



Co-occurrence of contaminants in marine fish from the North East Atlantic Ocean: Implications for human risk assessment

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ABSTRACT

Marine fish from the North East Atlantic Ocean (NEAO) are nutrient rich and considered a valuable economic resource. However, marine fish are also a major dietary source of several contaminants, including persistent organic pollutants (POPs) and heavy metals. Using one of the world's largest seafood datasets ($n > 25,000$ individuals), comprising 12 commercially important fish species collected during 2006–2019 in the NEAO, we assessed the co-occurrence of elements and POPs, and evaluated potential risks to human consumers. Several positive correlations between concentrations of mercury (Hg), dioxins, polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs) were observed. Concentrations of Hg, dioxins, PCBs and PBDEs increased from North to South and associations between marine sediment contamination, sea temperature, and fish Hg and POPs concentrations were identified using multi-linear regression (MLR) models. In general, Hg concentrations in fillet and liver of fish were positively associated with increases in both sediment contamination and sea temperature. POPs concentrations in both fillet and liver were positively associated with increases in sediment contamination, and only POPs concentrations in the liver of benthopelagic and demersal species were found to be positively correlated with sea temperature. Using a probabilistic approach to estimate human contaminant exposure from seafood, we showed that intake of pelagic species posed the highest risk of dioxins and dioxin-like PCBs (DL-PCBs) exposure, while intake of benthopelagic and demersal species posed the highest risk of Hg exposure. This study can serve as a model to further understand the distribution, co-occurrence, and trends of contaminants in seafood harvested from the NEAO and their potential risks to human consumers.

1. Introduction

Global fish production was estimated to be ~179 million tons in 2018 and the importance of seafood to ensure global food security will likely increase with human population growth (FAO, 2020). Seafood safety research has also garnered significant interest from policy makers, scientists, and food security professionals. Ocean health has become a paramount issue in the context of the United Nations Sustainable Development Goals (SDGs) which aim to end hunger and malnutrition by 2030 (Bank et al., 2020). Seafood provides nutrients, such as high

quality marine proteins, long chain omega-3 polyunsaturated fatty acids, as well as essential vitamins and elements (Khalili Tilami and Sampels, 2018; Mohanty et al., 2014; Nøstbakken et al., 2021). However, the occurrence of pollutants such as protein-binding methylmercury (MeHg), and lipid soluble persistent organic pollutants (POPs, e.g. dioxins (sum of dioxins and furans, PCDD/PCDF), polychlorinated biphenyls (PCB) or polybrominated diphenyl ethers (PBDE)) may pose a significant health concern for consumers (Azad et al., 2019b; Boitsov et al., 2019; Huang et al., 2020; Nøstbakken et al., 2018, 2021). Fish from the North East Atlantic Ocean (NEAO) are affected by a wide array

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of environmental stressors, including local and global pollution and climate change effects such as increases in sea temperature and ocean acidification (Bank et al., 2020).

Bioaccumulation of contaminants varies across fish species and geographical areas within the NEAO (Azad et al., 2019b; Boitsov et al., 2019; Nøstbakken et al., 2018). Previous surveillance studies have described geographical and seasonal variations in contaminant concentrations in single fish stocks from the NEAO (Frantzen et al., 2015; Frantzen et al., 2011; Julshamn et al., 2013a; Julshamn et al., 2013b; Julshamn et al., 2013c). In general, concentrations of most pollutants in marine fish are known to be higher in southern areas of the NEAO compared to the north (Azad et al., 2019b; Nøstbakken et al., 2018). However, no study has combined data for a large array of contaminants in marine fish taxa from the NEAO to assess their combined geographical variation together with comprehensive data on sediment concentrations, salinity, and sea temperature. Increased sea temperature can affect the ocean's biological pump and influence trophic transfer dynamics of carbon (Davison et al., 2013; Irigoien et al., 2014) and nutrients (Colombo et al., 2020) as well as bioaccumulation regimes of contaminants. However, information on how increases in sea temperature across latitudinal gradients may influence bioaccumulation of co-occurring contaminants is not well understood. To address variability in risk of contaminant exposure due to seafood consumption, probabilistic approaches including surveillance data have been used to assess risk–benefit dynamics in human consumers (Moreno-Ortega et al., 2017; Morisset et al., 2013). This approach has not been utilized for data from marine fish harvested in the NEAO, and likely provides a better understanding of potential effects on human health by assessing exposure risk of multiple contaminants concomitantly.

The objectives of this study were to: (1) investigate patterns of co-occurrence and geographical variation of seafood elements and contaminants including total arsenic (As), total mercury (Hg), selenium (Se), and selected POPs (dioxins, PCBs, and PBDEs) in a wide array of marine fish taxa from the NEAO, and to assess the potential relationships with environmental factors such as sediment contaminant concentrations, sea temperature, and salinity, and (2) examine human exposure patterns of these contaminants via seafood consumption. A dataset comprising 12 commercially important fish species ($n > 25,000$ individuals) sampled from the NEAO was chosen as a model system to test the following hypotheses and *a priori* predictions, which were based on knowledge derived from previous investigations comprising the dataset and now evaluated using a more mechanistic approach. Specifically, we predicted that: (1) different contaminants will co-vary in marine fish and that contaminant concentrations will vary geographically (e.g. latitude) and are driven by abiotic factors including sediment contamination, sea temperature and salinity, and (2) risk of human exposure to contaminants from marine fish varies across geographical regions, fish life history strategies, and patterns of habitat use. Lastly, using a probabilistic approach, and exposure estimations from selected contaminants along with NEAO fisheries and trade information, we also evaluated human exposure scenarios.

2. Materials and methods

2.1. Study area and fish sampling

The data presented in this study comprises results from analyses of 25,631 individual samples from 12 commercially important marine teleost species including Atlantic cod (*Gadus morhua*), Atlantic halibut (*Hippoglossus hippoglossus*), Atlantic herring (*Clupea harengus*), Atlantic mackerel (*Scomber scombrus*), Atlantic salmon (*Salmo salar*), common ling (*Molva molva*), European hake (*Merluccius merluccius*), Greenland halibut (*Reinhardtius hippoglossoides*), haddock (*Melanogrammus aeglefinus*), plaice (*Pleuronectes platessa*), saithe (*Pollachius virens*) and tusk (*Brosme brosme*) collected mostly from Norwegian fisheries areas in the NEAO (Table S1). Wild fish were sampled using different sampling gear

including long line, gill net, purse seine and pelagic trawl between 2006 and 2019 by the Norwegian reference fleet composed of commercial fishermen collaborating with researchers. Sampling was also conducted on scientific cruises with research vessels owned by the Institute of Marine Research (IMR) or hired fishing vessels. Wild fish were caught in the northeastern parts of the Atlantic Ocean covering most of the important fishing areas (from 20 °W to 42 °E and 50 °N to 80 °N). Fish samples caught in polluted harbors and wreck sites were excluded in this study, however, contaminant concentrations in fish from some of the fjords included are known to be elevated either due to either local point sources or long-range transport (Azad et al., 2019a, 2021). Most fish were frozen whole at -20 °C before being shipped to the laboratory. The NEAO was divided into five regions; the Barents Sea, the Norwegian Sea, the North Atlantic, the North Sea and Skagerrak as defined by Azad et al. (2019b), as these regions are distinct ecosystems characterized with climate, geographical and pollution features, possibly associated with concentrations of elements and POPs in biota. The Barents Sea is considered to be the most pristine (Azad et al., 2019b), whereas Skagerrak is the most polluted region (Magnusson et al., 1996). The Barents Sea is the northernmost Arctic shelf sea with low temperatures. The Barents Sea is the northernmost Arctic shelf sea with low temperatures, while sea temperatures in the Norwegian Sea, North Atlantic, North Sea and Skagerrak are higher since sea temperatures increase with decreasing latitude (Eldevik et al., 2014). The Norwegian Sea, separating the North Sea from the Barents Sea, which are both rather shallow ocean areas, features a steep slope along the continental edge, descending swiftly to abyssal depths.

2.2. Laboratory analyses

Weight and length of each fish were recorded, and the liver was removed before filleting, using clean equipment, and employing trace metal clean sampling techniques. Skin-free fillet and liver samples were homogenized prior to analysis, and homogenized fillet samples were lyophilized, except for some fillet samples from Atlantic mackerel that were not freeze dried due to high oil content (Frantzen et al. (2010)). Samples were analyzed for a wide array of elements including Hg, As and Se, as well as the POPs classes of dioxins (PCDD/F), DL-PCB, PCB6, and PBDE7. The method used for determination of elements is a Nordic, European, and US standard for Hg, As, cadmium (Cd) and lead (Pb) (CEN, 2009; Julshamn et al., 2013d) and analyses were performed using inductively coupled plasma-mass spectrometry (ICP-MS). The methods for determination of PCDD/F, DL-PCB, PCB6 and PBDEs have been modified over time, and these methods have been described in detail in other peer reviewed publications (Berntssen et al., 2005; Lundebye et al., 2017). PCDD/F and non-ortho PCB analyses were performed by high-resolution gas chromatography/high resolution mass spectrometry HRGC-HRMS. PCB6, mono-ortho PCBs and PBDEs were determined by gas chromatography-tandem mass spectrometry (GC–MS–MS) or GC–MS. More details of the analytical methods are described in Supplementary Text S1.

2.3. Geographic assessment of marine sediments, sea temperature, and salinity

Inorganic environmental elements and POPs marine sediment data (2003–2019) were obtained from MAREANO's Chemistry Program (<https://www.mareano.no/kart-og-data/kjemidata>, see more details in Supplementary Text S2). Data included Hg, As, Se, PCB6 and PBDE7 concentrations. For the dioxin-like polychlorinated biphenyls (DL-PCB), only PCB-105, –118 and –156 were measured and the sum of these congeners were considered as DL-PCB indicator for the sediments. Of PBDEs, PBDE –209 is often the main congener in the sediments, but this congener was not included in the current analysis. Only PBDE7 (including PBDE-28, –47, –99, –100, –153, –154 and –183) were selected for the analysis since they are the most common PBDE

congeners reported in seafood species (Lundebye et al., 2017; Nøstbakken et al., 2018). The spatial distribution of sea temperature and salinity in the NEAO from 2005 to 2019 was obtained from IMR oceanographic data (see more details in Supplementary Text S2). The sediment contaminant concentrations, sea temperature and salinity data were incorporated into the seafood dataset to evaluate the associations of combined abiotic factors with element and POP concentrations. Geographical plots of elements and POPs in seafood species harvested from the NEAO and in sediments were performed in Program R (R Core Team, 2020) using the “rnatuarearth” and “rnatuarearthdata” and “gam” packages.

2.4. Statistical analyses

All seafood element and POPs data were reported on a wet weight basis. Statistical analyses were performed using raw upper bound (UB) data, where concentrations below the limit of quantification (LOQ), were substituted with the LOQ-value. For element analysis, > 40% and >85% of the results for Cd and Pb, respectively, were below the LOQ and therefore these elements were excluded from subsequent analyses.

Concentration data of elements and POPs were not normally distributed and were log transformed to obtain an approximate normal distribution. To analyze the relationship between elements and POPs we used Pearson correlation coefficients. Fish species have different ranges of weight and length and can be exposed to different concentrations of specific elements or POPs (Azad et al., 2019b; Nøstbakken et al., 2018). Therefore, to analyze correlation relationships across these variables, in all fish species, scaling was applied using log-transformed data of each species to normalize the range of the element/POP concentrations, using the following formula:

$$x_{s,i} = \ln(c_i) - \overline{\ln(c_i)} \quad (1)$$

where $x_{s,i}$ is the scaled value of the log-transformed concentration of one individual of the fish species i with concentration c_i , $\overline{\ln(c_i)}$ is the mean value of the log-transformed concentration of the fish species i .

For combined datasets across all fish species, scaling of fish length was applied. Fish length in each fish species was scaled with its mean value using the formula outlined here:

$$FL_{s,i} = FL_i / \overline{FL_i} \quad (2)$$

where FL_i and $\overline{FL_i}$ are the individual fish length and the mean fish length of the specific fish species i . Subsequently to the scaling procedure, correlation analyses, analysis of variance (ANOVA) or analysis of covariance (ANCOVA) were performed. For ANOVA or ANCOVA tests, distribution and normal Q-Q plots of residuals (the differences between values and the mean of all values for the group) were carried out to examine data normality. Fish length was correlated with contaminant concentrations in fish fillets and therefore, ANCOVA with scaled fish length as a covariate (i.e., to control for the effects of fish length variation) was used to compare contaminant concentrations of fish fillets from different geographical areas by latitude. Unlike contaminant concentrations in fillets, contaminant concentrations in liver was only weakly correlated with fish length. Therefore, ANOVA was applied to evaluate contaminant concentrations in fish liver samples. Statistical significance was accepted at $p < 0.05$ and all analyses were performed using program R (R Core Team, 2020) operated in RStudio (version 1.3.959; RStudio Team, 2015).

Multiple linear regression (MLR) models for concentrations of elements and POPs (i.e., As, Se, Hg, dioxins, PCBs or PBDEs) in fillet or liver samples in association with latitude and fish length are described as follows:

$$x = A_0 + \sum A_m * p_m \quad (3)$$

where x is the predicted scaled concentration of the element or POP, p_m

is the independent variable, i.e. latitude or fish length, A_0 and A_m are the model coefficients.

Important abiotic factors including sea temperature, salinity, and degree of sediment contamination governed by geographical factors including latitude could be associated with concentrations of elements and POPs in seafood. To evaluate associations of abiotic factors and fish length with Hg and POPs concentrations in fillet or liver samples we also used MLR's (Eq. (3)) with combined sea temperature, salinity or sediment contaminant concentrations, and fish length as independent variables. Notably, latitude was correlated with abiotic factors including temperature and contaminant levels in sediments. Therefore, to avoid multicollinearity of independent variables, the abiotic factors and latitude were not simultaneously incorporated into the model.

We used the Bayesian model selection inference technique, Akaike's Information Criterion (AIC), in addition to r-squared (r^2) values to evaluate goodness of fit of several candidate models with either single or multiple variables as described in Eq. (3), (Burnham and Anderson, 2002; Burnham et al., 2011). For linear regression models, r^2 refers to the squared Pearson correlation coefficient values between element or POPs concentrations and the selected independent variables. For MLR models, r^2 indicates the square of the correlation between the fitted linear model and the response data. A model with $r^2 < 0.1$ (or $|r| < 0.32$) implied weak correlation of the predicted model and was not considered in subsequent analyses. Moreover, r^2 can be used to compare among goodness of fit for MLR models describing elements and POPs.

To evaluate the important variation of element or POP concentrations with respect to geographical variation (latitude) or abiotic factors (sea temperature, salinity, or sediment contamination), sensitivity analyses were performed. The relative sensitivity of elements or POPs with respect to variation of the variable p_m was defined using the following formula:

$$\frac{\partial x}{\partial p_m} = A_m \quad (4)$$

Since x is the scaled value of the log transformed concentration c of the element or POP, $\partial x = \frac{\partial c}{c}$. Therefore, Eq. (4) can be rewritten as follows:

$$S_{c,p} = \frac{\partial c}{c} = A_m * \partial p_m \quad (5)$$

where $S_{c,p}$ is the relative change of contaminant concentration in relation to variation of the abiotic factor ∂p_m . A high value of A_m indicates that the concentration of the element or POP is highly influenced by a small change in the independent variable p_m .

2.5. Probabilistic exposure analysis based on fish consumption

Human exposure from ingestion of contaminants such as Hg, dioxins, DL-PCBs, and PBDEs in NEAO seafood was evaluated in a risk assessment context. The human exposure assessment, presented here, focused only on fillet data since fish livers, in general, are not regularly consumed (Guil-Guerrero et al., 2011). Briefly, the probabilistic distribution parameters of contaminant concentrations including Hg, dioxins, and DL-PCBs in fillets of each fish species were estimated by fitting the log-normal distribution to the data of contaminant concentrations. Using Monte-Carlo simulations, a dataset (100,000) of fillet contaminant concentrations containing all fish species was randomly generated from their probabilistic distribution parameters. We considered a total of 12 commercially important marine fish species from the NEAO as the main source of seafood consumption for the risk assessment section of this study. The production volumes of the fish species i harvested in the NEAO was also accounted for in the analyses (see more details in Supplementary Text S3, Table S2). The probabilistic distributions of weekly intake $WI(c)$ of Hg ($\mu\text{g}/\text{kg}$ body weight) or dioxins and DL-PCBs ($\text{pg TEQ}/\text{kg}$ body weight) by seafood consumption (defined in

Supplementary Text S3) were computed. For the risk assessment of PBDEs, a margin of exposure (MOE) approach was used as suggested by the European Food Safety Authority (EFSA, 2011) and lower MOE values indicate higher risk. When MOE is lower than the threshold set by EFSA for a given contaminant contribution source, mitigating action is recommended. We assessed PBDE 47, 99, and 153 at the upper confidence limit of 95% (see more details in Supplementary Text S3). Risk assessment analyses were also performed using program R including packages “fitdistrplus” and “ggridges”. The package “fitdistrplus” was used to estimate parametric distributions. Subsequently, Monte-Carlo simulations were performed also using program R. The package “ggridges” was used to visualize results.

3. Results

3.1. Co-occurrence of elements and POPs in NEAO marine fish

Overall, significant variation in concentrations of elements and POPs among fish species from the NEAO was observed (Fig. 1 and Supplementary Table S3). The geometric mean Hg concentrations ranged from 5.23×10^{-2} to 2.44×10^{-1} mg/kg and 1.14×10^{-2} to 0.39 mg/kg in fillet and liver, respectively. The geometric mean concentrations of As in fillet ranged from 1.71 to 18.1 mg/kg, whereas geometric mean values in liver samples varied from 4.02 to 11.5 mg/kg. The geometric mean Se concentrations in liver samples ranged from 1.09 to 3.69 mg/kg, an order of magnitude larger than in fillets (2.58×10^{-1} to 5.22×10^{-1} mg/kg). The geometric mean fillet concentrations of the sum of dioxins varied from 2.27×10^{-2} to 0.937 pg TEQ/g whereas the sum of dioxins and DL-PCBs ranged from 4.97×10^{-2} to 2.54 pg TEQ/g. The geometric mean concentrations of PCB6 and PBDE7 in fillet ranged from 5.4×10^{-1} to 20.9 ng/g and 1.9×10^{-2} to 1.09 ng/g, respectively. The geometric mean concentrations of dioxins and DL-PCB, PCB6 and PBDE7 in the liver samples ranged from 4.09 to 39.4 pg TEQ/g, 15.5 to 468 ng/g and 0.97 to 21.5 ng/g, respectively, from one to two orders of magnitude larger than those in fillet samples. All geometric mean concentrations of Hg and POPs in fillet were below the EU's maximum levels (European Union, 2020) (Supplementary Text S4). However, the mean TEQ levels of sum dioxins and DL-PCB in liver of common ling and tusk both exceeded the EU maximum level of 20 pg TEQ/g applying to the total TEQ of dioxins and DL-PCB in fish liver. The EU maximum level of 200 ng/g for PCB6 in fish liver was exceeded by mean concentrations only in ling and tusk.

To explore the relationships between different elements and POPs, Pearson correlation matrices were computed using log-transformed and scaled concentrations across all fish species including both fillet and liver samples (Fig. 1B & 1C). Strong significant correlations between POPs including PCDD/F, DL-PCB, PCB6 and PBDE7 in the fillet (r of 0.73–0.91, $p < 0.05$) and the liver (r of 0.71–0.90, $p < 0.05$) were observed. Also, Hg concentrations were significantly correlated with POPs in fillets (r of 0.41–0.51, $p < 0.05$) and liver samples (r of 0.50–0.59, $p < 0.05$). As and Se concentrations exhibited weak correlations with Hg in both fillet and liver samples and did not correlate well with POPs concentrations in either tissue types. The relatively strong correlations between POPs and Hg concentrations indicated co-occurrence of these contaminants.

3.2. Fat content, Hg, and POPs in fish

Correlation analyses were performed to evaluate possible relationships between fat content, Hg, and POPs across and within species (Table S4). For all species, except Atlantic halibut and European hake, negative correlations between fat content and Hg were observed, with r ranging from -0.52 for fillets of wild Atlantic salmon to -0.072 in fillets of Atlantic herring (Table S4). For all species combined, a weak negative correlation ($r = -0.25$) between fat content and Hg in fillets was observed, whereas a moderate negative correlation ($r = -0.44$) between

fat content and Hg was detected in liver samples.

The relationships between fat content and POPs within each fish species and tissue types were highly variable. In fillets from semi-fatty species with mean fat contents ranging from 1.95% (European plaice) to 4.49% (Atlantic halibut), strong positive correlations between fat content and POPs were observed (r ranging from 0.46 to 0.83). Fatty pelagic species such as wild Atlantic salmon and Atlantic mackerel had mean fat content of 7.83% and 21.6% respectively and we found no correlation or weak negative to weak positive correlations between fat content and POPs (r from -0.12 to 0.21). In Atlantic herring (mean fat content 13.0%), weak positive correlations between fat content and POPs were observed (r ranging from 0.33 to 0.46). It is noteworthy that no strong relationships (i.e., low correlation values) were observed between fat content and POPs in fillets of pelagic species. Similarly, liver samples of lean fish species such as Atlantic cod, haddock, common ling and saithe had high fat content ranging from 51.0% in Atlantic cod to 58.2% in common ling (see Table S3), and weak negative correlations between fat content and POPs were observed (r of -0.37 to 0.06). For all species combined, moderate positive correlations (r of 0.30–0.43) between fat content and POPs in fillets were observed whereas fat content in liver samples showed a weak negative correlation with POPs (r of -0.22 to -0.06).

3.3. Geographical variation of elements and POPs in fish from NEAO

Fillet and liver samples from fish inhabiting the NEAO showed a decrease in concentrations of Hg, dioxins, PCB6, and PBDE7 from south to north (Fig. 2, Fig. S1). This was demonstrated by moderate negative Pearson correlation values for both fillet and liver Hg and for liver POPs with latitude (r value range from -0.48 to -0.35 and -0.65 to -0.44 , respectively, $p < 0.05$, Fig. 1B, Fig. 1C). This pattern was not observed for either As or Se. For analysis purposes, the NEAO was divided into five smaller regions, Skagerrak, North Sea, North Atlantic, Norwegian Sea and Barents Sea (Fig. 2). Fillet and liver samples in the Skagerrak region had the highest concentrations of Hg, DL-PCB, PCB6 and PBDE7, followed by those collected in the North Sea and then the Norwegian Sea. Samples from the Barents Sea had the lowest concentrations of these contaminants (Fig. 3). In addition, levels of elements and POPs in fillet and liver samples did not show a clear correlation with distance to shore/fjords (r value range from -0.17 to 0.053 in fillet and -0.33 to 0.13 in liver, respectively).

We used linear regression (LR) to quantify potential associations between fish elements and POPs concentrations, and latitude (Table S5). Neither As nor Se concentrations in fillet or liver samples exhibited a clear trend with latitude (r^2 model fit < 0.1 , Table S5), while concentrations of Hg in fillet (all species) and liver of benthopelagic and demersal fish and the different POPs sums in fillet of pelagic fish and in liver of benthopelagic and demersal species all showed clear latitudinal trends ($r^2 > 0.1$). Incorporating fish length into LR analyses of contaminants in fish fillets improved the r^2 model fit and the associated AICs values (Table S5). Hg concentrations in fillet (all species) and liver samples of benthopelagic and demersal species decreased with increasing latitude (negative A_{Lat} and r^2 model fit ranging from 0.11 to 0.47, Table S5). POPs concentrations in fillets of pelagic, but not benthopelagic and demersal species, significantly decreased with increasing latitude ($r^2 > 0.1$, Table S5). In all species where livers were analyzed (benthopelagic and demersal species), the LR models showed relatively moderate to high negative correlations (negative A_{Lat}) between liver POPs concentrations and latitude (r^2 of 0.15–0.50, Table S5). The model describing Hg concentrations in fillet samples exhibited the best goodness of fit with r^2 values ranging from 0.37 to 0.47 (Fig. S2, Table S5). For POPs, PBDE7 and dioxin concentrations in fish liver exhibited the best correlations with latitude (r^2 of 0.37–0.43 and 0.22–0.50, respectively), followed by liver PCB6 and DL-PCB concentrations (r^2 of 0.16–0.20 and 0.15–0.19, respectively). Hg and POPs concentrations in fillet samples decreased with increasing latitude (represented by

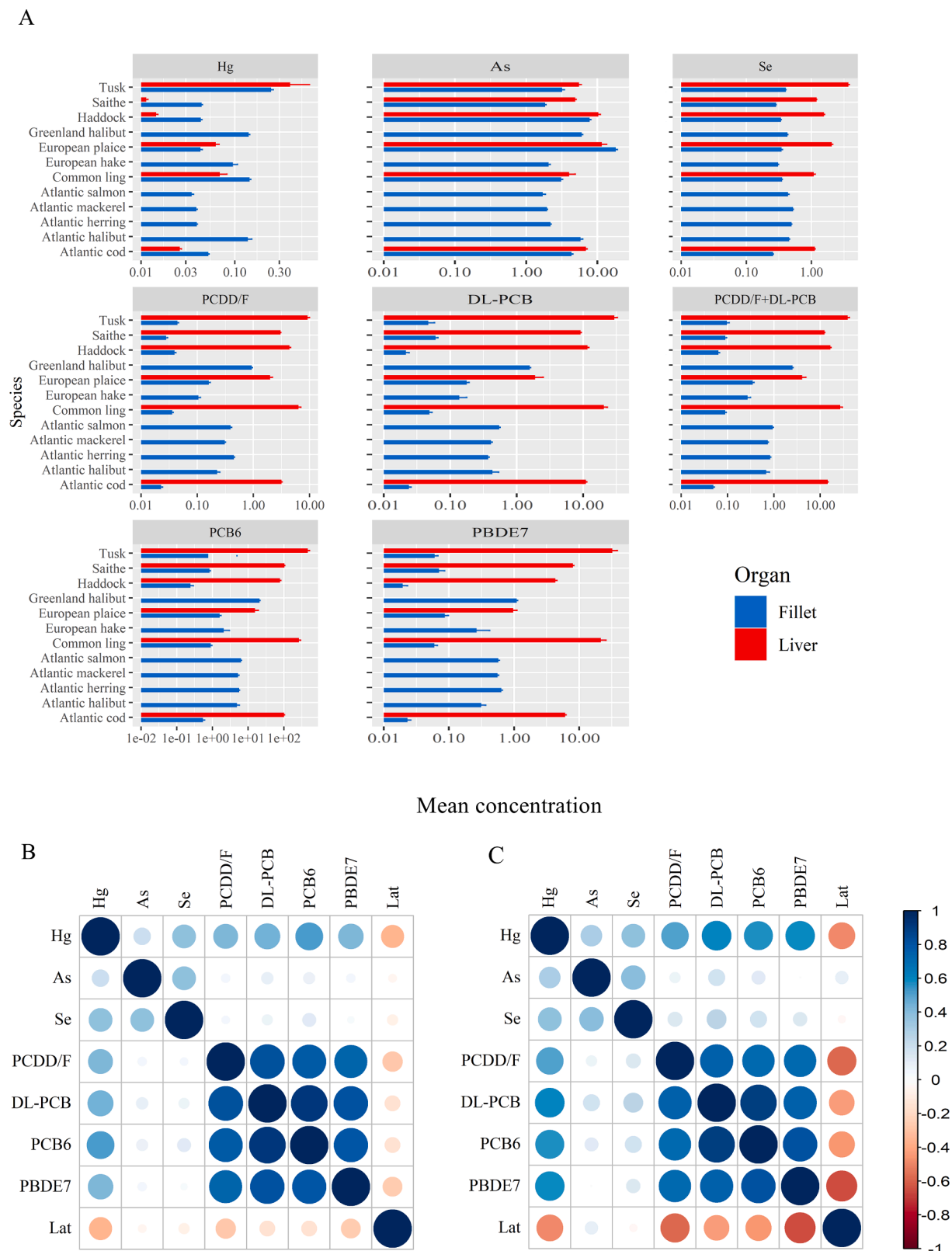


Fig. 1. A) Geometric means and standard errors (error bars) of elements and POPs concentrations in fillets and liver samples of different fish species from NEAO sampled during 2006–2019. B) and C) are Pearson correlations between contaminants and latitude (Lat) in fish fillets and liver samples, respectively. As, Se, Hg, PCDD/F, DL-PCB, PCB6, PBDE7 and Lat denote arsenic (mg kg^{-1}), selenium (mg kg^{-1}), mercury (mg kg^{-1}), sum of dioxins and furans (pg TEQ g^{-1}), sum of dioxin-like polychlorinated biphenyls (DL-PCBs, pg TEQ g^{-1}), sum of non-dioxin-like PCBs (PCB6, ng g^{-1}), sum of polybrominated diphenyl ethers (PBDE7, ng g^{-1}) and latitude ($^{\circ}$ degree), respectively (see Table S3 for their values). In (B) and (C), the data were scaled with the log-transformed levels across all fish species (see Materials and Methods). Color bar and circle size in B and C indicate the level of correlation.

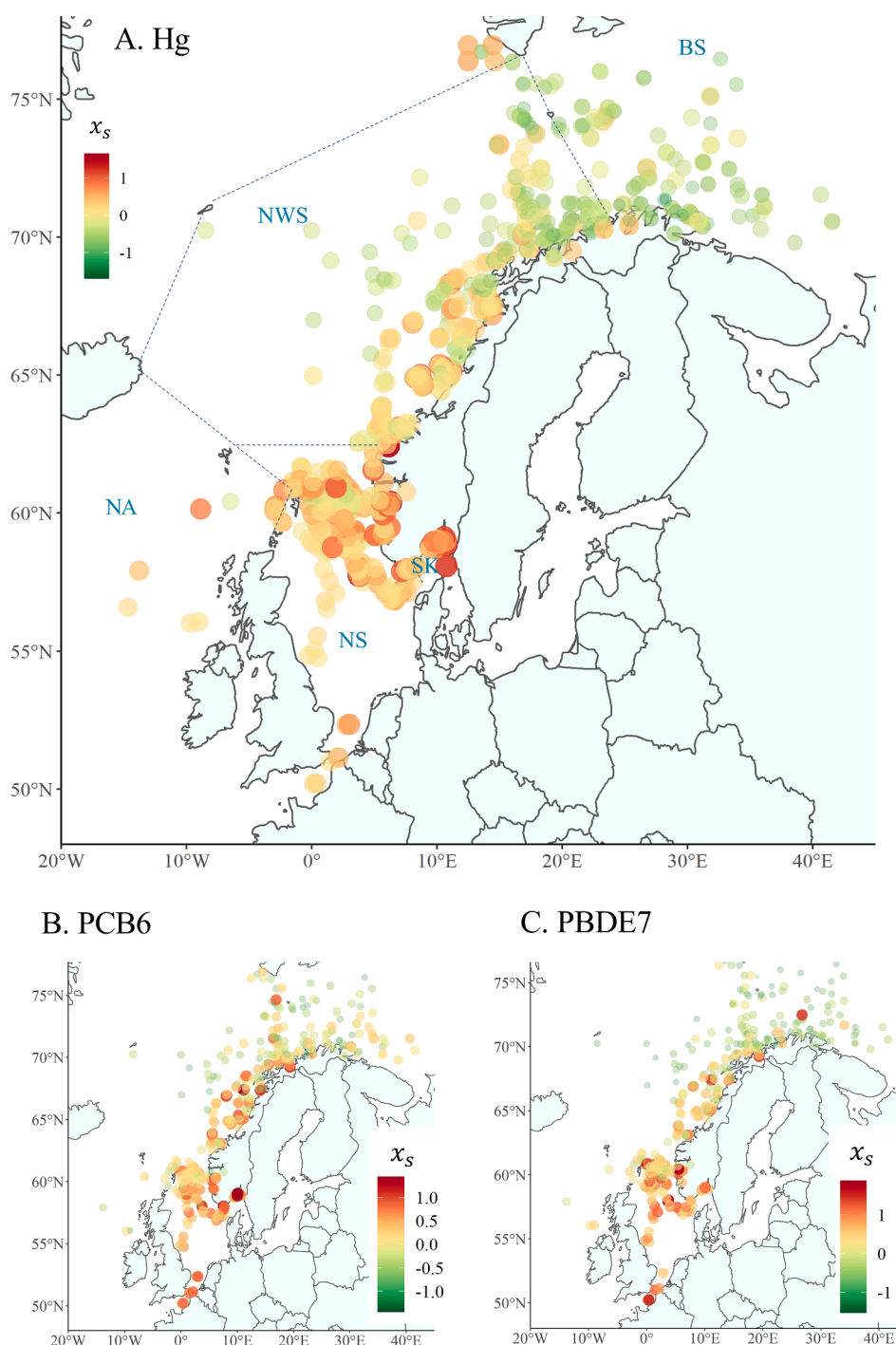


Fig. 2. Distribution of Hg, PCB6 and PBDE7 in seafood (both fillet and liver) from the NEAO, 2006–2019. x_s is the scaled value of the log-transformed level across all fish species (see Equation (1)). BS, NWS, NA, NS and SK denote the Barents Sea, the Norwegian Sea, the North Atlantic, the North Sea and Skagerrak, respectively. Color and circle size correspond to the scaled value of the log-transformed level.

negative sign of A_{Lat} , p -values < 0.05, Table S5), but significantly increased with increasing fish length (positive sign of A_{FL} , p -values < 0.05, Table S5). To analyze the sensitivity of Hg and POPs with latitude, we evaluated latitude coefficients of the LRs (Table S5). Concentrations of Hg in fillets of fish from different habitats significantly decreased between 4.51% and 6.82% per increased degree latitude. Highest sensitivity in POPs concentration to latitude was found for liver PBDE7 (9.30–9.49% decrease per increased degree latitude), followed by liver dioxin concentrations (4.59–8.30% decrease per increased degree latitude), and liver PCB6 of benthopelagic species (4.85–5.94% decrease per increased degree latitude) (Table S5). The decreasing concentrations

in fish with increasing latitude may be due to variation of different abiotic factors such as sea temperature, salinity, and/or contaminant concentrations in seawater and sediments.

3.4. Sea temperature, salinity, Hg/POPs in sediments and Hg/POPs in marine fish

Concentrations of Hg, As, Se, DL-PCBs, PCB6 and PBDE7 in sediment samples from the Barents Sea, Norwegian Sea, North Sea and Skagerrak are shown in Fig. S3. Since there was variation in sediment composition, we used the average values of contaminant concentrations representing

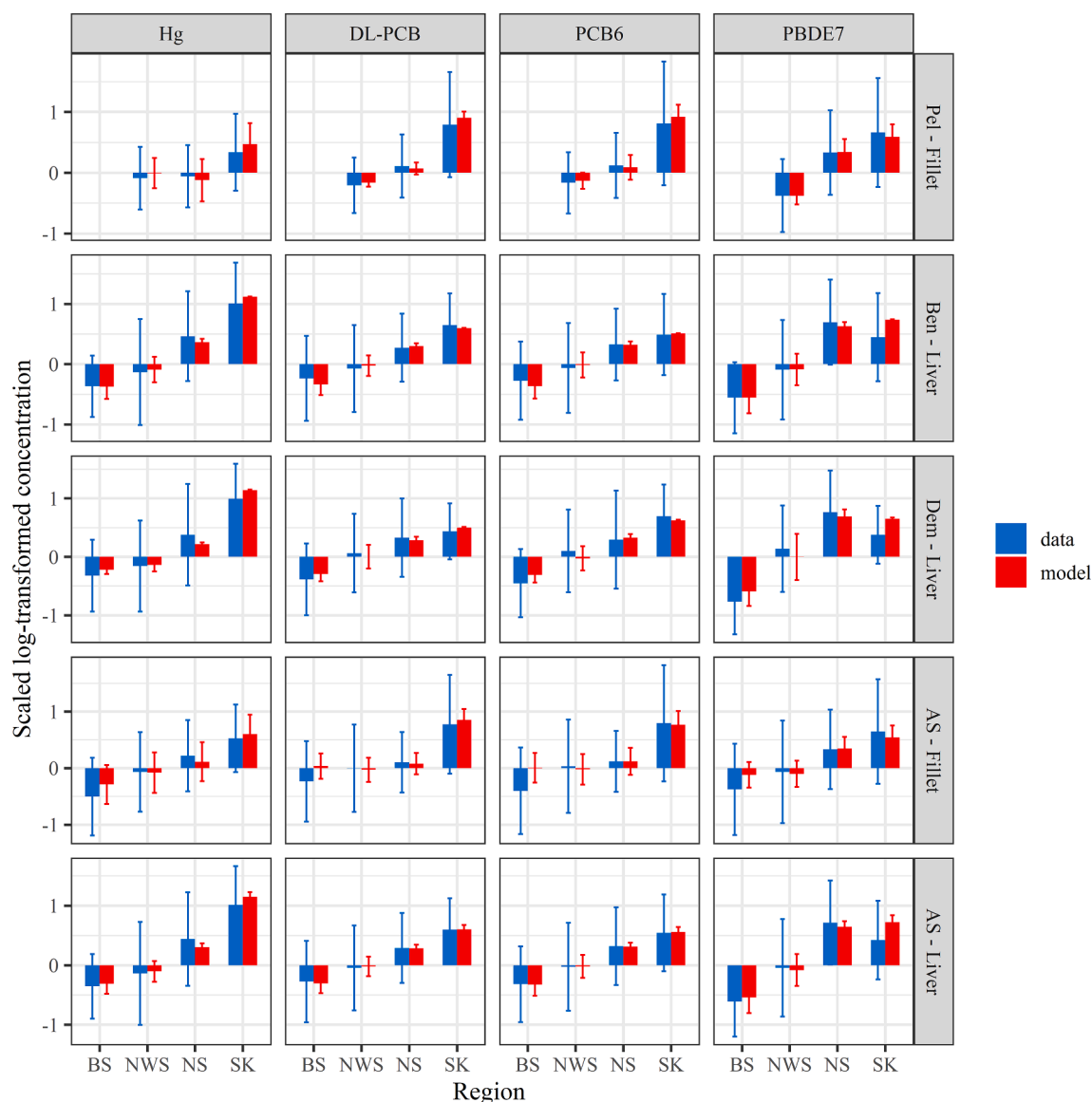


Fig. 3. Variation in mercury (Hg) and POPs concentrations in marine fish collected from four different regions. Results are shown for Hg, sum of dioxin-like PCBs (DL-PCB), sum of non-dioxin like PCBs (PCB6) and sum of 7 polybrominated diphenyl ethers (PBDE7), respectively. Values are presented in scaled log-transformed concentration (means \pm standard deviations). Blue color presents data sampled from the four different sea regions while red color presents the fitted model at the same geographical positions of the collected samples. Model parameters were described in Table S7. BS, NWS, NS and SK denote the Barents Sea, the Norwegian Sea, the North Sea and Skagerrak, respectively. Pel, Ben, Dem and AS denote the pelagic, the benthopelagic, the demersal and all species, respectively. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

different sea regions in our analyses (Fig. S4). There were no measured sediment data for As, Hg and POPs in the North Atlantic or for Se in the North Atlantic, the North Sea and Skagerrak. Concentrations of Hg, As, Se and PCB6 in surface sediments were the lowest in the Norwegian Sea and moderately increased from the Norwegian Sea to the Barents Sea whereas concentrations of DL-PCBs and PBDE7 in the Norwegian Sea and the Barents Sea were similar. Highest concentrations of Hg, DL-PCBs, PCB6 and PBDE7 were observed in sediment samples collected in Skagerrak, followed by those collected in the North Sea and the Barents Sea. Notably, relatively few sediment samples were taken in the North Sea and the Skagerrak compared to the Norwegian Sea and the Barents Sea, and in the Norwegian Sea, sediment samples were mainly taken on the continental shelf or continental slope and not in the deep parts of the Norwegian Sea. As expected, the mean temperatures of the seawater column decreased with increasing latitude and were also higher in shallow sea regions along the coast and within fjords compared to deeper offshore areas (Fig. S5). Salinity did not vary latitudinally, but

varied slightly between open ocean, coast, and fjord and with sea depth (Fig. S6).

Three abiotic factors comprising (a) degree of sediment contamination, (b) sea temperature, and (c) salinity were analyzed together with the marine fish dataset using MLR models to evaluate their associations with Hg and POPs in seafood. In our study, normalization of POPs concentrations to fat content generally reduced goodness of fit (r^2) of the model (Table S6), and therefore, was not considered further in subsequent analyses. When either sea temperature or sediment-associated contaminants were excluded from the MLR model, r^2 values of the MLR model were reduced considerably (r^2 values reduced by up to 0.11 and 0.15 for the MLR models describing Hg and POPs, respectively) (Table S6). However, excluding salinity from the MLR model mostly did not affect the r^2 values of the MLR model (variation of r^2 value $<$ 0.03) (Table S6). Furthermore, excluding salinity from the MLR model applied for contaminants of all fish species combined did not significantly change the values of sea temperature and sediment-associated

contaminant coefficients (See Fig. S7). Therefore, of the investigated factors, sediment contaminants and sea temperature were thus identified as the primary abiotic factors associated with seafood contaminants (Table S7). Using the MLR model for all fish species combined, Hg levels in fillet and PBDEs levels in liver showed the best association with sea temperature and sediment-associated contaminants since r^2 values computed from the model (0.31 and 0.34 for Hg and PBDEs, respectively) were higher than those computed from the model of the remaining contaminants in fillet and liver samples for all fish species combined (Table S7). Fillets from pelagic species and liver samples from benthopelagic and demersal species were analyzed separately due to their high concentrations of POPs. Higher r^2 values in fillets of pelagic species improved the model fit for POPs. Variation in seafood contaminant concentrations measured and modelled from four different NEAO regions and the measured and model fitted seafood Hg and POPs concentrations data were in good accordance (Fig. 3).

Using the MLR model describing Hg and POP concentrations in both fillet and liver samples, coefficients of sediment contaminants A_{sed} of the models were shown to be significantly positive, suggesting that contaminant concentrations in both tissue types were positively associated with changes in sediment contaminants (Table S7). The average concentrations (dry weight) of Hg, DL-PCBs, PCB6 and PBDEs in surface sediments collected in the NEAO were 3.16×10^{-2} mg/kg, 2.73×10^{-1} µg/kg ($\sim 8.20 \times 10^{-3}$ pg TEQ/g), 1.07 µg/kg and 5.68×10^{-1} µg/kg, respectively. Variation of Hg or POPs in seafood with respect to variation of sediment contaminants was further evaluated using MLR models. Hg concentrations in fillet and liver of seafood increased by 25.5% and 45.2%, respectively, with a doubling of sediment-associated contaminants. Similarly, with a doubling of sediment POP concentrations, POPs in fillet increased between 17.8% and 52.7% while POPs in liver increased between 5.3% and 21.7%. POP concentrations in fillet thus were more sensitive to sediment-associated contaminants compared to liver samples. Associations between Hg concentrations in fillets for pelagic, benthopelagic, and demersal fish species and sediment-associated Hg showed a similar trend ($S_{c, sed}$ of 25.5%, 31.7% and

29.5% for pelagic, benthopelagic and demersal fish species, respectively). Additionally, Hg concentrations in liver of demersal fish species ($S_{c, sed} = 52.2\%$) and POP concentrations in fillets of pelagic fish species ($S_{c, sed}$ from 24.0 to 52.7%) were the measurements most influenced by changes in sediment contaminant concentrations (Fig. 4A).

In general, Hg levels in both fillet and liver samples were positively associated with changes in sea temperature as illustrated by the significant positive coefficients of temperature A_T in the MLR models (Table S7). Concentrations of Hg in fillet and liver samples of fish from all habitats increased from 5.3% to 15.4% per degree Celsius (Fig. 4B). POP concentrations in fillets from all fish species and pelagic species were non-significantly related to sea temperature except for DL-PCB in all fish species which showed slight and significantly negative associations with sea temperature. Conversely, POPs concentrations in liver of benthopelagic and demersal species were significantly and positively associated with sea temperature. Concentrations of POPs in liver of benthopelagic and demersal species increased between 9.7% and 19.6% with an increase of 1 °C in sea temperature. Among the different POPs, concentrations of PBDE7 in liver of benthopelagic and demersal species were most strongly associated with temperature ($S_{c, T}$ from 19.3 to 19.6%) followed by concentrations of PBDE7 in liver of all fish species combined ($S_{c, T}$ of 16.9%) (Fig. 4B).

3.5. Risk estimation and assessment of seafood consumption

According to fish capture data from FAO (2006–2019) Atlantic herring (38.6%), Atlantic cod (22.2%) and Atlantic mackerel (18.4%) were the most captured among the studied fish species ($n = 12$) (Table S2). Capture data was used to estimate relative consumption rate of each species, and species-specific variation in Hg and dioxin and DL-PCB concentrations were characterized using probabilistic distribution parameters (Table S8). Using Monte-Carlo simulations, frequency distributions corresponding to weekly intakes of Hg and dioxin and DL-PCBs from fish from different habitats were calculated assuming fish fillet intakes of 200 g, 300 g and 400 g per week, respectively (Fig. 5A Fig. 5B)

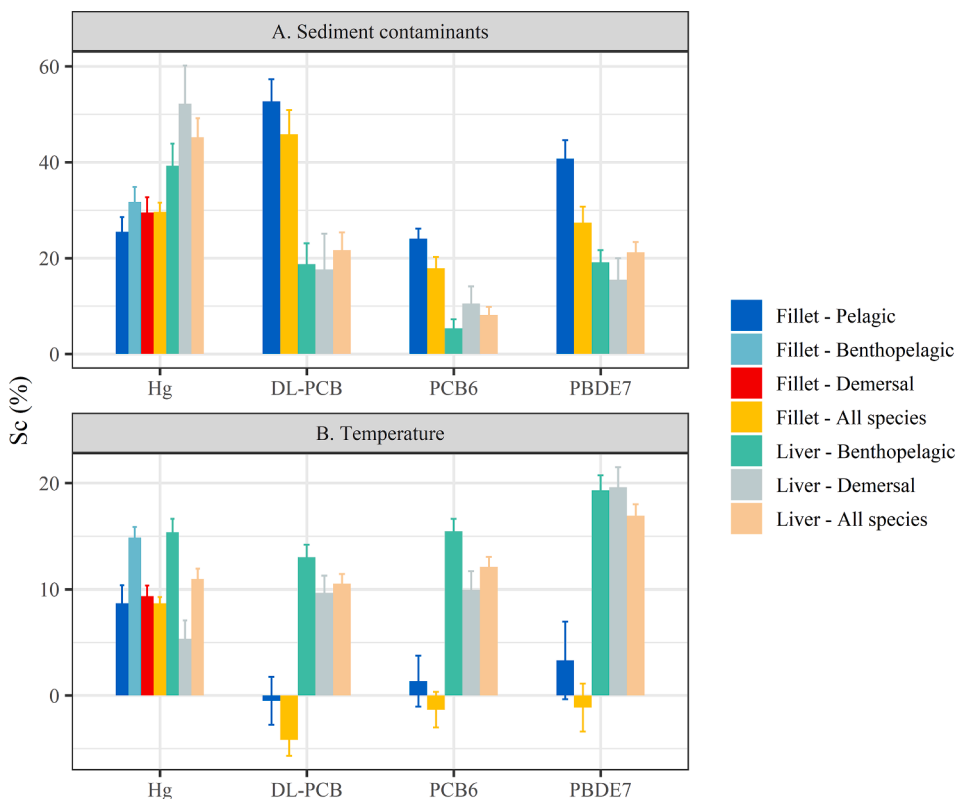


Fig. 4. Relative change of contaminant levels in seafood (S_c) (means \pm 95% confidence intervals) in relation to variation of sediment-associated contaminant (A) and temperature (B). In (A), $S_{c,C} = A_{sed} \cdot \partial c_{sed}$ where A_{sed} is the sediment-associated concentration coefficient of the MLR model (see Table S7); ∂c_{sed} is the variation of sediment-associated concentration. A double concentration increase of averaged sediment-associated concentration in the NEAO was selected ($\partial c_{sed} = c_{sed}$) in the study. In (B), $S_{c,T} = A_T \cdot \partial T$, where A_T is the temperature coefficient of the MLR model (see Table S7); ∂T is the temperature variation ($\partial T = 1^\circ\text{C}$).

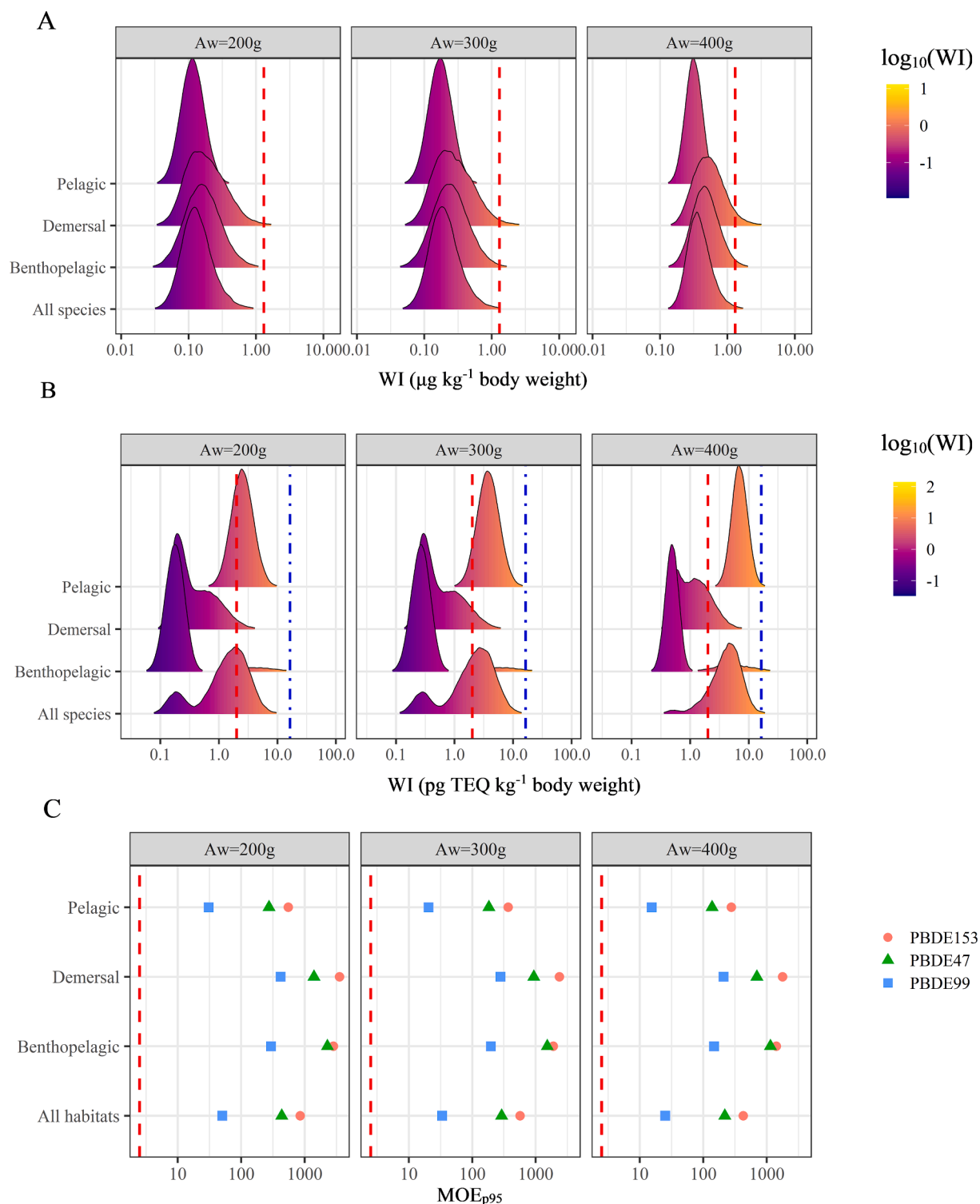


Fig. 5. Probabilistic exposure analysis of mercury (Hg) and POPs intake via seafood consumption. A) Total weekly intake (WI) of Hg. B) WI of sum dioxins (dioxins and DL-PCBs). C) Margin of exposures (MOE) calculated with upper confidence limits (95%) of PBDE47, PBDE99, PBDE153 due to fish consumption. Dashed red lines in (A), (B) and (C) are the tolerable weekly intake (TWI) and threshold of MOE set by EFSA (see more details in Supplementary text S3). Dash-dotted blue line is criterion of dioxins and DL-PCBs set by JECFA (a provisional tolerable monthly intake (PTMI) of 70 pg TEQ/kg body weight per month, this equates to TWI of 16.3 pg TEQ/kg body weight per week). A_w is the amount of fish consumption per week (g). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

to account for varying degrees of consumption. Seafood is considered to be an important source of dietary MeHg (Berry and Ralston, 2008; Moreno-Ortega et al., 2017) and as shown in Fig. 5A, consumers of marine fish from NEAO were at low risk of exceeding EFSA's tolerable

weekly intake (TWI) for MeHg of 1.3 $\mu\text{g}/\text{kg}$ body weight (EFSA, 2012). The probability of consumers exceeding the TWI was 1.0% with fish intake up to 400 g/week. If either benthopelagic or demersal species were considered as the sole fish source with an intake of 400 g/week, the

probability of consumers exceeding the TWI for MeHg was 2.0% or 5.4%, respectively (Fig. 5A).

The predicted probability of exceeding the TWI of sum dioxin and DL-PCBs recently reduced to 2 pg TEQ kg⁻¹ body weight by EFSA (EFSA, 2018), ranged from 34.8% to 87.0% for a 70 kg person consuming 200 g to 400 g fish fillet per week (Fig. 5B). The highest risk was found from consumption of pelagic species (i.e., fatty fish species such as Atlantic mackerel and Atlantic herring), followed by consumption of benthopelagic and demersal species. The wide variation with two peaks in the weekly intake of dioxin and DL-PCBs from benthopelagic species was primarily driven by the consumption of Greenland halibut fillets which exhibited relatively high POPs concentrations. Exposure to contaminants due to seafood intake varied with geographical regions. For example, Atlantic mackerel caught in Skagerrak contained about twice the concentration of dioxin and DL-PCBs compared to those caught from the other regions. Consuming 200 g fillet of mackerel from the Skagerrak area would lead to 86.4 % probability of exceeding the TWI for dioxins and DL-PCBs (Fig. S8), versus approximately 50% for other regions. To evaluate PBDE exposure, margin of exposure (MOE) was calculated as suggested by EFSA (2011). We assessed PBDE 47, 99, 153 at the upper confidence limit of 95% (see Supplementary Text S3 for definition). PBDE 99 constituted the highest risk within this group, indicated by the lowest MOE values compared with the other congeners (Fig. 5C). The MOEs for PBDE 47, 99, and 153 assessed for all habitats were, however, well above the MOE threshold of 2.5 set by EFSA (EFSA, 2011), suggesting low overall risk to human health from the intake of PBDEs from seafood harvested from the NEAO.

4. Discussion

In this study, we report co-occurrence of Hg and selected POPs in 12 NEAO marine fish species. Concentrations of Hg and POPs, but not As and Se, were negatively correlated with increasing latitude. This was in accordance with previous investigations, based on some but not all of the same individual fish, which found that PBDEs and Hg concentrations in most fish species were also negatively correlated with latitude (Azad et al., 2019a; Nøstbakken et al., 2018). In general, As in marine organisms appears mainly in organo-arsenic forms with low toxicity (Francesconi, 2010) and is not of anthropogenic origin. Although As and Hg patterns in sediment are similar, the patterns between the two elements in marine fish were quite different. In our study, As concentrations in either fillet or liver samples did not exhibit a clear declining trend with increasing latitude while Hg concentrations in both fillet and liver samples decreased considerably with increasing latitude. Se and MeHg exhibit mutual de-toxication mechanisms (Gajdosechova et al., 2016). Hence the presence of Se may affect bioavailability and toxicity of MeHg in seafood (Azad et al., 2019a). It is noteworthy that As and Se do not exhibit the same properties, and how they vary is likely governed by different factors including physiological regulation. General pollution of the sea from rivers, industry, and other regional pollution sources in Europe and the Baltic Sea may partly explain the gradient of Hg and POPs in seafood from south to north. As POPs are fat soluble and lean fish species store their fat reserves in the liver (Boitsov et al., 2019; Julshamn et al., 2004), concentrations of POPs in liver samples were much higher compared to fillets for those species where both tissue types were analyzed (i.e., mainly for lean fish taxa). POPs concentrations in fillets of lean species were much lower than those in fillets of fatty species and correlations between POPs concentrations and fat content were complex. Nevertheless, we found no or weak correlations between fat content and POPs in high fat content organs such as fillets of pelagic fatty fish and liver samples from benthopelagic and demersal lean species. Species-specific variation in POPs concentrations may, to a large extent, be determined by fat content variation whereas intra-species variation likely depends on additional factors. Other biotic factors such as food web complexity, prey contaminant profiles, metabolism and growth rate may also influence the geographical variations in

contaminant concentrations. However, these factors were not considered in this study and overall, their quantitative contributions are unknown and beyond the scope of this investigation. A moderate to strong correlation between fat content and POPs in semi-fat species of European plaice and Atlantic halibut might be partially explained by seasonal fluctuation rates, especially since reproduction of these demersal fish species in winter and spring can result in lower fat content and at the same time a reduction in POPs. However, more data on these specific fish species should be collected and analyzed to formally evaluate these hypotheses. In Greenland halibut high POPs concentrations were found despite high catch latitudes (north of 62 °N). These individuals may be influenced by species-specific metabolism dynamics, and high concentrations may be related to the high fat content, high trophic position, age, and size. Pelagic species which are predominantly plankton feeders are also subjected to seasonal effects due to periodic changes in food availability, and gonad maturation and spawning (Frantzen et al., 2011). For example, for Norwegian spring spawning herring, when fat is metabolized during the winter starvation period, contaminants are retained (Frantzen et al., 2011). Conversely, spawning resulted in decreased fat and POPs concentrations in fillets, likely due to a transfer of fat with associated contaminants to gonads. Depending on the season, POPs concentrations in herring thus either increased or decreased with fat content. This shows that for fatty fish there is no simple relationship between fat metabolism and POPs bioaccumulation, which is likely why we found a weak or no correlation between POPs concentrations and fat content in fatty fish species (Table S4).

By including abiotic factors, we identified associations of sediment contaminants and sea temperature with concentrations of Hg, dioxin, PCB6 and PBDE7 in seafood using MLR models. For POPs analyses, the model was applied to fillet and liver samples of all fish species combined in addition to the fillet of pelagic species and the liver of benthopelagic and demersal fish species which contained high concentrations of POPs. Normalization of POPs concentrations to fat content in these samples introduced significant bias to the MLR model and hence reduced model goodness of fit. Sediment contaminants could also be considered as a proxy for seawater pollution and are often monitored to assess contaminants including PCBs, polycyclic aromatic hydrocarbons (PAHs), pesticides, and metals (Everaert et al., 2017). In our study, high levels of pollution were observed in the Skagerrak region as confirmed by high concentrations of sediment contaminants, especially Hg and POPs, and also in the North Sea the concentrations of these contaminants in sediments were higher compared to the northern regions of the study area. Elevated concentrations of Hg and POPs in seafood samples from Skagerrak and along the coast of southern Norway originate from a complex combination of non-point source and point sources of pollution (Azad et al., 2019a; Magnusson et al., 1996). Although pelagic fish generally migrate extensively, we still found significantly higher concentrations of contaminants in fish fillets collected in Skagerrak compared to those collected in the Norwegian Sea and the North Sea. Using MLR models, we confirmed positive associations of sediment contaminants with contaminants measured in marine fish at a broad spatial scale. Modelling combined with measurements has suggested a weak latitudinal gradient of Hg concentrations in the surface sediments in the offshore NEAO (Everaert et al., 2017). While a lower impact from population densities and industrial and agricultural activities in the north might result in low environmental contaminants in the Barents Sea compared to areas further south, Hg and PCB6 concentrations of surface sediments were found to increase moderately from the Norwegian Sea to the Barents Sea of the NEAO. The observed co-variation of sea temperature with contaminants is likely confounded by other factors not measured here, and causal correlation may partly lie in the association between sea temperature, human population density, and distance to significant regional industries. However, considering our analyses, this does not seem to be the whole explanation. Increasing concentrations of these contaminants in the north of the NEAO may be originating from ice or atmospheric inputs explained by the phenomenon known as the

“grasshopper effect”, in accordance with previous studies (Everaert et al., 2017; Lohmann et al., 2009; Wania and Mackay, 1993). This effect is enabling some POPs to be transferred from industrial and agricultural areas at lower latitudes to remote areas further north including remote Arctic ecosystems.

In our study, contaminant concentrations in seafood taxa collected in the Barents Sea were significantly lower than those collected in the Norwegian Sea in contrast to observations for sediments which showed similar or higher concentrations of contaminants in sediments from the Barents Sea compared to the Norwegian Sea. This suggests a lower degree of contaminant exposure and bioaccumulation in marine fish from the Barents Sea which has low sea temperatures. Additionally, organic matter mass (e.g., measured as total organic carbon) of Barents Sea sediments was greater compared to those from the Norwegian Sea, suggesting a slow decomposition rate of organic matter in the Barents Sea due to lower sea temperature in addition to either higher productivity rates in the Barents Sea ecosystem or long-range, atmospheric and ice transport. Little is known about the effects of sea temperature on contaminant bioaccumulation in wild, marine fish. Higher sea temperatures have been reported to promote MeHg bioaccumulation in marine organisms such as brittle star (*Ophiothrix fragilis*) and juvenile seabass (*Dicentrarchus labrax*) (Hutchins et al., 1996; Maulvault et al., 2016). Furthermore, data on organic contaminants and Hg in Lake Ontario salmon in relation to air temperature have also been reported (French et al., 2006). However, analysis of marine fish contaminants in relation to air sea exchange of contaminants (Bank et al., 2021) and air temperature is complex and highly uncertain since seawater temperature response to climate change is likely slower than that of air due to much higher heat capacity of the seawater compared to the air and the fact that seawater serves as a large sink of heat transfer. In our study, we found that associations between seawater temperature and fish contaminant levels were contaminant specific. By using the MLR model, Hg concentrations in fillet and liver of fish species were found to associate significantly and positively with changes in seawater temperature. POPs concentrations in liver samples of benthopelagic and demersal species also exhibited significantly positive associations with changes in seawater temperature. However, POPs concentrations in fillets, especially pelagic species, exhibited no association or even slightly negative association (i.e., DL-PCB in fillets of all fish species combined) with changes in seawater temperature. Bioaccumulation of POPs in seafood is inherently complex and likely also governed by physiological and metabolic factors regarding the regulation partitioning kinetics of POPs. Regulation and partitioning of POPs in different fish organ types varies. For example, concentrations of dioxins, DL-PCB, PCB6 and PBDE7 in the liver in our study were found to be from one to two orders of magnitude larger than those in the fillets and were species dependent. Part of this difference is due to fat being stored in the liver of the lean fish species and likely related to levels of specific lipoproteins that can transport POPs to adipose tissue (Lind and Lind, 2020). Further investigation of advanced kinetics is likely required to reveal such complex mechanisms of organ-specific POPs bioaccumulation in relation to abiotic factors. Both Hg and POP concentrations in seafood fillet and liver samples were significantly and positively associated with increases in sediment contaminants. Collectively, these results suggest regional scale marine environmental pollution dynamics are important abiotic drivers of Hg (Bank et al., 2021) and POPs concentrations in NEAO marine fish. This investigation is the first to quantify effects of abiotic drivers on contaminant concentrations of seafood using databases of commercially important seafood species in combination with sediment contaminants, sea temperature, and salinity data from the NEAO. Since global anthropogenic emissions of Hg have been relatively stable from approximately 2011 (Streets et al., 2019), sea temperature increase due to ocean warming may be an important driver for increasing MeHg concentrations in some species of marine fish (Schartup et al., 2019). However, further research is required to evaluate this hypothesis.

Linking contaminant exposure to environmental conditions in

marine fish harvested from the NEAO to human exposure via seafood consumption, exposure of contaminants from seafood intake varied with geography, fish habitat, and species. Commercial seafood species harvested from the NEAO, in general, were not found to constitute a high risk of MeHg or PBDE exposure to human consumers. Our results for PBDEs are in good accordance to those described by Nøstbakken et al. (2018). However, a considerable proportion of the seafood consuming population, in particular for those consuming pelagic fish, would exceed the TWI for dioxins and DL-PCBs of 2 pg TEQ/kg body weight per week set by EFSA (EFSA, 2018). Using the tolerable intake set by the Joint FAO/WHO Expert Committee on Food Additives (JECFA, 2010) (provisional tolerable monthly intake 70 pg/kg bw per month), consumption up to 400 g fish fillet per week was not found to be a high risk regarding dioxin and DL-PCBs exposure. This study also shows the need for high resolution consumption data of seafood for risk assessment purposes, particularly for exposure to those contaminants where seafood is reported to be among the largest sources.

We identified the following limitations of our study. First, the data compiled in this study have been collected through different baseline studies and other monitoring programs, resulting in non-random sampling which may not be entirely representative of the fishery and seafood distribution in the NEAO. Also, sampling was unbalanced across the study period and the study area because of variable sampling periods for different species and differences in the species distribution and habitat. However, the extremely large number of fish analyzed (Tables S1 and S9) mitigate these effects and limits both type I and type II statistical errors. Within the scope of this study, there was no possibility to evaluate variation within and between species caused by several factors including effects of diet, physiology, growth rate, energetics, etc. Also, temporal trends and seasonal variation were not considered in this study. Furthermore, our study focused on risks of contaminants in fish fillets including Hg and POPs due to consumption of seafood from the NEAO rather than of seafood from other sources and areas. More accurate data on seafood consumption profiles are required to predict exposure concentrations of contaminants for vulnerable groups including young children, pregnant women as well as for individuals involved in subsistence or recreational fishing including indigenous people. Nevertheless, this investigation is built on one of the world's largest seafood databases and uses chemical information from accredited laboratory methods and will serve as an effective model to further understand the distribution and co-occurrence of seafood contaminants in the context of the observed increases in sea temperature and the potential resulting human health risks.

5. Conclusions

Analyzing a large seafood dataset from the NEAO, we observed positive correlations between concentrations of Hg, dioxins, PCBs, and PBDEs, which bioaccumulate in marine food webs. Concentrations of Hg, dioxins, PCBs and PBDEs increased from North to South, and habitat specific associations of Hg and POPs with sea temperature and marine sediment contaminants were highlighted. Hg in fish fillets and liver were positively associated with changes in either sediment contaminants or sea temperature. POPs in both fillets and liver samples were positively associated with increases in sediment contaminants, and only POPs in livers of benthopelagic and demersal species were positively associated with increases in sea temperature. Using a probabilistic approach to estimate human contaminant exposure from seafood, we determined that Hg exposure was highest from the intake of benthopelagic and demersal species. On the other hand, the risk from dioxin and dioxin-like PCBs (DL-PCBs) is the greatest health concern for those consuming large amounts of fillet of pelagic species.

CRedit authorship contribution statement

Quang Tri Ho: Formal analysis, Visualization, Data curation,

Writing – original draft, Writing – review & editing. **Michael S. Bank:** Conceptualization, Methodology, Visualization, Data curation, Formal analysis, Resources, Supervision, Writing – original draft, Writing – review & editing. **Atabak M. Azad:** Investigation, Writing – review & editing. **Bente M. Nilsen:** Investigation, Writing – review & editing. **Sylvia Frantzen:** Investigation, Writing – review & editing. **Stepan Boitsov:** Formal analysis, Writing – review & editing. **Amund Maage:** Investigation, Writing – review & editing. **Tanja Kögel:** Investigation, Writing – review & editing. **Monica Sanden:** Writing – review & editing. **Livar Frøyland:** Funding acquisition, Project administration, Writing – review & editing. **Rita Hannisdal:** Investigation, Writing – review & editing. **Helge Hove:** Investigation, Writing – review & editing. **Anne-Katrine Lundebye:** Investigation, Writing – review & editing. **Ole Jakob Nøstbakken:** Conceptualization, Writing – review & editing. **Lise Madsen:** Supervision, Conceptualization, Writing – original draft, Writing – review & editing.

Declaration of Competing Interest

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

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Appendix A. Supplementary material

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envint.2021.106858>.

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