Research Article

Congener-specific accumulation of persistent organic pollutants in marine fish from the Northeast Atlantic Ocean

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HIGHLIGHTS

- POP congener profiles in fish from the NEAO were explored for the first time.
- POP congener profiles were more influenced by fish species than by geography.
- Light congeners varied less with latitude than heavy congeners.
- POPs concentrations increased with fat content but levelled off at around 10% fat.
- POPs with higher degrees of chlorination/bromination exhibited higher fat affinity.

GRAPHICAL ABSTRACT
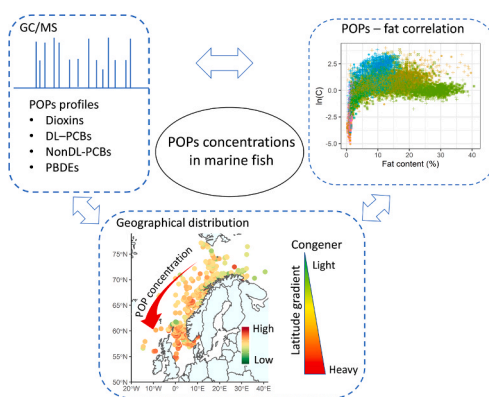
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ABSTRACT

Bioaccumulation of persistent organic pollutants (POPs) in marine fish may pose a health risk to human consumers. Using data from ~8400 individuals of 15 fish species collected in the North-East Atlantic Ocean (NEAO),

we assessed concentrations of individual POP congeners, including dioxins, polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs). POPs analyses were performed with accredited methods using high-resolution gas chromatography/high-resolution mass spectrometry, gas chromatography/tandem mass spectrometry (GC-MS/MS) and GC/MS. The results showed that POPs congener composition profiles were more influenced by fish species than by geography. However, due to long range transport from emissions at lower latitudes, lighter congeners made a larger contribution to the total POPs concentrations in the northernmost areas compared to southern regions. A model was developed to elucidate the relative effects of several factors on POPs concentrations and showed that variation among and within fish species was associated with fat content, fish size, trophic position, and latitude. For the first time, POPs concentrations were shown to increase non-linearly with fat content, reaching an asymptotic plateau when fat content was > 10%. This study explored detailed POP congener profiles and the factors associated with POPs accumulation in commercially relevant fish harvested from the NEAO.

1. Introduction

Consumption of marine fish can provide high quality proteins, essential vitamins, minerals, trace elements, and long chain omega-3 polyunsaturated fatty acids [15,46,47], and marine foods play an important role in ensuring global food security. However, the occurrence of globally distributed pollutants such as persistent organic pollutants (POPs) in marine fish may also pose a health concern for consumers [23,50,8]. The co-occurrence of nutrients and contaminants have been used to support several risk-benefit approaches and analyses of marine fish consumption [13,17,18,19,49,53].

POPs comprise several groups of organic compounds including polychlorinated dibenzo-p-dioxins/ polychlorinated dibenzofurans (PCDD/F, further called "dioxins"), polychlorinated biphenyls (PCBs), and polybrominated diphenyl ethers (PBDEs). According to the Stockholm convention, POPs are prioritized in listed groups of elimination, restriction, and unintentional production [22]. PCBs and PBDEs are among the POPs identified where measures are needed to eliminate production, while for dioxins there is a goal of continuing minimization and, where feasible, complete elimination. Dioxins, PCBs and PBDEs are lipophilic [11,33], and can bioaccumulate and undergo bio-magnification in marine food webs [11,1]. The EU has set maximum levels for food safety applying to fish for sum of dioxins and dioxin-like PCBs (DL-PCBs) and the sum of six non-dioxin-like (nonDL-PCBs or PCB6) [20].

POPs in marine fish are mainly determined by dietary exposure pathways and are bioaccumulated at varying concentrations throughout their lifespan [26,28,34,8]. Hence, trophic position (position or level in the food chain), size, age, and fat content are important factors determining the concentrations of POPs in fish fillets. Lean fish species, such as cod or saithe, store lipids and POPs mainly in liver tissue and fillet concentrations are often low [35]. Pelagic fatty fish such as mackerel, herring, and salmon often contain higher concentrations of POPs in fillets from individuals harvested from the NEAO [49,50] and other marine areas such as the Baltic Sea, Gulf of Finland and the Bothnian Sea [36,59]. Humans consuming fatty fish in accordance with recommended intake may exceed the tolerable weekly intake value (TWI) of dioxins and DL-PCBs set by the European Food Safety Authority (EFSA) [49]. Although POPs concentrations are associated with fat content in marine fish, the relationships between POPs concentrations and fat content are inherently complex [23,50]. For example, in Norwegian spring spawning herring, the relationship between fat content and fillet POPs concentrations varies seasonally and is dependent on feeding dynamics and spawning cycles [23]. Moreover, the relationship between fat content and PBDE levels can vary by tissue type and individual PBDE congeners [50].

Concentrations of dioxins and DL-PCBs expressed as upper bound (UB) sums of TEQ values are reported as critical values in the context of seafood safety [19]. Moreover, understanding congener profiles of commercially important fish species and their geographical and intra-species variation in the NEAO is important to contribute to the knowledge of POPs exposure, bioaccumulation, and toxicity from different sea regions and to estimate potential risks to human consumers [28,49]. However, evaluating variation in marine fish species POP congeners can be challenging because concentrations of several congeners of PBDEs, dioxins and DL-PCBs are often lower than their limits of quantification (LOQ) [12,49,50]. When many congeners of dioxins and DL-PCBs have concentrations below their LOQs, using UB sums may overestimate the total sum TEQ concentrations [49]. A robust statistical method such as regression on order statistics (ROS) is therefore required and can be applied to estimate concentrations of these censored contaminants in marine fish [49,50,9]. An iterative principal components analysis (PCA) imputation technique [30] can also be used to estimate censored data with high correlations between congeners.

The aim of this study was to assess concentrations of different congeners of dioxins, DL-PCBs, nonDL-PCBs and PBDEs in selected, commercially relevant, marine fish, and to investigate the drivers of POP congeners variation in fillets sampled from the NEAO. A dataset comprising 15 commercially relevant fish species (n = ~8400 individuals) sampled from the NEAO was used to test the following hypothesis and a priori prediction: POP congener profiles in fish fillets are species- and sea region specific in the NEAO. We also developed a mechanistic model to evaluate associations between POP congeners and abiotic and biotic drivers including fat content, species, trophic position, fish size, and geographic latitude.

2. Materials and methods

2.1. Study area and fish sampling

The data presented includes results from POP analyses of ~8400 samples from 15 commercially relevant marine teleost species including anglerfish (*Lophius piscatorius*), Atlantic cod (*Gadus morhua*), Atlantic halibut (*Hippoglossus hippoglossus*), Atlantic herring (*Clupea harengus*), Atlantic mackerel (*Scomber scombrus*), beaked redfish (*Sebastes mentella*), golden redfish (*Sebastes norvegicus*), common ling (*Molva molva*), European hake (*Merluccius merluccius*), Greenland halibut (*Reinhardtius hippoglossoides*), haddock (*Melanogrammus aeglefinus*), plaice (*Pleuronectes platessa*), saithe (*Pollachius virens*), pollack (*Pollachius pollachius*), and tusk (*Brosme brosme*) collected mostly from the Norwegian fishing regions of the NEAO (Table S1, Fig. 1). Sampling methods have been previously described in detail elsewhere [23,3,50]. Fish species, year of sampling, and references for earlier studies describing the samples are shown in Table S1. Briefly, fish sampling was carried out on scientific cruises with research vessels owned by the Institute of Marine Research (IMR), hired fishing vessels, or the Norwegian reference fleet composed of commercial fishermen collaborating with IMR researchers. The sampling took place in fjords and in coastal and offshore areas of the NEAO covering most of the important fishing areas (Fig. 1). For analysis purposes, sampling areas were divided into five sub-areas: Barents Sea, Norwegian Sea, North Atlantic, North Sea and Skagerrak. We used ocean basin boundaries defined by Azad et al. [3], except for the border between the Barents and the Norwegian Seas, which was defined in Nøstbakken et al. [50]. Sampling gear used to catch fish included long

line, gill net, purse seine and pelagic trawl [23,3,35,50]. Most fish were frozen whole at − 20 °C before being transported to the laboratory.

2.2. Laboratory analyses

At the laboratory, individual fish were thawed, measured for length and weight, and dissected as described elsewhere [23,3,35]. In general, skin-free fillet samples (n = ~8400) were homogenized and lyophilized before analysis using trace metal clean techniques [23,3,35,4,50]. In addition, liver samples of cod and saithe (n = 149) were homogenized and analysed (Table S1). More details of the analytical methods for dioxins (dioxins and furans), dioxin-like PCBs (DL-PCBs), non-dioxinlike PCBs (nonDL-PCBs) and polybrominated diphenyl ethers (PBDEs) are given in Supplementary Text S1. Procedures of sample extraction, clean-up, and analysis have been modified over time and have been described in detail in previous studies [23,43,50,52,6]. Samples were analysed for seven congeners of dioxins (polychlorinated dibenzo-p-dioxins, PCDDs) including 2,3,7,8-TCDD, 1,2,3,7,8-PeCDD, 1, 2,3,4,7,8-HxCDD, 1,2,3,6,7,8-HxCDD, 1,2,3,7,8,9-HxCDD, 1,2,3,4,6,7, 8-HpCDD and OCDD, ten furans (polychlorinated dibenzofurans, PCDFs) including 2,3,7,8-TCDF, 1,2,3,7,8-PeCDF, 2,3,4,7,8-PeCDF, 1,2, 3,4,7,8-HxCDF, 1,2,3,6,7,8-HxCDF, 1,2,3,7,8,9-HxCDF, 2,3,4,6,7, 8-HxCDF, 1,2,3,4,6,7,8-HpCDF, 1,2,3,4,7,8,9-HpCDF and OCDF, twelve DL-PCBs, i.e. four non-ortho PCBs (PCB-77, −81, −126, and −169), and eight mono-ortho PCBs (PCB-105, −114, −118, −123, −156, −157, −167 and −189), six nonDL-PCBs, i.e. PCB-28, − 52, − 101, − 138, − 153 and − 180. Notably, eight indicator congeners of PBDEs which the European Food Safety Authority (EFSA) have advised to monitor are PBDE-28, − 47, − 99, − 100, − 153, − 154, − 183 and − 209 [16,17]. Seven out of these eight indicator congeners (PBDE7) were analysed in this study, but not the heaviest congener, PBDE − 209. Briefly, wet and homogenized liver sample or dried homogenized muscle sample was mixed with hydromatrix and ^{13}C-labelled internal standards were added (27 standards for dioxins, furans and dioxin-like PCBs, CIL, and one standard for PBDEs). The mixture was transferred to an Accelerated Solvent Extractor 300 (ASE, Dionex Corp.) or a

Pressurised Liquid Extraction System (PLE, FMS Inc.), with a layer of acidic silica gel and extracted with hexane under elevated pressure and temperature (100 °C, 1500 psi). Subsequently, the extract was purified using PowerPrep (FMS Inc.) over three columns packed with multilayer silica, basic alumina and carbon, respectively, and eluted with different solvents. Two fractions were collected. Fraction 1 contained PBDEs, PCB6 and mono-ortho PCBs and fraction 2 contained dioxins and non-ortho PCBs.

Dioxin and non-ortho PCB congeners were analysed by high resolution gas chromatography/high resolution mass spectrometry (HRGC-HRMS). NonDL-PCBs, mono-ortho PCBs and PBDEs were analysed by gas chromatography/ tandem mass spectrometry (GC-MS/MS) or GC/ MS. Concentrations of congeners of dioxins and DL-PCBs are here reported using either raw concentrations in pg g^{-1} wet weight (ww) or concentrations in terms of TEQ (pg TEQ kg^{-1} ww) according to Van den Berg et al. [57]. The analytical methods are accredited according to the ISO 17025 standard. Trueness was maintained by annual participation in inter-laboratory proficiency tests, with results well within accepted limits (−2 < Z-score < 2). With every sample batch, blanks and spiked control material salmon were prepared and analysed to ensure internal reproducibility, with resulting relative standard deviations less than 14% for PCDD, PCDF and non-ortho PCB congeners, 11% for mono-ortho PCB and ndl-PCB congeners, and 10% for PBDEs. Recovery efficiencies of ^{13}C labelled standards between 30% and 130% were accepted, but 82% of the samples had recoveries between 60% and 120% for almost all the congeners.

The analytical method's limits of quantification (LOQs) vary between congeners and depend on the fat content of the sample. LOQs in lean and oily fish muscle varied from 0.008 to 0.13 pg g^{-1} for dioxins, non-ortho PCBs, nonDL-PCBs, from 2.4 to 16 pg g^{-1} for mono-ortho PCBs and from 0.001 to 0.013 pg g^{-1} for PBDEs.

2.3. Estimation of censored data and statistical analyses

Prior to conducting statistical analyses, censored data were defined as concentrations below limit of quantification (LOQ) (Table S3). POP
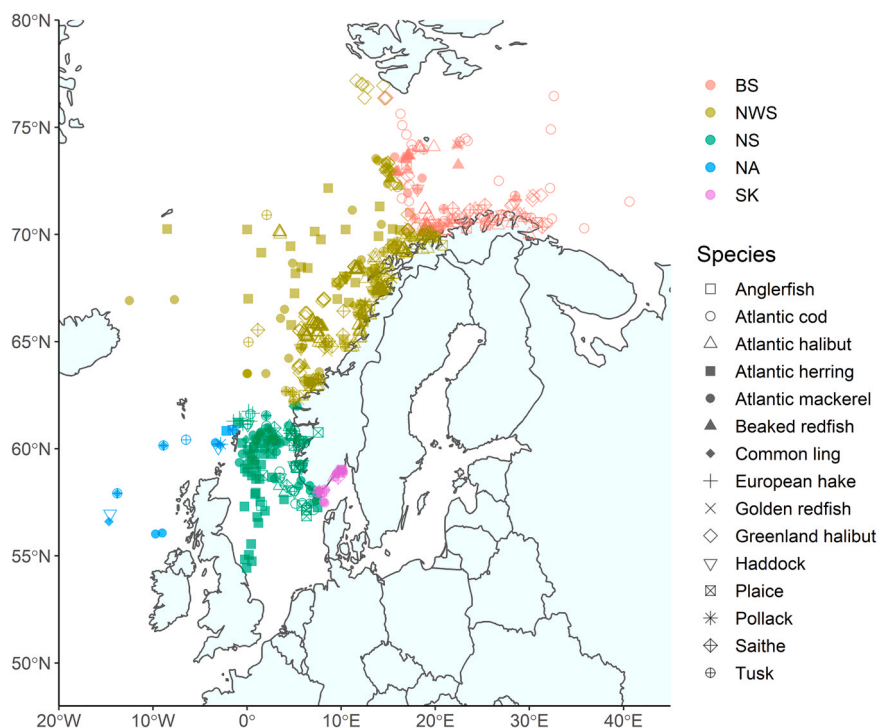

Fig. 1. Sampling area of fish species in different sea regions of the NEAO. Each fish species is denoted by a specific symbol with different colours in different sea regions. BS, NWS, NS, NA and SK denote the Barents Sea, the Norwegian Sea, the North Sea, the North Atlantic Sea and Skagerrak, respectively.

congeners were classified into four groups; dioxins, DL-PCBs, nonDL-PCBs and PBDEs, and within each group, the congener present at the highest concentration was selected as a proxy (see Supplementary Text S2). Censored data of the proxies were estimated by means of the semi-parametric Regression on Order Statistics (ROS) approach [50,55]. The missing values of the remaining congeners were estimated based on correlations between proxies and the remaining congeners using an iterative principal components analysis (PCA) imputation approach (see Supplementary Text S2, Figs. S1–3). The missing values of censoring proxies were modelled using package "NADA" while missing values of censoring of the remaining congeners were estimated using an iterative PCA imputation approach described in the package "MissMDA" in R (R Core Team, 2020) operated in RStudio (version 1.3.959; R StudioTeam, 2015).

Congener profiles of each fish species were visualized using the median of congener concentrations in addition to cluster analysis performed and interpreted in the form of dendrograms (hierarchical clustering sorting based on the average distance of subtrees at every merging point) using package "pheatmap" in R.

2.4. Association of POP concentrations with abiotic and biotic factors

Concentration data of POPs were not normally distributed and were log transformed to obtain an approximate normal distribution. To evaluate the relationships between POPs and environmental and biological factors, we used nonlinear models (NLMs) with the POPs concentrations as dependent variables, and fat content, trophic position, fish length, and latitude as independent variables as follows:

$$\ln(C_i) = \ln\left(\frac{C_{Fat}^n}{C_{Fat}^n + K_m^n}\right) + A_o + A_{TP} \bullet TP + A_{dFL} \bullet dFL - A_{Lat} \bullet Lat \quad (1)$$

where C_i is the predicted level of congener i in the fillet, C_{fat} (%) is the fat content; n is the order coefficient; K_m is the apparent parameter describing association of POPs to fat content; TP is the literature based trophic position of the fish (see Table S2), and Lat is the latitude. A_o, A_{TP}, A_{dFL} and A_{Lat} are the model coefficients.

dFL is the variation of fish length within a fish species, defined as follows:

$$dFL = FL - \overline{FL} \quad (2)$$

where FL and $\overline{FL}$ are the individual fish length and the mean fish length, respectively, of the fish species.

In the model equation, the first nonlinear term analogous to the Hill description refers to the concentration of POPs that is associated to fillet tissue fat [24,25]. Notably, two biological factors, fish length and trophic position were highly correlated. Therefore, to avoid multicollinearity of independent variables, fish length and trophic position (TP) were not evaluated concomitantly in the model. Instead, TP appearing in the third term accounted for biomagnification of POP congener concentrations due to variation in trophic position among fish species while dFL in the fourth term accounted for bioaccumulation of the POP congener concentrations due to variation in intraspecific fish length. Biomagnification patterns are often characterised by increasing concentration of POPs with increasing TP [14,60]. The last term describes the association of POPs with latitude. The minus sign of the last term refers to a decrease in POPs with increasing latitude, which has been shown previously [28].

Fat-weight concentrations of POP congeners were also calculated, and we used a multi-linear model to analyse association of fat-normalized POP concentrations with selected biotic and abiotic variables (see Supplementary Text S3).

2.5. Parameter estimation

The parameters of the model including K_m, A_o, A_{TP}, A_{dFL}, A_{Lat}

described by Eq. 1 were estimated by minimising the sum of square differences between the log-transformed POP concentrations for different fat content values, trophic position, fish length, and latitude as predicted by the model (Eq. 1) and the measured log-transformed individuals using a non-linear estimation programme written in R with package "nlme", operated in RStudio (version 1.3.959; RStudio Team, 2015). In the model, Akaike's Information Criterion (AIC) was computed for model selection purposes using a priori derived combinations of variables, residual sum of squares estimates, and r-squared (r^2) values [10].

3. Results

3.1. Concentrations of POPs in fillets of marine fish species from the NEAO

Prior to conducting analyses, censored data, defined as concentrations below LOQ, were identified (Table S3). The congeners 2378-TCDF, PCB-118, − 138, − 153 and PBDE-47 were easily detected (detection, i. e., results $\geq$ LOQ, $\geq$ 76.9%) and present in the highest concentrations within their respective contaminant group and were therefore chosen as proxies within their group (Tables S3 and S4). POPs detection rate varied between different fish samples. Generally, POP concentrations in fatty fish such as Greenland halibut and Atlantic halibut were high with high detection rates while POP concentrations in lean fish were low and concentrations of several POP congeners (i.e., 1234678-HpCDD, 1234678-HpCDF, 1234789-HpCDF, 123478-HxCDD, 123789-HxCDF, OCDD, OCDF and PBDE-183) were mostly below the LOQs (Tables S3 and S4). Censored data were estimated as shown in Supplementary Text S2. Concentrations of POPs in fillets of different fish species in NEAO are summarized in Table S4A. The highest median raw concentrations of sum dioxins (4.4×10^{-3} ng g^{-1}), sum DL-PCBs (3.99 ng g^{-1}), sum nonDL-PCBs (17.3 ng g^{-1}) and sum PBDEs (1.03 ng g^{-1}) in fish fillets were all found in the fatty fish species Greenland halibut. The second highest concentrations were found in the fatty fish Atlantic halibut (e.g. median sum DL-PCB 2.1 ng g^{-1}), followed by the fatty fish species herring (sum DL-PCB 0.95 ng g^{-1}) and mackerel (sum DL-PCB 0.80 ng g^{-1}). The semi-fatty species (i.e., redfish, hake, and plaice) had intermediate concentrations of all POPs groups (e.g. median sum DL-PCB 0.46–0.58 ng g^{-1}). The lowest median concentrations of the different POPs were observed in the lean fish species cod (lowest sum dioxins, 0.71×10^{-4} ng g^{-1}; Table S4A), anglerfish (lowest sum DL-PCBs 3×10^{-2} ng g^{-1}), pollack (lowest sum nonDL-PCBs 1.39×10^{-1} ng g^{-1}) and haddock (lowest sum PBDE 1.03×10^{-2} ng g^{-1}).

Median TEQ concentrations of the sum of dioxins and of the total sum of dioxins and DL-PCBs in fillet samples of Greenland halibut were 0.80 and 2.4 pg TEQ g^{-1}, respectively and hence below the EU maximum levels (MLs) of 3.5 and 6.5 pg TEQ g^{-1}, respectively, applicable for fish meat traded for human consumption [20]. All median concentrations of the sum of nonDL-PCBs (PCB6) in fillets were below the ML of 75 ng g^{-1}. However, for Greenland halibut, 8.6%, 15.3% and 5.0% of the individual fish had concentrations of the sum of dioxins, the sum of dioxins and DL-PCBs and PCB6 above the MLs, respectively (Table S4C). For Atlantic halibut, 1.2%, 2.2% and 1.2%, respectively, of B-cut (the muscle tissue most representative as food) had concentrations of the sum of dioxins, the sum of dioxins and DL-PCBs and PCB6 above the MLs while 5.4%, 15.6% and 8.1% of fatty samples I-cut (see Supplementary Text S1) had concentrations of the sum of dioxins, the sum of dioxins and DL-PCBs and PCB6 above the MLs, respectively. Less than 0.7% of mackerel and herring were above the MLs. All fillet samples of semi-fatty and lean fish species had concentrations well below the MLs.

The median concentrations of sum of dioxins and DL-PCB, sum of nonDL-PCBs and sum of PBDEs in the liver samples of cod and saithe ranged from 10.6 to 11.3 pg TEQ g^{-1}, 61.0–71.0 ng g^{-1} and 3.62–7.39 ng g^{-1}, respectively. In the liver samples with high fat content (median fat content 56.3% and 56.8% for cod and saithe,

respectively), the concentrations were two orders of magnitude higher than the concentrations in fillet samples from the same fish (Table S4B).

3.2. Inter-specific and geographical variation of POP congeners

To study the relative contribution of individual congeners among fish species, the calculated congener composition profile was visualized in terms of percentage contribution of each congener to the total sum of congeners within each group (Fig. 2). In general, 2378-TCDF (34.1–69.8% of sum dioxins) and PCB-118 (53. to 64.5% of sum DL-PCBs) were the predominant congeners of dioxins and DL-PCBs,

respectively. PCB-138 and 153 were the predominant congeners of nonDL-PCBs (PCB-138 + 153 from 54.9% to 78.1% of sum nonDL-PCBs) while PBDE-47 (48.1–73.7% of sum PBDEs) was the predominant PBDE congener. Beaked redfish, golden redfish, haddock, tusk, hake, Atlantic halibut, Greenland halibut, plaice and herring had similar congener composition profiles, differing from the cluster comprising ling, saithe, cod, mackerel, and pollack. Cod, mackerel and pollock had relatively lower dominance of PBDE-47 and higher percentage of PBDE-99 compared to most other species. Furthermore, Atlantic halibut and Greenland halibut, herring and plaice formed a separate cluster relative to most other species, where 23478-PeCDF contributed more and 2378-
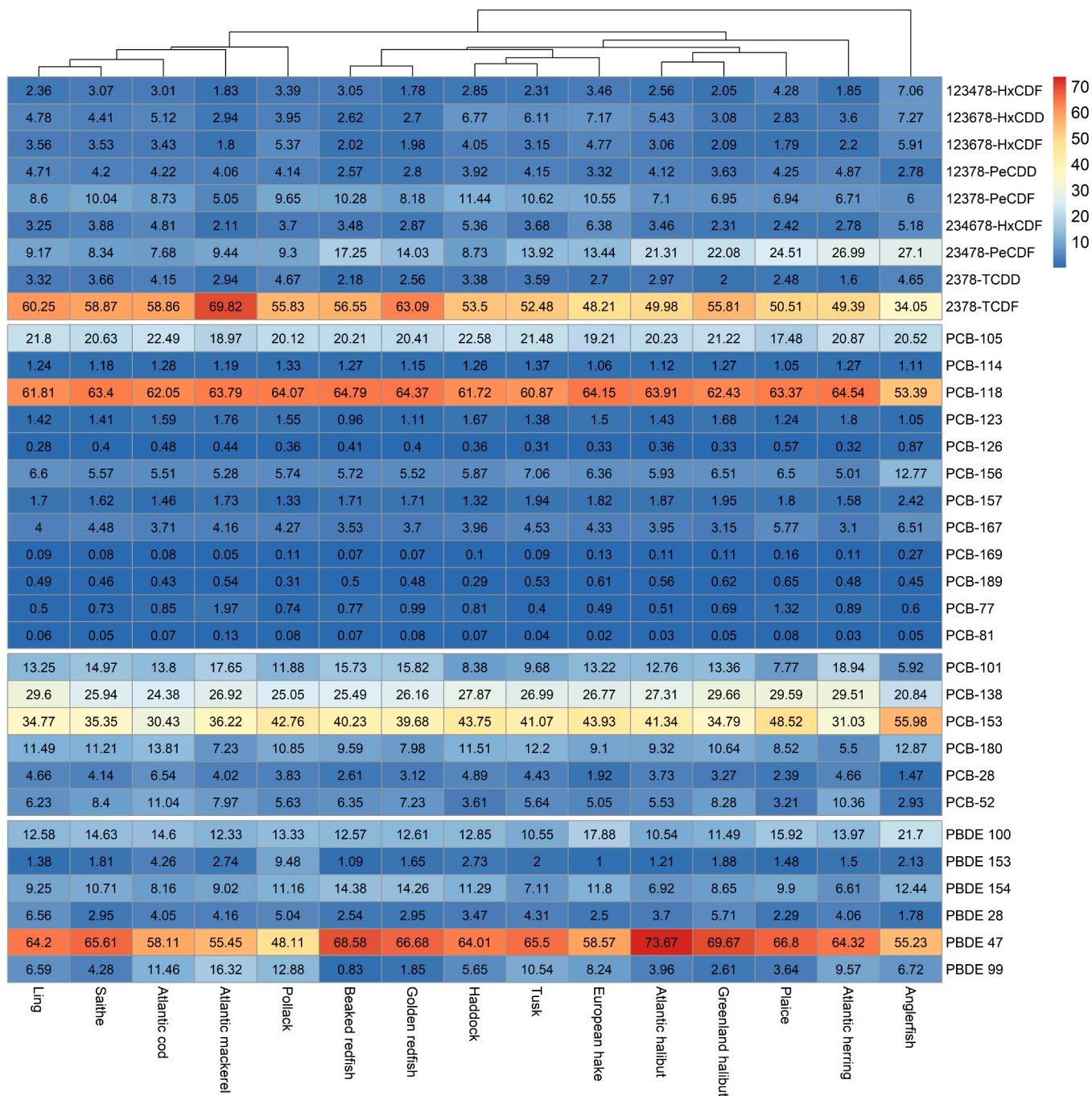
Fig. 2. Congener composition profiles (%) of dioxins, DL-PCBs, nonDL-PCBs and PBDEs in fillet of different fish species collected from the NEAO. Colours indicate the composition (%) of congeners. The contribution of congener i is defined as $C_{c,i}(\%) = \frac{C_i \cdot 100}{\sum C_i}$ where C_i is the concentration of congener i and $\sum C_i$ is the sum of the concentration of the congeners within the group. Congener profiles of each fish species were visualized using the median of congener concentrations and cluster analysis was performed and interpreted in the form of dendrograms.

TCDF contributed less (Fig. 2). Anglerfish had a distinct profile compared to other species. For instance, compared with other species, anglerfish had relatively low percentages of PCB-118, PCB-138 and 2378-TCDF, and relatively high percentages of PCB-153 and 23478-PeCDF. The congener profiles of POPs in liver of cod and saithe were like those of fillets, with some exceptions (Fig. S4). Percentages of 23478-PeCDF in liver of cod and saithe were relatively low while percentages of PCB-153 were relatively high compared to fillet. Percentages of PBDE-47 and PBDE-99 in liver of cod were relatively higher and lower than those in fillet, respectively.

In terms of TEQ concentrations, the congeners 23478-PeCDF, 2378-TCDF, 12378-PeCDD, 2378-TCDD all contributed significantly to the total toxicity of sum dioxins (Fig. S5), whereas for raw concentration profiles, 2378-TCDF was much more dominant (Fig. 2). Within the DL-PCB group, TEFs of PCB-126 (TEF 0.1) and PCB-169 (TEF 0.03) are much larger than those of the remaining congeners (TEF of 3×10^{-5} to 1×10^{-4}). Due to the high TEF-value, PCB-126 contributed substantially to the total TEQ of sum DL-PCBs (Fig. S5), even though PCB-118 had the highest raw concentrations (Fig. 2). Regarding TEQ-concentrations, PCB-126 was the single most dominant congener of the total sum dioxins and DL-PCB for all species (Fig. S5).

Fatty fish species including Atlantic halibut, Greenland halibut, herring, and mackerel were the most sampled species from the different NEAO regions. To explore the geographical variation of congener composition, data from these fatty species were analysed for each geographic region (Fig. 3). The composition profiles of mackerel in the Barents Sea and the Norwegian Sea were different from those collected in the other sea regions while the profiles in the North Sea and the North Atlantic clustered together and were distinct from the Skagerrak region. Similarly, the profile of herring in the Norwegian Sea clustered separately from those of herring in the other sea regions. The profiles of herring from the North Sea and the North Atlantic were similar and clustered into a subgroup differing from those from the Skagerrak region. Greenland halibut sampled from the Barents Sea had different congener profiles compared to the Norwegian Sea, whereas the profiles of Atlantic halibut from the Barents Sea and the Norwegian Sea were similar and clustered together.

The congener profiles of mackerel from the Norwegian Sea and the Barents Sea differed from those of mackerel from all other sea areas by having relatively higher portions of 2378-TCDF, PCB-101 and PBDE-28 and relatively lower 23478-PeCDF, PCB-138 and PBDE-99. Similarly, herring from the Norwegian Sea had a different congener profile than herring from all other sea areas due to relatively higher portions of 2378-TCDF, PCB-28, − 52, − 101 and − 105 and PBDE-28 and relatively lower portions of PCB-138 and − 153 and PBDE-99. For Atlantic halibut sampled only in two sea areas, the Norwegian Sea and the Barents Sea, the congener profiles were mostly similar, but Atlantic halibut from Barents Sea had slightly higher portion of the dioxin congener 2378-TCDF and lower 123678-HxCDD, 234678-HxCDF and 12378-PeCDF compared with Atlantic halibut from the Norwegian Sea. For Greenland halibut, also sampled only in the two northernmost sea areas, the Barents Sea fish had relatively higher contributions from 2378-TCDF, PCB-52 and − 101 and PBDE-47 compared with those from the Norwegian Sea.

For both mackerel and herring, the congener profiles of fish sampled in the Skagerrak were distinct from individuals from the North Sea and the North Atlantic (Fig. 3). However, the congener profiles of mackerel and herring from the Skagerrak were distinct from the other two areas in different ways. While Skagerrak mackerel had higher portions of 23478-PeCDF, PCB-153 and PBDE-100 than mackerel from the North Sea and the North Atlantic, herring from Skagerrak had relatively lower portions of these congeners than herring from the two other sea areas. While mackerel from the Skagerrak had lower portions of 123678-HxCDD, 123678- and 234678-HxCDF, PCB-138 and PBDE-28 and − 47 than mackerel from the North Sea and North Atlantic, herring from Skagerrak had relatively larger contribution from most of these. Additionally,

herring from Skagerrak had relatively lower portions of 2378-TCDF, PCB-180, and PBDE-154, and higher portions of 123478-HxCDF, 12378-PeCDF, and PCB-101.

In general, light congeners such as 2378-TCDF, PCB-28, PCB-52, PBDE − 28 and − 47 were relatively higher in the northernmost sea regions compared to southern areas of the NEAO (Fig. 3 & S6). The composition of congener profiles of DL-PCBs in fatty fish fillets were less variable across sea regions and fish species compared to those of dioxins, nonDL-PCBs and PBDEs (Fig. 3). Within each fatty fish species, congener profiles of POPs in the Barents Sea and the Norwegian Sea were similar and the profiles in the North Atlantic Sea were more like the North Sea. For all substance groups, fish from Skagerrak had somewhat distinct congener profile in comparison to all other sea regions.

POPs fillet concentrations in each species tended to decrease from south to north. POPs concentrations in fish sampled in Skagerrak were highest, followed by those collected in the North Atlantic, the North Sea, and the Norwegian Sea (Fig. S7). POPs concentrations in fish sampled from the Barents Sea were lowest.

3.3. POPs concentrations and biotic/abiotic factors

Correlation analyses were performed to evaluate possible relationships between latitude, fat content, fish length, trophic positions, and POPs (Fig. S8). Positive correlations (r from 0.30 to 0.45) between fat content and all POPs were observed while negative correlations (r from −0.31 to −0.19) between latitude and all POPs were detected. Positive correlations (r from 0.26 to 0.36) were also observed between fish length, fish weight, trophic position and levels of DL-PCBs and nonDL-PCBs, whereas for dioxins and PBDEs there was only a weak relationship or no correlation with these factors. Since POPs are fat soluble, we evaluated the relationship between fillet POP concentrations and fat content. The relationships between fillet POPs concentrations and fat content in large and small fish (see Table S2) from different sea areas are shown in Fig. 4. Fillets of lean fish such as anglerfish, cod, haddock, ling, pollack, saithe and tusk with low fat content in the fillet (< 2.5%) contained low concentrations of POPs regardless of fat content. Semi-fatty and fatty fish such as beaked redfish, golden redfish, plaice, hake, Atlantic halibut, and Greenland halibut with mean fat content > 2.5% contained greater concentrations of POPs compared to lean fish and POP concentrations increased with increasing fat content up to about 10% fat, before levelling off (Fig. 4). The fatty fish species, Atlantic halibut, Greenland halibut, mackerel, and herring, had fat content mean values ranging from 9.8% in Atlantic halibut (average value of both B- and I-cuts, see Supplementary Text S1) to 19.8% in mackerel. In fillet samples with fat contents > 10%, POPs concentrations in large fatty fish species (i.e., Atlantic halibut and Greenland halibut) were higher than in small fatty fish species (i.e., herring, mackerel) with similar fat content from the same sea region.

Because POPs concentrations did not increase linearly with increasing fat content and reached an asymptotic relationship at ≥ 10% fat, we evaluated whether an increase in fat content might cause a dilution of POPs concentrations. To evaluate this hypothesis, POPs concentrations were normalised to fat content for fatty fish species (Figs. S9-S10). In fatty fish species, fat-normalized concentrations of POPs decreased with increasing fat content, likely because of a POPs dilution effect when fat content is high. A similar pattern was also detected for other semi-fatty species including beaked redfish, and golden redfish, but not for plaice (data not shown). However, fat-normalized concentrations of POPs in Atlantic halibut increased slightly in individuals sampled from the Barents Sea, or levelled off for samples in the Norwegian Sea, with increasing fat content (Fig. S10).

To understand the associations between POPs and biotic/abiotic drivers, different models were tested with POPs concentrations as dependent variables and fat content, fish length, trophic level, and latitude as independent variables (Table S5). The model assuming a linear association of fat content with POPs, including fat normalized
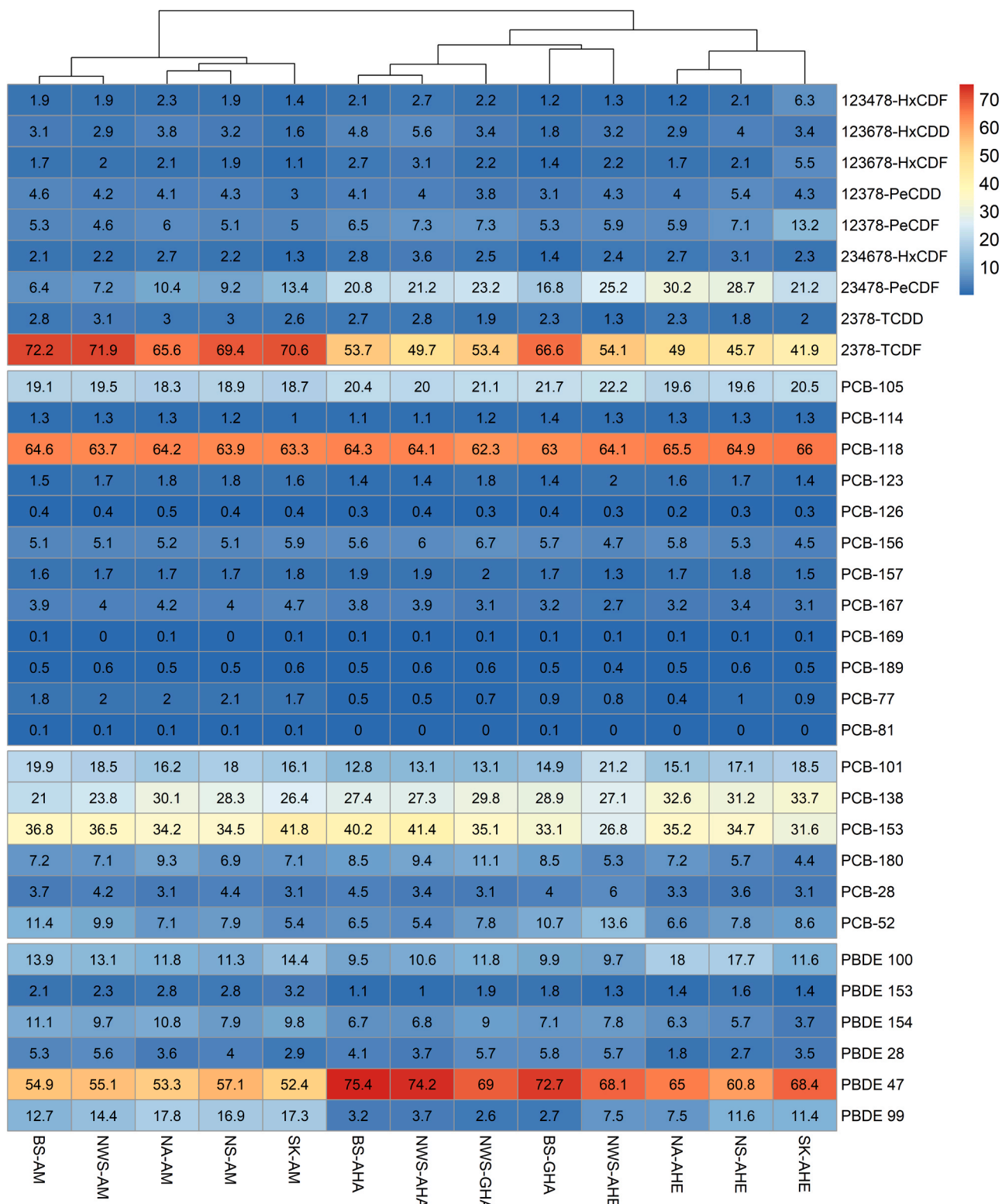
Fig. 3. Congener composition (%) of dioxins, DL-PCBs, nonDL-PCBs and PBDEs in fillets of fatty fish collected from different areas of the NEAO. A cluster analysis was performed for the fish species Atlantic halibut (AHA), Atlantic herring (AHE), Atlantic mackerel (AM), and Greenland halibut (GHA) sampled in the different sea regions, the Barents Sea (BS), the Norwegian Sea (NWS), the North Sea (NS), the North Atlantic Sea (NA) and Skagerrak (SK), respectively. The contribution of congener i in the fish fillet is calculated as $C_{c,i}(\%) = \frac{C_i \cdot 100}{\sum C_i}$ where C_i is the congener concentration expressed on a wet weight basis, and $\sum C_i$ is the sum of the concentrations of all congeners in the group.
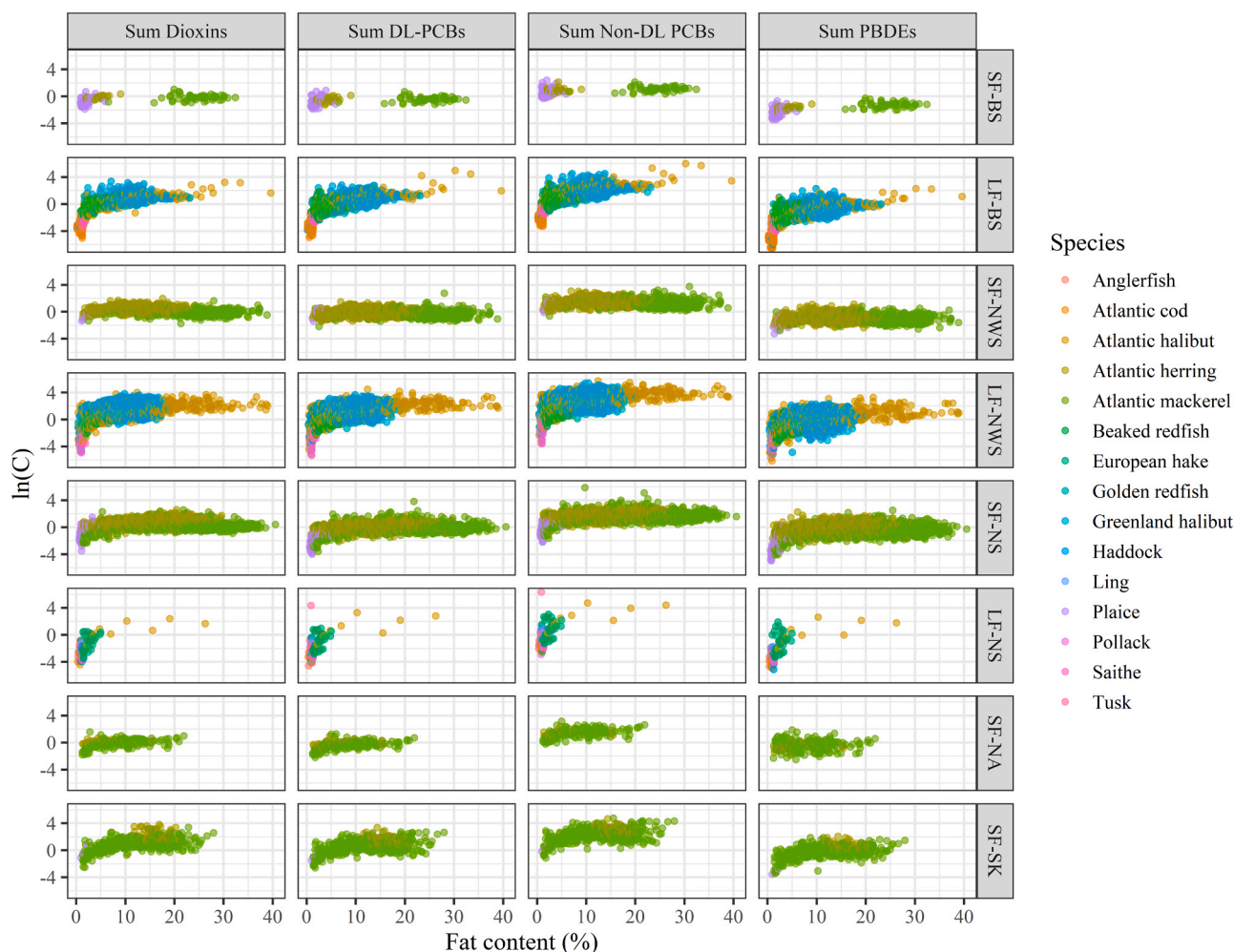
Fig. 4. Relationship between log transformed wet weight concentrations of POPs (dioxins (pg g^{-1}), DL-PCBs (ng g^{-1}), nonDL-PCB (ng g^{-1}) and PBDE (ng g^{-1})) and fat content (%) in the fillet of different fish species in the NEAO. SF denote small fish species (herring, mackerel, and plaice) and LF denote large fish species (anglerfish, cod, Atlantic halibut, beaked redfish, hake, golden redfish, Greenland halibut, haddock, ling, pollack, saithe and tusk). BS, NWS, NA and NS denote the Barents Sea, the Norwegian Sea, the North Atlantic and the North Sea, respectively. The different coloured symbols represent different species as indicated. POPs of large fish species in the North Atlantic and Skagerrak were not shown due to their low number of samples in these sea regions (< 50).

concentrations or wet weight concentrations, did not show a strong goodness of fit for the model (r^2 of 0.087–0.4, Table S5). When a nonlinear term analogous to the Hill description referring to the concentration of POPs (expressed in wet weight) associated with fat in fillet tissue (Eq. 1, see Material and Methods) was incorporated in the model, r^2 model fit and the associated AIC values improved significantly (i.e., a change in r^2 from 0.294 to 0.559, Table S5). The best model fit (high r^2, lowest AIC values) was obtained for all POPs groups expressed in wet weight and when both fish length and trophic level predictors were included in the model. The second-order, non-linear model (NLM) mostly showed a better fit compared to the first order and third order models, with r^2 values ranging from 0.508 to 0.543 for sum of congeners of DL- PCBs, nonDL-PCBs, and PBDEs (Table S5, Fig. S11). For dioxins, the second- and third-order models showed similar fit. Values of the model parameters estimated by the second order model are shown in Table S6. Notably, partial correlation coefficients between the model variables and POPs were also calculated (Table S7). The coefficients were low, confirming high variation of POPs concentrations and a non-linear relationship with fat content.

When using the second order NLM describing POP concentrations in fillets, where K_m illustrates the association of POPs to fat content, estimated K_m values were highly variable across the different POP congeners (Fig. 5). As a general pattern, K_m decreased with increasing degree

of chlorination or bromination. For dioxin congeners, except for 2378-TCDD (K_m=4.09%), K_m decreased with increasing chlorination from 2378-TCDF to 234678-HxCDF. Similarly, for the DL-PCB congeners K_m decreased with increased chlorination from PCB-81 to PCB-169, and for nonDL-PCBs from PCB-52 to PCB-180. K_m values of the PBDEs congeners decreased with increasing bromination from PBDE 28 to PBDE 154, except PBDE 99 which had a K_m of 8.22%, the highest value of all the analysed POPs congeners. Nevertheless, K_m values were similar between the sums of dioxins, DL-PCBs, nonDL-PCBs and PBDEs, with mean values of 3.77%, 3.61%, 3.82% and 3.88%, respectively. When K_m values for the nonDL-PCBs and the PBDEs were plotted against K_{OW} values from the literature [37,60], there were negative correlations between K_m and K_{OW} (Fig. S12).

The model also showed that the sum of PBDEs varied the most with latitude ($S_{c,Lat} = -7.21\%/Lat$), followed by sum nonDL-PCB ($S_{c,Lat} = -5.06\%/Lat$) and sum dioxin ($S_{c,Lat} = -4.69\%/Lat$) while sum DL-PCB ($S_{c,Lat} = -3.87\%/Lat$) varied the least with latitude (Table S8). These results were consistent with those described in the previous section since congener composition profiles of DL-PCBs in fatty fish fillets were the least variable across sea regions. For all the POP congeners a negative sensitivity to latitude was found. The light congeners 2378-TCDF, PCB-28, PCB-52, PBDE-28 showed lower sensitivity to latitude compared to the remaining congeners within their respective POP groups ($S_{c,Lat}$
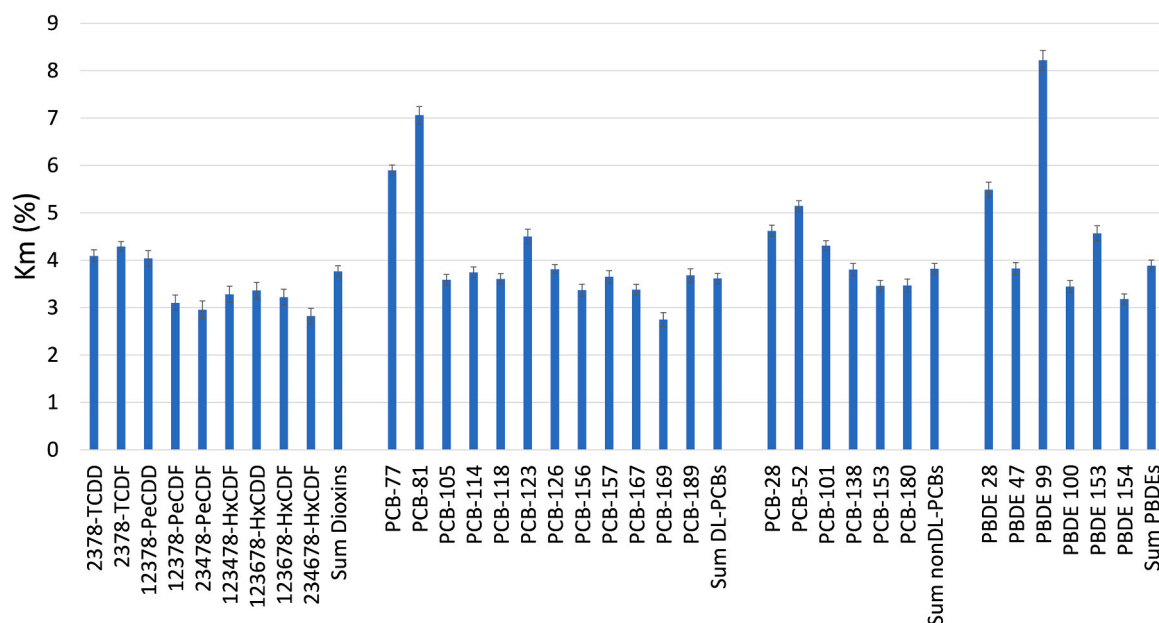
Fig. 5. Estimated affinities to fat content (K_m) of different congeners and sums of dioxins, DL-PCBs, nonDL-PCBs and PBDEs. Values were expressed as mean ± standard error.

values −4.09, −1.1, −0.76 and −3.32%/Lat, respectively).

4. Discussion

4.1. POPs congener composition and species variation

This study, for the first time, explored detailed POPs congener profiles in marine fish using a large dataset of 15 commercially relevant fish species from the NEAO. The congener compositions of the studied POPs, dioxins, DL-PCBs, nonDL-PCBs and PBDEs, varied between fish species and geographical areas. The effect of fish species on POP congener composition was a stronger driver than the effects of sea region since POPs congener composition profiles were more influenced by species than by geography. Several explanations may be suggested for the observed wide range of inter-species variation. For example, different habitats such as fjord, coast, or open sea or different life histories (including age, growth rate, age at maturity), habitat use patterns, and feeding strategies (i.e., demersal versus pelagic) could be possible explanations. In addition, differences in congener profiles for benthic and pelagic invertebrates and fish was reported for PBDEs [54]. The ratio between different congeners may also be driven by trophic position due to differences in congener biomagnification factors, as has been reported for PBDEs [54,63]. Some of the similarities observed in our study could perhaps be a result of similar habitat use or prey selection. However, herring and mackerel, both pelagic plankton feeders mostly sampled in the open ocean, had very different congener profiles, especially when considering fish from the same region. Conversely, congener profiles of cod, saithe, and mackerel were relatively similar, despite having different feeding strategies and habitat use patterns. While mackerel is strictly pelagic, cod, and saithe are benthopelagic, and mackerel was predominantly sampled in the open North Sea and in the coastal, Skagerrak region. Cod was sampled predominantly in the open sea, but also from coastal and fjord ecosystems. Habitat and feeding strategies alone could not explain the observed variation in congener profiles between species. Notably, lean fish species had very low fillet concentrations of POPs, and many congeners had censored data. Because of this, and despite the use of predicted concentrations of the censored congeners, the profiles for these species may be more uncertain and therefore should be interpreted with caution. Nevertheless, cod and saithe had relatively similar POP congener profiles in both fillet and liver (Fig. S4).

4.2. POPs congener composition and potential sources between sea regions

There were some distinct geographical patterns of congener composition when fatty fish species were considered. Except for Greenland halibut, fatty fish from the Barents Sea had relatively similar POP profiles compared to those from the Norwegian Sea while fish from the North Sea had comparable POPs profiles compared to those sampled from the North Atlantic. Mackerel and herring from Skagerrak had dioxin profiles which were different from samples collected from other areas, although in these two species different congeners of PeCDF were more prevalent in Skagerrak relative to the other areas. Gradients of POPs in the ocean were found due to historical pollution in the south, especially in Skagerrak [45] because of prevailing ocean currents [42]. However, higher prevalence of the lighter congener 2378-TCDF in fish fillets collected in the northernmost sea areas might be best explained by the higher capacity for long range atmospheric transport of this congener. 2378-TCDF has a relatively higher vapour pressure (1×10^{-5} Pa) compared to the remaining dioxin congeners (vapour pressures from 5.4×10^{-8} to 6.2×10^{-6} Pa) [39]. Our model also showed that this congener had the lowest sensitivity to latitude among the different dioxin congeners. In herring, the percentages of the heavier PCDFs were higher in Skagerrak compared to other sea regions. Due to proximity, Skagerrak is more influenced by point sources, pollution from the southern coast of Norway and central Europe and may also be more affected by the historically dioxin-contaminated Baltic Sea [5]. DL-PCB composition profiles showed little variation across sea regions. Also, low sensitivity to latitude was found for most light PCB congeners (PCB-28, −52, −101, −105, −115 and −118), with PCB-77 and PCB-81 as the only exceptions. The results confirmed their increasing distribution northwards likely by atmospheric transport due to their relatively high evaporation capacity. Higher occurrence in the northernmost sea areas reduced the observed gradient effects across latitudes. Large variation of most PBDEs with latitude (Table S8) might result from historical pollution in the south, especially Skagerrak, and relatively less atmospheric transport and deposition, since most PBDEs, except PBDE 28, have low vapour pressures ($< 1 \times 10^{-4}$ Pa) [2]. This was also confirmed by PBDE-28 showing lower sensitivity to latitude compared to the other PBDE congeners. Moreover, sources of the long range transported pollution have been shown to vary between different air monitoring

stations in Norway and the Arctic [56] and between different ocean currents [42], which might also explain some of the observed geographical variations.

4.3. Abiotic and biotic factors governing POPs in marine fish

Species-specific POPs profiles may be affected by multiple biotic and abiotic factors, at different spatiotemporal scales, resulting from different species-specific metabolic rates, lipid metabolism, and subsequently highly variable POPs accumulation rates [28,38,44,49,50,9]. In our study, we used non-linear models to evaluate the relationships between POPs concentrations, fat content, trophic position, fish length, and latitude (Eq. 1). The best model fit was obtained for all POPs wet weight concentrations when both fish length and trophic position were included in the model. The model which included a nonlinear term for the association between wet weight concentrations of POPs and fat content (Eq.1) gave the best model fit compared to the model using a linear term. Fat content and POPs concentrations were positively but not linearly correlated, and at high fat concentrations ($\geq$10%) there was no correlation between POPs and fat content. Moreover, fat normalized POPs concentrations in fillets of fatty fish species were negatively correlated with fat content, indicating that POPs are likely being diluted when fillet fat increases rapidly. This effect is most likely explained by plankton feeding strategies common in the fatty pelagic species such as mackerel or herring, that are subject to large seasonal variation in fat content due to seasonality in food availability, gonad maturation, and spawning [29,31]. A baseline study of Norwegian spring spawning herring caught throughout the year reported no overall correlation between POPs concentrations and fat content [23]. Increasing POPs concentrations were observed as fillet storage fat was metabolised during winter starvation, leading to maximum observed POPs concentrations (wet weight) in late spring and at intermediate fat content (9.9%). During the summer feeding season, fat was accumulated more rapidly (from 4% to 17%) than POPs, leading to only a weak increase in POPs concentrations on wet weight basis and decreasing concentrations on a fat weight basis. Mackerel and Greenland halibut may be subject to some of the same effects as shown for herring. In Atlantic halibut, however, fat-normalized POPs concentrations were stable or increased slightly with greater fat content, indicating no dilution effect. Atlantic halibut is an apex predator, and adults feed on other large fish such as redfish or tusk [27], which may have high body burdens of POPs. Thus, exposure to and intake of POPs per unit fat is likely to be greater in Atlantic halibut compared to plankton feeding pelagic fish such as herring or mackerel, which may at least partly explain the lack of an observed dilution effect. Moreover, Atlantic halibut are large bodied (females up to 300 kg, males up to 50 kg) and may reach older ages and larger sizes prior to spawning, and males and females have very different growth rates, ages, and sizes at maturation [27]. The fish size range of Atlantic halibut analysed in this study ranged from 44 to 250 cm, and high levels of POPs in large fish with high fat content might offset potential dilution effects. However, when Atlantic halibut were divided into large (length $\geq$116 cm) and small size categories (length <116 cm), both groups showed similar patterns where fat-normalized POPs concentrations were stable or increased slightly with greater fat content. Since this species is demersal, fat variation driven by seasonality might be less pronounced compared with pelagic fish species. Notably, POPs accumulation is highly nuanced, species-specific, and associated with growth rate variation, life history, habitat, prey use, and spawning patterns. These factors, collectively, may offer the best explanation of the observed differences in POPs across species, particularly for the fatty species.

Our non-linear model confirmed that trophic position is the main driver of biomagnification of POPs in fish, where POPs concentrations increase with increasing TP, as has been shown previously for many POPs [38,51,60,9]. Trophic position and fish size is often strongly correlated, since large fish naturally consume larger prey at higher

trophic positions compared with smaller fish. However, the model indicates an additional effect of size not caused by trophic position. The effect of fish size is most likely due to higher age, where older fish have accumulated contaminants over a longer period than younger fish. The last term in Eq.1 represents the relative variation of a specific POP congener with respect to latitude (see $S_{C,Lat}$ Table S8) and the model confirms that most light congeners varied less with latitude than heavy congeners. K_m illustrates the association of POPs to fat content, where a higher K_m indicated a lower association with fat content. Generally, K_m decreased with increasing degree of chlorination or bromination and showed a negative correlation with the log-transformed n-octanol water partition coefficient, $\ln(K_{OW})$, for PCBs and PBDEs from the literature [37,60]. Lipoprotein particles are reported to be the main transport mechanism of fat-soluble POPs [41,48], but highly chlorinated PCBs were more likely to be associated with lipoproteins compared to less chlorinated PCBs [40,48]. Lower chlorinated PCB congeners are excreted to a greater extent than those with higher chlorination [61]. Our finding of lower K_m with a higher degree of chlorination and bromination in fish species from NEAO thus provides support for higher chlorinated congeners having a higher affinity to fat and being more easily bioaccumulated and biomagnified [37]. This also likely influences how POPs congener composition varies between species.

4.4. Food safety aspects of POPs in marine fish from NEAO

Although median TEQ concentrations of the sum of dioxins and the sum of dioxins and DL-PCBs in fillet samples of Greenland halibut and Atlantic halibut were below the MLs set by EU for fish meat traded for human consumption, a considerable portion of these fish species had concentrations of the sum of POPs above the MLs. Up to 15.3% of Greenland halibut and 15.6% of Atlantic halibut I-cut had concentrations of the sum of dioxins and DL-PCBs above the MLs while 4.9% of Greenland halibut and 8.1% of Atlantic halibut I-cut had concentrations of the sum the sum PCB6 above the MLs. A risk assessment of contaminant intake due to fish consumption has not been made here. However, in a previous study [28], we estimated that a considerable portion of the seafood consuming population, in particular those consuming fatty fish, would exceed the TWI for dioxins and DL- PCBs of 2 pg TEQ/kg body weight per week set by EFSA [19], while their intake would be below the tolerable intake set by the Joint FAO/WHO Expert Committee on Food Additives (provisional tolerable monthly intake 70 pg kg^{-1} body weight per month) [32]. In general, there is a low risk of exposure to PBDEs from consumption of seafood from the NEAO [28,50]. In another recent study described by Zhu et al. [64], PBDEs were the predominant halogenated organic pollutants in the Beibu Gulf and their content in marine fish might not pose a risk to human health. Nevertheless, regularly consuming large amounts of fatty fish would result in greater risk of health effects from POPs exposure, compared with leaner fish species, and consuming higher TP fatty fish would result in greater risk than the lower TP species. However, while Greenland halibut and Atlantic halibut were the species with the highest concentrations of POPs, their captured volumes are relatively low (2020: 120,000 and 10,000 tons, respectively, FAO [21]). Herring and mackerel exhibit lower concentrations of POPs, but their capture volumes are greater (2020: 1.5 and 1.0 mill. tons, respectively, FAO [21]) and therefore these species may contribute as much or more to the total intake of POPs in humans. We should also bear in mind that, when considering health effects of seafood consumption, the potentially negative effects of contaminant exposure should be weighed against potentially positive effects of nutrients of fatty fish such as omega-3 fatty acids [49,58,62]. Such effects have not been considered in this study.

4.5. Study limitations

Our study has some important limitations regarding species distributions, sampling history, and bias. The large baseline studies and

follow-up monitoring provide a solid basis for studying contaminant concentrations and congener composition across several commercially relevant fish species from a very large geographical area such as the NEAO. The sampling effort, however, varied considerably across species. Particularly, fillet sample sizes of fatty fish were larger compared to lean fish, because when analysing for POPs in lean fish species, liver tissues were traditionally chosen [35]. In several of the lean fish species included here only composite samples were analysed, resulting in a lower number of analysed samples which may mask the potential effects from individual variation. Moreover, in some baseline studies, geography and seasonality dynamics were well covered, while for others there was geographical or seasonal sampling bias. The concentrations in lean fish fillets of several POP congeners, especially dioxins, were often well below LOQs, making it difficult to identify their associations with biotic and abiotic factors. Therefore, the results regarding congener profile variation were more uncertain for lean fish compared to fatty fish, despite estimation of missing values by implementation of combined ROS and PCA models. Because of this, lean fish species were not considered when comparing congener profiles between geographical areas, only when comparing the different species. Furthermore, temporal trends were not accounted for in this study. Atmospheric deposition of POPs in Norwegian areas has decreased since the early 1990s [7], which possibly could be reflected in the levels and congener profiles measured in marine fish. In a recent study from the Barents Sea, decreasing temporal trends of POPs in cod and haddock from 1992 to 2015 were demonstrated, with a slower rate of decline in the 2000s [8]. In our study, the different fish species were sampled over different time periods, and how this might affect the between-species variations in POPs concentrations and congener profiles is unknown and falls outside the scope of this paper. We are also left with some important questions regarding the bioaccumulation regimes related to physiological and toxicokinetic dynamics including POPs organ-specific transport, storage, excretion, and potential and unknown effects of spawning. Nevertheless, this study demonstrates a purposeful method to report variation and bioaccumulation of POPs congener profiles in commercially relevant marine fish species from the NEAO.

5. Conclusions

This study evaluated ~8400 fillet samples from 15 fish species sampled in the NEAO, a unique dataset for studying variations of POP congeners between species and geographical areas. Fatty fish species with high TP exhibited the highest POPs fillet concentrations. Fish from the Norwegian Sea and the Barents Sea had relatively similar composition profiles and the same was found for the North Sea and the North Atlantic. Skagerrak had its own congener profile, probably due to a larger influence from more regional sources compared to other areas. POPs concentrations generally decreased from south to north in the NEAO while low variation of some light congeners in fish with latitude was found due to their long-range atmospheric transport northwards. An unexpected finding in this study was that POP concentrations increased with increasing fat content but levelled off at approximately $\geq$ 10% fat, likely due to a dilution effect of POPs at high fat content. This study can serve as an effective model to further understand the potential distribution and accumulation mechanisms of POPs in marine fish harvested from the NEAO.

CRediT authorship contribution statement

Quang Tri Ho: Conceptualization, Methodology, Formal analysis, Writing – original draft, Writing – review & editing. **Sylvia Frantzen:** Conceptualization, Resources, Investigation, Methodology, Formal analysis, Writing – original draft, Writing – review & editing. **Bente M. Nilsen:** Conceptualization, Resources, Data interpretation, Writing – review & editing. **Ole Jakob Nøstbakken:** Conceptualization, Methodology, Writing – review & editing. **Atabak M. Azad:**

Conceptualization, Resources, Data interpretation, Writing – review & editing. **Arne Duinker:** Conceptualization, Resources, Writing – review & editing. **Lise Madsen:** Supervision, Conceptualization, Writing – original draft, Writing – review & editing. **Michael S. Bank:** Conceptualization, Resources, Supervision, Data interpretation, Formal analysis, 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.

Data Availability

Data will be made available on request.

Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2023.131758.

References

[1] Alexander, J., Benford, D., Boobis, A., Bergman, Å., 2011. Scientific opinion on Tetrabromobisphenol A (TBBPA) and its derivatives in food: EFSA panel on contaminants in the food chain (CONTAM). EFSA J 9 (12), 2477.

[2] Al-Omran, L.S., 2018. Physiochemical properties and environmental levels of legacy and novel brominated flame retardants, Flame Retardants. IntechOpen.

[3] Azad, A.M., Frantzen, S., Bank, M.S., Nilsen, B.M., Duinker, A., Madsen, L., Maage, A., 2019. Effects of geography and species variation on selenium and mercury molar ratios in Northeast Atlantic marine fish communities. Sci Total Environ 652, 1482–1496.

[4] Bank, M.S., Frantzen, S., Duinker, A., Amouroux, D., Tessier, E., Nedreaas, K., et al., 2021. Rapid temporal decline of mercury in Greenland halibut (*Reinhardtius hippoglossoides*). Environ Pollut 289, 117843.

[5] Berge, J.A., Hylland, K., Schlabach, M., Ruus, A., 2011. Accumulation of polychlorinated dibenzo-p-dioxins and Furans in Atlantic cod (*Gadus morhua*)—cage experiments in a Norwegian Fjord. J Toxicol Environ Health, Part A 74 (7–9), 455–465.

[6] Berntssen, M.H., Lundebye, A.K., Torstensen, B.E., 2005. Reducing the levels of dioxins and dioxin-like PCBs in farmed Atlantic salmon by substitution of fish oil with vegetable oil in the feed. Aquacult Nutr 11 (3), 219–231.

[7] Bohlin-Nizzetto, P., Aas, W., Halvorsen, H.L., Nikiforov, V., Pfaffhuber, K.A., 2021. Monitoring of environmental contaminants in air and precipitation. Annual report 2020. NILU rapport.

[8] Boitsov, S., Grøsvik, B.E., Nesje, G., Malde, K., Klungsøyr, J., 2019. Levels and temporal trends of persistent organic pollutants (POPs) in Atlantic cod (*Gadus morhua*) and haddock (*Melanogrammus aeglefinus*) from the southern Barents Sea. Environ Res 172, 89–97.

[9] Borgå, K., Kidd, K.A., Muir, D.C., Berglund, O., Conder, J.M., Gobas, F.A., Kucklick, J., Malm, O., Powell, D.E., 2012. Trophic magnification factors: considerations of ecology, ecosystems, and study design. Integr Environ Assess Manag 8 (1), 64–84.

[10] Burnham, K.P., Anderson, D.R., 2002. A practical information-theoretic approach. Model Selection and Multimodel Inference, second ed. Springer, New York, p. 2.

[11] Bureau, S., Zebühr, Y., Broman, D., Ishaq, R., 2004. Biomagnification of polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs) studied in pike (*Esox lucius*), perch (*Perca fluviatilis*) and roach (*Rutilus rutilus*) from the Baltic Sea. Chemosphere 55 (7), 1043–1052.

[12] Castro-Jiménez, J., Banaru, D., Chen, C.-T., Jiménez, B., Muñoz-Arnanz, J., Deviller, G., Sempere, R., 2021. Persistent organic pollutants burden, trophic

magnification and risk in a pelagic food web from coastal NW Mediterranean Sea. Environ Sci Technol 55 (14), 9557–9568.

[13] Chen, M., Wong, W., Chung, S., Tran, C., Chan, B., Ho, Y., Xiao, Y., 2014. Quantitative risk–benefit analysis of fish consumption for women of child-bearing age in Hong Kong. Food Addit. Contam Part A 31 (1), 48–53.

[14] Corsolini, S., Sarà, G., 2017. The trophic transfer of persistent pollutants (HCB, DDTs, PCBs) within polar marine food webs. Chemosphere 177, 189–199.

[15] Dale, H.F., Madsen, L., Lied, G.A., 2019. Fish–derived proteins and their potential to improve human health. Nutr Rev 77 (8), 572–583.

[16] EFSA, 2006. Advice of the scientific panel on contaminants in the food chain on a request from the commission related to relevant chemical compounds in the group of brominated flame retardants for monitoring in feed and food. EFSA J 328, 1–4.

[17] EFSA, 2011. Scientific opinion on polybrominated diphenyl ethers (PBDEs) in food. EFSA J 9 (5), 2156.

[18] EFSA, 2012. Scientific Opinion on the risk for public health related to the presence of mercury and methylmercury in food. EFSA J 10 (12), 2985.

[19] EFSA, 2018. Risk for animal and human health related to the presence of dioxins and dioxin-like PCBs in feed and food. EFSA J 16 (11), e05333.

[20] EU, 2022. Commission regulation (EC) No 1881/2006 of 19 December 2006 setting maximum levels for certain contaminants in foodstuffs (Text with EEA relevance). Official Journal of the European Union. Consolidated version 07.01.2022.

[21] FAO, 2022. Fisheries & Aquaculture - Global capture production Quantity (1950 - 2020), https://www.fao.org/fishery/statistics-query/en/capture.

[22] Fiedler, H., Kallenborn, R., Boer, Jd, Sydnes, L.K., 2019. The Stockholm convention: a tool for the global regulation of persistent organic pollutants. Chem Int 41 (2), 4–11.

[23] Frantzen, S., Mage, A., Iversen, S.A., Julshamn, K., 2011. Seasonal variation in the levels of organohalogen compounds in herring (*Clupea harengus*) from the Norwegian Sea. Chemosphere 85 (2), 179–187.

[24] Gesztelyi, R., Zsuga, J., Kemeny-Beke, A., Varga, B., Juhasz, B., Tosaki, A., 2012. The Hill equation and the origin of quantitative pharmacology. Arch Hist Exact Sci 66, 427–438.

[25] Goutelle, S., Maurin, M., Rougier, F., Barbaut, X., Bourguignon, L., Ducher, M., Maire, P., 2008. The Hill equation: a review of its capabilities in pharmacological modelling. Fundam Clin Pharm 22 (6), 633–648.

[26] Goutte, A., Chevreuil, M., Alliot, F., Chastel, O., Cherel, Y., Eléaume, M., Massé, G., 2013. Persistent organic pollutants in benthic and pelagic organisms off Adélie Land, Antarctica. Mar Pollut Bull 77 (1–2), 82–89.

[27] Haug, T., 1990. Biology of the Atlantic halibut, *Hippoglossus hippoglossus* (L., 1758). Adv. Mar. Biol. Elsevier, pp. 1–70.

[28] Ho, Q.T., Bank, M.S., Azad, A.M., Nilsen, B.M., Frantzen, S., Boitsov, S., Maage, A., Kögel, T., Sanden, M., Frøyland, L., 2021. Co-occurrence of contaminants in marine fish from the North East Atlantic Ocean: implications for human risk assessment. Environ Int 157, 106858.

[29] Holst, J.C., Røttingen, I., Melle, W.., 2004. The Herring. In: Skjoldal, H.R. (Ed.), The Norwegian Sea Ecosystem. Tapir Academic Press, Trondheim, pp. 203–226.

[30] Isaak, D.J., Luce, C.H., Horan, D.L., Chandler, G.L., Wollrab, S.P., Nagel, D.E., 2018. Global warming of salmon and trout rivers in the Northwestern U.S.: Road to ruin or path through purgatory? Trans Am Fish Soc 147 (3), 566–587.

[31] Iversen, S.A.., 2004. Mackerel and horse mackerel. In: Skjoldal, H.R. (Ed.), The Norwegian Sea Ecosystem. IMR/Tapir academic press, Trondheim, pp. 289–300.

[32] JECFA, 2010. Joint FAO/WHO expert consultation on the risks and benefits of fish consumption. FAO Fisheries and Aquaculture Report No 978. Rome.

[33] Jones, K.C., De Voogt, P., 1999. Persistent organic pollutants (POPs): state of the science. Environ Pollut 100 (1–3), 209–221.

[34] Julshamn, K., Lundebye, A.K., Heggstad, K., Berntssen, M.H.G., Boe, B., 2004. Norwegian monitoring programme on the inorganic and organic contaminants in fish caught in the Barents Sea, Norwegian Sea and North Sea, 1994–2001. Food Addit Contam 21 (4), 365–376.

[35] Julshamn, K., Duinker, A., Berntssen, M., Nilsen, B.M., Frantzen, S., Nedreaas, K., Maage, A., 2013. A baseline study on levels of polychlorinated dibenzo-p-dioxins, polychlorinated dibenzofurans, non-ortho and mono-ortho PCBs, non-dioxin-like PCBs and polybrominated diphenyl ethers in Northeast Arctic cod (*Gadus morhua*) from different parts of the Barents Sea. Mar Pollut Bull 75 (1), 250–258.

[36] Karl, H., Ruoff, U., 2007. Dioxins, dioxin-like PCBs and chloroorganic contaminants in herring, Clupea harengus, from different fishing grounds of the Baltic Sea. Chemosphere 67 (9), S90–S95.

[37] Kelly, B.C., Ikonomou, M.G., Blair, J.D., Gobas, F.A., 2008. Bioaccumulation behaviour of polybrominated diphenyl ethers (PBDEs) in a Canadian Arctic marine food web. Sci Total Environ 401 (1–3), 60–72.

[38] Kobayashi, J., Yoshimoto, M., Yamada, K., Okamura, K., Sakurai, T., 2019. Comparison of trophic magnification factors of PCBs and PBDEs in Tokyo Bay based on nitrogen isotope ratios in bulk nitrogen and amino acids. Chemosphere 226, 220–228.

[39] Li, X.W., Shibata, E., Nakamura, T., 2005. Thermodynamic prediction of vapor pressures for polychlorinated dibenzo-p-dioxins, polychlorinated dibenzofurans, and polybrominated dibenzo-p-dioxins. Environ Toxicol Chem 24 (9), 2167–2177.

[40] Lind, P.M., Lind, L., 2020. Are persistent organic pollutants linked to lipid abnormalities, atherosclerosis and cardiovascular disease? A review. J Lipid Atheroscler 9 (3), 334–348.

[41] Ljunggren, S.A., Helmfrid, I., Norinder, U., Fredriksson, M., Wingren, G., Karlsson, H., Lindahl, M., 2017. Alterations in high-density lipoprotein proteome and function associated with persistent organic pollutants. Environ Int 98, 204–211.

[42] Lohmann, R., Markham, E., Klanova, J., Kukucka, P., Pribylova, P., Gong, X., Pockalny, R., Yanishevsky, T., Wagner, C.C., Sunderland, E.M., 2020. Trends of diverse POPs in air and water across the western atlantic ocean: strong gradients in the ocean but not in the air. Environ Sci Technol 55 (14), 9498–9507.

[43] Lundebye, A.-K., Lock, E.-J., Rasinger, J.D., Nøstbakken, O.J., Hannisdal, R., Karlsbakk, E., Wennevik, V., Madhun, A.S., Madsen, L., Graff, I.E., 2017. Lower levels of persistent organic pollutants, metals and the marine omega 3-fatty acid DHA in farmed compared to wild Atlantic salmon (*Salmo salar*). Environ Res 155, 49–59.

[44] Madgett, A.S., Yates, K., Webster, L., McKenzie, C., Brownlow, A., Moffat, C.F., 2022. The concentration and biomagnification of PCBs and PBDEs across four trophic levels in a marine food web. Environ Pollut 309, 119752.

[45] Magnusson, K., Ekelund, R., Dave, G., Granmo, Å., Förlin, L., Wennberg, L., Samuelsson, M.-O., Berggren, M., Brorström-Lundén, E., 1996. Contamination and correlation with toxicity of sediment samples from the Skagerrak and Kattegat. J Sea Res 35 (1–3), 223–234.

[46] Markhus, M., Rasinger, J., Malde, M., Frøyland, L., Skotheim, S., Braarud, H., Stormark, K., Graff, I., 2015. Seafood consumption predicts the maternal docosahexaenoic acid status 3-, 6-and 12 months postpartum: 149/201. Ann Nutr Metab 67.

[47] Markhus, M.W., Kvestad, I., Midtbø, L.K., Nerhus, I., Ødegaard, E.R., Graff, I.E., Lie, Ø., Dahl, L., Hysing, M., Kjellevold, M., 2018. Effects of cod intake in pregnancy on iodine nutrition and infant development: study protocol for Mommy's Food-a randomized controlled trial. BMC Nutr 4 (1), 1–8.

[48] Noren, K., Weistrand, C., Karpe, F., 1999. Distribution of PCB congeners, DDE, hexachlorobenzene, and methylsulfonyl metabolites of PCB and DDE among various fractions of human blood plasma. Arch Environ Contam Toxicol 37, 408–414.

[49] Nøstbakken, O., Rasinger, J., Hannisdal, R., Sanden, M., Frøyland, L., Duinker, A., Frantzen, S., Dahl, L., Lundebye, A.-K., Madsen, L., 2021. Levels of omega 3 fatty acids, vitamin D, dioxins and dioxin-like PCBs in oily fish; a new perspective on the reporting of nutrient and contaminant data for risk–benefit assessments of oily seafood. Environ Int 147, 106322.

[50] Nøstbakken, O.J., Duinker, A., Rasinger, J.D., Nilsen, B.M., Sanden, M., Frantzen, S., Hove, H.T., Lundebye, A.-K., Berntssen, M.H.G., Hannisdal, R., Madsen, L., Maage, A., 2018. Factors influencing risk assessments of brominated flame-retardants; evidence based on seafood from the North East Atlantic Ocean. Environ Int 119, 544–557.

[51] Rebryk, A., Haglund, P., 2022. Comprehensive non-target screening of biomagnifying organic contaminants in the Baltic Sea food web. Sci Total Environ 851, 158280.

[52] Reksten, A.M., Bøkevoll, A., Frantzen, S., Lundebye, A.-K., Kögel, T., Kolås, K., Aakre, I., Kjellevold, M., 2020. Sampling protocol for the determination of nutrients and contaminants in fish and other seafood–The EAF-Nansen Programme. MethodsX 7, 101063.

[53] Skåre, J.U., Brantsæter, A.L., Frøyland, L., Hemre, G.I., Knutsen, H.K., Lillegaard, I. T.L., Andreassen, Å.K., Elvevoll, E.O., Andersen, L.F., Hjeltnes, B., 2014. Benefit-risk assessment of fish and fish products in the Norwegian diet–an update. Opinion of the Scientific Steering Committee of the Norwegian Scientific Committee for Food Safety. VKM Report.

[54] Sørmo, E.G., Jenssen, B.M., Lie, E., Skaare, J.U., 2009. Brominated flame retardants in aquatic organisms from the North Sea in comparison with biota from the high Arctic marine environment. Environ Toxicol Chem 28 (10), 2082–2090.

[55] Tekindal, M.A., Erdoğan, B.D., Yavuz, Y., 2017. Evaluating left-censored data through substitution, parametric, semi-parametric, and nonparametric methods: A simulation study. Interdiscip Sci Comput Life Sci 9 (2), 153–172.

[56] Ubl, S., Scheringer, M., Stohl, A., Burkhart, J.F., Hungerbuhler, K.., 2012. Primary source regions of polychlorinated biphenyls (PCBs) measured in the Arctic. Atmospheric Environment 62, 391–399.

[57] Van den Berg, M., Birnbaum, L.S., Denison, M., De Vito, M., Farland, W., Feeley, M., Fiedler, H., Hakansson, H., Hanberg, A., Haws, L., 2006. The 2005 World Health Organization reevaluation of human and mammalian toxic equivalency factors for dioxins and dioxin-like compounds. Toxicol Sci 93 (2), 223–241.

[58] VKM, A.L., Berstad, P., Bukhvalova, B., Carlsen, M., Dahl, L., Goksøyr, A., Jakobsen, L., Knutsen, H., Kvestad, I., Lillegaard, I., 2022. Benefit and risk assessment of fish in the Norwegian diet. Scientific Opinion of the Scientific Steering Committee of the Norwegian Scientific Committee for Food and Environment. VKM Report.

[59] Vuorinen, P.J., Keinänen, M., Kiviranta, H., Koistinen, J., Kiljunen, M., Myllylä, T., et al., 2012. Biomagnification of organohalogens in Atlantic salmon (*Salmo salar*) from its main prey species in three areas of the Baltic Sea. Sci Total Environ 421, 129–143.

[60] Walters, D.M., Mills, M.A., Cade, B.S., Burkard, L.P., 2011. Trophic magnification of PCBs and its relationship to the octanol− water partition coefficient. Environ Sci Technol 45 (9), 3917–3924.

[61] WHO, 2000. Air quality guidelines for Europe. Copenhagen: WHO Regional Office for Europe.

[62] WHO, 2011. Report of the joint FAO/WHO expert consultation on the risks and benefits of fish consumption. 25-29 January 2010. World Health Organization, Rome, Italy.

[63] Zhang, Y., Wang, W., Song, J., Ren, Z., Yuan, H., Yan, H., Zhang, J., Pei, Z., He, Z., 2016. Environmental characteristics of polybrominated diphenyl ethers in marine system, with emphasis on marine organisms and sediments. Biomed Res Int 2016.

[64] Zhu, L., Wang, C., Huang, L., Ding, Y., Cheng, Y., Rad, S., Xu, P., Kang, B., 2022. Halogenated organic pollutants (HOPs) in marine fish from the Beibu Gulf, South China Sea: levels, distribution, and health risk assessment. Mar Pollut Bull 185, 114374.