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Rapid temporal decline of mercury in Greenland halibut (*Reinhardtius hippoglossoides*)[☆]

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ABSTRACT

Mercury (Hg) pollution in the ocean is an issue of global concern, however bioaccumulation regimes of this ubiquitous pollutant in marine apex predators have important knowledge gaps. Our fish length and stable isotope ($\delta^{15}\text{N}$ and $\delta^{13}\text{C}$) normalized data of Greenland halibut (GH) (*Reinhardtius hippoglossoides*) showed that Hg bioaccumulation in fillet tissue decreased by ~35–50 %, over a ten-year period from 2006 to 2015 ($n = 7$ individual sampling years). Hg was predominantly in the methylmercury form (>77 %). Results from a Bayesian information theoretic model showed that GH Hg concentrations decreased with time and its associated declines in Hg air emissions, estimated trophic position, and a potentially lower degree of demersal prey use as indicated by temporal trend shifts in nitrogen ($\delta^{15}\text{N}$) and carbon ($\delta^{13}\text{C}$) stable isotope values. GH trophic shifts accounted for about one third of the observed temporal reduction in Hg. Our study demonstrates the importance of simultaneously considering Hg emissions, food web dynamics and trophic shifts as important drivers of Hg bioaccumulation in a marine, deep water fish species and highlights the effectiveness of Hg regulations on ocean apex predator Hg concentrations and overall seafood safety.

1. Introduction

Mercury (Hg) pollution is a significant environmental and public health issue that has garnered sustained interest from scientists, health officials and international policymakers. In August 2017, the United Nations Minamata Convention on Mercury (UN-MCM) entered into force as a multinational agreement to reduce Hg emissions and releases to protect the environment and human health (UNEP, 2013). Temporal analyses of Hg in marine fish species consumed by humans are an important aspect of the effectiveness evaluation of the UN-MCM (Bank et al., 2020). One of the primary challenges of evaluating the effectiveness of the UN-MCM is to distinguish temporal changes in biotic Hg concentrations resulting from measures undertaken by the UN-MCM from shifts caused by other factors such as changes in climate and contaminant food web pathways unrelated to policy measures (Bank

et al., 2020).

The Greenland halibut (GH) (*Reinhardtius hippoglossoides*) is a long-lived, slow-growing flatfish inhabiting deep, cold waters throughout large geographical areas of both the Northwest and Northeast Atlantic Ocean (Bowering and Needreass, 2000; Albert, 2016; Dwyer et al., 2016). This circumpolar, apex predator (trophic level 4.4 ± 0.1 SE; Froese and Pauly, 2000) has the potential for long range migration, exhibits opportunistic feeding strategies and extensive habitat use throughout much of the vertical range of the water column (i.e., GH is more of a free-swimming species than other flatfish) and is exposed to both anthropogenic and natural sources of Hg (Boje, 2002; Vollen and Albert, 2008). Therefore, GH is a good eco-indicator species of marine Hg, and trophic and ecosystem shifts (Solmundsson, 2007; Vollen and Albert, 2008; Stasko et al., 2016; Giraldo et al., 2018). GH is also an important commercial seafood species in Iceland, Canada, Norway,

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USA, Faroe Islands, Greenland, and Russia (Barbeaux et al., 2013; Giraldo et al., 2018). Total catch volume of GH in Norwegian waters in 2018 exceeded 17,000 metric tons, having an estimated value of ~600 million NOK (Sandberg and Steinseide, 2019).

Previous studies on GH fillets (Julshamn et al., 2006; Julshamn et al., 2011) reported elevated levels of total Hg with some samples exceeding the regulatory limit for Hg which in Europe is set at 0.5 mg Hg/kg wet weight for most species including GH (EU, 2019). These initial findings identified the need for additional regional investigations of this species in the North East Atlantic to track Hg concentrations over time in the context of Hg emissions and food web pathways.

Marine ecosystems have complex food webs, water circulation systems, and biogeochemistry regimes and ocean ecosystems are not polluted uniformly by Hg at a global scale (Lamborg et al., 2014). Recently investigators have shown contemporary declines in Hg in marine predators inhabiting different ocean basins for seafood species such as bluefish (*Pomatomus saltatrix*) and Atlantic bluefin tuna (*Thunnus thynnus*), and also for several species of southern ocean squids (*Alluroteuthis antarcticus*, *Bathyteuthis abyssicola*, *Filippovia knipovitchi*, *Galioteuthis glacialis*, *Gonatus antarcticus*, *Kondakovia longimana*, *Psychroteuthis glacialis* and *Slosarczykovia circumantarctica*) (Cross et al., 2015; Lee et al., 2016; Grieb et al., 2020; Seco et al., 2020).

Marine apex predators such as GH are considered important drivers of ecosystem structure and function as well as indicators of environmental change, trophic shifts, and benthic-pelagic coupling dynamics (Griffiths et al., 2017; Giraldo et al., 2018; Hazen et al., 2019). Trends in environmental change related to important marine pollutants such as Hg are most accurately depicted when changes in food web pathways are evaluated simultaneously with changes in air emissions and contaminant concentrations in targeted indicator species and marine apex predators such as GH (Braune et al., 2014; Bank et al., 2020). Diet is the main exposure pathway for Hg in marine organisms (Wang and Wong, 2003) and stable isotope analyses of carbon and nitrogen have been used extensively to identify food web pathways and trophic niches of biota such as GH (Michener and Lajtha, 2007; Dennard et al., 2009). The simultaneous assessment of both $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ is a reliable tool for evaluating spatiotemporal trophic dynamics of marine apex predators (Braune et al., 2014; Vo et al., 2011). Trophic position can be estimated using $\delta^{15}\text{N}$ (with higher values observed for fish at higher trophic position) (Post, 2002). Energy flow dynamics and the degree of demersal or pelagic prey use can be estimated, along with trophic shifts, using $\delta^{13}\text{C}$ values from GH muscle tissue (with higher values observed at higher trophic position and greater use of demersal prey) (Post, 2002; Dennard et al., 2009).

Hg concentrations in GH fillet tissue from the Norwegian Sea were investigated over a ten-year period ($n = 7$ individual sampling years) from 2006 to 2015. To investigate potential relationships between temporal changes in Hg concentrations in GH fillet samples, declining Hg emissions and food web pathways, we evaluated *a priori* predictions using Bayesian statistical model selection techniques. The models consider a suite of independent predictor variables including year (to reflect environmental changes such as Hg emissions and ecosystem dynamics), fish sex, length, fat content, total Hg (as a proxy for methylmercury (MeHg)), and carbon and nitrogen stable isotopes (to estimate energy flow and trophic position). Specifically, we predicted that Hg bioaccumulation would be influenced by fish size, changes in Hg emissions, food web position and/or complexity and benthic-pelagic feeding strategies. We also predicted that trophic shifts in food web position and energy sources of GH over time could lead to changes in individual fish Hg concentrations. Finally, we discuss the role of biomonitoring GH in the context of the effectiveness evaluation of the UN-MCM (UNEP, 2013; Bank et al., 2020).

2. Materials and methods

2.1. Study area and wild fish sampling

We sampled GH from the Norwegian Sea from 2006 to 2015 (Fig. 1, Table 1) by using gillnets (July, August, October 2006–2008) or longline fishing (March–July 2011–2015). In total, 625 fish were collected, 50 fish from one position each year in 2006–2008, 100 fish from four positions in 2011 and 125 fish from five positions each year in 2013–2015 (Fig. 1, Table 1). In 2006–2008, fish length, weight and sex were determined, and muscle samples were excised shortly after sampling. Muscle samples were wrapped in aluminum foil and frozen at $-20\text{ }^{\circ}\text{C}$ before shipment to the laboratory. In 2011 and 2013–2015, whole fish were frozen and shipped to the laboratory where fish length, weight and sex were determined, and muscle samples were excised. Skinless and boneless muscle samples of approximately 200 g from each fish were taken from the upper side of the fish, from approximately 10 cm behind the head at both sides of the central line towards the tail. The muscle samples were homogenized using a food processor that was cleaned between samples, and the wet homogenates were either frozen (samples from 2011) or freeze-dried and ground to a fine powder (all other samples) before further analyses. Dry matter content was determined for freeze-dried samples and all data are reported on a wet weight (ww) basis.

2.2. Air emissions of Hg in Norway

Data on air emissions of Hg were retrieved from the Norwegian Environment Agency (NEA) web site (<https://www.norskeutslipp.no/en/Frontpage/>). The total annual emissions to air are calculated by Statistics Norway and the NEA and this inventory includes emission data from all relevant companies and industries in Norway. For the largest point sources, the emission data from the companies' obligatory annual reports to the authorities are utilized in the inventory, and for smaller emission sources not subject to obligatory reporting, the company activity data and emissions factors are used to calculate the emissions. Data are expressed as kg of Hg/year and are freely available at the agency website listed above.

2.3. Laboratory and analytical methods

Fat content was determined after extraction with ethyl acetate by a gravimetric method as described by Frantzen et al. (2011). Total Hg was determined by inductively coupled plasma-mass spectrometry (ICP-MS) after microwave digestion of freeze-dried or wet muscle samples. The method is a European and US standard for Hg, arsenic, cadmium, and lead (CEN, 2009; Julshamn et al., 2013) and was described in detail by Julshamn et al. (2007). Briefly, 0.2 g freeze-dried or wet and homogenized muscle sample was digested with 2.0 ml concentrated (65 %) nitric acid and 0.5 ml 30 % hydrogen peroxide in a microwave oven (Milestone MLS-1200 MEGA or Milestone ultraWAVE Microwave Digestion). Total Hg determination was performed by quantitative ICP-MS (Agilent 7500c, Agilent 7500cx or Thermo iCap-Q with collision cell and ICP ChemStation or Qtegra software) using an external calibration curve. Rhodium was used as the internal standard to correct for instrumental drift and gold was added to stabilize the Hg-ions. The limit of quantification (LOQ) for Hg was 0.03 mg/kg dw from 2006 until 2010 when the laboratory instrumentation was changed and LOQ was reduced to 0.005 mg/kg dw. Trueness of the method was controlled by concomitant analysis of the certified reference materials (CRMs) CRM 1566 (oyster tissue) from the National Institute of Standards and Technology (Gaithersburg, USA) and CRM TORT-2 or TORT-3 (lobster hepatopancreas) from the National Research Council (Ottawa, Canada), and by regular participation in proficiency tests. Analysis of CRMs showed that the recovery of Hg ranged from 80 % to 120 % for the whole period of analysis (2006–2015), and the results of proficiency tests

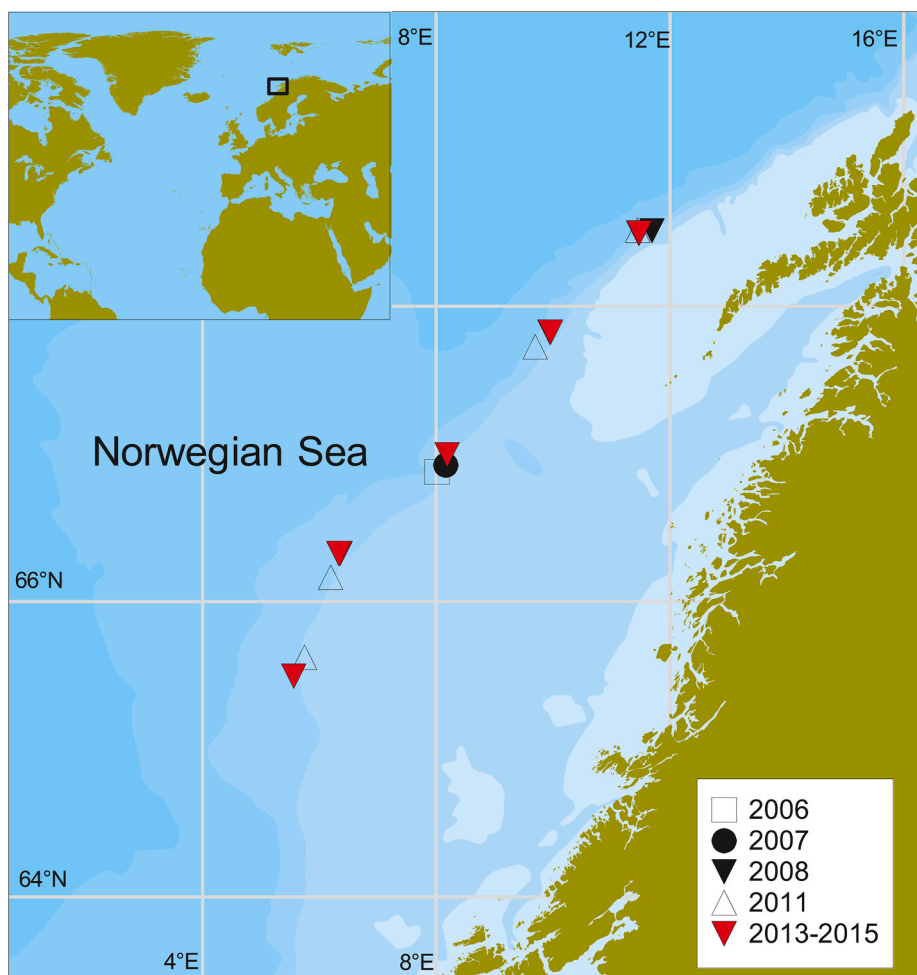


Fig. 1. Sampling locations of Greenland halibut collected during the investigation from 2006 to 2015 in the Norwegian Sea, Norway. Fifty fish were collected from one position each year in 2006, 2007 and 2008, 25 fish were collected from each of four positions in 2011, and 25 fish were collected from each of five positions each year in 2013, 2014 and 2015. In the period 2013–2015, the same five positions were sampled each year.

showed satisfactory results with Z-scores between -1.33 and 0.76 for various seafood matrices. The repeatability (% RSD_r) of the method was 8.2% and the reproducibility (% RSD_R) was 17% for Hg in fish muscle (Julshamn et al., 2007).

To evaluate % MeHg in GH muscle tissue, Hg speciation analyses in 20 muscle samples from fish of varying size and fat content, carefully selected to represent fish from all sampling years and the entire sampling area, were performed by gas chromatography (GC) coupled to ICP-MS (GC-ICP-MS: RSH Triplus/Trace Ultra GC-XII Series ICPMS, Thermo Scientific, USA). Sample preparation and analytical protocols were adapted from previously established methods (Clémens et al., 2011). Briefly, Hg species were extracted from 0.2 g of freeze-dried and homogenized fish muscle in 5 mL of tetramethylammonium hydroxide solution (25% TMAH in H_2O , ACS reagent grade, Sigma Aldrich). This alkaline digestion was completed using a Pyrex vessel and an Explorer-Discover SP-D instrument (CEM Corporation, USA). In order to prevent the endogenous speciation of the Hg compounds of the fish specimens, a soft extraction method was applied: 1 min of warming up to 75°C and 4 min at 75°C with continuous sample homogenization by magnetic stirring. Prior to GC injection, aliquots of the digested extracts were derivatized at $\text{pH } 3.9$ using sodium tetraethylborate solution (NaBEt_4 , 5% v:v in water, Merseburger Spezialchemikalien, Germany), in order to produce volatile ethylated forms of Hg, then extracted in isooctane by mechanical shaking (elliptic table, 20 min , 400 rpm) and readily separated by GC. Quantification was performed by species-specific isotope dilution by spiking known amounts and

concentrations of isotopically enriched standard solutions (Me^{201}Hg and $^{199}\text{Hg(II)}$, ISC Science, Spain) after the microwave extraction step. The detection limit of the method was 0.2 ng Hg g^{-1} , for both Hg species. The analytical results were continuously quality-checked by controlling the Hg background blanks in the reagents involved and by analysing a reference material certified for Hg speciation (ERM CE-464 Tuna Fish) in triplicate. Blanks equivalent concentrations were found to average $1.2 \pm 0.1\text{ ng Hg g}^{-1}$, for Hg(II), whereas MeHg remained undetectable. Relative standard deviations of MeHg and Hg(II) concentrations have been evaluated to 0.9 and 2.5% , respectively, from triplicate preparations and analyses of ERM CE-464. Recoveries of the certified concentrations for ERM CE-464 reached $99 \pm 1\%$ and $100 \pm 1\%$ for MeHg and Hg(II), respectively.

Stable isotope signatures of carbon and nitrogen, and their percentages for C:N ratio determination, were measured at the Institute for Energy Technology (Kjeller, Norway) using a Eurovector EA3028 Elemental Analyzer and Horizon isotope ratio mass Spectrometer (EA-IRMS). Stable isotopes were calculated as $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ using Eq. (1):

$$\delta X = \left(\left(R_{\text{sample}} / R_{\text{standard}} \right) - 1 \right) \times 1000 \quad (1)$$

where X is ^{13}C or ^{15}N and R is $^{15}\text{N}/^{14}\text{N}$ or $^{13}\text{C}/^{12}\text{C}$ for sample and standard. Results were reported in parts per mil (‰) relative to the international standards atmospheric N_2 in air and Vienna Pee Dee Belemnite (VPBD) for nitrogen and carbon, respectively. The accuracy and precision of the method was determined using an internal standard

Table 1

Temporal variation in total Hg (THg) and biological parameters of Greenland halibut sampled during 2006–2015. Statistics are given per year for fish length, weight, sex, fat content and THg concentration in fillet. For a subset of samples, lipid-corrected $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ are given.

Year	Fish length (cm) N mean \pm SE (median) min - max	Fish weight (g) N mean \pm SE (median) min - max	Sex N (% F)	Fat content (g/100 g) N mean \pm SE (median) min - max	THg (mg/kg ww) N mean \pm SE (median) min - max	Lipid-corrected $\delta^{13}\text{C}$ N mean \pm SE (median) min - max	$\delta^{15}\text{N}$ N mean \pm SE (median) min - max
2006	50 67.7 \pm 1.0 (66) 58–86	50 3009 \pm 179 (2668) 1635–7415	50 (92)	50 12.5 \pm 0.3 (13) 4.5–17	50 0.30 \pm 0.02 (0.25) 0.030–0.95	50 −19.05 \pm 0.04 (−19.1) −19.9–−18.5	50 14.36 \pm 0.16 (14.1) 12.8–17.6
2007	50 64.4 \pm 0.7 (65) 41–74	50 2970 \pm 105 (2970) 665–5030	50 (88)	50 11.9 \pm 0.3 (12) 6.6–17	50 0.22 \pm 0.01 (0.22) 0.060–0.45	48 −19.02 \pm 0.04 (−19.0) −19.5–−18.1	48 13.86 \pm 0.14 (13.6) 12.4–17.3
2008	50 67.7 \pm 0.6 (68) 59–74	50 3259 \pm 97 (3215) 1950–5050	50 (94)	50 12.7 \pm 0.3 (12) 8.6–18	50 0.25 \pm 0.01 (0.24) 0.080–0.70	50 −19.15 \pm 0.05 (−19.2) −20.3–−18.3	50 13.36 \pm 0.18 (13.1) 11.6–18.1
2011	100 64.1 \pm 0.6 (64) 53–85	75 2706 \pm 125 (2608) 1190–6271		100 10.9 \pm 0.2 (11) 4.0–15	100 0.15 \pm 0.01 (0.14) <0.002–0.69	43 −19.34 \pm 0.05 (−19.3) −20.0–−18.8	43 13.02 \pm 0.14 (12.8) 11.9–16.5
2013	125 59.7 \pm 0.7 (60) 44–78	125 1996 \pm 77 (1830) 703–4910	125 (62)	125 10.3 \pm 0.2 (10) 2.4–17	125 0.13 \pm 0.01 (0.11) 0.023–0.41	116 −19.70 \pm 0.04 (−19.7) −21.3–−17.3	116 13.13 \pm 0.11 (12.8) 11.4–17.0
2014	125 62.1 \pm 0.7 (62) 45–91	125 2228 \pm 88 (2023) 769–7002	125 (68)	125 9.41 \pm 0.22 (10) 2.0–16	125 0.13 \pm 0.01 (0.11) 0.021–0.42	125 −19.55 \pm 0.03 (−19.6) −20.4–−17.9	125 12.99 \pm 0.07 (12.9) 11.5–16.7
2015	125 62.3 \pm 0.6 (63) 48–81	125 2343 \pm 88 (2315) 938–5766	125 (86)	125 9.58 \pm 0.23 (10) 1.4–19	125 0.16 \pm 0.01 (0.14) 0.030–0.41	123 −19.54 \pm 0.04 (−19.5) −20.7–−18.6	123 13.19 \pm 0.10 (13.0) 11.6–17.9
All years	625 63.0 \pm 0.3 (63) 41–91	600 2476 \pm 42 (2385) 665–7415	525 (78)	625 10.6 \pm 0.1 (11) 1.4–19	625 0.17 \pm 0.00 (0.16) 0.002–0.95	555 −19.44 \pm 0.02 (−19.4) −21.23–−17.3	555 13.30 \pm 0.05 (13.1) 11.4–18.1

(IFE trout) calibrated against international certified reference materials (IAEA-N-1 and IAEA-N-2 for $\delta^{15}\text{N}$ and USGS-24 for $\delta^{13}\text{C}$). Average ($\pm 1\text{SD}$) values for IFE trout were: $\delta^{15}\text{N}_{\text{AIR}}$: 11.6 ‰ \pm 0.2 and $\delta^{13}\text{C}_{\text{VPDB}}$: 20.2 ‰ \pm 0.2. Average ($\pm 1\text{SD}$) results for 58 analyses of the IFE trout standard analyzed together with the samples, were: $\delta^{15}\text{N}_{\text{AIR}}$: 11.5 ‰ \pm 0.2 and $\delta^{13}\text{C}_{\text{VPDB}}$: 20.1 ‰ \pm 0.1. Lipid correction of $\delta^{13}\text{C}$ -values was performed as described by Kiljunen et al. (2006) using Eqs. (2) and (3):

$$L = \frac{93}{1 + (0.246 \times (C : N) - 0.775)^{-1}} \quad (2)$$

$$\delta^{13}\text{C}' = \delta^{13}\text{C} + D \times \left(I + \frac{3.90}{1 + 287/L} \right) \quad (3)$$

where L is the lipid content of the sample and $\delta^{13}\text{C}'$ is the lipid-corrected value of the sample; C and N are the proportions of carbon and nitrogen measured in the sample; $\delta^{13}\text{C}$ is the measured value of the sample; D is the isotopic difference between protein and lipid (assigned a value of 7 ‰) and I is a constant (assigned a value of 0.048).

2.4. Statistical analyses

All data were evaluated for normality using Lillefors test and equality of variances using Levene's test and Hg concentration data were subsequently \log_{10} transformed to improve normality. Temporal comparisons of Hg in GH fillets were made using Hg-concentrations normalized for length or $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$. Normalization was performed using analysis

of covariance (ANCOVA) with fish length (cm) as covariate or both $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ as covariates, to control for the effects of these variables on Hg bioaccumulation across years. The latter ANCOVA was also performed using a subset of the dataset including fish with length 60–70 cm ($n = 332$ fish). We used linear regression to evaluate trends between GH total Hg concentrations (\log_{10} transformed) and time (year of sample collection), length (cm), $\delta^{13}\text{C}$, $\delta^{15}\text{N}$ and fat content as predictor variables. To evaluate the relationship between Hg air emissions (kg/year) and GH Hg tissue concentrations we used general linear regression with GH Hg least square means (\log_{10} transformed) as the dependent variable, air emissions as the independent variable and year as the unit of replication. Student's t-test was used to compare Hg concentrations between sexes. All statistical tests were performed using Statistica version 13.4 with R integration (TIBCO Software Inc., 2018) and statistical significance for all analyses was accepted at $P \leq 0.05$ (Zar, 2010). We then used Akaike's Information Criterion (AIC) and AIC weights (w_i) to evaluate *a priori* models using single variables and combinations of variables (Burnham and Anderson, 2002). AIC values and w_i were calculated using Eqs. (4) and (5):

$$AIC = n^* \ln(RSS/n) + 2^*K \quad (4)$$

$$w_i = \frac{\exp\left(-\frac{1}{2}\Delta_i\right)}{\sum_{r=1}^R \exp\left(-\frac{1}{2}\Delta_r\right)} \quad (5)$$

The residual sum of squares (RSS) divided by sample size (n) from the linear regression models is used here to estimate model likelihood which is considered a suitable measure of model fit and K represents the number of model parameters plus the intercept and variance from regression analyses. AIC estimates the “best model” in comparison to the other estimated models, including a global model comprised of all significant variables (sampling year, fish length, $\delta^{13}\text{C}$, $\delta^{15}\text{N}$). AIC values with the lowest score are the most parsimonious and models with a difference of >2 , generally, are believed to be substantially different for comparative purposes (Burnham and Anderson, 2002). AIC models were conducted on a modeling data set ($n = 512$ individual fish) that contained no missing values since AIC is sensitive to missing data (Burnham and Anderson, 2002). This modeling data set excluded 2011 since sex data were not available and due to several missing cases of carbon and nitrogen stable isotope values.

3. Results and discussion

3.1. Temporal variation in Hg concentrations and biological parameters

Total Hg concentrations determined in fillets of GH sampled in the Norwegian Sea (Fig. 1) from the years 2006–2015 averaged 0.17 mg/kg ww (range = 0.002–0.95 mg/kg) (Table 1). This was in accordance with earlier results for GH from the same area (Julshamn et al., 2011) and confirms that GH accumulates relatively high levels of Hg as expected for a slow-growing, long-lived, apex predator fish species (Albert et al., 2009; Albert, 2016; Dwyer et al., 2016). The mean concentration was

nonetheless well below the EU’s regulatory limit of 0.5 mg/kg ww applying to Hg in fish muscle (EU, 2019), and only seven of the 625 fish (1.1 %) had concentrations above this limit (Fig. 2).

In order to estimate % MeHg in GH fillets, Hg speciation analysis was performed on skin-free, boneless fillet samples ($n = 20$). Individual fish samples for Hg speciation analyses were selected to be generally representative of all sampling years and areas, and the results showed that MeHg comprised $>77\%$ of the total Hg (mean = 93.5 %, SE = 1.5 %).

There was a significant decrease in total Hg concentrations in GH over the ten years of investigation. The fish were collected at different seasons in some of the years, but an analysis of a larger GH data set from the Norwegian Sea and Barents Sea showed no seasonal GH Hg trends (SI Text 1, Fig. S2). In the years 2006–2008, annual mean concentrations of total Hg in GH varied between 0.22 and 0.30 mg/kg ww, while between 2011 and 2015, mean concentrations varied only between 0.13 and 0.17 mg/kg ww (Table 1). In the same period, a decrease in length, fat content, $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ were observed in the sampled fish (Table 1). The decrease in fish size may be due to variation in fishing gear used in the different years. Sampling in 2006–2008 was performed using gillnet whereas longline was used in 2011 and 2013–2015. Earlier studies have shown that gillnet may selectively catch slightly larger fish than longline (Huse et al., 1999). $\delta^{15}\text{N}$ decreased from 14.4 ‰ to about 13 ‰, corresponding to a shift of approximately 0.4 trophic levels (Post, 2002). Annual mean lipid-corrected $\delta^{13}\text{C}$ values decreased from about -19% to about -19.6% (Table 1), and the values were generally consistent with the average diet being predominantly pelagic (Hobson et al., 2002).

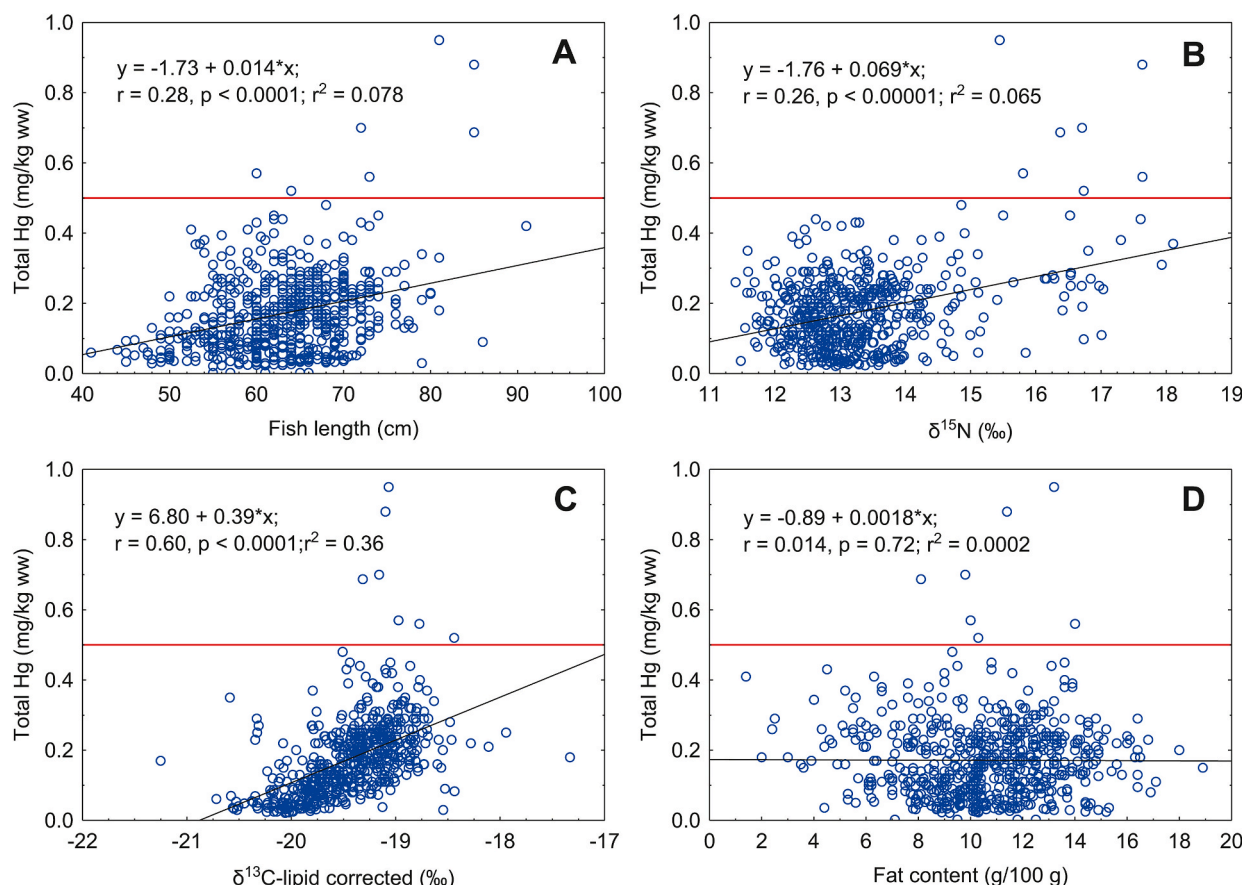


Fig. 2. Relationships between Greenland halibut total Hg (THg) concentrations, fish size, fat content and food web parameters. Linear regression between THg concentrations (mg/kg ww) and (A) fish length (cm, $n = 625$), (B) $\delta^{15}\text{N}$ (‰, $n = 555$), (C) lipid-corrected $\delta^{13}\text{C}$ (‰, $n = 555$) and (D) fat content (g/100 g, $n = 625$) in muscle of GH sampled in the Norwegian Sea during 2006–2015. The red horizontal line in each panel indicates the EU and Norway’s maximum level for THg in fish muscle for human consumption. Results of linear regression using \log_{10} -transformed THg concentrations are presented and raw data are shown in the graph. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

This was also in accordance with other results reported for GH, where values of $\delta^{13}\text{C}$ between -19.05‰ and -18.62‰ were found (Dennard et al., 2009). The reduction in lipid-corrected $\delta^{13}\text{C}$ in our study may be additional evidence for a trophic decline, since $\delta^{13}\text{C}$ also has been shown to increase with increasing trophic level (up to 1.5‰ per trophic position, Sweeting et al., 2007). The decline in $\delta^{13}\text{C}$ may, however, also indicate a modest temporal shift in the GH diet from a predominantly pelagic diet with some use of demersal prey to a more exclusively pelagic diet with less use of demersal prey over the course of the investigation period (Sweeting et al., 2007).

Linear regression analysis showed that the \log_{10} transformed Hg concentration increased with increasing length ($P < 0.0001$, $r^2 = 0.078$), $\delta^{15}\text{N}$ ($P < 0.00001$, $r^2 = 0.065$) and lipid-corrected $\delta^{13}\text{C}$ ($P < 0.0001$, $r^2 = 0.36$), but there was no relationship between \log_{10} transformed Hg and fat content ($P = 0.72$, $r^2 = 0.0002$) (Fig. 2), nor between \log_{10} transformed Hg and C:N ratio ($P = 0.64$ and $r^2 = 0.0004$). No difference in mean Hg concentration (\log_{10} transformed) was detected between the sexes of GH ($t = 1.75$, $df = 510$, $P = 0.08$).

Results from the Bayesian information theory AIC model showed that total Hg concentrations in GH were driven by a regime of conditions rather than by a single variable (Table 2). The model best explaining the variation in the data ($w_i = 0.82$) included year, $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$, followed by a model with length, $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ ($w_i = 0.17$), and the global model including the following four variables, length, year, $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ ($w_i = 0.01$). These results were supported by results of ANCOVA (Fig. 3, Table 3). The length normalized data from the ANCOVA analyses showed that total Hg concentrations in muscle tissue of GH decreased by $\sim 50\%$ over the ten years of the investigation (Fig. 3A). When Hg data were normalized by $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ using ANCOVA with $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values as covariates, the time trend decrease was reduced to $\sim 35\%$ (Fig. 3B) over the study period. Since length and $\delta^{13}\text{C}$ were not independent variables (Fig. 4A) they could not be included in the same ANCOVA analysis and were tested separately. However, to test the additional effect of fish size on the $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ adjusted Hg concentrations, we also performed the latter ANCOVA using a subset of the fish within the size class range of 60–70 cm which was equally present in all years. The results showed that the year-to-year variation for the $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ adjusted Hg concentrations in fish within this limited size class range was almost identical to the results for the complete dataset (compare Fig. 3B and C), indicating that variation in fish size, because of potential size class sampling bias, did not contribute significantly to the

Table 2

Poly-parameter Akaike's Information Criterion (AIC) model of predictors of total Hg in Greenland halibut sampled from 2006 to 2015 in Norway. Sample size ($n = 512$) reflects all cases in the data set used for the model that had no missing values. RSS represents residual sum of squares from the general linear regression models. K represents the number of fitted parameters plus the intercept and variance terms.

Model	K	n	RSS	AIC	Δ_i	w_i
Length	3	512	42.58	-1267.27	208.89	0.00
$\delta^{13}\text{C}$	3	512	30.27	-1441.95	34.21	0.00
$\delta^{15}\text{N}$	3	512	43.77	-1253.16	223.0	0.00
Fat Content	3	512	46.80	-1218.93	257.22	0.00
Year	3	512	38.16	-1323.38	152.78	0.00
Sex	3	512	46.69	-1220.15	256.01	0.00
Year + Length	4	512	36.56	-1343.29	132.87	0.00
Year + Fat Content	4	512	37.19	-1334.63	141.53	0.00
Year + $\delta^{13}\text{C}$	4	512	28.94	-1463.06	13.10	0.00
Year + $\delta^{15}\text{N}$	4	512	37.39	-1331.88	144.28	0.00
Year + Sex	4	512	38.16	-1321.40	154.76	0.00
Year + $\delta^{13}\text{C}$ + $\delta^{15}\text{N}$	4	512	28.80	-1465.51	10.65	0.00
Year + $\delta^{13}\text{C}$ + $\delta^{15}\text{N}$	5	512	28.10	-1476.16	0.00	0.82
Year + Length + $\delta^{13}\text{C}$	5	512	28.72	-1464.94	11.22	0.00
Year + Length + $\delta^{15}\text{N}$	5	512	35.48	-1356.67	119.49	0.00
Year + Length + $\delta^{13}\text{C}$ + $\delta^{15}\text{N}$	5	512	28.27	-1472.96	3.20	0.17
Global ^a	6	512	28.52	-1466.43	9.73	0.01

^a The global model includes all significant variables ($\delta^{13}\text{C}$, $\delta^{15}\text{N}$, length, and year).

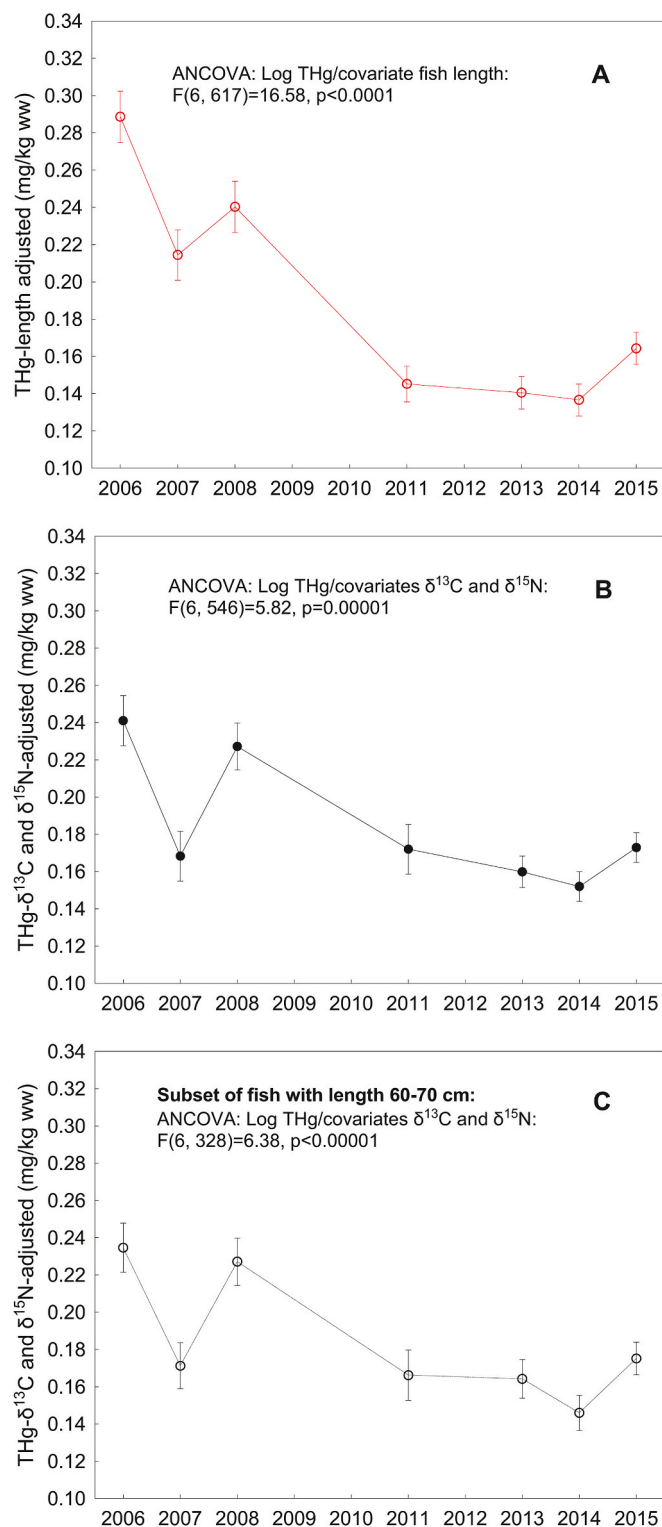


Fig. 3. Temporal trends of total Hg (THg) in muscle of Greenland halibut collected in the Norwegian Sea during 2006–2015. Annual mean \pm standard error (SE) of (A) length-adjusted THg-concentrations (mg/kg ww), (B) THg-concentrations (mg/kg ww) adjusted for lipid-corrected $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ in all fish, and (C) THg-concentrations (mg/kg ww) adjusted for lipid-corrected $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ in a subset of fish with length 60–70 cm. THg-concentrations were adjusted for length (A) or $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ (B, C) using ANCOVA. Data analyses were performed on \log_{10} -transformed Hg concentration data and raw data are presented in the graphs.

Table 3

Temporal trends in Greenland halibut Hg concentrations with normalization of length (cm), $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$. Analysis of covariance (ANCOVA) tests performed on \log_{10} -transformed THg data from filets of GH sampled during 2006–2015 in the Norwegian Sea.

Variable	F-Statistic	Df	P
Length	16.58	6, 617	<0.00001
$\delta^{13}\text{C}$	8.92	6, 547	<0.00001
$\delta^{15}\text{N}$	20.90	6, 547	<0.00001

observed variation in Hg-concentrations over time. The regime of conditions explaining the variation in total Hg in GH thus included the following trends: 1) Hg decreasing with time and its associated environmental conditions (sampling year), 2) Hg decreasing with lower trophic position and a possible shift to a more pelagic based diet ($\delta^{15}\text{N}$ and $\delta^{13}\text{C}$) (Table 2, Fig. 3).

3.2. Trophic shifts and food web dynamics

These results show that a large part of the observed decrease in GH Hg during 2006–2015, and particularly between 2008 and 2013 (2011 was excluded in the AIC model), can be explained by a combination of Hg decreasing with time, which may be due to reduced levels of environmental Hg, and a significant shift in trophic dynamics, both of which occurred concomitantly over the study period. The GH trophic shifts accounted for about one third of the observed temporal reduction in Hg. This shift in trophic dynamics includes a decrease in trophic position of the GH and possibly also a change to a more pelagic diet over the course of the investigation as indicated by the concomitant decrease in stable isotope values over the study period (Table 1, Fig. 3). During the same

time period, mean size of the fish also decreased (Table 1) and although linear regression analysis showed no relationship between $\delta^{15}\text{N}$ and fish length ($P = 0.84$, $r^2 = 0.0001$), the lipid-corrected $\delta^{13}\text{C}$ decreased with decreasing fish length ($P < 0.0001$, $r^2 = 0.14$) (Fig. 4). A weak positive statistical relationship was detected between fat content and fish length ($P < 0.0001$, $r^2 = 0.059$), but no statistical relationship was observed for C:N ratio and fish length ($P = 0.15$, $r^2 = 0.004$), indicating that fish length did not introduce any bias to the mathematical lipid correction of $\delta^{13}\text{C}$ where lipid content was determined from the C:N ratio (Eq. (2)). A change in diet might be suspected to be related to change in size, as GH may shift from a more pelagic prey base to a more benthic one as they grow older (Vollen et al., 2004; Solmundsson, 2007). Vollen and Albert (2008) showed that, in terms of weight, the contribution of cephalopods, shrimps, and most other small crustaceans decreased with increasing length of the GH predator. On the other hand, the contribution of herring (*Clupea harengus*) and most other fish increased with increasing predator length, while demersal fish like eelpouts and flatfish were only found in GH specimens larger than 65 cm, and redfish (*Sebastes* spp.) were eaten only by larger individuals generally >75 cm. However, in this study a subsample of the dataset with size range between 60 and 70 cm GH, equally present in all years, showed no evident change in the year-to-year variations in $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ compared to the complete dataset, suggesting that the size differences observed in this study had little effect on the trophic shift parameters (Fig. S3).

Changes in both $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ during the study period (Table 1) indicate a decrease in the average trophic position and possibly a simultaneous shift to a more pelagic diet. Linear regression showed only a very weak relationship between $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ ($P = 0.0025$, $r^2 = 0.016$) (Fig. 4C). However, a simultaneous change in these two factors makes sense, since both are influenced by trophic position, and because trophic position and feeding habitat are intricately linked. Pelagic fish

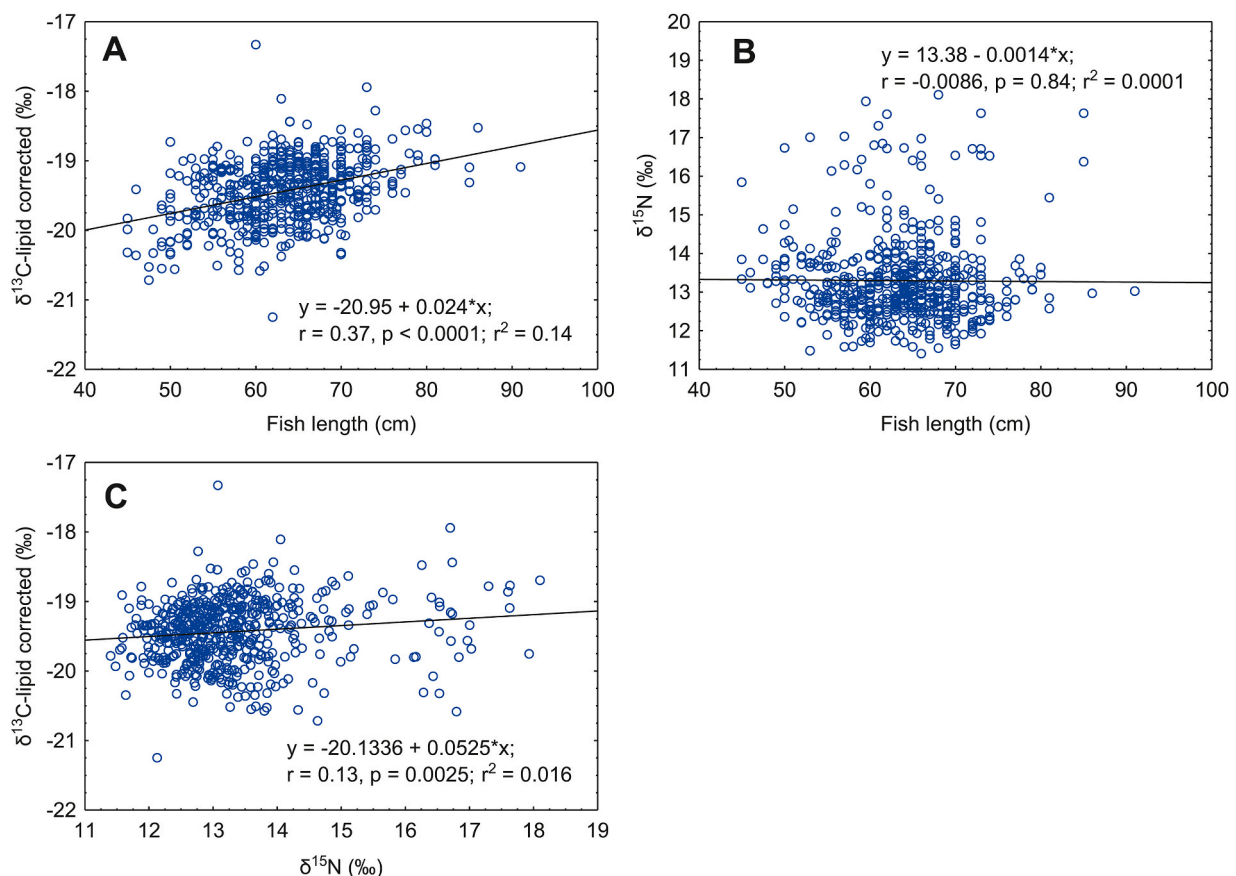


Fig. 4. Relationship between fish size and food web dynamics. Linear regression between (A) lipid-corrected $\delta^{13}\text{C}$ (‰) and fish length (cm), (B) $\delta^{15}\text{N}$ (‰) and fish length (cm) and (C) lipid-corrected $\delta^{13}\text{C}$ (‰) and $\delta^{15}\text{N}$ (‰) in muscle of GH sampled in the Norwegian Sea during 2006–2015.

that occur in large volumes in the Norwegian Sea, such as herring, mackerel (*Scomber scombrus*) or blue whiting (*Micromesistius poulassou*), are plankton feeders (i.e., having a trophic level of approximately 3), whereas a benthic diet will include demersal fish and invertebrates mainly feeding on other benthic organisms, which may result in a slightly higher average trophic level for GH. GH is known to be a generalist and opportunistic species, feeding on a wide array of available prey items (Solmundsson, 2007; Vollen et al., 2004; Dwyer et al., 2016; Giraldo et al., 2018). Thus, diet shifts may be a result of changing availability of pelagic prey and an increased abundance of pelagic fish at water depths inhabited by GH. Blue whiting abundance increased in the Norwegian Sea from 2010 to 2018 (ICES, 2019), however, it is unknown if this population change was related to the overall decrease in Hg concentrations in GH.

3.3. Changes in environmental Hg concentrations

In addition to the effects of shifts in trophic dynamics, time itself had a substantial effect on Hg levels in GH which was not due to size or feeding mode (Table 2, Fig. 3, Table 3), but may be due to reduced environmental levels of regional Hg. Data from the Norwegian Environment Agency show a linear decrease of about -3.6% per year in Norwegian Hg air emissions during the period 1994–2015 (Fig. 5) (Norwegian Environment Agency, 2020). This corresponds well with the decrease in the $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ normalized Hg concentrations of about -3.5% per year during 2006–2015 (Fig. 5B). Also, the length normalized Hg concentrations of GH mirrored the declines of regional Hg (Fig. 5A). Linear regression analysis with data from Fig. 5 showed a significant relationship between length normalized \log_{10} -transformed Hg concentrations of GH and total Hg air emissions ($P = 0.008$; $r^2 = 0.79$, $y = -1.14 + 0.001x$), and a similar trend between $\delta^{15}\text{N}$ and $\delta^{13}\text{C}$ normalized Hg concentrations and total Hg air emissions ($P = 0.059$; $r^2 = 0.54$, $y = -0.94 + 0.0005x$). This data should however be interpreted cautiously especially given the potential for lag times in responses of marine apex predators to global scale pollutants such as Hg (Vo et al., 2011). The national total releases (to air, water, and soil) of Hg in Norway were also reported to decrease from approximately 6 tons in 1985 to 0.5 tons in 2017 (Miljostatus.no, 2020). Norwegian national monitoring of Hg in air and precipitation showed a slow but steady decrease between 2006 and 2014 (Bohlin-Nizzetto et al., 2015). These reductions likely result from measures to reduce the total emissions of Hg, undertaken by national and regional levels of government in the Northeast Atlantic through the system of North Sea Conferences (OSPAR, 2020). Moreover, global emissions have been estimated to

have decreased by $\sim 20\%$ between 1990 and 2010 with larger decreases in emissions in Europe and North America more than outweighing increased emissions in Asia (Zhang et al., 2016).

Several recent investigations have reported declines (approximately -1.5 and -2.2% per year) of atmospheric total gaseous Hg concentrations over the North Atlantic Ocean (Slemr et al., 2011; Soerensen et al., 2012). Inventories of anthropogenic emissions were not in accordance with these declines and investigators have postulated that these declines may have been driven by decreased Hg concentrations via evasion in the upper North Atlantic Ocean or by changes in terrestrial surface–atmosphere fluxes (Slemr et al., 2011; Soerensen et al., 2012). However, significant uncertainties remain regarding the underlying mechanism of these observed declines.

Our investigation is one of a series of studies showing declining Hg concentrations in seafood species in different parts of the world, including bluefin tuna and bluefish from the Northwest Atlantic. These declining fish concentrations of Hg have been linked to regional declines in Hg emissions and deposition (Cross et al., 2015; Lee et al., 2016) but neither of these studies have considered changes in fish diet concomitantly with changes in Hg air emissions. Our research shows that the rapid temporal decline of Hg concentrations in GH is likely best explained and driven by a complex set of interwoven ecosystem-scale processes involving both changes in food web dynamics and declining air emissions of Hg (Table 2, Figs. 3, Fig. 5).

3.4. Study limitations

This investigation and supporting data have important limitations that should be considered. Specifically, since our food web modeling relies on the use of bulk $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ isotope estimates it is important to recognize that interpretation of the isotope changes is not straightforward for any study that uses these techniques to estimate spatial or temporal changes in food web dynamics. This applies especially when fish stomach contents data or information from compound-specific isotope analysis of amino acids (CSIA-AA) are lacking (Ishikawa, 2018). $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ bulk isotope values may change because of spatiotemporal changes in baseline values, and even though our study was conducted within a limited geographical area, changes in baseline values cannot be excluded. For $\delta^{13}\text{C}$, the change in baseline may also be partly driven by the Suess effect (i.e., a change in the ratio of the atmospheric concentrations of carbon isotopes, ^{13}C and ^{14}C , as a result of anthropogenic activities such as the burning of fossil fuels; Keeling et al., 1979; Olsen et al., 2006), however this effect is not well understood regarding its impacts on the ocean and marine food webs. Here we

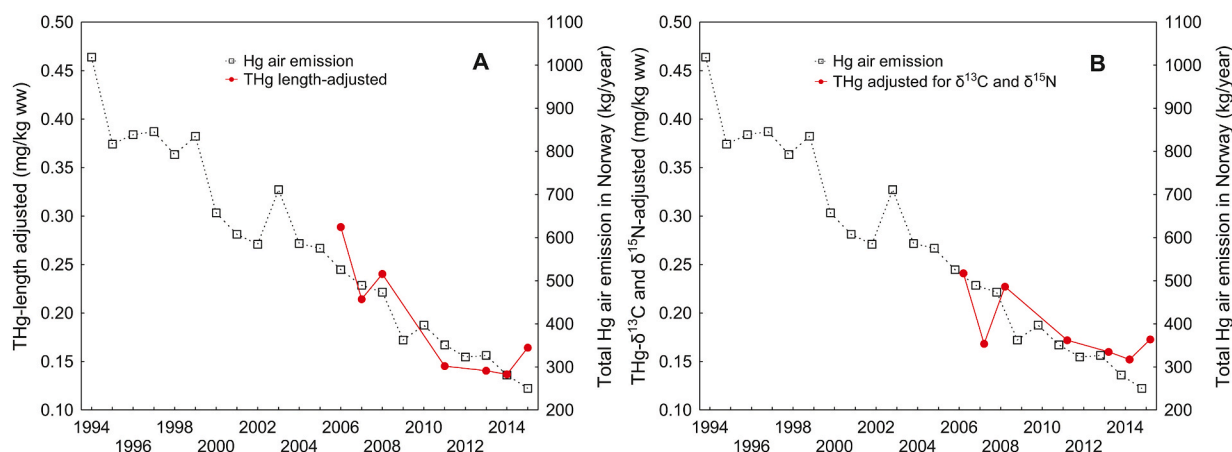


Fig. 5. Temporal trends in total Hg air emissions and total Hg (THg) in Greenland halibut fillet tissue. Annual means of total Hg air emission in Norway (kg/year; black open symbols, right axis) and THg-concentrations in muscle of GH (mg/kg ww; red closed symbols, left axis). THg-concentrations in GH muscle (least square means from ANCOVA) were adjusted for fish length in (A) and for lipid-corrected $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ in (B). Data for Hg air emissions are from Norwegian Environment Agency (2020). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

propose a set of hypotheses to help disentangle the observed decline in both $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ values. In addition to our interpretation, the observed isotopic changes may also be explained by the following plausible scenarios either individually or interactively: 1) a significant decline in trophic level within a common food web e.g. including regular feeding on mainly pelagic prey over the entire time period of this investigation, but shifting to lower trophic level prey items over time, 2) a shift to a more pelagic food web that does not necessarily translate to a change in trophic position, but rather is driven by changes in bulk stable isotope values related to different $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ baselines between the two food webs, and 3) a stable diet over time, but with temporal changes in the isotopic baselines driven by abiotic conditions or changes at the base of the food web (Lorrain et al., 2020). Another consideration is that our GH Hg data utilizes total Hg as a surrogate for MeHg. However, our high-resolution simultaneous GC-ICP-MS isotopic dilution Hg speciation results for fillet tissue of 20 fish of varying size and fat content, carefully selected to represent all sampling years and the entire sampling area of this study, showed that % MeHg was >77 % in all measured samples. Therefore, total Hg is a suitable and robust proxy for MeHg in GH fillet tissue. These findings demonstrate that fillet Hg was primarily in the highly neurotoxic MeHg form and our findings are also in good accordance with other marine fish studies that also used simultaneous GC-ICP-MS isotopic dilution techniques where % MeHg was reported to be >97 % in two snapper species (red snapper - *Lutjanus campechanus*, grey snapper - *Lutjanus griseus*) from the Gulf of Mexico (Bank et al., 2007). Future research on temporal dynamics of Hg in marine fish, such as GH, will likely benefit from the use of CSIA-AA (Ishikawa, 2018) and possibly Hg stable isotopes (Bergquist and Blum, 2007) to further elucidate MeHg source apportionment and the importance of energy flow and trophic dynamics.

3.5. Biomonitoring of apex predators in the context of the UN-MCM

Although some questions remain regarding the drivers of Hg bioaccumulation in GH, we feel this species is an excellent candidate for long-term biomonitoring of Arctic and sub-Arctic marine ecosystems, especially in the context of the newly established UN-MCM (UNEP, 2013). A critical component of Article 19 of this convention will be the collection and interpretation of large-scale, marine fish biomonitoring data and the establishment of baseline estimates of Hg for seafood species commonly consumed by humans. Here we have supplied reliable and reproducible temporal trend data for GH, from an accredited laboratory, prior to the establishment of the UN-MCM. Our data set therefore serves as a baseline and an effective model for the evaluation of the effectiveness of the UN-MCM for three reasons: a) in contrast to Hg modeling studies, which often operate using unrealistic assumptions (Bank et al., 2020), our investigation utilizes a large set of empirical data and information from an existing national seafood safety surveillance program with official laboratory accreditation, b) our sampling approach allows for temporal trend evaluation and assessments of changes in Hg levels in this population over time scales relevant to the convention (i.e. starting within six years after the entry into force of the convention, as stated in Article 22.1) and c) our Hg temporal assessment also evaluates concomitant changes in food web structure, trophic shifts and declining Hg emissions over the period of investigation which took place before the UN-MCM entered into force. Collectively these factors are critical for standardization of global fish biomonitoring programs in support of the effectiveness evaluation of the UN-MCM. We also recommend that long-term marine fish biomonitoring programs use standardized sampling approaches (Bank et al., 2014). Specifically, we recommend incorporating assessments of Hg emission data where possible, and to utilize carbon and nitrogen stable isotope data to estimate energy flow and trophic dynamics, respectively, as opposed to solely relying on Hg fish concentration data. Speciation data of Hg may also be required for some taxa as well, especially for those species that occupy lower trophic positions. By using such an approach, this study

has demonstrated a rapid decline in Hg concentrations in GH which may reflect both shifts in food web structure and trophic position as well as declining emissions due to the effectiveness of pollution mitigation measures.

4. Conclusions

Our GH Hg data normalized for fish length or $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ showed that fillet Hg concentrations in this species decreased, rapidly, by ~35–50 % over a ten-year period from 2006 to 2015. Total Hg in fillet tissue was predominantly in the MeHg form (>77 %). A Bayesian information theory model showed that decreasing GH Hg was associated with declines in Hg air emissions, decreasing trophic position, and possibly lower demersal prey use. Overall, our study demonstrates the importance of simultaneously considering Hg emissions, food web dynamics, and trophic position as important drivers of Hg bioaccumulation in GH and highlights the effectiveness of Hg regulations on ocean apex predator Hg concentrations and overall seafood safety.

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Author contributions

M.S.B. and B.M.N. conceived the project, designed the study, analyzed data, and were the co-primary authors. S.F., A.D., K.N., and A. M. assisted with sample collection, data analysis and interpretation, and writing. E.T. and D.A. oversaw and conducted laboratory work on mercury speciation and assisted with writing.

Data availability

Data related to this paper may be requested from the corresponding author.

Credit author statement

Michael S. Bank: Conceptualization, Investigation, Formal analysis, Writing-Original draft preparation, Reviewing, and Editing, Sylvia Frantzen: Formal analysis, Investigation, Writing-Reviewing, and Editing, Arne Duinker: Formal analysis, Investigation, Writing-Reviewing, and Editing, David Amouroux: Formal analysis, Writing-Reviewing, Emmanuel Tessier: Formal analysis, Writing-Reviewing, Kjell Nedreaas: Formal analysis, Investigation, Writing-Reviewing, Amund Maage: Formal analysis, Investigation, Writing-Reviewing, and Bente M. Nilsen: Conceptualization, Investigation, Formal analysis, Writing-Original draft preparation, Reviewing, and Editing.

Declaration of competing interest

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

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envpol.2021.117843>.

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