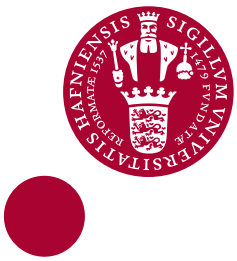

Effects of geography and species variation on selenium and mercury molar ratios in Northeast Atlantic marine fish communities

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Published in:
Science of the Total Environment

DOI:
10.1016/j.scitotenv.2018.10.405

Publication date:
2019

Document version
Publisher's PDF, also known as Version of record

Science of the Total Environment 652 (2019) 1482–1496

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Effects of geography and species variation on selenium and mercury molar ratios in Northeast Atlantic marine fish communities

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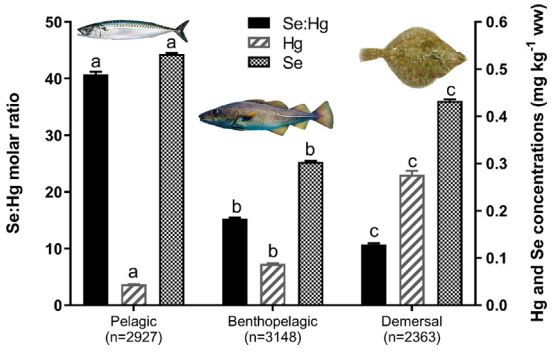
HIGHLIGHTS

- MeHg is the primary contaminant of concern for seafood consumption advisories.
- Selenium and mercury molar ratios were investigated in fish from the North East Atlantic Ocean.
- Hg concentrations in similar species were higher in coastal areas compared to offshore.
- In offshore areas mercury in fish increased from north to south.
- Two servings of tusk, blue ling, and Atlantic halibut exceeded the tolerable weekly intake of MeHg.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 21 July 2018
Received in revised form 26 October 2018
Accepted 29 October 2018
Available online 1 November 2018

Editor: Mae Sexauer Gustin

Keywords:
Mercury
Selenium
Marine
Norway
Seafood safety

ABSTRACT

Methylmercury (MeHg) is a potent neurotoxin that bioaccumulates in seafood. Co-occurrence of selenium (Se) may affect the bioavailability and toxicity of MeHg in organisms. Here we report the concentrations of total mercury (Hg) and Se in 17 teleost fish species (n = 8459) sampled during 2006–2015 from the North East Atlantic Ocean (NEAO) and evaluate species variation and effects of geography. Mean Hg concentration ranged from 0.04 mg kg^{-1} ww in Atlantic mackerel (*Scomber scombrus*) and blue whiting (*Micromesistius poutassou*) to 0.72 mg kg^{-1} ww in blue ling (*Molva dypterygia*). Se concentrations were less variable and ranged from 0.27 mg kg^{-1} ww in Atlantic cod (*Gadus morhua*) to 0.56 mg kg^{-1} ww in redfish (*Sebastes* spp.). The mean Se:Hg molar ratio ranged from 1.9 in blue ling to 43.3 in mackerel. Pelagic species had the lowest Hg concentrations and the highest Se:Hg ratios, whereas demersal species had the highest Hg concentrations and the lowest Se:Hg ratios. Se and Hg concentrations were positively correlated in 13 of the 17 species. Hg concentrations increased from the North to South in contrast to the Se:Hg molar ratio which exhibited the opposite trend. Fish from fjord and coastal areas had higher concentrations of Hg and lower Se:Hg molar ratios compared to fish sampled offshore. All species had average Se:Hg molar ratios >1 and Hg concentrations were largely below the EU maximum level of 0.5 mg kg^{-1} ww with few exceptions including the deep water species tusk (*Brosme brosme*) and blue ling sampled from fjord and coastal habitats. Our results show that two fillet servings of tusk, blue ling or Atlantic

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https://doi.org/10.1016/j.scitotenv.2018.10.405

halibut (*Hippoglossus hippoglossus*) exceeded the tolerable weekly intake of MeHg although the surplus Se may possibly ameliorate the toxic effects of MeHg. However, some individuals with selenium deficiencies may exhibit greater sensitivity to MeHg.

1. Introduction

Seafood is the main dietary source of methylmercury (MeHg) exposure for humans (Berry and Ralston, 2008; Hrenchuk et al., 2011; Rice et al., 2000) and MeHg is a primary contaminant of concern for seafood consumption advisories. During the past 150 years, human activities, mostly gold mining and coal combustion, have dramatically increased the concentrations of anthropogenic mercury (Hg) in the environment, although some recent studies have shown a decreasing trend in atmospheric Hg concentration (Zhang et al., 2016) and in Hg concentrations in fish from the North Atlantic Ocean (Cross et al., 2015; Lee et al., 2016). Hg is a natural element existing in all major compartments of the earth, and can easily be emitted to the atmosphere due to its volatility. Hence, Hg can travel long distances and be deposited from the atmosphere to remote areas (Fitzgerald et al., 1998) and therefore, all organisms are exposed to Hg to some degree (Lorey and Driscoll, 1999; Sonke et al., 2013; Streets et al., 2011).

Fish are mainly exposed to MeHg through their diet (Lindqvist et al., 1991), and factors such as trophic level, age and foraging depth may affect the MeHg concentrations in marine fish (Choy et al., 2009). Further, when species from extensive geographical areas are compared environmental factors that vary across broad spatial areas may influence the overall bioaccumulation regime of marine fish. Temperature is one of the most important environmental parameters that can directly affect MeHg bioaccumulation by increasing the rate of Hg elimination (Trudel and Rasmussen, 2006).

Compared with MeHg, inorganic Hg is assimilated less efficiently from ingested food (Dutton and Fisher, 2010) and the ratio of MeHg to total Hg typically increases with food web position (Lavoie et al., 2013). Heavy metals, as well as other contaminants present in seafood, can accumulate in the human body. High levels of seafood consumption may result in an elevated body burden of MeHg as has been reported for the Seychelles (Davidson et al., 1998), Faroe Islands (Grandjean et al., 1997) and French Guiana (Bourdineaud et al., 2008). Seafood consumption varies within and among European countries and MeHg exposure can be influenced by seafood species specific consumption rates (Agostoni et al., 2014). Hg contamination in seafood is regulated and in Europe the maximum level of Hg has been set by the European Union at 0.5 mg kg^{-1} ww for most of the marine fish species and at 1.0 mg kg^{-1} ww for large predatory species (EU Commission, 2006). The European Food Safety Authority (EFSA) has set the tolerable weekly intake (TWI) for MeHg at 1.3 μg kg^{-1} body weight.

Dietary intake of seafood, in particular fish with high MeHg concentrations may cause adverse effects in humans (Karagas et al., 2012; Oken et al., 2005). Both the Seychelles and the Faroe studies investigated the harmful effects of prenatal and postnatal MeHg exposure in 5.5 and 7 year old children. The Seychelles study found no significant negative effects of either prenatal or postnatal MeHg exposure, but the Faroe study found neurophysiological dysfunctions related to language, attention and memory at comparable MeHg exposure levels (Davidson et al., 1998; Grandjean et al., 1997). Although in Faroe Island, pilot whale is a popular seafood with Se:Hg molar ratio less than one (Julshamn et al., 1987; Ralston et al., 2016). However, the Seychelles Child Development Study was followed up by a cohort study where some delayed neurotoxic effects were found (Davidson et al., 2006). Recently the Seychelles investigators updated the ocean fish consumption effect on the same cohort at 17 years and found consistent positive nutritional effects from prenatal seafood exposure (Davidson et al., 2011). Additionally, other recent epidemiological studies, reported the beneficial effects of fish consumption on child neurodevelopmental outcomes (Avella-Garcia and Julvez, 2014; Golding et al., 2017; Hibbeln et al., 2007; Julvez et al., 2016; Llop et al., 2016).

The trade-off between beneficial nutrients and contaminants is still an issue of significant debate within the scientific community. However, several clinical studies have shown that health benefits from consuming a variety of seafood species in the recommended amounts outweigh the health risks associated with MeHg (Mozaffarian, 2009; Mozaffarian and Rimm, 2006; Mozaffarian et al., 2011). Fish is a high quality protein source and contains relatively high concentrations of long chain polyunsaturated fatty acids (LC n-3 PUFA), including eicosapentaenoic acid (EPA) and docosahexaenoic acid (DHA) with well documented health benefits (Mozaffarian and Rimm, 2006). These include improvement of blood lipid profiles, potential reduced risk of cardiovascular disease, lower potential for high blood pressure and stroke. A balanced seafood diet may also enhance eye and brain development (Dewailly et al., 2003; Ginsberg and Toal, 2009; Virtanen et al., 2008). Moreover, selenium (Se) and Hg co-exposure in seafood is a classic example of the trade-offs between nutrients and the bioavailability of toxic substances. The protective and antagonistic effects of Se against Hg toxicity have been addressed in several studies using Se:Hg molar ratios (Parizek and Ostadalova, 1967; Ralston et al., 2008; Siscar et al., 2014).

The protective effect of Se against Hg toxicity may be linked to different roles of Se including: 1) Hg has a higher affinity for Se than for the thiol group of amino acids (Berry and Ralston, 2008), 2) formation of stable MeHg-selenocysteine compounds may block Se bioavailability due to MeHg exposure and the antioxidant activities of selenoenzymes may be inhibited or lowered. However, available Se from the diet or body supply may compensate for the reduced Se in HgSe or MeHg-selenocysteine and preserve the Se dependent enzyme function in the central nervous system (Peterson et al., 2009; Spiller, 2018), 3) enhance demethylation of MeHg to the inorganic form and redistribution of Hg to less sensitive organs (Spiller, 2018) and 4) a reduction in the Hg uptake in the gastrointestinal tract (Spiller, 2018).

The molar ratio of Se:Hg is suggested as an important human risk factor and a ratio above 1 may provide protection against MeHg toxicity in humans and fish (Burger and Gochfeld, 2012; Peterson et al., 2009; Ralston, 2008). However, due to the biochemical interactions of Se with other components, it is difficult to determine the actual effectiveness of Se amelioration on Hg toxicity in seafood and consumers. The underlying mechanisms of Hg-Se interactions are not fully understood and practical information on the protective ratio is lacking. Still, the Se:Hg molar ratio may provide a relatively more accurate, and physiologically relevant, indicator for MeHg toxicity in the body than MeHg concentrations alone. Recently, a Health Benefit Value of Se (HBV_{Se}) has been suggested as an index to better estimate the health risk associated with Hg reflecting the biochemical mechanisms of MeHg toxicity and the interactions with Se. Thus, fish with positive HBV_{Se} values would provide surplus Se while negative values would indicate a relative deficiency in Se (Ralston et al., 2016).

Here we evaluate variation in Hg and Se concentrations and Se:Hg molar ratios across a latitudinal gradient in NEAO marine fish communities to assess species differences and the effects of geography on Se and Hg dynamics and exposure. We present Hg and Se data from several commercially important fish species in NEAO collected during 2006–2015. To our knowledge, this is the first extensive study analyzing the NEAO marine fish community for Hg and Se from a large sampling area encompassing Arctic, subarctic and temperate zones of the NEAO. Data from this investigation were used to test the following hypotheses

and a priori predictions on length normalized fish concentrations: 1) individuals of the same species inhabiting coastal areas would have greater concentrations of Hg compared to offshore environments, 2) fish species from geographical areas in the southern region of our study area would have greater concentrations of Hg compared to more northerly sampling sites, 3) demersal fish species would have greater concentrations of Hg compared to benthopelagic and pelagic species and 4) concentrations of Hg and Se in fish fillets would be positively correlated across species. We integrate these hypotheses and incorporate them into our interpretations of Se:Hg molar ratios using geography, species variation and coastal vs. offshore habitat comparisons as potential drivers. Additionally, we also conducted an exposure assessment of MeHg based on the European consumption rate of fish species from the NEAO and used TWI metrics established by EFSA.

2. Materials and methods

2.1. Study area and sample collection

Fish samples (n = 8459) comprising 17 commercially important marine teleost species including Atlantic cod (*Gadus morhua*), Atlantic halibut (*Hippoglossus hippoglossus*), Atlantic herring (*Clupea harengus*), Atlantic mackerel (*Scomber scombrus*), blue ling (*Molva dypterygia*), blue whiting (*Micromesistius poutassou*), common ling (*Molva molva*), European eel (*Anguilla anguilla*), European hake (*Merluccius merluccius*), Greenland halibut (*Reinhardtius hippoglossoides*), haddock (*Melanogrammus aeglefinus*), plaice (*Pleuronectes platessa*), pollack (*Pollachius pollachius*), redfish (*Sebastes* spp.), saithe (*Pollachius virens*), tusk (*Brosme brosme*) and wolffish (*Anarhichas* spp.) were collected from Norwegian fisheries areas in NEAO (Table S1; Fig. 1). Fish were sampled using different sampling gears including long line, gill net, purse seine and pelagic trawl between 2006 and 2015 by the authorized Norwegian reference fleet research vessels of the Institute of Marine Research (IMR), Bergen, Norway or local professional fishermen along the coastal areas of Norway. The Hg concentrations of a few fish species including cod, herring and Greenland halibut have been reported previously but without discussion of the selenium content (Frantzen et al., 2015; Julshamn et al., 2013a; Julshamn et al., 2013b; Julshamn et al., 2011; Julshamn et al., 2006). Fish were caught from different parts of NEAO covering most of the important fishing areas (from 22.9°W to 41.6°E and 50.2°N to 75.6°N). The study area is delineated by the Svalbard Islands in the north, Yuzhny Island in the east, Strait of Dover in

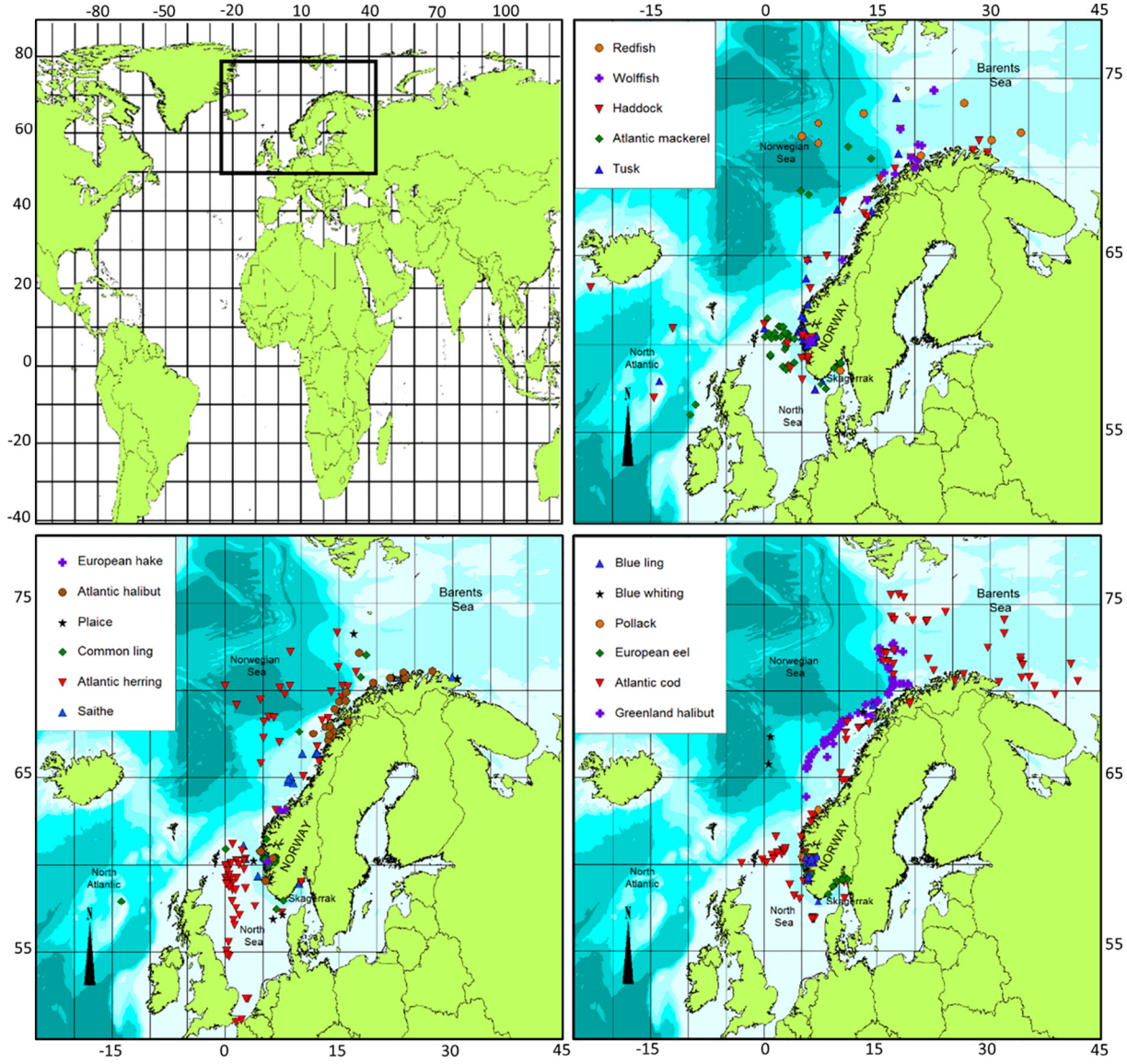

Fig. 1. Sampling sites of fish species analyzed in this study from NEAO collected during 2006–2015. The position of the study area in the world map is highlighted on the top left map in black rectangle. To avoid overlap, different species are showed in three maps.

the south and Iceland in the west, representing a major part of the NEAO (Fig. 1). This large area was divided into 2 primary habitats, 1) offshore ecosystems and 2) fjords and coastal areas. To ease the geographical comparison, the offshore area was divided into five smaller areas including the Barents Sea (BS), the Norwegian Sea (NO), the North Atlantic (NA), the North Sea (NS) and Skagerrak (SK), an arm of the NS. The borders between areas and the study area are described in more detail in the supplementary materials.

2.2. Sample preparation

All fish were shipped whole and frozen to the Institute of Marine Research where individual fish were registered in the Laboratory Information Management System (LIMS) and weight and length were recorded. Hg and Se were analyzed in fillet, since fish fillet is an important storage compartment for MeHg and the main tissue consumed by humans. One side fillet (bone and skin free) was homogenized except for 1) Greenland halibut for which the fillet sample was taken from the upper side of the fish with a cut from the middle of the fish towards the tail (Julshamn et al., 2006) and 2) Atlantic halibut for which the fillet sample was taken from a special cut of the upper part of the pectoral area (i.e., B cut area – see Nortvedt and Tuene (1998) for more details). A subsample was freeze dried and dry matter was recorded as g per 100 g and then samples were ground to a powder before analytical measurements. In the available data there were some composite samples that were excluded from the data set except for common ling, eel, Greenland halibut and tusk (composite samples were 113 of 1968) in order to increase the sampling points and cover larger geographical distribution of those species. The differences in mean and standard error of Se:Hg molar ratio, Se and Hg concentrations (with and without composite samples) for these four species are presented in the Supplementary materials section (Table S2).

2.3. Analytical methods

The concentration of elements was determined using inductively coupled plasma-mass spectrometry (ICP-MS) following microwave digestion. First, weighed samples were digested using concentrated (65%) HNO_3 and 30% H_2O_2 in a microwave oven (Milestone Microwave digestion system MLS-1200 MEGA Microwave Digestion Rotor - MDR 300/10). Hg and Se were determined using quantitative ICP-MS (Agilent 7500 with collision cell and ICP-ChemStation software). A standard curve was used to determine the concentration of Hg and Se. Germanium (Ge), thulium (Tm) and rhodium (Rh) were used either individually or in combination as an internal standard, and gold was added to stabilize the Hg signals. The method is a Nordic and European standard for these two elements (CEN, 2009; NMKL, 2007) and is described in detail by (Julshamn et al., 2007). MeHg was measured using an isotope dilution method and gas chromatography coupled with ICP-MS and details of this method are presented in (Valdersnes et al., 2012).

2.4. Quality assurance

The ICP-MS method is accredited according to ISO 17025 for Hg and Se. The accuracy and precision of the method has been tested by analyzing certified reference materials and the recoveries of both Hg and Se ranged from 80% to 120% for the whole period of analysis (2006–2015). Certified reference materials (CRM) 1566 (oyster tissue) from the National Institute of Standards and Technology (Gaithersburg, USA) and lobster hepatopancreas (TORT-2, TORT-3) from the National Research Council (Ottawa, Canada) were used for measurement quality control by including them in each sample run.

Reproducibility (% RSD) from five day analyses of reference materials showed a variation in the results <10% on analysis values above limit of quantification (LOQ) of the method. The LOQ of the method for Hg and Se were 0.03 and 0.1 mg kg^{-1} dry weight from 2006 until 2010 when the laboratory instrumentation was changed and LOQs were reduced to 0.005 and 0.01 mg kg^{-1} dry weight for Hg and Se, respectively.

The internal method reproducibility for MeHg (RSD) was between 1 and 12% and the *Z*-score for different CRM's was better than |1.5| and the method was validated in different seafood matrices (Valdersnes et al., 2012).

2.5. Mercury in sediment

Hg concentrations in sediment samples collected from NEAO between 62.3 and 76.6°N latitude and 4.3 and 37.2°E longitude have been analyzed in the MAREANO project and was included to determine the spatial distribution of seabed Hg pollution. This data set is accessible online from the MAREANO project website (www.mareano.no downloaded on 07.02.2018 for this study). The sediment samples were collected mostly with a sediment multi-corer and in some cases with Van Veen grab or box corer during 2003–2015. Hg concentrations were measured using Cold Vapor Atomic Absorption Spectrometry (CV-AAS) in freeze-dried samples.

2.6. Statistical analysis

Prior to all correlation and analysis of variance (ANOVA) or analysis of covariance (ANCOVA) tests, outliers were removed from the data using Grubbs test. Outliers were found in 8 of 17 species and in total 21 of 8459 measurements (<1%) were removed as outliers. In order to improve the assumption of normal distribution, all statistical analyses were conducted on log-transformed data (Zar, 2010).

Geographical variation within each species (different offshore areas and offshore versus fjords and coast) were investigated using ANCOVA followed by Tukey unequal sample HSD post-hoc test, with length as a covariate for each species. To show the North-South gradient, least squares means adjusted for length, derived from Generalized Linear Model (GLM) and ANCOVA models, were used. To compare the Se:Hg ratio, Hg and Se concentrations in fish from different habitats, ANOVA was conducted followed by Tukey unequal sample HSD post-hoc test to determine the binary differences between groups. Linear regression tests were used to examine the relationship between Se:Hg molar ratio, Hg and Se concentrations and fish length. Pearson correlation (r) tests were used to examine the relationship between Hg concentrations and latitude of sampling as well as sediment Hg concentration and geographical location expressed as latitude and longitude. Statistical significance was accepted at $P < 0.05$ (Zar, 2010). All statistical analyses were performed using STATISTICA 13 (Statsoft Inc., Tulsa, USA) or GraphPad Prism 7.02 (GraphPad software Inc., San Diego, CA, USA).

2.7. Se:Hg molar ratio calculation

The Se:Hg molar ratio was calculated for all fish individuals. First, the concentration of Se and Hg (mg kg^{-1} ww) were divided by the molar masses 78.96 and 200.59 g mol^{-1} respectively and then the Se:Hg molar ratio was calculated using the following formula:

$$\text{Se : Hg molar ratio} = \left(\text{mmol Se kg}^{-1}\text{ww}\right) / \left(\text{mmol Hg kg}^{-1}\text{ ww}\right)$$

All Se:Hg molar ratio means reported in this study were averaged from specimen values for each species, area and habitat.

2.8. Selenium health benefit value

Selenium health benefit value (HBV_{Se}) has been suggested as an evaluation index showing the Se amount provided in fish after sequestration of

Hg and was calculated using the following formula (Ralston et al., 2016):

$$HBV_{Se} = \frac{Se - Hg}{Se} \times (Se + Hg)$$

Se = Selenium content in molar concentration.
Hg = Mercury content in molar concentration.

3) The amount of fish that can be consumed safely per week was calculated using the following formula:

$$A = \frac{W \times I}{C}$$

A = the amount of fish that can be safely consumed per week (g).
W = average body weight of consumer (70 kg).
I = TWI of MeHg (1.3 μg kg^{-1} body weight).
C = MeHg concentration in fish fillet (mg kg^{-1} ww).

3. Results and discussion

3.1. Inter- and intraspecies variation in Se:Hg molar ratios, Hg and Se concentrations

The mean Hg concentrations ranged from 0.04 to 0.72 mg kg^{-1} ww with the lowest concentration in mackerel and blue whiting and the highest in blue ling (Table 1). Most blue ling were sampled from fjords and coastal areas (55 out of 79) where many sampled individuals had high concentrations of Hg. However, the Hg concentrations, both for arithmetic and length adjusted means, in 12 samples of blue ling from the Norwegian Sea were also higher than the other species from the same area (Table 2; Fig. 2B). Our data show that the observed high concentrations of Hg in blue ling was independent of geography and possibly driven by trophic position or energy sources. Based on average Hg concentrations, we grouped sampled fish into three categories: 1) Highly contaminated species with mean Hg concentration higher than 0.5 mg kg^{-1} ww, i.e. only blue ling. 2) moderately contaminated species with mean Hg concentration between 0.3 and 0.5 mg kg^{-1} ww including Atlantic halibut and tusk, and 3) low contaminated species with mean Hg concentration lower than 0.3 mg kg^{-1} ww, including the rest of species (Table 1).

The mean Se concentrations ranged from 0.27 mg kg^{-1} ww in cod to 0.56 mg kg^{-1} ww in redfish. Hg concentrations exhibited higher variation (~18 fold between the lowest and the highest) than Se concentrations (~2 fold). Similar patterns of variation for Hg and Se have been reported in marine fish from other areas (Burger and Gochfeld, 2012; Polak-Juszczak, 2015). The difference in variation is likely a result of Se being an essential trace element with a regulated pattern of uptake and excretion (Thiry et al., 2012). The range between essential, beneficial and toxic concentrations of Se for living organisms is narrow and in general Se concentrations often tend to show lower overall variability compared to Hg.

Blue ling, tusk and hake had the lowest mean Se:Hg molar ratios of 1.9, 5.1 and 5.4, respectively, whereas mackerel had the highest Se:Hg ratio followed by blue whiting and herring (43.3, 41.6 and 39.3 respectively, Table 1). Variation in Hg concentrations caused most of the variation in Se:Hg ratio for most species, although species such as wolffish, redfish and Atlantic halibut had higher Se:Hg molar ratios as a result of higher Se concentrations (Table 1).

All species showed significant geographical variation ($P < 0.05$). Additionally, individuals from the same species sampled from different offshore areas were also significantly different for Se:Hg molar ratio, and

Table 1
Mean Se:Hg molar ratio, Hg and Se concentrations (mg kg^{-1} ww), HBV_{Se}, Hg intake as percentage of TWI (TWI %), consumption limit per week, landed catch from Norwegian fisheries and percentage of total catch (% Catch) for fish species from NEAO. TWI % and HBV_{Se} were calculated from mean values. Species are sorted according to Hg concentrations. Data are from NEAO sampled during 2006–2015. Colors represent low risk (green), moderate risk (yellow) and high risk (red).

Species	N	Se:Hg molar ratio	Hg	Se	HBVSe	TWI % (2 servings)	TWI % (4 servings)	Consumption limit per week (g)	Landed catch from Norwegian fisheries (in tons, 2017)*	% catch
Blue whiting	75	41.6	0.04	0.48	6.11	15	30	2241	399210	20.6
Atlantic mackerel	1042	43.3	0.04	0.55	7.00	16	32	2114	221588	11.4
Atlantic herring	1810	39.3	0.05	0.52	6.60	17	34	2019	526167	27.2
Plaice	198	23.2	0.06	0.38	4.76	23	45	1510	848	0.04
Haddock	245	17.4	0.07	0.32	3.97	26	52	1317	113776	5.9
Saithe	439	16.9	0.07	0.29	3.59	26	53	1295	177196	9.2
Atlantic cod	2105	16.4	0.08	0.27	3.44	28	56	1208	412441	21.3
Wolffish	89	21.3	0.09	0.44	5.57	35	69	983	6451	0.3
European eel	185	11.2	0.11	0.30	3.73	40	80	851	12	0.001
Redfish	185	22.9	0.13	0.56	7.05	48	96	710	22582	1.2
Pollack	58	8.1	0.14	0.38	4.65	52	104	652	2028	0.1
Greenland halibut	546	10.3	0.14	0.42	5.23	54	108	631	16687	0.9
European hake	92	5.4	0.19	0.34	4.12	72	145	469	5307	0.3
Common ling	294	7.7	0.22	0.41	5.00	82	164	415	18481	1.0
Atlantic halibut	53	9.7	0.38	0.48	5.45	142	283	240	2648	0.1
Tusk	943	5.1	0.44	0.49	5.46	163	327	208	10191	0.5
Blue ling	79	1.9	0.72	0.38	2.09	270	540	126	244	0.01
All species#	8438	17.7	0.17	0.41	5.08	65	130	521	1935857	100

#Means of all species were averaged for Se:Hg molar ratio, Hg and Se and TWI % and safe consumption limit were calculated based on mean of all species.
*Numbers obtained from www.fiskeridir.no.

Table 2
Mean, standard error (SE) and quartile range for Se:Hg molar ratio, Hg and Se concentrations and length of fish species from different areas of NEAO sampled during 2006–2015. Since some species had missing length data, N is presented separately for fish with and without length data.

Species	Area	N[a]	N[b]	Se:Hg molar ratio				Hg (mg kg^{-1})				Se (mg kg^{-1})				Length (cm)			
				Mean	SE	Q25	Q75	Mean	SE	Q25	Q75	Mean	SE	Q25	Q75	Mean	SE	Q25	Q75
Atlantic cod*	BS	507	507	24.7	0.6	15.2	30.5	0.03	0.001	0.02	0.04	0.25	0.002	0.23	0.27	64.8	0.6	55	73
	NO	472	471	21.1	0.6	11.6	25.4	0.04	0.001	0.02	0.05	0.23	0.002	0.21	0.25	65.6	0.5	57	73
	NA	25	25	8.9	0.5	7.1	9.8	0.09	0.01	0.07	0.10	0.28	0.01	0.26	0.28	60.8	0.7	59	62
	NS	490	490	9.4	0.3	5.2	11.7	0.11	0.003	0.06	0.14	0.28	0.002	0.25	0.31	64.8	0.8	50	80
	SK	23	23	8.0	0.8	4.6	10.7	0.16	0.02	0.09	0.19	0.38	0.01	0.34	0.39	53.4	2.3	47	59
	FC	588	588	11.9	0.3	5.8	15.7	0.11	0.004	0.05	0.14	0.31	0.003	0.27	0.36	58.7	0.5	50	67
Atlantic halibut	NO	13	12	15.9	2.6	5.4	20.3	0.20	0.07	0.05	0.24	0.47	0.03	0.40	0.47	96.9	17.6	65	97
	FC	40	9	7.6	1.5	1.8	8.3	0.44	0.06	0.14	0.76	0.48	0.02	0.39	0.55	93.9	11.0	78	93
Atlantic herring*	NO	798	798	51.1	0.9	31.8	66.1	0.04	0.001	0.02	0.05	0.61	0.01	0.51	0.69	31.4	0.1	30	33
	NS	963	960	30.7	0.5	18.3	39.8	0.05	0.001	0.03	0.06	0.46	0.003	0.38	0.51	27.2	0.1	26	30
	FC	49	49	17.4	1.1	11.6	22.2	0.06	0.003	0.04	0.08	0.38	0.01	0.35	0.40	28.1	0.2	27	30
Atlantic mackerel*	NO	77	77	36.8	1.2	31.0	40.6	0.04	0.001	0.04	0.05	0.60	0.01	0.55	0.64	38.4	0.2	38	40
	NA	134	134	29.8	0.9	22.2	36.2	0.06	0.001	0.04	0.07	0.61	0.01	0.53	0.67	35.4	0.2	33	37
	NS	647	647	49.3	1.0	31.9	61.2	0.03	0.001	0.02	0.04	0.54	0.004	0.47	0.60	31.9	0.2	28	36
	SK	184	184	34.7	2.0	14.0	48.6	0.07	0.004	0.03	0.09	0.54	0.01	0.46	0.61	32.8	0.4	28	37
Blue ling*	NO	12	12	3.2	0.3	2.8	3.4	0.27	0.02	0.22	0.32	0.31	0.01	0.29	0.34	93.9	3.9	85	101
	SK	12	12	1.5	0.1	1.3	1.6	0.52	0.03	0.44	0.56	0.29	0.01	0.28	0.30	110.5	1.5	107	113
	FC	55	53	1.8	0.1	1.0	2.3	0.87	0.08	0.49	1.13	0.41	0.01	0.34	0.50	94.5	1.7	87	101
Blue whiting	NO	75	50	41.6	2.0	23.6	56.3	0.04	0.003	0.02	0.07	0.48	0.01	0.41	0.54	22.0	0.4	19	25
Common ling*	NO	75	75	10.1	0.5	7.0	12.1	0.12	0.01	0.08	0.15	0.38	0.01	0.36	0.42	87.9	1.2	81	94
	NA	23	22	5.9	0.4	4.2	7.1	0.25	0.02	0.16	0.35	0.50	0.02	0.44	0.54	76.4	2.1	69	82
	NS	132	106	6.5	0.3	3.8	8.4	0.20	0.01	0.11	0.26	0.39	0.01	0.34	0.42	82.2	1.8	69	93
	FC	64	59	8.0	0.8	2.5	13.1	0.37	0.05	0.08	0.50	0.47	0.01	0.41	0.51	75.8	2.0	68	84
European eel	FC	185	88	11.2	0.8	5.1	13.2	0.11	0.01	0.05	0.15	0.30	0.01	0.18	0.38	58.1	1.2	51	67
European hake	FC	92	92	5.4	0.3	3.9	6.0	0.19	0.01	0.13	0.24	0.34	0.004	0.32	0.37	75.0	1.2	67	81
Greenland halibut	NO	546	525	10.3	0.3	5.7	12.0	0.14	0.004	0.07	0.19	0.42	0.01	0.29	0.47	62.3	0.4	57	68
Haddock*	BS	12	12	17.3	1.6	13.4	19.8	0.04	0.003	0.03	0.04	0.22	0.01	0.20	0.24	56.0	0.8	54	58
	NO	65	65	19.7	1.0	14.2	23.3	0.05	0.004	0.03	0.06	0.29	0.01	0.25	0.32	55.0	0.5	53	58
	NA	24	24	14.0	1.9	6.1	22.9	0.10	0.01	0.05	0.15	0.38	0.02	0.33	0.43	54.8	2.1	49	65
	NS	24	24	6.4	0.7	4.6	6.8	0.15	0.02	0.11	0.18	0.32	0.01	0.27	0.36	53.3	1.0	51	57
	FC	120	120	19.0	1.1	10.7	23.6	0.06	0.004	0.03	0.07	0.32	0.01	0.27	0.38	50.9	0.7	46	56
Plaice*	BS	25	25	29.4	3.1	19.2	36.3	0.06	0.01	0.03	0.05	0.42	0.03	0.31	0.45	42.0	1.0	39	45
	NO	49	24	30.9	2.1	19.1	41.0	0.04	0.004	0.02	0.04	0.31	0.01	0.26	0.35	41.3	0.7	39	43
	NS	124	123	18.9	0.7	13.2	24.1	0.07	0.005	0.04	0.09	0.39	0.01	0.30	0.46	29.3	0.5	26	32
Pollack	FC	58	57	8.1	0.5	5.4	9.9	0.14	0.01	0.10	0.18	0.38	0.01	0.34	0.40	56.5	0.8	53	61
Redfish*	BS	56	56	32.2	2.7	17.2	45.2	0.06	0.01	0.03	0.08	0.54	0.01	0.45	0.62	41.2	0.5	39	44
	NO	123	100	19.7	1.6	7.6	22.5	0.13	0.01	0.06	0.18	0.57	0.01	0.51	0.64	34.4	0.2	33	36
	SK	6	6	2.5	0.6	1.4	2.6	0.67	0.12	0.47	0.94	0.54	0.05	0.46	0.62	29.2	1.2	27	30
Saithe*	BS	48	25	37.5	1.6	30.2	43.7	0.02	0.001	0.02	0.03	0.30	0.01	0.28	0.32	41.9	0.6	40	44
	NO	122	97	11.3	0.7	5.7	14.7	0.11	0.01	0.05	0.12	0.28	0.003	0.26	0.30	60.7	1.2	52	68
	NS	75	50	11.0	0.5	8.6	13.6	0.07	0.004	0.05	0.08	0.26	0.003	0.24	0.28	47.9	0.6	45	51
	FC	194	194	17.6	0.6	11.0	23.1	0.06	0.003	0.03	0.07	0.30	0.004	0.26	0.32	46.8	0.9	37	54
Tusk*	NO	124	124	9.2	0.4	6.8	10.4	0.14	0.01	0.10	0.16	0.42	0.004	0.39	0.44	49.4	0.6	45	54
	NA	25	25	6.5	0.7	3.8	8.4	0.23	0.03	0.12	0.29	0.44	0.01	0.41	0.45	57.0	1.5	50	63
	NS	465	465	5.1	0.1	3.6	6.3	0.27	0.01	0.19	0.34	0.47	0.003	0.42	0.52	49.2	0.4	43	54
	SK	45	45	3.6	0.2	2.6	4.1	0.44	0.03	0.29	0.55	0.53	0.02	0.45	0.58	61.0	1.1	56	66
	FC	284	272	3.4	0.2	1.4	4.8	0.85	0.05	0.24	1.20	0.56	0.01	0.43	0.66	63.0	0.8	54	72
Wolffish	BS	36	36	33.5	4.7	11.6	49.7	0.05	0.01	0.02	0.06	0.41	0.04	0.23	0.53	66.8	1.9	58	77
	NO	51	42	23.4	3.9	6.9	21.6	0.12	0.01	0.06	0.14	0.79	0.13	0.35	0.59	74.3	3.1	60	90
	FC	14	6	29.9	10.2	8.2	38.1	0.13	0.02	0.09	0.19	1.22	0.31	0.31	2.50	66.3	2.4	63	70
All species#	BS	684	661	26.7	0.6	15.9	31.8	0.04	0.001	0.02	0.04	0.29	0.004	0.23	0.30	61.1	0.6	49	70
	NO	2594	2473	27.6	0.5	9.4	39.4	0.08	0.002	0.03	0.10	0.45	0.004	0.28	0.57	50.5	0.4	33	64
	NA	231	231	21.0	0.9	8.3	29.5	0.10	0.01	0.05	0.11	0.52	0.01	0.42	0.61	46.7	1.0	34	60
	NS	2920	2865	24.9	0.47.3	7.3	36.1	0.10	0.002	0.03	0.13	0.44	0.002	0.34	0.52	41.0	0.3	28	49
	SK	270	270	25.0	1.6	5.1	40.3	0.17	0.01	0.03	0.24	0.52	0.01	0.44	0.59	42.6	1.2	29	51
	FC	1739	1591	10.7	0.2	4.0	14.7	0.27	0.01	0.05	0.22	0.37	0.003	0.29	0.42	59.4	0.4	48	69

N[a] number of samples with Se:Hg molar ratio, Hg and Se concentrations data.
N[b] number of samples with length data.
BS: Barents Sea; NO: Norwegian Sea; NA: North Atlantic; NS: North Sea; SK: Skagerrak; FC: fjords and coastal areas.
* Species with significant differences in length between areas (ANOVA-test; $P < 0.05$).
\# Means of individuals.

Hg and Se concentration with the exception of Se concentrations in blue ling and wolffish (ANCOVA; $P < 0.05$: Fig. 2, Table S3). The Se:Hg molar ratio varied between 51.1 in herring from the Norwegian Sea and 1.5 in blue ling from Skagerrak (~34 fold). The mean Hg concentrations varied from 0.02 mg kg^{-1} ww in saithe from the Barents Sea to 0.87 mg kg^{-1} ww in blue ling from fjords and coastal areas (~44 fold). The mean Se concentration varied between 0.22 in haddock from the Barents Sea to 1.22 in wolffish from fjords and coastal area (~6 fold: Table 2).

The highest variation for each species in terms of difference between lowest and highest Se:Hg molar ratio between areas was found in redfish (~12.9 fold) followed by saithe (~3.4 fold) and cod (~3.1 fold, Table 2). Also, Hg concentrations in redfish had the greatest differences between areas (~11.2 fold), followed by tusk (~6.1 fold), saithe (~5.5 fold) and cod (~5.3 fold, Table 2). Redfish also had the highest Hg concentrations among all species from offshore areas (0.67 mg kg^{-1} ww from Skagerrak, Table 2).

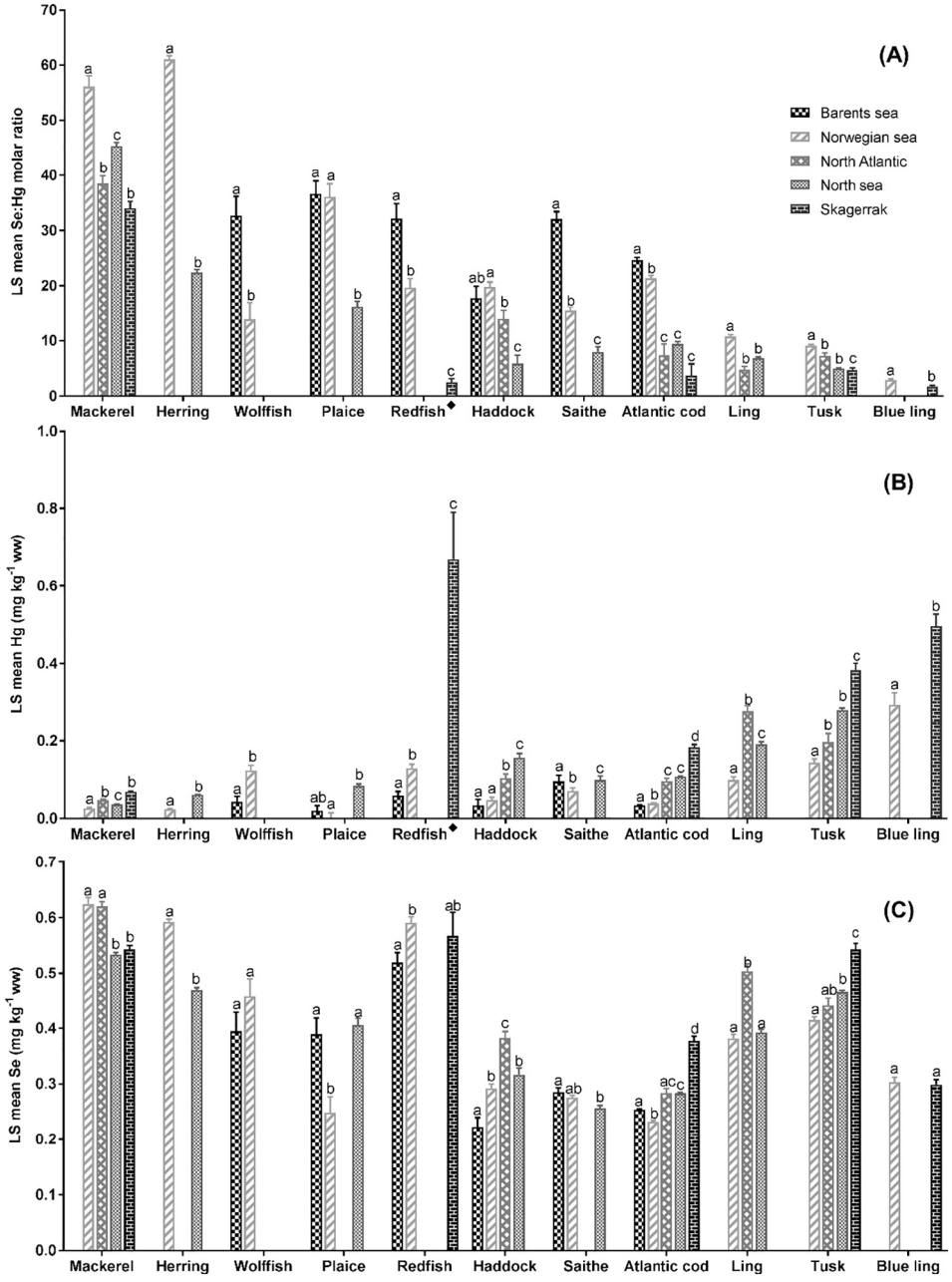

Fig. 2. Least squares mean (length adjusted) of Se:Hg molar ratio, Hg and Se concentrations in fish species from different offshore areas of NEAO sampled between 2006 and 2015. Areas are sorted from north to south. Error bars represent +1 standard error. Post hoc comparison (ANCOVA; $P < 0.05$) between areas are shown by letters above error bars. For redfish, Se:Hg molar ratio and Hg results (♦) are presented as arithmetic means for better graphical illustration since LS means was negative for some areas due to large variation in length of fish between areas and the area with largest fish was lowest in Hg.

Burger and Gochfeld (2012) studied saltwater teleost fish species from the North West Atlantic Ocean (NWAO) and found mean Hg concentrations ranging from 0.01 to 0.52 mg kg^{-1} (Fig. 3), whereas the measured Hg concentrations in species from NEAO, in this study, varied between 0.04 and 0.72 mg kg^{-1}. The Se concentrations in fish from NWAO (0.18–0.48 mg kg^{-1} ww) were lower compared with NEAO (0.27 to 0.56 mg kg^{-1} ww). Burger and Gochfeld (2012) found a mean Se:Hg molar ratio < 5 in fish from NWAO for 11 of 19 species, whereas blue ling was the sole species with a mean Se:Hg molar ratio < 5 in our study, demonstrating that fish with similar Hg concentrations from NWAO had a lower Se:Hg molar ratio (Fig. 3). These results and comparisons suggest that for fish at the same Hg concentration, variations in the Se:Hg molar ratio may also become pronounced when widespread species distributions are considered and evaluated.

3.2. Se and Hg in fish from different NEAO habitats

In order to assess the impact of habitat on Hg concentration, different species were grouped into three major habitat use categories as either pelagic (3 species), benthopelagic (4 species) or demersal (10 species, Table 3). The mean Se:Hg molar ratio, Hg and Se concentrations were significantly different between habitats in all binary comparisons (Se:Hg molar ratio: $F(2, 8435) = 3243.2$, $P < 0.0001$; Hg concentration: $F(2, 8435) = 1846.5$, $P < 0.0001$; Se concentration: $F(2, 8435) = 3083.7$, $P < 0.0001$).

Hg concentrations were observed in the following order for each habitat category: demersal > benthopelagic > pelagic, and demersal fish species on average (0.28 mg kg^{-1} ww) had about three times higher Hg concentrations than benthopelagic species (0.09 mg kg^{-1}

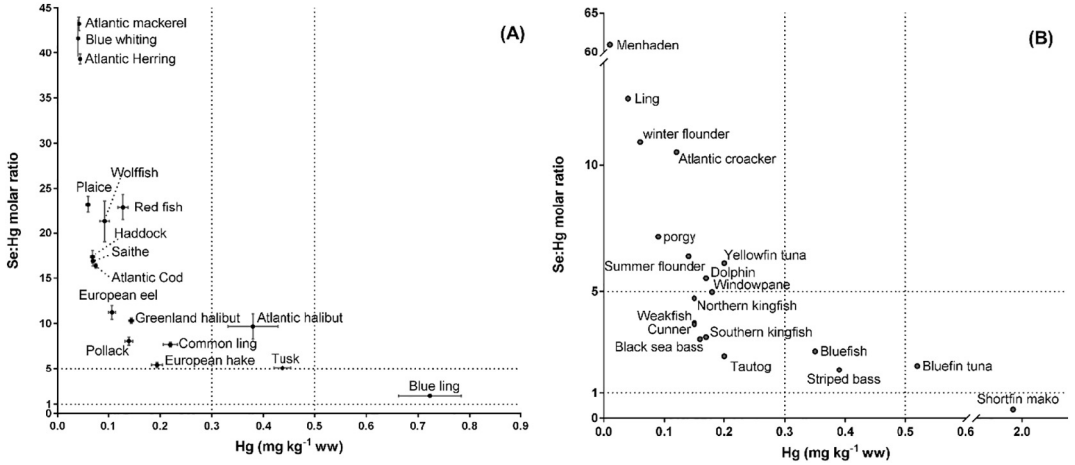

Fig. 3. Relationship between mean Se:Hg and mean Hg in fish from NEAO sampled during 2006–2015 (A) and in fish from NWAO redrawn from Burger and Gochfeld, 2012 (B). The vertical lines are placed at 0.5 and 0.3 mg kg^{-1} ww, the EU and the US maximum levels for Hg in muscle meat of most fish species. The horizontal lines are placed at 1, where below this value Hg exceeds the Se in mole and the suggested safe ratio, and 5 for comparative purposes. Error bars represent ±1SE for both axes.

ww) and more than six times higher than pelagic species (0.04 mg kg^{-1} ww). The Se:Hg molar ratio followed the opposite order of Hg concentration. Pelagic species had the highest ratio (40.8), >2.5 times higher than benthopelagic (15.3) and >3.5 times higher than demersal species (10.7). The Se concentration was highest in the pelagic group (0.53 mg kg^{-1} ww), followed by the demersal (0.43 mg kg^{-1} ww) and benthopelagic group (0.30 mg kg^{-1} ww) and the difference between the highest and lowest groups was <2-fold (Fig. 4). Saei-Dehkordi et al. (2010) measured Hg concentrations in 15 fish species from the Persian Gulf and reported the highest concentrations in demersal species (similar to this study) and lowest in benthopelagic, while pelagic species were intermediate. The pelagic group in the Persian Gulf included high trophic level and predatory species such as Spanish mackerel (*Scomberomorus commerson*), barracuda (*Sphyraena jello*), cobia (*Rachycentron canadum*) and long tail tuna (*Thunnus tonggol*), whereas pelagic species in this study mostly comprised low trophic level species such as mackerel, blue whiting and herring. Thus, variation observed between different habitats may likely be more related to the differences in life histories and trophic position of fish from different habitats than from a habitat effect alone. In general, food sources, and hence contaminant concentrations, vary in different marine habitats and geographical areas. In NEAO, pelagic species are mostly zooplankton feeders and at the lowest trophic level among fish species (Bachiller et al., 2016), while demersal species mostly include more long lived and deep water dwelling predatory species that feed on other fish species with some degree of cannibalism (Jaworski and Ragnarsson, 2006). Although some demersal species like plaice feeds on benthic invertebrates and thus belongs to a lower trophic position (McMeans et al., 2010).

The effect of forage depth was not investigated in this study, but species such as tusk, common ling, blue ling and Greenland halibut, having some of the highest Hg concentrations (Table 1) all inhabit deep sea environments (>150 m). The effect of forage depth on Hg accumulation in marine fish from different ecosystems has been reported in previous studies (Choy et al., 2009; Madigan et al., 2018; Magalhães et al., 2007). These studies showed that Hg concentrations were higher in species and individuals feeding at greater depths.

3.3. Se and Hg antagonism in fish species from NEAO

Mean Se and Hg concentrations showed weak to moderate positive correlation (Pearson r range = 0.24 to 0.70) in most species (13 of 17 species), while no significant correlation was observed in mackerel, herring, saithe or pollack (Table S4, Fig. S1). The strongest correlation was found in blue whiting, caused by two separate batches of samples

Table 3
Percent of specimens with Se:Hg molar ratio of 0–1, 1–5 or > 5 and Hg concentration (mg kg^{-1} ww) ≥0.3 or ≥0.5. Habitat data are collected from www.imr.no and www.fishbase.com. The species are sorted based on Hg concentration. Data are from NEAO sampled during 2006–2015.

Species	N	Habitat	Se:Hg molar ratio			Hg concentrations (mg kg^{-1} ww)	
			0–1	1–5	>5	Hg ≥ 0.3	Hg ≥ 0.5
Blue whiting	75	Pelagic	0.0	0.0	100.0	0.0	0.0
Atlantic mackerel	1042	Pelagic	0.0	0.6	99.4	0.0	0.0
Atlantic herring	1810	Pelagic	0.0	0.1	99.9	0.1	0.0
Plaice	198	Demersal	0.0	2.0	98.0	0.5	0.0
Haddock	245	Demersal	0.0	6.5	93.5	1.2	0.0
Saithe	439	Benthopelagic	0.2	9.8	90.0	0.9	0.7
Atlantic cod	2105	Benthopelagic	0.0	11.4	88.6	1.8	0.1
Wolffish	89	Demersal	0.0	4.5	95.5	3.4	1.1
European eel	185	Demersal	0.5	22.7	76.8	4.3	0.5
Redfish	185	Demersal	0.0	7.0	93.0	7.6	2.2
Pollack	58	Benthopelagic	0.0	19.0	81.0	1.7	0.0
Greenland halibut	546	Benthopelagic	0.0	17.6	82.4	8.4	1.1
European hake	92	Demersal	0.0	46.7	53.3	9.8	2.2
Common ling	294	Demersal	1.0	31.6	67.3	19.4	7.5
Atlantic halibut	53	Demersal	3.8	49.1	47.2	45.3	34.0
Tusk	943	Demersal	3.9	52.8	43.3	42.1	20.1
Blue ling	79	Demersal	17.7	81.0	1.3	81.0	59.5
All species	8438		0.7	14.2	85.1	8.0	3.5

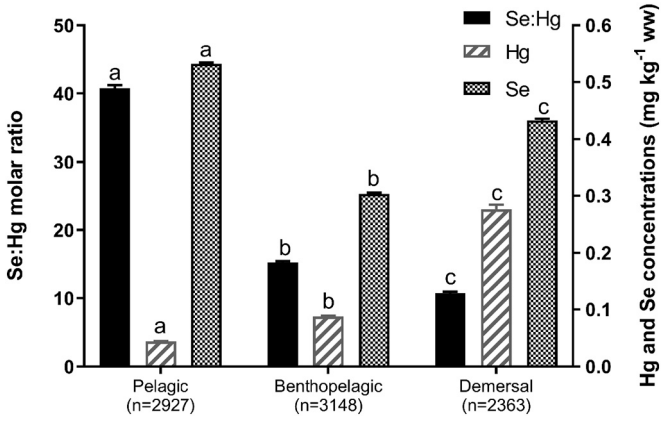

Fig. 4. Mean +1SE of Se:Hg molar ratio (left Y axis), Hg and Se concentrations (right Y axis) in fish species from different habitats of NEAO sampled between 2006 and 2015. Different letters above the columns denote significant differences between habitats (ANOVA; $P < 0.05$).

with different sizes (22 vs 30.9 cm) and Hg concentrations driving this correlation. The correlation in each size class, when analyzed separately, was not significant. Excluding blue whiting, the best correlation was found in plaice, tusk and blue ling (Pearson r range = 0.63 to 0.66).

The protective effect of Se against Hg toxicity has been reported in a variety of organisms and is most commonly linked to the antagonistic effect of Hg and Se (Khan and Wang, 2009). If Se plays an important role in ameliorating Hg toxicity due to antagonism between these two elements, a correlation between Hg and Se in the wild species can be expected. This may be due to upregulation of Se to ameliorate the Hg toxicity and to replace the reduced Se body burden after formation of Hg-Se. The other possible reason is that fish receive a significant part of Hg as Hg-Se compounds (methylmercuric selenide and MeHg selenocysteinate, selenoprotein P-bound HgSe clusters) in their diet from consuming lower trophic marine organisms (Khan and Wang, 2009). In species with low concentrations of Hg, particularly the pelagic species, no correlation was observed between Hg and Se concentrations, but a tendency towards stronger correlation was observed when the concentration of Hg was higher. These findings support a possible antagonistic effect of Se against Hg in wild fish species collected from our large study area, indicating a potential interaction between Se and Hg.

As fish and seafood contain both nutrients and contaminants, potential health benefits from the nutrients should be considered simultaneously along with the contaminants. A correlation between Hg and Se at higher concentrations of Hg may have implications for human risk assessment, food security and environmental management. Since Se may ameliorate MeHg toxicity, it is conceivable that the Se:Hg molar ratio may be used as a better indicator when assessing seafood safety that may be more informative than evaluating fish MeHg concentration alone.

3.4. Effects of geography

Nine of the 17 species investigated in this study were sampled from both offshore and fjord and coastal areas of the NEAO, whereas 11 of 17 species were sampled from different offshore areas (Fig. 2, Fig. 5). In most species, fish from fjord and coastal areas had higher Hg concentrations than fish sampled from offshore areas. When offshore areas were compared, fish from the south, i.e. the Skagerrak and the North Sea had higher Hg concentrations than fish from Norwegian Sea and the Barents Sea located in the northerly sector of our study area (Table 2, Fig. 1).

Fish length also varied in 10 of 12 species between geographical areas. The exceptions were Atlantic halibut and wolffish (ANOVA; $P < 0.05$, Table 2). Fish size (length) is a well-established covariate of Hg concentration and the high assimilation efficiency of MeHg (>95%) combined with a very long half-life of MeHg (3.3 years) lead to bioaccumulation of MeHg over time (Van Walleghem et al., 2013). Therefore, MeHg concentrations are expected to be higher in older and larger individuals compared with younger, smaller individuals of the same species.

Hg concentrations increased with length in most species sampled during the investigation (Table S5) while no significant correlations were found for blue whiting, wolffish, plaice and blue ling. When all individuals from all areas were considered, Hg concentration was not correlated with length in plaice and redfish and Hg concentrations decreased with length for these species. However, when linear regression was conducted for different areas separately, Hg concentrations showed an increasing trend with length in all areas for both species (Fig. S2). The Se:Hg molar ratio decreased significantly with length in most species (R^2 between 0.05 and 0.76; $P < 0.05$) except blue whiting, wolffish and eel (no relationship observed). Similarly, when all individuals from all areas were considered, no correlation between the Se:Hg ratio and length was found in plaice and in redfish, the Se:Hg ratio increased with length. However, when areas were analyzed separately, the Se:Hg ratio in both plaice and redfish decreased with length in all areas (Fig. S3). Selenium concentrations increased with length in some species including blue whiting, herring, Greenland halibut and tusk and decreased with length in mackerel, wolffish, haddock, cod, pollack and blue ling. Thus, when comparing Hg and Se concentrations and the Se:Hg molar ratio between areas, fish size was taken into account. In order to remove the effect of size when evaluating geographical trends, least squares means adjusted for mean length of each species were compared using ANCOVA. When comparing fillet Hg concentrations after adjusting for length, there was still a clear gradual increasing trend from north towards south in offshore areas, and Hg concentrations were higher in most species from fjords and coastal areas compared with offshore areas (Figs. 2, 5).

Pearson correlation showed a significant weak to moderate negative correlation (Pearson r range = −0.11 to −0.67) between logHg concentration in fish fillets and sampling latitude in 12 of 13 species (Table S6). The only exception was Greenland halibut, where no correlation was found. In cod and haddock we observed a strong correlation ($r = -0.67$, $P < 0.0001$ and $r = -0.60$, $P < 0.0001$) across a latitudinal gradient of 19.1 and 15.2°, respectively, covering a large range of the study area (Table S6). The slopes of the regression equations were between −0.005 in herring and -0.12 in Atlantic halibut. Se:Hg molar ratio varied significantly in all 11 species when samples from different offshore areas were compared (Fig. 2A), demonstrating a northward gradual increase in Se:Hg molar ratio for all species from NEAO. Se concentrations also varied significantly, but not with a clear latitudinal trend for most species (Fig. 2C) and variations in Se:Hg molar ratios were driven by variation in Hg concentrations rather than Se concentrations.

Se concentrations varied between areas in three different ways. In pelagic species including mackerel and herring, Se concentration varied in the opposite direction of Hg concentration, decreasing from north to south areas. In saithe and blue ling, Se concentrations were unrelated to the Hg concentrations, and in the rest of the species such as wolffish, cod and tusk, Se concentrations followed the Hg concentrations, increasing from north towards south (Fig. 2).

It is important to note that samples investigated in this study were collected over an extensive time period spanning 10 years during 2006–2015. Some studies showed a decline (−2.5% per year) in atmospheric Hg from the North Atlantic during 1990–2009 (Mason et al., 2001; Soerensen et al., 2012). Additionally, a decreasing trend of Hg concentrations is reported in Atlantic bluefin tuna (*Thunnus thynnus*) at −2.4% per year during 2004–2012 (Lee et al., 2016) and in coastal bluefish (*Pomatomus saltatrix*) at approximately −1% per year from 1972 to 2011 (Cross et al., 2015). A large part of the data set presented in this investigation were derived from different baseline studies. However, when samples of each species from different areas were compared, the sampling time overlapped in most cases or the maximum difference in sampling time between areas was only three years. Therefore, sampling in different years was shown to have a negligible effect on Hg variation when fish from different areas were compared.

3.4.1. Mercury in the NEAO environment

In most of the sampled species from NEAO we observed a gradual increasing trend in Hg concentrations from north to south and this may be driven by an increase in effects of populated and industrialized areas in the southern region of our study area (Fig. 1). The Skagerrak and the North Sea are more impacted by industrialization and terrestrial run off in comparison to the more northerly areas such as Barents Sea and the northern Norwegian Sea, which are considered to be more pristine. Thus, the correlation between Hg concentrations in sediment and latitude of sampling location was used as a proxy to evaluate the influence of anthropogenic contamination on Hg concentrations in fish. A very weak correlation (slope = 0.009; $r = 0.11$; $P < 0.0001$; n = 2003) was found between sediment Hg concentrations and latitude (Fig. S4), showing a very small increase towards the north, the opposite trend as found in fish, however this analysis had poor explanatory power

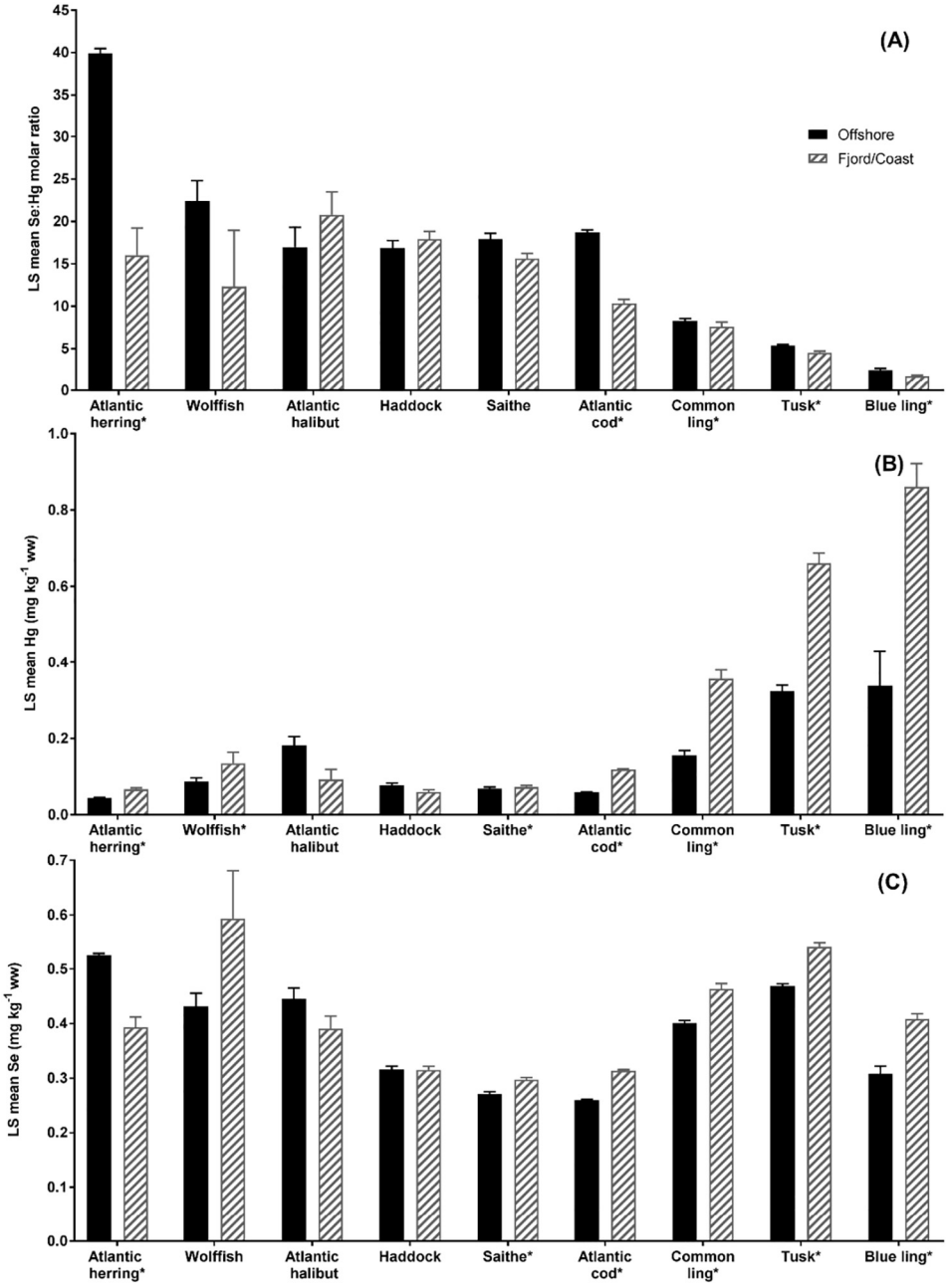

Fig. 5. Least squares mean (length adjusted) of Se:Hg molar ratio, Hg and Se concentrations in fish species from offshore and fjord and coastal areas of NEAO sampled between 2006 and 2015. Error bars represent +1SE. Asterisks (*) indicate species with significant difference between the areas (ANCOVA test; $P < 0.05$).

with only 11% variance explained. Hg concentrations in sediment and longitude were not correlated (slope = 0.0002; $r = 0.008$; $P > 0.05$; n = 2003) (Fig. S4).

Air sea exchange of Hg is considered an important component of the global Hg cycle. It is estimated that the open ocean receives the majority of total Hg input from the atmosphere (Mason and Sheu, 2002; Soerensen et al., 2010). In NWAO, Fitzgerald et al. (1974) investigated the Hg concentration in seawater between Halifax and Bermuda and reported no latitudinal trend in this area. Hg concentration in sediment may reflect Hg concentration in seawater (Gworek et al., 2016) however this relationship is highly variable and inconsistent. No comprehensive study on Hg and MeHg concentrations in NEAO seawater has been undertaken. It is possible that other abiotic and biotic factors rather than environmental Hg concentrations are the main drivers for the observed geographical trends in Hg concentrations in fish species from NEAO.

3.4.2. Latitudinal changes in light and temperature and their effects on Hg concentrations in biota

Photoperiod, sea temperature and photosynthesis dynamics are important environmental parameters that vary across broad latitudinal ranges. In the southern part of the NEAO the planktonic bloom starts earlier in spring than in the northern part. There is a negative correlation between bloom timing and its duration and the blooming period in the northern sector of our study area starts later and is shorter, compared with the southern areas (Friedland et al., 2016).

Thirty-one years of data on seawater surface temperature measurements in the North Atlantic showed a decreasing gradient on both sides

of the Atlantic Ocean (Baumann and Doherty, 2013). In NWAO, the temperature decreased 0.91 °C per degree latitude (in the range of 26–60°N) while in NEAO this decreased only 0.34 °C per degree latitude in the range of 37–70°N on average. Lower temperatures as well as shorter periods of effective light in the northern areas will shorten the period of primary production in which carbon from the environment is pumped into the biomass at the base of the food web (phytoplankton) and may influence MeHg and Hg cycling and biomagnification dynamics. According to the growth bio-dilution theory (Trudel and Rasmussen, 2006), MeHg incorporation from seawater to the first trophic level biomass is higher in southern areas where the production period is relatively prolonged. In northern areas, where the planktonic primary production takes place over a shorter time period but at a higher rate, MeHg incorporation to phytoplankton and biomagnification at higher trophic levels are reduced. Additionally, experimental mesocosm studies on freshwater taxa have shown that increased algal bloom intensity will reduce the MeHg bioaccumulation at higher trophic levels through a bio-dilution effect of MeHg in algae and lead to a two- to three fold reduction in zooplankton MeHg concentration (Pickhardt et al., 2002). During the shorter algal bloom period at the northern latitudes, the primary productivity may be particularly high due to the longer photoperiod. However, MeHg assimilation efficiency will increase in lower temperatures mainly due to lower elimination and longer half-life of MeHg (Lavoie et al., 2013; Trudel and Rasmussen, 2006). In contrast to the findings of this study, this could lead to increasing Hg concentrations northwards, but in NEAO this effect may be confounded by other biological/ecological changes.

In estuarine fish higher temperature has been reported to increase the Hg accumulation in mummichogs (*Fundulus heteroclitus*) in both experimental (12 °C temperature range) and in situ (2.6 °C temperature range) sampling approaches, potentially as a result of increased metabolic rates and energy budgets (Dijkstra et al., 2013). Considering the large variability in seawater temperature between the north and south regions of our study area (NEAO, approximately 8.5 °C; 25° latitude and 0.34 °C change per latitude degree), temperature may be an important driver of increased Hg bioaccumulation in fish samples from the southern region of our study area (Fig. 1).

Fish growth efficiency may also affect Hg concentrations in fish fillets. Higher growth rates and food conversion efficiency have been reported in the Atlantic halibut populations in northern parts of their range compared to southern regions of the NEAO (Jonassen et al., 2000). Counter gradient growth capacity has also been reported in striped bass (*Morone saxatilis*) and Atlantic silverside (*Menidia menidia*) from NWAO as a compensatory mechanism for the short growth period in northern latitudes (Conover et al., 1997; Conover and Present, 1990). Higher growth efficiency in fish from northern areas would result in an increase in body mass from the same amount of ingested food compared with southern areas, which may result in lower MeHg accumulation due to potential bio-dilution effects (Trudel and Rasmussen, 2006; Ward et al., 2010). Considering that most of the Hg is assimilated from food, a higher growth efficiency in the northern areas may lead to lower Hg concentrations for the second trophic level (zooplankton) potentially resulting in lower Hg exposure and bioaccumulation in higher trophic positioned fish from northern areas.

Methylation of inorganic Hg into MeHg is the mechanism that makes it more bioavailable to biota and this process takes place in both sediment and in the open water column (Ullrich et al., 2001). It is also reported from field studies that higher temperatures, as a result of seasonal changes, can increase methylation rates and elevate the concentration of the more labile MeHg (Hammerschmidt and Fitzgerald, 2004; Korthals and Winfrey, 1987; Wright and Hamilton, 1982). Other studies have also shown latitudinal trends with Hg concentration in wild fish populations. Cutshall and Pearcy (1978) reported an increasing trend with latitude, in Pacific hake (*Merluccius productus*) from the North Pacific Ocean, whereas Hall et al. (1976) reported that Hg concentrations decreased towards the north in Pacific halibut (*Hippoglossus stenolepis*) from Washington State towards the Bering Sea in the North Pacific Ocean. Baumann et al. (2017) performed a comprehensive study on bioaccumulation of Hg in Atlantic silverside populations and showed a latitudinal increase in Hg concentration along NWAO coast between 38.4 and 45.2°N, contrary to the findings in this study. The authors suggested that higher ingestion and higher MeHg assimilation are the main reasons for higher Hg in the northern populations. The main difference between this study and Baumann et al. (2017) was different latitudinal ranges. We studied different marine fish species (oceanic) between 50.2°N and 75.6°N whereas they analyzed a low trophic level fish from more southern latitudinal range from 38.4°N to 45.2°N in estuarine habitats, which are dramatically different compared to offshore ecosystems and fjord and coastal areas. We postulate that it is likely that the difference in temperature and light regimes between the extreme north and south sampling were less pronounced in their study area compared with this study and thus higher Hg assimilation efficiency in lower temperature outweighs the other driving parameters in NWAO.

3.4.3. Offshore versus fjord and coastal areas

Nine of the species investigated in this study were sampled in both offshore areas and in fjord and coastal ecosystems. After adjusting for fish length, the Se:Hg molar ratio was significantly higher in the samples of all species from offshore areas than in the same species sampled from fjords and coastal areas except for wolffish, Atlantic halibut, haddock and saithe (Table S7; Fig. 5A). The largest difference between these two areas was found for herring and cod. The samples from offshore areas contained significantly lower Hg concentrations in seven of nine species, except for Atlantic halibut and haddock which were not significantly different. Blue ling, tusk, common ling and cod showed the largest variation in Hg concentrations between offshore and fjord and coastal areas. Se concentrations were higher in fish from fjord and coastal areas in most species (6 of 9 species) and varied mainly in accordance with Hg concentrations. The exceptions were for herring from offshore areas which contained higher Se and Atlantic halibut and haddock where no significant differences were found (Table S5; Fig. 5).

In general species such as herring, with low Hg concentrations, had large differences in their Se:Hg ratios as Se concentrations varied in the opposite direction as the Hg concentrations. In species with higher Hg contamination, such as blue ling, tusk and common ling, Se concentrations varied in the same direction as Hg and were higher in samples from fjord and coastal areas. Thus the Se:Hg molar ratio values did not exhibit considerable variation between offshore and fjord and coastal areas (Fig. 5).

Fjord and coastal areas are more affected by anthropogenic activities than the open ocean due to centralization of industries and households and the fact that in Norway, >80% of population lives <20 km from the coast (NMFA, 2017). Hence, these areas are expected to be more contaminated by Hg than offshore areas. Fjord and coast also receive more runoff from terrestrial catchments and likely deliver more organic matter and atmospherically deposited Hg compared with offshore areas (Everaert et al., 2017; Grigal, 2002). Therefore, more Hg is bound to organic matter in fjord and coastal areas (Jonsson et al., 2014). Fjord and coastal areas have relatively limited water exchange than offshore areas with higher water circulation due to oceanic currents. Furthermore, the addition of organic matter from terrestrial environments and Hg-organic matter compounds may lead to an enhancement of higher Hg methylation (Jonsson et al., 2014). MeHg originating from atmospheric and terrestrial sources has greater bioavailability compared with MeHg produced in marine sediments (Jonsson et al., 2014).

Salinity is an important factor determining Hg methylation in sediment. Within the natural salinity range (0.03–2.4%), Hg methylation may be reduced by more than half in high salinity sediments (Compeau and Bartha, 1987). Additionally, freshwater inputs from terrestrial catchments lead to fjord and coastal areas generally having lower salinity than offshore areas, although the water in deep parts

(below the halocline) may not be lower in salinity but may be influenced by water residence times. Considering all factors, Hg methylation is possibly occurring at a higher rate and may therefore exist in more bioavailable and labile forms in fjords and coastal areas potentially leading to higher MeHg accumulation in fish inhabiting these environments. Additionally, in the inner sectors of fjord ecosystems dissolved oxygen may also be lower compared to the open ocean which may also contribute to enhanced methylation efficiency.

3.5. MeHg to THg ratio

We measured MeHg concentrations in 278 samples comprising five species. The percent mean ± SD of Hg present as MeHg was >93% for all measured species (Greenland halibut 104 ± 12; n = 71, tusk 97 ± 10; n = 118, saithe 93 ± 5; n = 44, cod 104 ± 12; n = 30 and blue ling 100 ± 5; n = 15. These five species represent benthopelagic (cod, Greenland halibut and saithe) and demersal (tusk and blue ling) as well as both lean (cod, tusk and saithe) and oily fish (Greenland halibut). These findings are in good agreement with the general assumption that the MeHg fraction in marine fish fillets is approximately 95% of the measured total Hg (Bank et al., 2007; Bloom, 1992; Razavi et al., 2014). It is well established that the MeHg to THg ratio varies according to the trophic position of marine organisms and that this ratio increases along the food web due to higher assimilation efficiency of MeHg and consumption of more contaminated prey and higher MeHg ratio in higher trophic position organisms has been reported by others (Lavoie et al., 2010; Lavoie et al., 2013). Therefore, THg serves as a good proxy for MeHg in fish fillets from species that inhabit higher trophic positions in marine ecosystems. For exposure assessment, a conservative assumption was made and 100% of THg in fish fillets was assumed to be in the MeHg form.

3.6. Comparison with reference levels

Different reference values for Hg in seafood and fish are set by guidelines authorized by different countries in the world including 0.3 mg kg^{-1} ww in USA (EPA, 2001) and 0.4 mg kg^{-1} ww in Japan (Marumoto and Imai, 2015; Ministry of Health and Welfare, 1973). In the EU the maximum level for Hg in muscle meat from fish for human consumption is 0.5 mg kg^{-1} ww for most fish species including most of those investigated in this study. The exceptions are wolffish, eel, Atlantic halibut and redfish, where the EU maximum level is 1.0 mg kg^{-1} ww (EU Commission, 2006). Among all individual fish investigated, 8.0% and 3.5% contained Hg concentrations equal or above 0.3 and 0.5 mg kg^{-1} ww, respectively (Table 3). None of the samples from herring, plaice, haddock, blue whiting or mackerel had Hg concentrations above 0.5 mg kg^{-1} ww and none of the samples of blue whiting and mackerel had Hg concentrations above 0.3 mg kg^{-1} ww. Blue ling and tusk had the highest portion of specimens with Hg concentrations above the 0.3 and 0.5 mg kg^{-1} ww reference values. For blue ling, 81% and 60% of the fish were above the two reference values, while 42% and 20% of the tusk were above these values respectively.

Se:Hg molar ratio and Se have no regulated reference levels. However, it has been suggested that fish with a molar ratio above 1.0 may be protective, although considerable uncertainties regarding the level of protectiveness still exist (Burger, 2012; Peterson et al., 2009; Ralston, 2008; Ralston et al., 2016). In this study all species had a mean Se:Hg molar ratio above one and considering all individual fish, 0.7% had ratios below one. Only common ling, Atlantic halibut, tusk and blue ling had equal or >1% of samples with a molar ratio below one.

3.7. Hg exposure assessment from NEAO fish consumption

Fish and other types of seafood provide healthy nutrients including essential fatty acids (EFA), and consumption of seafood therefore is advised (Kris-Etherton et al., 2009). The US Environmental Protection Agency (EPA), have issued a recommendation of 340 g seafood consumption per week for pregnant women (EPA, 2004). An EFSA panel on contaminants concluded that consumption of one to two servings of seafood per week in general for adults and three to four servings per week during pregnancy are associated with better health outcomes (Agostoni et al., 2014). Most of the European countries recommend two servings of at least 150 g per week, although the recommended amount varies from 100 g per week up to 200 g per day (Agostoni et al., 2014). Hg exposure was calculated based on two servings of fish (as a general recommendation) equal to 340 g (170 g per serving) fish per week for adults (70 kg) and four servings equal to 680 g of fish consumption for pregnant women (Table 1).

For a person of 70 kg and a consumption of 340 g fish per week, TWI for Hg will be exceeded if the Hg concentration in the fish is higher than 0.27 mg kg^{-1} ww. Thus, considering the average Hg concentration of the fish species analyzed here, two servings of Atlantic halibut, tusk or blue ling and even only one serving of blue ling would lead to a dietary intake of Hg exceeding the TWI (Table 1). Four servings of pollack, Greenland halibut, hake, common ling, Atlantic halibut, tusk and blue ling would lead to Hg intake exceeding the TWI if other sources of MeHg exposure are excluded.

Blue ling and tusk from fjord and coastal areas were the most Hg contaminated species in this study (0.87 and 0.85 mg kg^{-1} ww, respectively, Table 2). One serving of blue ling and tusk from this area per week would lead to Hg intake of 163% and 159% of TWI, respectively. Excluding other factors for MeHg exposure, intake of these species (from fjord and coastal areas) should not exceed 107 and 105 g per week for a 70 kg adult. Considering the geographical variation in Hg concentration in these two species and more sensitive consumers (pregnant women and children) consumption of tusk and blue ling caught from fjords and coastal areas in the south of Norway may lead to high levels of MeHg exposure. However, the Norwegian Food Safety Authority has issued warnings against consumption of deep water species including tusk and blue ling from some of the large fjords in western Norway.

Most of the consumption of fish comes from commercial fisheries, and catch volume of the different species gives some information about the consumption of the different species by the general population. The species with the highest catch volumes, such as mackerel, herring, cod, haddock and saithe, all had relatively low concentrations of Hg, and a 70 kg person could consume more than a kilogram per week of these species without exceeding the TWI (Table 1). The most highly contaminated species constitute a very small portion of the annual catch from NEAO. Atlantic halibut, tusk and blue ling, having mean concentrations of Hg above 0.3 mg kg^{-1}, all constituted <1% of the annual catch. The catch volumes of the species with a risk after four servings per week (pollack, Greenland halibut, hake and common ling) were below 3% in 2017. Therefore, these species were not considered as a great risk to the general consumers at a large scale. However, local recreational fishermen, and their families, living in the fjord and coastal areas catching deep water species such as tusk and blue ling may well exceed the TWI for Hg, and may be considered at risk of greater MeHg exposure if they consume these species regularly.

We next calculated the weekly consumption limits (i.e. the amount that can be consumed without exceeding the TWI, for a 70 kg adult) using the mean Hg values for each species. The consumption limits of blue whiting, mackerel and herring were high (2241 g, 2114 g and 2019 g respectively) whereas the limits for consumption of Atlantic halibut, tusk and blue ling were low (240 g, 208 g and 126 g respectively). These calculations are based on TWI for Hg exposure only. Hence, the concomitant exposure of other potentially associated contaminants in fish such as dioxins, polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers and other persistent organic pollutants (POPs), are not considered here.

Further, as these calculations do not take the interaction between Se and Hg into account we calculated the health benefit value (HBV_{Se}) for each species (Table 1). HBV_{Se} varied between 2.1 in blue ling and 7.1 in

redfish when overall mean concentrations of Hg and Se were used. No negative HBV_{Se} values were found. Hence, all species provided more Se than Hg at the molar concentrations and consumption of these species thus provides a surplus of Se, potentially ameliorating the adverse effects of MeHg. The majority of the epidemiological studies that reported adverse health effects of MeHg due to high levels of seafood consumption, were conducted in populations consuming species with negative HBV_{Se} values. In the Faroes study, pilot whale was a significant part of the diet with a negative HBV_{Se} (−18.6 to −82.3) (Julshamn et al., 1987; Ralston et al., 2016), but in the Seychelles study, where only oceanic fishes were consumed (no marine mammals), with the similar Hg exposure level as the Faroes study, no clear health effect of Hg was found. In a New Zealand study, another cohort study showing negative health effect of maternal Hg exposure in children, shark species with negative HBV_{Se} (−120) was consumed frequently (Ralston et al., 2016). However, several studies have demonstrated positive HBV_{Se} in oceanic fish corresponding to more Se than Hg in molar concentration. Negative values for HBV_{Se} are only reported in pilot whale, mako shark, other shark species and swordfish (Ralston et al., 2016) and it seems there is a connection between consumption of species with negative HBV_{Se} and potential health effects from MeHg exposure.

The Recommended Dietary Allowance (RDA) for Se for adults and pregnant women is 55 and 60 μg day^{-1} respectively and the upper intake level for adults is set at 400 μg day^{-1} (IOM, 2000). Two servings of fish species from NEAO per week would cover 24–49% of the RDA (adults) while four servings of fish with the highest Se concentration is still well below the upper intake assuming all Se intake is from fish.

4. Conclusions

The large variation in Hg concentrations is the main driving factor for the observed level of Se:Hg ratio variability. A gradual increasing trend of Hg concentrations from north to south was observed, where fish from southern areas had higher concentrations of Hg and lower Se:Hg molar ratios compared to fish from northern sectors of the study area. Generally, fish from fjord and coastal areas had higher Hg and therefore a lower Se:Hg molar ratio compared with fish collected from offshore areas. The majority of species sampled in this investigation showed a positive correlation between Hg and Se concentrations and this relationship was strongest for species with higher Hg concentrations. Surplus Se may reduce MeHg toxicity although substantial uncertainty still exists in understanding the relationships between Se and Hg interactions and human health. All species had on average Se:Hg molar ratios above 1.9 and HBV_{Se} above 2.1 emphasizing the excess Se after sequestration of Hg. Generally, fish from NEAO can be considered safe regarding Hg contamination except for some deep water species including Atlantic halibut, Greenland halibut, tusk and blue ling especially fish from southern sections of our study area and fjord and coastal ecosystems. Two servings of Atlantic halibut, tusk and blue ling exceed the Hg TWI and therefore this is an important consideration for children, pregnant women and women of child bearing age. Further research is required to address the detailed mechanisms causing the protective effect of Se on MeHg toxicity in different Se:Hg molar ratios and to achieve more fine scale risk-benefit information from Se:Hg molar ratios with regard to human health risk assessments. Providing more data on fish nutrients and elaborating on the interaction between contaminants and nutrients will improve risk communication and enable authorities to provide more specific and meaningful advisories.

Acknowledgements

The authors wish to thank the technical staffs in sample reception and laboratory departments at the Institute of Marine Research (previously NIFES) for sample collection, preparation and metal analysis. Data presented in this article originated from several biomonitoring projects and several organizations contributed funding including: Norwegian Ministry of Trade, Industry and Fisheries, Norwegian Food Safety Authority, Institute of Marine Research, The Norwegian Seafood Research Fund and the Norwegian Herring Sales Organization. Photos used in the graphical abstract were reprinted with permission Atlantic mackerel (photo credit: Lorentzen E. A.), plaice (photo credit: Kvalsund M.), Atlantic cod (photo credit: Paulsen O.).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2018.10.405.

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