofjord gulls (Figure 1). The δ^{15}N values of blue muscle collected from areas close to each colony was used as food web baseline in the trophic level calculation, ensuring a local signal. However, in the urban colony, blue mussel is likely not an accurate food web baseline for the terrestrial Oslofjord gulls because they likely have a high share of terrestrial food items in their diet. The calculated trophic level was used as an environmental variable in the PCA, but when directly comparing between the colonies, the comparison was limited to the urban marine Oslofjord and remote Hornøya gulls to avoid the potential bias in the trophic level calculations. Individual trophic level and δ^{15}N values can be found in Supporting Information, Table S1. The differences in trophic levels between the colonies placed the remote Hornøya gulls approximately one trophic level above the urban Oslofjord marine feeding gulls (Hornøya mean trophic level = 4.3 ± 0.1, Oslofjord marine mean trophic level = 3.4 ± 0.2). In the remote colony, the herring gulls were observed predating on eggs and other seabirds such as black-legged kittiwakes (*Rissa tridactyla*), which will increase both δ^{15}N and δ^{13}C values compared to fish feeders. In comparison to the remote herring gulls, the diet of gulls nesting close to human settlements shows a different diet which includes anthropogenic waste, terrestrial invertebrates, mammals, and plants (Ewins et al., 1994; Laurich et al., 2019; Mendes et al., 2018). The larger size of the remote Hornøya females compared to urban Oslofjord gulls can also explain the difference observed in trophic level, because larger gulls can predate on birds and larger fish, both food sources leading to higher δ^{15}N values (Leat et al., 2019; Nogales et al., 1995).

Contaminant concentration: location comparison

Based on knowledge of emissions and distribution in the environment, we expected to detect all the analyzed contaminant groups in both colonies, but with higher concentrations in the urban Oslofjord colony compared to the remote Hornøya gulls due to closeness to previous and current releases.

An initial PCA was run on individual contaminant concentrations (Supporting Information, Figure S2). Based on chemical properties and intercorrelations in the initial ordination, contaminants were grouped for further ordination and statistical analysis. The resulting groups were the PCB homologue groups tetra, penta, hexa, and hepta CB, cVMS (D4, D5, and D6), PBDE 99 and 153, PBDE 47 and 100, and PFOS (Figure 2). The correlations of PBDE 99 and 153, and PBDE 47 and 100 might be due to debromination mechanisms, because PBDE 153 debrominates to PBDE 99, and PBDE 100 debrominats to PBDE 47 (Wei et al., 2013), justifying the grouping of these contaminants in the final PCA. The PCA plot shows that concentrations did not differ between herring gull males and females from the remote Hornøya, and the sexes were pooled for further colony comparison. The final PCA with the grouped contaminants showed higher concentrations of POPs and PFOS in the remote Hornøya gulls, and higher detection frequency of cVMS in the urban Oslofjord terrestrial feeding gulls (Figure 2). Two urban Oslofjord marine feeding individuals had similar PCB and PFOS concentrations to those found in the remote Hornøya gulls and were closely positioned in the PCA. One urban marine individual had similar cVMS concentrations to the urban terrestrial individuals, showing individual variation in contaminant concentrations within the feeding groups. Based on the RDA, trophic level and colony were the only significant explanatory variables.
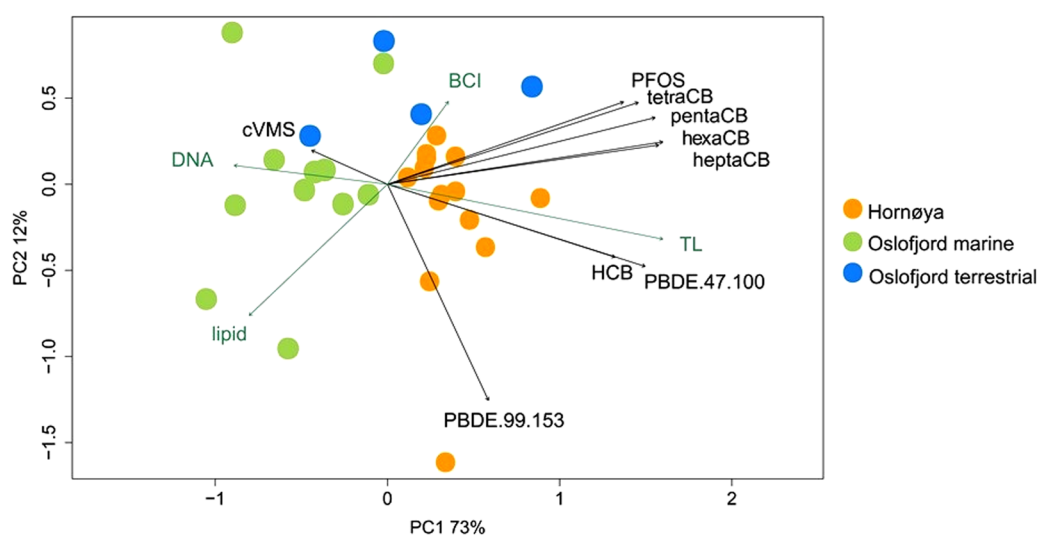

FIGURE 2: Principal component analysis (PCA) triplot based on contaminant concentrations (log10 (X + 1), ng/g wet wt) in individual herring gulls (1–30) positioned according to contaminant loading (response variables). Oslofjord (all females, other diet n = 11 [green], marine n = 4 [light blue]); Hornøya (all marine, females n = 7 [red], males n = 8 [dark blue]). Explanatory variables (DNA damage, lipid content, body condition index, trophic level) are shown as passive vectors (green), with significant explanatory variables (trophic level) marked with a yellow box. The proportion of variance explained by PC1 and PC2 is shown on the x and y axes, respectively. cVMS = cyclic volatile methyl siloxane; BCI = body condition index; PFOS = perfluorooctane sulfonic acid; TL = trophic level; HCB = hexachlorobenzene; PBDE = polybrominated diphenyl ether.
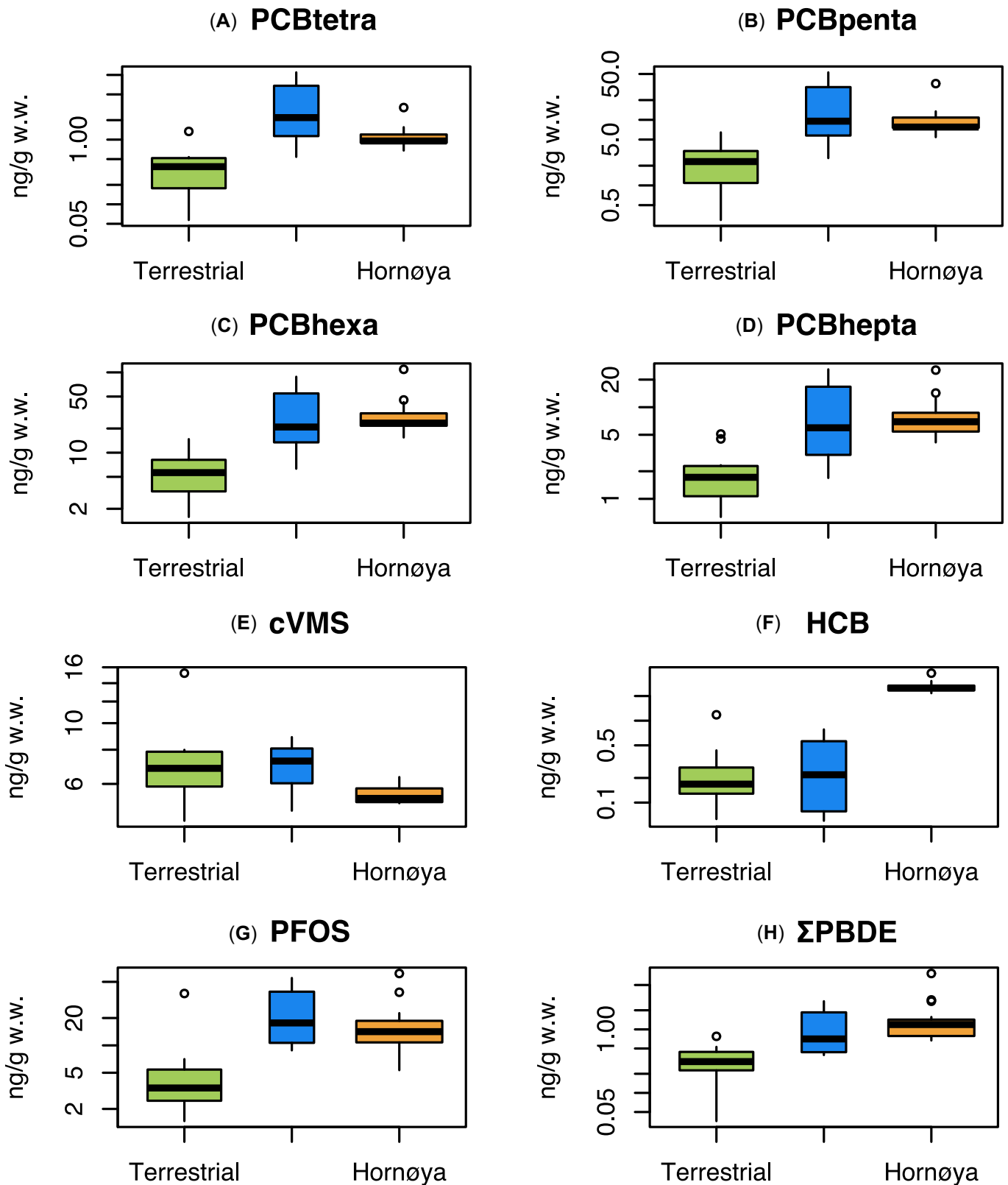
FIGURE 3: Concentrations (ng/g wet wt) of polychlorinated biphenyl ether (PCB) homologs (tetra-, penta-, hexa-, hepta-), cyclic volatile methyl siloxanes (cVMS), hexachlorobenzene (HCB), polybrominated diphenyl ether (PBDE), and perfluorooctane sulfonic acid (PFOS) in Oslofjord (all females, other diet [$n = 11$], marine [$n = 4$]) and Hornøya gulls (all marine, $n = 15$). The width of the boxes corresponds to the number of samples in each group. The boxes stretch from first to third quartile, with the horizontal line representing the measurement median. The whiskers extend to 1.5 times the interquartile range and the points are extreme values, also defined as outliers.

All the PCB homologue groups and PFOS correlated in the PCA, with higher concentrations in the remote Hornøya gulls than in the urban Oslofjord gulls. Hexachlorobenzene (HCB) and both PBDE groups, PBDE 47 and 100, and PBDE 99 and 153 also had higher concentrations in the remote than the urban colony gulls. Hexachlorobenzene was correlated to PBDE 47 and 100, and PBDE 99 and 153 was close to orthogonal to the PCB and PFOS groups, indicating no correlation. The PBDE 99 and 153 vector showed high concentrations in two remote Hornøya females. In a study of two urban Oslofjord herring gull colonies in 2017, the year after sampling for the present study, PBDE 99 and PBDE 153 also deviated from the other lipophilic POPs studied (Knudtzon et al., 2021). Furthermore, in a study of herring gulls from 1998 in the outer Oslofjord, which is also an urban area, PBDE 99 and PBDE 153 correlated positively (Sørmo et al., 2011).

As indicated by the PCA, the remote Hornøya gulls and urban Oslofjord marine feeders had similar concentrations of PCBs (Supporting Information, Table S2 and Figure 3). Considering the higher trophic level of the remote Hornøya gulls, this could indicate that the urban environment of the Oslofjord

colony results in increased exposure from lower trophic levels compared to the remote Hornøya birds.

Within the urban Oslofjord gulls, the concentrations of PCB, ∑PBDE, and PFOS were higher in the marine than in the terrestrial feeders (Table 1, Figure 3, and Supporting Information, Table S2). This was expected, because higher concentrations of legacy POPs in general are associated with marine feeding and higher $\delta^{15}N$ values as a result of distribution and accumulation of these contaminants in marine food webs and a more lipid-rich diet in the marine-feeding gulls leading to higher exposure (Bustnes et al., 2013). In the urban Oslofjord, higher concentrations in marine feeders can be explained by PCBs still being present in the inner Oslofjord due to leaching from legacy contamination (Breivik et al., 2004). However, the trend of higher concentrations of POPs with higher trophic position has not always been observed for PBDEs. For example, Elliott et al. (2009) found that while PCB concentrations increased with trophic position and marine input in the diet of bald eagle nestlings, PBDE concentrations increased with marine input, but not with trophic position. Also, Elliott et al. (2021) found that for PBDEs, spatial stable isotopes, especially $\delta^{34}S$, were

TABLE 1: Contaminant concentrations (ng/g wet wt) in herring gulls, mean ± standard deviation (SD) and range (min–max)

Mean ± SD min–max	Oslofjord		Hornøya	
	Nonmarine $n = 11$	Marine $n = 4$	Females $n = 7$	Males $n = 8$
HCB	0.28 ± 0.32 0.063–1.2	0.32 ± 0.33 0.060–0.78	2.5 ± 0.32 2.2–3.8	2.7 ± 0.50 2.2–3.8
PCB 47 (tetra)	0.13 ± 0.093 0.021–0.35	1.2 ± 1.5 0.14–3.5	0.34 ± 0.12 0.21–0.54	0.38 ± 0.24 0.25–0.96
PCB 66 (tetra)	0.28 ± 0.26 0.037–0.99	2.7 ± 3.2 0.40–7.5	0.71 ± 0.22 0.47–1.0	0.88 ± 0.53 0.58–2.2
PCB 99 (penta)	0.82 ± 0.62 0.091–2.0	5.9 ± 7.1 0.78–16	2.5 ± 0.99 1.5–4.3	3.3 ± 2.4 2.0–9.1
PCB 105 (penta)	0.41 ± 0.32 0.039–1.0	3.4 ± 4.3 0.45–9.8	1.4 ± 0.35 0.93–2.0	2.1 ± 1.7 1.1–6.3
PCB 118 (penta)	1.4 ± 1.1 0.17–3.4	9.4 ± 12 1.4–27	4.6 ± 1.4 3.0–7.1	6.9 ± 5.5 4.0–20
PCB 128 (hexa)	0.32 ± 0.23 0.060–0.79	2.8 ± 3.9 0.32–8.6	1.6 ± 0.57 0.99–2.7	2.4 ± 2.2 1.4–7.7
PCB 138 (hexa)	2.6 ± 1.8 0.51–5.7	14 ± 16 2.7–38	9.9 ± 3.7 6.4–17	15 ± 912 8.7–43
PCB 153 (hexa)	3.7 ± 2.4 1.0–8.4	17 ± 17 3.3–42	13 ± 5.9 8.1–26	20 ± 16 11–58
PCB 180 (hepta)	0.74 ± 0.62 0.20–1.9	3.2 ± 3.2 0.39–7.9	4.3 ± 1.5 2.7–7.2	6.1 ± 5.0 2.8–18
PCB 183 (hepta)	0.35 ± 0.32 0.064–1.1	1.6 ± 1.9 0.28–4.4	2.1 ± 1.6 0.92–5.6	2.4 ± 1.1 1.2–4.3
PCB 187 (hepta)	0.95 ± 0.57 0.30–2.1	5.0 ± 5.8 1.0–14	0.87 ± 0.31 0.52–1.5	1.2 ± 0.95 0.64–3.5
PFOS	6.7 ± 10 1.5–37	25 ± 21 8.8–5	15 ± 11 5.3–38	21 ± 17 10–62
PBDE 47	0.16 ± 0.086 0.018–0.28	0.81 ± 0.85 0.21–2.1	1.05 ± 0.68 0.48–2.3	0.83 ± 0.41 0.45–1.7
PBDE 99	0.12 ± 0.097 0.0070–0.37	0.14 ± 0.078 0.065–0.25	0.60 ± 1.3 0.036–3.6	0.10 ± 0.079 0.039–0.27
PBDE 100	0.040 ± 0.023 0.0060–0.071	0.17 ± 0.17 0.051–0.43	0.36 ± 0.30 0.14–0.80	0.25 ± 0.12 0.12–0.47
PBDE 153	0.029 ± 0.017 0.0050–0.069	0.027 ± 0.016 0.010–0.047	0.19 ± 0.34 0.020–0.95	0.083 ± 0.092 0.026–0.30

The Oslofjord birds, all females, are split into nonmarine feeders and marine feeders based on their stable isotope values. Hornøya birds are both males and females. The PCB homologues are included in brackets and indicate the number of chlorine substitutions on the biphenyl.
PBDE = polybrominated diphenyl ether; PCB = polychlorinated biphenyl ether; PFOS = perfluorooctane sulfonic acid; HCB = hexachlorobenzene.

TABLE 2: Individual concentrations or limit of quantification of cyclic volatile methyl siloxanes D4, D5, and D6 in herring gull blood from Hornøya and the Oslofjord sites

Location	D4	D5	D6
Hornøya	2.5	<LOQ (1.6)	2.26
Hornøya	2.01	<LOQ (1.6)	<LOQ (2.0)
Hornøya	2.44	<LOQ (1.6)	<LOQ (2.0)
Hornøya	1.53	<LOQ (1.6)	NA
Hornøya	1.56	<LOQ (1.6)	2.44
Hornøya	1.81	<LOQ (1.6)	2.37
Hornøya	1.53	<LOQ (1.6)	<LOQ (2.0)
Hornøya	<LOQ (1.5)	<LOQ (1.6)	<LOQ (2.0)
Hornøya	1.57	<LOQ (1.6)	<LOQ (2.0)
Hornøya	<LOQ (1.5)	<LOQ (1.6)	<LOQ (2.0)
Hornøya	1.55	<LOQ (1.6)	<LOQ (2.0)
Hornøya	1.7	<LOQ (1.6)	<LOQ (2.0)
Hornøya	1.98	<LOQ (1.6)	2.44
Hornøya	1.62	<LOQ (1.6)	2.09
Oslo	2.24	2.71	<LOQ (2.5)
Oslo	2.8	2.56	<LOQ (2.5)
Oslo	2.37	4.02	<LOQ (2.5)
Oslo	<LOQ (2.0)	2.77	<LOQ (2.5)
Oslo	<LOQ (2.0)	2.78	<LOQ (2.5)
Oslo	<LOQ (2.0)	1.42	2.45
Oslo	<LOQ (2.0)	1.73	<LOQ (2.5)
Oslo	2.02	1.67	<LOQ (1.1)
Oslo	<LOQ (2.0)	<LOQ (1.3)	<LOQ (1.1)
Oslo	<LOQ (2.0)	1.43	<LOQ (1.1)
Oslo	2.61	10.11	<LOQ (2.5)
Oslo	3.04	2.45	<LOQ (2.5)
Oslo	2.21	1.68	<LOQ (2.5)

For the cyclic volatile methyl siloxanes (cVMS), not all samples were analyzed In the Oslofjord, $n = 13$ for all cVMSs, on Hornøya, $n = 14$ for D4 and D5, and $n = 13$ for D6.

more important for explaining concentrations than trophic position, as indicated by $\delta^{15}N$. A difference between accumulation of PCBs and PBDEs in herring gulls could be reflected by the lack of correlation between PCBs and PBDE 99 and 153, and the weak correlation between PCB and PBDE 47 and 100 in the PCA in our study. This could be a result of different sources of exposure for PCBs and PBDEs. The difference between the marine and terrestrial feeders observed in our study illustrates the impact of diet within the area, and the lower concentrations in the terrestrial feeders likely reflect lower contamination of these banned legacy contaminants in the urban area compared to the marine environment. However, within the urban colony, the PBDE groups 99 and 153, and 47 and 100 did not correlate with either of the stable isotopes (Supporting Information, Table S3), supporting the results of Sørmo et al. (2011) which indicated little correlation of the stable isotopes and PBDEs 99 and 153.

Hexachlorobenzene concentrations were similar in the urban terrestrial and marine gulls (Table 1, t-test: $t = 0.072$, $p = 0.94$, $df = 13$), but concentrations were higher in the remote colony individuals (Table 1 and Figure 3; Welch test: $t = -11$, $p < 0.0001$, $df = 14$), illustrating a clear difference between the urban and remote locations. Hexachlorobenzene is easily transported with long-range transport, and a south–north gradient of HCB concentrations in environment and biota increasing with increasing latitude has been observed (Beyer et al., 2000; Sun et al., 2020).

Cyclic volatile methyl siloxane concentrations were generally low, with many samples below or close to the LOQ. For D5, none of the 14 samples were above LOQ in the remote colony, while 12 out of 13 were quantified in the urban colony, with mean concentration 2.8 ng/g wet weight (LOQ 1.6 and 1.3 ng/g wet wt in the remote and urban samples, respectively; Table 1). Limits of quantification and individual concentrations of D4, D5, and D6 can be found in Table 2. Cyclic volatile methyl siloxanes have previously been linked to proximity to human settlements, and concentrations of cVMSs are reportedly higher in water, sediment, and aquatic biota closer to urban areas (McGoldrick et al., 2014; Sparham et al., 2008; Warner et al., 2010). The high detection frequency of D5 in the urban compared to the remote colony indicates that this is also the case in the present study.

In addition to location and trophic level, migration and overwintering location can contribute to the observed contaminant concentrations in the birds (Leat et al., 2013). However, as a result of relatively rapid clearance of POPs from
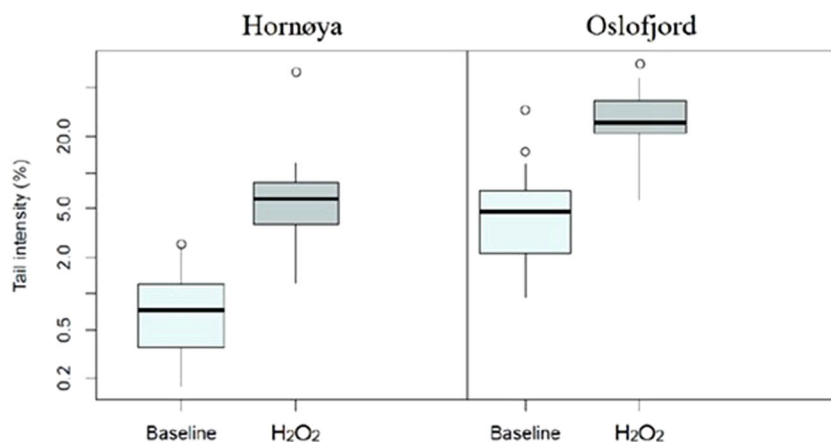

FIGURE 4: DNA damage (tail intensity) for both locations, Oslofjord ($n = 15$) and Hornøya ($n = 13$), showing response to treatments: baseline (no treatment), H₂O₂ treated, and recovery (incubated in phosphate-buffered saline at room temperature for 4–8 h after exposure to H₂O₂). The boxes include data from first to third quartiles, with the line representing the measurement median. The whiskers extend to 1.5 times the interquartile range and the points are extreme values, also defined as outliers.

blood, blood could be a relatively good indicator matrix of recent, local contaminant exposure (Miller et al., 2020).

DNA strand breaks

Because male and female herring gulls from the remote Hornøya did not differ in DNA strand breaks, neither in the baseline nor the H_2O_2 treatment (Supporting Information, Figure S3 and Table S4), the sexes were grouped. When comparing DNA strand breaks in baseline and H_2O_2 treatment between the marine and terrestrial feeding urban gulls, results were also similar (Supporting Information, Figure S4 and Table S4), and the feeding groups were therefore combined.

There was higher DNA damage in the urban Oslofjord than the remote Hornøya colony for all treatments (Figure 4 and Supporting Information, Table S4). The similar levels of DNA damage between the marine and terrestrial urban gulls indicate that the amount of DNA damage detected is not affected by the concentrations of the contaminants included in our study. This was supported by testing association between PC1 and PC2 site scores from the contaminant concentration PCA and baseline DNA damage. For PC1, which explained 73% of total variation in the PCA, there was a negative relationship with baseline DNA damage (linear model: Supporting Information, Table S5 and Figure S5). Principle component 2 (explained 12% of total PCA variation) showed a slight increase with baseline DNA damage, but with a high p value (linear model: Supporting Information, Table S5 and Figure S5).

The observed DNA damage could be due to other contaminants not included in our study, like polycyclic aromatic hydrocarbons, which are known to cause genotoxicity (Cachot et al., 2006). There are also possible synergistic effects from the exposure mixture these urban gulls experience compared to the remote colony. In addition to contaminants, several other stressors contribute to increased oxidative stress and DNA damage, such as available food sources and quality of diet, proximity to human activities, predation, and reproduction (Bourgeon et al., 2012). Lower quality food as well as increased disturbance from human activities and pressures on habitats could be expected in the urban colony, possibly contributing to the observed higher levels of DNA damage in the urban birds. The urban Oslofjord colony is located in a nature reserve with restricted access of the public during the breeding period, but the total amount of disturbance of the colony is likely higher in the heavily trafficked Oslofjord than at remote Hornøya. The urban and remote colonies were sampled at roughly the same time in the breeding season, close to egg hatching, meaning that there should be little difference between them when considering reproductive stress.

For both locations, there was an increase in DNA damage from baseline treatment to peroxide (Figure 4). The induced oxidative damage was more prominent in the urban gulls than in the remote gulls. A possible reason for this is that the urban gulls, already having more DNA damage to repair due to the elevated baseline DNA damage, were less equipped to repair additional oxidative damage (Alonso-Alvarez et al., 2004).

In conclusion, our study showed that although concentrations of PCBs, PBDEs, and PFOS were similar between the remote Hornøya and the urban Oslofjord marine herring gulls, the apparently lower trophic level in the urban gulls indicates higher exposure in the urban area, also for these contaminants. The similar concentrations of the emerging cVMS in the marine and terrestrial urban Oslofjord gulls indicate similar exposure in the urban environment regardless of food source. Levels of DNA damage were highest in the urban Oslofjord area, apparently regardless of dietary input. The most likely explanation for this is that the presence of other genotoxic contaminants and stressors in urban environments produce greater effects on oxidative stress and DNA damage. White blood cells from herring gulls with initially higher DNA damage deteriorated most after exposure to oxidative stressor, H_2O_2. This was likely because the already heightened oxidative damage in the cells caused a poorer response to the addition of more oxidants, because the antioxidant and repair systems were already fully mobilized in the cells.

Author Contributions Statement—**Ellen Kristine Keilen**: Formal analysis; Investigation; Software; Visualization; Writing—original draft; Writing—review & editing. **Helene Skjeie Thorstensen**: Formal analysis; Investigation; Software; Visualization; Writing—original draft; Writing—review & editing. **Katrine Borgå**: Conceptualization; Investigation; Supervision; Validation; Writing—review & editing. **Ketil Hylland**: Conceptualization; Investigation; Supervision; Validation; Writing—review & editing. **Morten Helberg**: Conceptualization; Data curation; Investigation; Methodology; Resources; Supervision; Writing—review & editing. **Nicholas Warner**: Investigation; Methodology; Resources; Writing—review & editing. **Kine Bæk**: Investigation; Methodology; Resources; Writing—review & editing. **Tone Kristin Reiertsen**: Investigation; Methodology; Resources; Writing—review &

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