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Mercury bioaccumulation pathways in tusk (*Brosme brosme*) from Sognefjord, Norway: Insights from C and N isotopes☆

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ABSTRACT

Seafood is the main source of methylmercury (MeHg) exposure for humans and elevated total mercury (Hg) concentrations have been reported in marine fish from Norwegian fjords compared with offshore areas. Hg in tusk fillets (n = 201) and liver samples (n = 177) were measured in individuals from different habitats including offshore, coastal area, outer and inner Sognefjord. Specifically, the effects of habitat, energy sources and trophic complexity on Hg bioaccumulation pathways in tusk (*Brosme brosme*) were investigated using stable isotopes of carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$). The concentrations of Hg in tusk increased from offshore towards inner Sognefjord. While Hg concentrations in sediment were at background levels, tusk fillet samples from 7 of 8 sites in Sognefjord had higher Hg levels than the maximum level set by European Union. Based on these findings, human consumption advice for tusk from Sognefjord was issued by the Norwegian Food Safety Authority. $\delta^{13}C$ values in tusk successfully discriminated individuals from different habitats and were positively correlated to Hg concentrations in tusk across individuals, sites and habitats, outlining the potential importance of terrestrial carbon and most likely the atmospheric deposition of Hg from the catchment to the overall Hg bioaccumulation and exposure regime in tusk. Additionally, we postulate that the effects of terrestrial carbon sources increased towards inner Sognefjord and likely influenced Hg bioavailability throughout the food web. In contrast, $\delta^{15}N$ values were patchy throughout the fjord system and although trophic position explained some of the Hg variation between individual fish, it was not correlated with Hg variation across sites and habitats. Our results suggest that tusk can accumulate high levels of Hg in fjord ecosystems and that catchment runoff is likely an important driver of Hg bioaccumulation in this species.

1. Introduction

Mercury (Hg) is a global pollutant and an important environmental and public health issue (Mason et al., 2012). Monomethylmercury (MeHg), the most toxic and bioavailable form, has a very long half-life in fish and biomagnifies with increasing trophic position in marine food webs. In remote regions, such as Norway, atmospheric deposition is the main source of Hg in most areas (Fitzgerald et al., 1998; Berg et al., 2006) although some point sources do exist (Azad et al., 2019a). While natural sources of Hg exist, anthropogenic sources are the main contributors of Hg to the environment (UNEP, 2013). Mercury is highly volatile and has a long atmospheric residence time. Hg can be transported long distances and deposited directly into marine ecosystems or into terrestrial catchments, washed into streams, rivers and lakes and subsequently delivered to estuarine habitats, fjords and coastal areas (Schartup et al., 2015; Obrist et al., 2018).

Atmospheric Hg largely exists as gaseous (Hg^0) or inorganic forms in very low concentrations and long range, hemispheric transport has been documented (Fitzgerald et al., 1998). The majority of Hg (more than 95%) in the aquatic ecosystem exists as inorganic Hg (iHg) (Wiener et al., 2002), and can undergo methylation in several aquatic ecosystem compartments including sediments, the water column and within biota. However, despite intensive study, the fine scale chemistry of the Hg methylation

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phenomenon remains poorly understood. MeHg is produced mostly by anaerobic sulfate reducing and iron bacteria in low oxygen water (Topping and Davies, 1981; Sunderland et al., 2009) and sediments (Compeau and Bartha, 1987; Gilmour et al., 2013). Trophic transfer of MeHg is highly efficient and the majority of Hg in fish fillet exists in the MeHg form (Magalhães et al., 2007; Hong et al., 2012) and total Hg (Hg) can be used as a reliable proxy for MeHg in this tissue type. Moreover, a previous companion investigation reported that MeHg represented >95% of the Hg in tusk fillet tissue (Azad et al., 2019b).

Fillet and liver are important organs for Hg bioaccumulation in fish (Khadra et al., 2019; Looi et al., 2016). As a result of bioaccumulation, Hg concentrations in these organs usually increase with both age, weight and length of marine fish species (Magalhães et al., 2007; Suzuki et al., 1973), and fish length may be used as a proxy for fish age when studying bioaccumulation (Scudder et al., 2009). Due to its persistence and efficient trophic transfer, MeHg is known to biomagnify in the food chain, and the highest Hg levels are often found in top trophic level organisms including predatory fish species.

Fjords represent an important component of marine ecosystems along the coast of Norway and provide a wide array of ecosystem services including commercial and recreational fishing. Therefore, it is important to understand the ecological conditions and parameters driving Hg biomagnification in fjord food webs from the abiotic environment up to predatory species, the latter which are often suitable bioindicators of contaminants and environmental change (Hazen et al., 2019).

Tusk (*Brosme brosme*) is a deep sea, predatory fish that can bioaccumulate high levels of Hg compared to many other fish species in the Northeast Atlantic (Azad et al., 2019b). For example, in the inner parts of the Hardangerfjord ecosystem, a fjord system ca. 100 km south of Sognefjord polluted by industry and other anthropogenic Hg pollution sources, including a zinc plant, tusk have been found to have concentrations of Hg more than three times the EU maximum level of 0.5 mg kg^{-1} ww (EC, 2006; Azad et al., 2019a). Tusk from other fjord ecosystems, without pollution point sources, have also been reported to have elevated concentrations of Hg (Berg et al., 2000). Moreover, tusk is considered a good bioindicator species of MeHg contamination in fjord ecosystems since they inhabit deep water, use local resources, exhibit low vagility and are widely distributed throughout fjords and offshore habitats.

In a companion study (Azad et al., 2019a) we reported that in Hardangerfjord, Norway, Hg concentrations in marine biota, seawater and sediment increased towards the point source of Hg pollution in the inner part of fjord. However, another branch of the inner Hardangerfjord had high Hg concentrations in biota and much lower Hg in surface sediments. This raised the question whether Hg bioaccumulation in marine biota inhabiting fjords is largely driven by point sources of pollution or by spatial variability in ocean biogeochemistry and food web dynamics, caused by a freshwater input gradient towards the inner sectors of the fjord. In order to address this, we conducted a follow up study in Sognefjord, which has similar ecological conditions as Hardangerfjord including great depths, a sill at the mouth of the fjord, and large freshwater inputs but without any known significant point sources of Hg pollution.

We hypothesized that large amounts of freshwater delivered from local catchments, containing organic matter bound Hg, would create an observable spatial gradient in Hg bioavailability from offshore to inner Sognefjord. We also predicted that both carbon sources and trophic position would be important drivers of Hg bioaccumulation in demersal tusk. To investigate this, we measured Hg, $\delta^{13}C$ and $\delta^{15}N$ in tusk from different positions, from the open North Sea to the innermost part of Sognefjord. We also compared our findings of this study, which was conducted in a fjord with a Hg pollution regime without a known point source of Hg, with Hg concentrations in tusk inhabiting Hardangerfjord, a highly impacted and Hg polluted fjord with a local point source.

2. Materials and methods

2.1. Study area

Sognefjord is one of the longest and deepest fjords (1308 m at deepest point) in the world, located at 61°N on the western coast of Norway in the Northeast Atlantic Ocean (Fig. 1). Sognefjord stretches ~205 km inland. The Sognefjord profile is U shaped and in most parts the depth of the middle part is > 1000m. Sognefjord has a sill (~300m depth) close to the mouth and branches out into several side arms in the inner part (Fig. 1) and is considered a highly complex ecosystem with dynamic water residence times and circulation patterns.

2.2. Tusk and sediment sampling and preparation

Tusk were captured using long line fishing with Atlantic mackerel (*Scomber scombrus*) as bait at 200–1000m depths in Sognefjord (sites 1–8), the North Sea coast (site 9) and offshore in the North Sea (site 10) during March–August (Fig. 1). In Sites 1–9 tusk sampling took place in 2015 and in site 10 tusk was sampled in 2013. A total of 14–25 individual fish were sampled at each site. Sites 1–4 were defined as “inner Sognefjord”, sites 5–8 were defined as “outer Sognefjord”, site 9 was desginated as “North Sea coast” and site 10 was categorized as “offshore North Sea” (Fig. 1).

After sampling, whole tusk were frozen before being shipped to the laboratory at Institute of Marine Research (IMR), Bergen, Norway. Total weight and total length (measured from the tip of the snout to the end of the caudal fin) were recorded and individual fillet samples from one side (skin and bone free) were homogenized in a food processor prior to being lyophilized, homogenized and stored in a sterile, Hg free, plastic container. Tusk livers from fish sampled at all sites except site 10, were dissected out, homogenized, and analyzed, omitting lyophilization due to the high fat content. After lyophilization of the fillet samples to a constant mass, the water content (% moisture) was recorded. Concentrations were measured on the dried sample material and back calculated using the percent moisture data and presented on a wet weight (ww) basis.

Sediment samples were collected in the Sognefjord during a cruise aboard the *R/V Håkon Mosby* during July 2016. Sediment samples were taken from the deepest part of the fjord at each site (Fig. 1) using a multi-corer (KC Denmark, Silkeborg, Denmark) and transparent PVC tubes (diameter: 100 mm; Length: 600 mm). The redox potential (E_h) profiles were measured using a portable electrode and by inserting the probe in different depths along the core at 1, 3, 6, 9, 12, 15 and 27 cm to characterize the oxic and anoxic profiles. Sediment samples were lyophilized to a constant dry mass and homogenized prior to analyses.

2.3. Mercury concentration measurements in tusk

The concentrations of Hg in individual fillet and liver samples of tusk were determined using an inductively coupled plasma-mass spectrometer (iCapQ ICP-MS, ThermoFisher Scientific, Waltham, MA, USA) equipped with an autosampler (FAST SC-4Q DX, Elemental Scientific, Omaha, NE, USA) following wet digestion in a microwave oven (UltraWave, Milestone, Sorisole, Italy). The

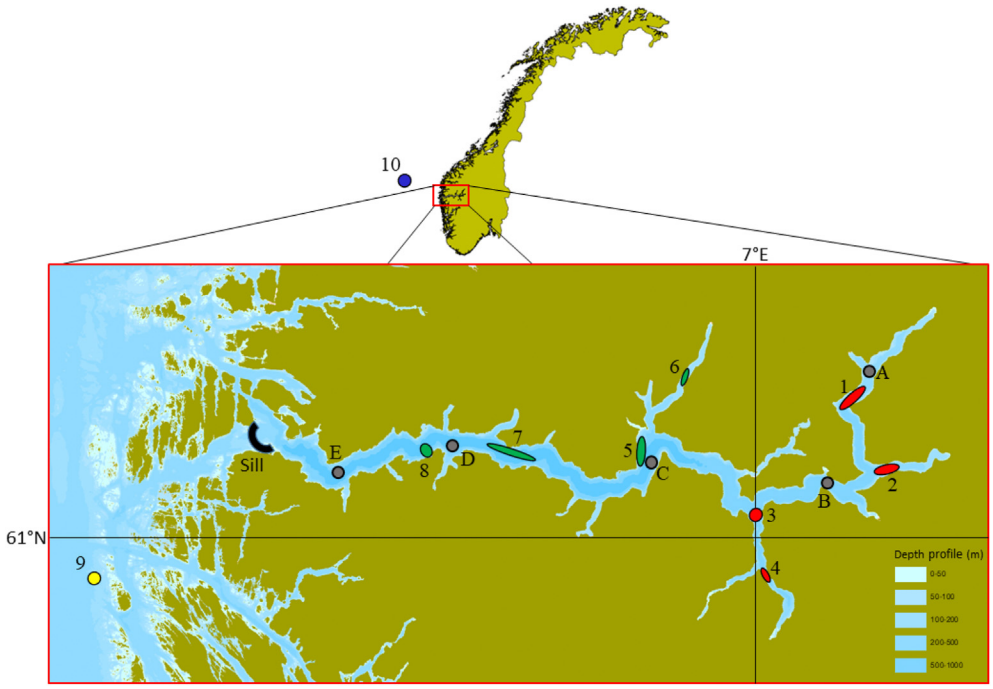

Fig. 1. Geographical location of sites (numbers 1–10) sampled for tusk during 2013 and 2015, and sediment sampling sites (grey circles, A to E) sampled in 2016 from Sognefjord, Norway. Tusk sampling areas are presented with color coding and categorized by habitat using the following scheme: red = inner Sognefjord; green = outer Sognefjord, yellow = North Sea coast, and blue = offshore North Sea. Site 10 is an offshore marine habitat sampling site (outside detailed map). The position of the study area in Norway is showed as a red rectangle. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

method is a CEN standard and is accredited for Hg according to NS-EN ISO 17025 (NMKL, 2007; CEN, 2009). The method was described in detail by Julshamn et al. (2007). Applied internal standards included rhodium (Rh), thulium (Tm) or germanium (Ge). The accuracy and precision of the method were controlled by analyzing certified reference materials (CRMs) including CRM1566 (oyster tissue) from the National Institute of Standards and Technology (Gaithersburg, USA) and lobster hepatopancreas (TORT-3) from the National Research Council (Ottawa, Canada) with the certified values of 0.037 ± 0.001 and 0.29 ± 0.02 mg kg^{-1} respectively. The mean ± SD recoveries of Hg were 0.03 ± 0.003, 87% recovery and 0.26 ± 0.08, 88% recovery for CRM1566 and TORT-3 respectively and well within the accredited range of 80%–120% for the whole period of analysis. Participation in proficiency tests during 2016 analyzing fish muscle or liver resulted in Z-scores between −0.78 and 0.13. The limit of quantification (LOQ) for Hg was 0.005 mg kg^{-1} dry weight (dw). Measurement uncertainties were 25% at 0.05–0.5 mg kg^{-1} and 20% at > 0.5 mg kg^{-1} Hg concentrations.

2.4. Mercury concentration measurements in sediment

Hg in sediment was measured using an AMA 254 trace mercury analyzer, (Altec, Czech Republic) by thermal decomposition, amalgamation and atomic spectrometry (US EPA method 7473). During the analyses, blank samples were analyzed with mean ± SD of 0.41 ± 0.16 ng and the mean value was subtracted from all samples.

Three certified reference materials (CRMs) including lichens BCR482 (0.48 ± 0.02), Tuna muscle ERM-CE464 (5.24 ± 0.1) and waterway sediment NIST1944 (3.4 ± 0.5) were used for measurement quality control. Results of Hg (mean ± SD) and recoveries for these CRMs were as follows: BCR482 0.45 ± 0.05 (95% recovery), ERM-CE464 4.88 ± 0.12 (93% recovery), and NIST1944 3.61 ± 0.65 (106% recovery). All sediment measurements were conducted at CNRS/University of Pau, Pau, France.

2.5. Stable isotope measurements of $\delta^{13}C$ and $\delta^{15}N$

A subsample of lyophilized and pulverized tusk fillet tissue and sediment samples were analyzed for stable isotopes of C and N using Eurovector EA3028 Elemental Analyser and Horizon isotope ratio mass Spectrometer (EA-IRMS). Stable isotopes (SI) were calculated as $\delta^{13}C$ and $\delta^{15}N$ using the following formula:

$$\delta X = \left(\left(R_{sample} \,/\, R_{standard}\right) - 1\right) * 1000$$

where X is ^{13}C or ^{15}N and R is $^{15}N/^{14}N$ or $^{13}C/^{12}C$ for sample and standard. Results are presented in ‰, and VPDB (Vienna Pee Dee Belemnite) and atmospheric nitrogen were used as international standards for carbon and nitrogen, respectively.

The $\delta^{15}N$ composition of IFE trout was calibrated using reference materials IAEA-N-1 and IAEA-N-2. The $\delta^{13}C$ composition of IFE trout was calibrated using the USGS-24 standard. Average (±1SD) values for IFE trout are as follows: $\delta^{15}N_{AIR}$: 11.60‰ ± 0.20 and $\delta^{13}C_{VPDB}$: -20.22‰ ± 0.19. Average (±1SD) results for 16 analyses of the IFE trout standard analyzed together with the samples were as follows: $\delta^{15}N_{AIR}$: 11.69‰ ± 0.06 and $\delta^{13}C_{VPDB}$: -20.23‰ ± 0.10. All carbon and nitrogen stable isotope analyses were conducted at the Institute for Energy Technology (IFE), Kjeller, Norway.

2.6. Statistical analyses

Data were log transformed to meet the assumptions of normal distribution and homogeneity of variances prior to statistical analyses as required. Analysis of covariance (ANCOVA) was used for

comparison of Hg concentrations across sampling sites and different habitats with length as a covariate to control for the effects of fish length on Hg bioaccumulation. One-way analysis of variance (ANOVA) was used to similarly compare spatial variation of parameters which were not correlated to size. For post-hoc comparisons, unequal sample Tukey-HSD tests were used to evaluate the effects of unequal sampling efforts and unbalanced design. Linear regression was used to test the relationship between fillet or liver Hg concentrations and tusk length. To evaluate the effects of trophic position and carbon sources on spatial variation of Hg in tusk, the relationships between LS mean Hg of tusk fillet from each site (corrected for length) and mean $\delta^{15}N$ and $\delta^{13}C$ were evaluated using linear regression. Nonparametric Kendal Tau test was used in a correlation matrix across the parameters measured in tusk fillets and liver samples and distances from the open ocean and the innermost point of the fjord. Distance from open ocean was calculated as distance from site 10 (Fig. 1) and distance from the inner fjord was calculated as distance from the inner most point of the Lustrafjord (site 1). All distances were measured as shortest distance through the fjord including all curviness. Statistical significance was accepted at $P < 0.05$ (Zar, 2010). Statistical analyses were performed using STATISTICA 13 (Statsoft Inc., Tulsa, USA) or Graphpad Prism 7.02 (Graphpad software Inc., San Diego, CA, USA).

3. Results

3.1. Mercury concentrations in tusk fillets and liver samples

Mean Hg concentration in tusk fillets varied between 0.24 mg kg^{-1} ww at site 9 on the North Sea coast and 1.2 mg kg^{-1} ww at site 1, the innermost site in Lustrafjord (Table S1, Fig. 2A). The mean liver Hg levels varied between 0.19 mg kg^{-1} ww in tusk collected at site 9 and 2.6 mg kg^{-1} ww in tusk collected at site 7 (Fig. 2B). In general, Hg concentrations in tusk from offshore and coastal areas were lower than in tusk sampled within Sognefjord.

Tusk length varied significantly across sites ($F(9, 191) = 9.08$; $P < 0.0001$; Fig. 2C) and there was a positive correlation between fillet Hg concentrations and fish length overall (Fig. 2E) and at all sites except site 9 (not shown). The liver Hg concentrations were not correlated to fish length at all sites ($P > 0.05$), and statistically significant relationships were observed only at sites 6 and 3 ($P < 0.05$; $R^2 = 0.22$ and 0.49). To remove the potentially confounding effect of fish length, ANCOVA was applied when comparing Hg in fillets across sites and habitats. This analysis showed that length adjusted fillet Hg concentrations varied significantly across habitats ($F(3, 196) = 67.9$; $P < 0.0001$) (Fig. 3C). The least square (LS) mean (adjusted for mean length) of Hg concentrations in tusk fillet (mg kg^{-1} ww) was highest in the inner

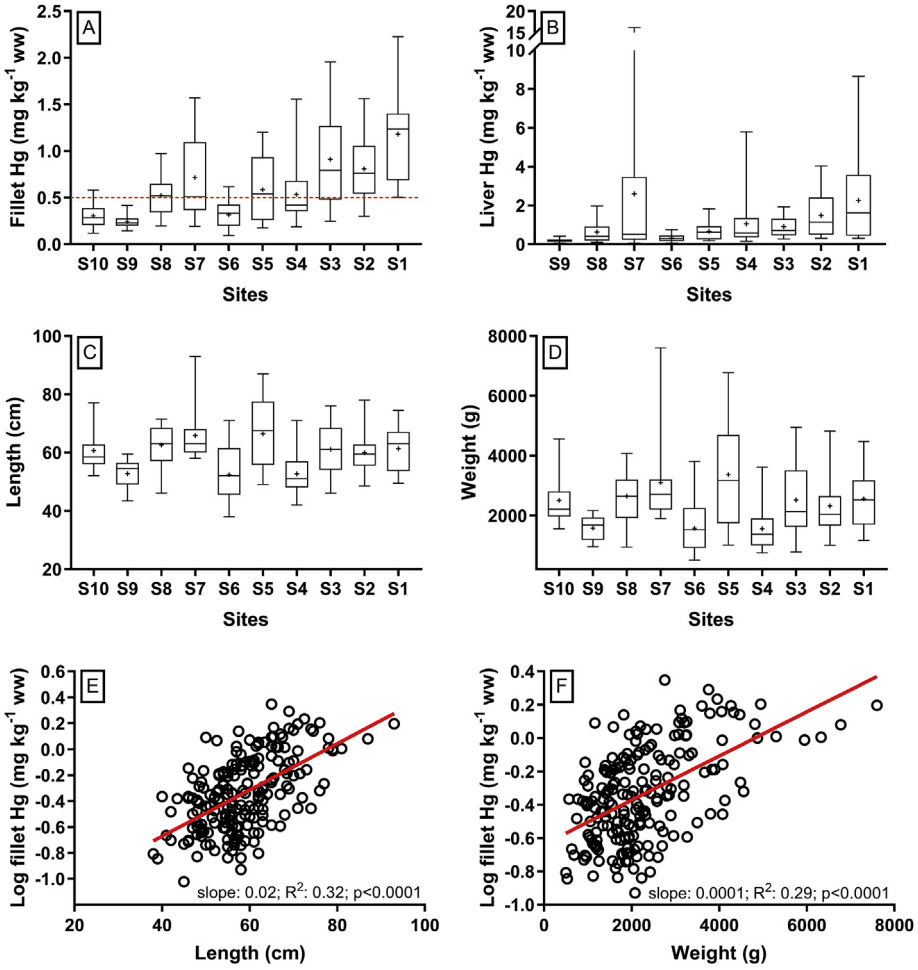

Fig. 2. Box plot (min-max, inter quartile, median and mean) of fillet Hg concentrations (A), liver Hg concentrations (B), fish length (C) and weight (D) and fillet Hg as a function of fish length (E) and weight (F) in tusk samples collected from Sognefjord during 2013 and 2015. The dashed red line in graph A represents the EU maximum limit of Hg (0.5 mg kg-1 ww). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

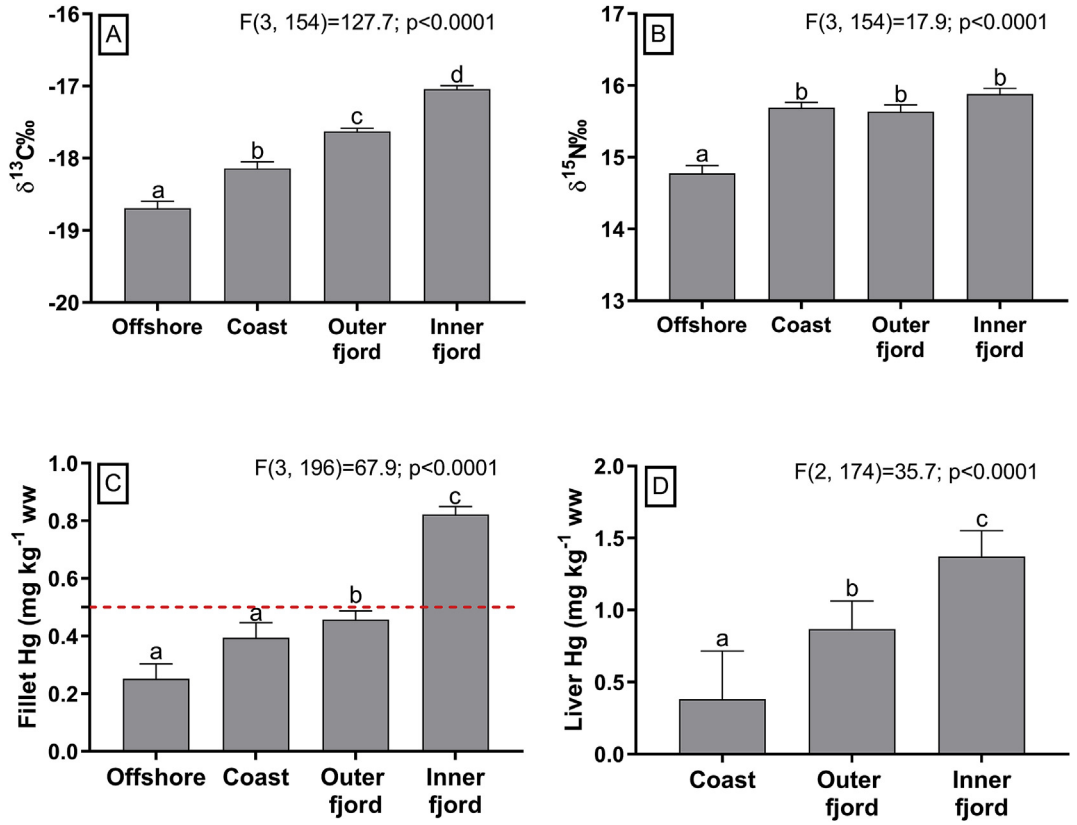

Fig. 3. Mean $\delta^{13}C‰$ values (A); $\delta^{15}N‰$ values (B); LS mean Hg concentrations in tusk fillets (C) and Hg concentrations in tusk liver samples (D) collected from Sognefjord during 2013 and 2015. Data are log transformed prior to analysis (except A and B) and raw data are presented in each graph. The dashed red line in graph C represents the EU maximum limit of Hg (0.5 mg kg^{-1} ww). Error bars represent one standard error. Letters represents significant differences between habitats. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

fjord (0.82; n = 81) followed by outer fjord (0.46; n = 71), coastal areas (0.39; n = 25) and offshore habitats (0.25; n = 24) respectively. Hg concentrations between coast and offshore were not different ($P > 0.05$). Similarly, the mean Hg concentrations (mg kg^{-1} ww) in liver of tusk from the different habitats were highest in inner fjord (1.36; n = 81), intermediate in the outer fjord (0.94; n = 71) and lowest in the coastal area (0.19; n = 25) (Fig. 3D). Hg concentrations in liver samples also varied significantly between all habitats (ANOVA: F (2, 174) = 35.7; $P < 0.0001$; Fig. 3D).

Length adjusted (LS mean) Hg concentrations in tusk fillets also varied significantly across individual sites (Fig. 4A). Site 1 (inner Sognefjord) had the highest LS mean Hg concentrations in tusk fillet and site 10 had the lowest (Fig. 4A). The outer Sognefjord sites (sites 5–8) had similar LS mean Hg concentrations which were substantially lower compared to the inner fjord sites. Tusk from coastal areas (site 9) had similar LS mean Hg fillet concentrations compared to outer Sognefjord (sites 5 and 8) and the offshore site (site 10) had the lowest observed LS mean Hg concentrations.

Except at site 7, where the largest tusk specimens were collected, liver sample Hg levels gradually increased from the coast towards the inner fjord sites (Fig. 2B), in the same manner as fillet Hg concentrations (Fig. 2A), with the exception of site 10 where no liver samples were taken. Hg concentrations in tusk liver were higher than in fillet at all the fjord sites. However, at the coastal site (site 9) the mean Hg concentration was higher in fillets compared to liver samples (Table S1). Lastly, LS mean Hg concentrations in fillet and mean Hg concentration in tusk liver samples were both negatively correlated with distance from the head of the fjord (Kendall tau = −0.82 and −0.61 respectively; Table S5).

3.2. Spatial variation in $\delta^{15}N$ and $\delta^{13}C$

Mean values of $\delta^{15}N$ measured in tusk fillets were 14.8 and 14.9‰ at sites 10 and 6, respectively, which was significantly lower than the values between 15.6 and 16.1‰ at all other sites (Table S1 and S2; Fig. 4B). Across habitats, mean $\delta^{15}N$ was significantly lower offshore compared to the coast and inner and outer fjord sectors, and the coastal, inner, and outer fjord areas were all statistically similar (Fig. 3B). Mean values of $\delta^{13}C$ varied between sites from −18.7‰ at the offshore site 10 to −16.9‰ at site 2 (Table S1), and there was a significant trend of increasing $\delta^{13}C$ (i.e., more positive values) from open ocean to the inner parts of the fjord. $\delta^{13}C$ increased between habitat types from offshore to coast to outer fjord to inner fjord (ANOVA F (3, 154) = 127.7, $P < 0.0001$) (Fig. 3A). Moreover, distance from the head of the fjord showed a strong negative correlation with $\delta^{13}C$ (Kendall tau = −0.87; Table S5), whereas no such correlation was observed for $\delta^{15}N$.

$\delta^{13}C$ values in tusk fillets had a weak relationship with individual fish length ($R^2 = 0.04$; $p < 0.05$) whereas $\delta^{15}N$ was moderately correlated with length ($R^2 = 0.23$; $p < 0.0001$; Figure S1). Tusk fillet $\delta^{13}C$ and $\delta^{15}N$ values were moderately correlated at the individual level ($R^2 = 0.32$; $p < 0.0001$) (Figure S1).

3.3. Mercury and $\delta^{15}N$ and $\delta^{13}C$ in tusk

Hg fillet concentrations were positively correlated with $\delta^{15}N$ values overall ($R^2 = 0.41$; $P < 0.0001$) and at each site ($R^2 = 0.28–0.78$) except for sites 1 and 3 (Table S3). Fillet Hg concentrations were also significantly positively correlated with $\delta^{15}N$

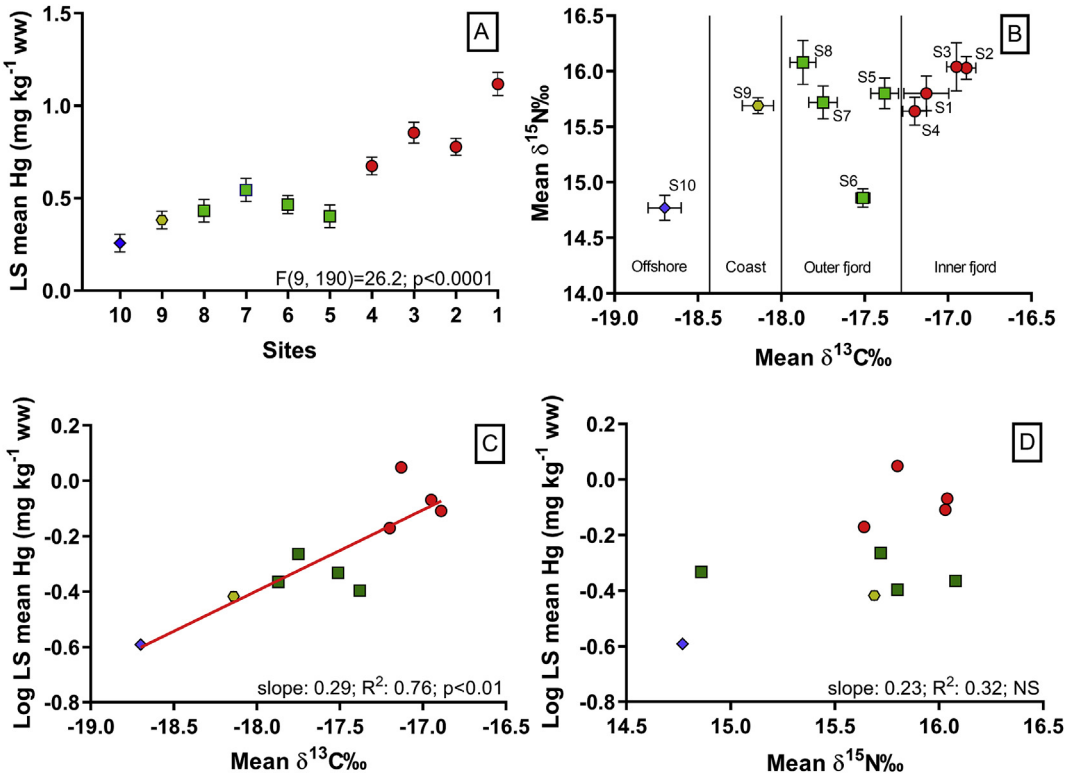

Fig. 4. LS mean Hg concentration (adjusted for length) across sampling sites (A), bi-plot of mean δ^{13}C‰ versus mean δ^{15}N‰ categorized by habitat type (B), and the relationship between LS mean Hg and mean δ^{13}C‰ values (C), and mean δ^{15}N‰ values (D), of tusk fillets collected from Sognefjord, Norway during 2013 and 2015. Color coding follows the habitat categorization scheme from graph (B) (red circles = inner Sognefjord; green squares = outer Sognefjord, yellow hexagon = North Sea coast; blue diamond = offshore North Sea. Error bars represent standard error. NS = not significant. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

within all four habitats ($R^2 = 0.16–0.61$) (Table S4). However, length adjusted LS mean fillet Hg concentrations for each site were not significantly correlated with mean δ^{15}N values (Table S5; Fig. 4D).

Tusk fillet Hg concentrations also showed a positive correlation with δ^{13}C overall ($R^2 = 0.44$, $P < 0.0001$) and within all habitats ($R^2 = 0.09–0.73$). Length adjusted LS mean fillet Hg of tusk from the different sites was strongly correlated with mean δ^{13}C, and this correlation explained 76% of the spatial variation across sites ($R^2 = 0.76$; Fig. 4C). Liver Hg level was moderately correlated to δ^{13}C overall ($R^2 = 0.32$; $P < 0.0001$) and δ^{15}N overall ($R^2 = 0.25$; $P < 0.0001$) (Figure S1). However, mean liver Hg concentrations at the different sites were not correlated with δ^{13}C or δ^{15}N values (Table S5).

3.4. Mercury and δ^{15}N and δ^{13}C in sediment

Redox potential (E_h) of surface sediments (top 1–3 cm) of all sampling varied between 32.3 and 174.0 mV. Redox potential values turned into negative values at 6 cm and had a decreasing trend from 6 to 27 cm depth (between −41.5 mV and −240.8 mV). Based on this, we classified sediments <3 cm core depth as the oxic layer, and between 3 and 6 cm as sub-oxic, and >6 cm core depth as the anoxic layer. Oxic layer sediments also had a different color (pale brown) compared to the anoxic layer (grey). Sub-samples from the oxic (1–3 cm) and anoxic layers (30–40 cm) were analyzed for Hg. Hg concentrations in the oxic sediment layer (1–3 cm) varied from 0.024 mg kg^{-1} dw at site A in the innermost part to 0.11 mg kg^{-1} dw at site C in the middle of the fjord (Fig. 5A). Oxic layer sediments from outer Sognefjord had higher Hg concentrations compared with the inner fjord sector. In both the oxic and the anoxic sediment layers δ^{13}C values increased from site A to site E (Fig. 5B), while δ^{15}N did not exhibit any spatial trend (Fig. 5C).

4. Discussion

4.1. Spatial distribution of mercury in tusk and sediment

Hg concentrations in both fillet and liver samples of tusk increased from the open North Sea towards the inner Sognefjord. Tusk, as a commercially important fish species in Norway and an important fish species for recreational fishing in the Sognefjord, had fillet Hg concentrations above the EU and Norway's maximum level (0.5 mg kg^{-1} ww) in most parts of the Sognefjord. Therefore, the Norwegian Food Safety Authority has previously issued consumption advice for this species from this area (https://www.matportalen.no), warning the public against consuming tusk from the greater part of the fjord.

The length adjusted Hg concentrations (LS mean) in fillets of tusk from the innermost site, Lustrafjord, (1.22 mg kg-1 ww) was only 14% lower than in fillet of tusk sampled from Sørfjord in Hardangerfjord (1.42 mg kg^{-1} ww), a nearby fjord which has a known point source of mercury (Azad et al., 2019a). The difference between the two sites was not significant (ANCOVA $F(1, 21) = 2.57$; $P > 0.05$; figure S2). Hg concentrations in tusk from the offshore area in this study were similar compared to tusk sampled from the North Sea (Azad et al., 2019b). The Hg concentration in liver samples showed a mean value of 2.26 mg kg^{-1} ww at the innermost site, which was much lower than in Sørfjord, where a concentration of around 8 mg kg^{-1} ww was found in a pooled liver sample.

Hg concentrations in sediment from Sognefjord

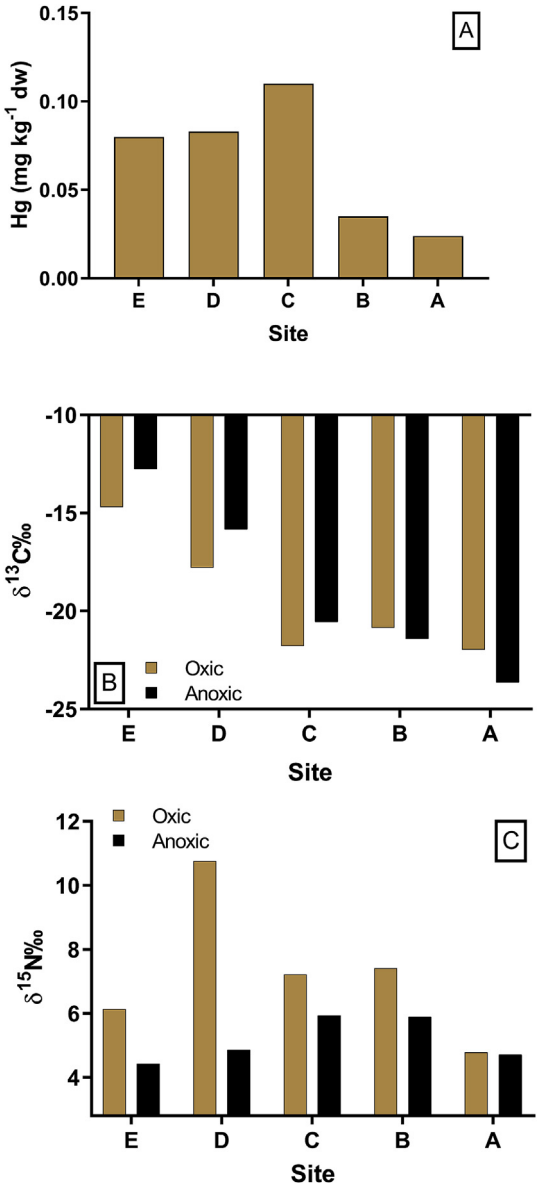

Fig. 5. Hg concentrations in the oxic section (A), stable isotopes $\delta^{13}C$ (B) and $\delta^{15}N$ (C) in oxic and anoxic sections of sediment samples from Sognefjord sampled in 2016. Sites locations are identified in Fig. 1.

(0.024–0.11 mg kg^{-1} dw) were comparable to average Hg concentrations in the Norwegian coastal zone (0.03 mg kg^{-1} dw) and offshore areas (0.02 mg kg^{-1} dw) and lower than average inner fjord Hg levels (0.13 mg kg^{-1} dw) as reported by Everaert et al. (2017). Low Hg sediment concentrations in samples from five different areas is an indication that the Sognefjord is not influenced by any significant point sources of Hg pollution. Everaert et al. (2017) further suggested that elevated Hg levels in sediments in inner parts of fjords are driven by anthropogenic point sources such as mining activities, and heavy metal industries such as the zinc smelter plant in Hardangerfjord. Sediments from Sørfjord, in the Hardangerfjord ecosystem with a well-known Hg point source, showed a Hg concentration as high as 2.26 mg kg^{-1} dw and MeHg of 8.4 μg kg^{-1} dw (Azad et al., 2019a). Thus, the sediments in inner Sognefjord had Hg concentrations about two orders of magnitude lower than in Sørfjord and were similar to background concentrations. In Sognefjord there are two relatively small industrial areas including Årdal (sampling site 2) and Høyanger (close to sampling site 7) (Fig. 1). According to the Norwegian Directorate of Environment (www.miljostatus.no) one industrial unit in Høyanger, located in the outer half of the fjord, has been emitting small amounts of mercury to the fjord in recent years. Higher Hg levels were observed in sediments sampled at the two outermost areas (sites C, D and E) compared to the more inner sites of the fjord, which could reflect emissions from industrial activities in Høyanger. However, overall, observed sediment Hg concentrations were relatively low throughout Sognefjord. Our results also suggest that local Hg pollution in Sognefjord is not significantly affecting the tusk population Hg bioaccumulation regime. We postulate that a significant part of Hg contamination in tusk is driven by local fjord conditions such as greater freshwater run-off, terrestrial organic matter input, water residence times, and ocean stratification dynamics.

4.2. Spatial trends of $\delta^{13}C$ in sediment and tusk

In sediment samples, $\delta^{13}C$ varied between −23.64‰ in the anoxic layer at the innermost site A to −12.76‰ in the anoxic layer at the outermost site E. Average $\delta^{13}C$ values for terrigenous influenced areas are generally around −24‰ and for typical marine sediment approximately −20‰ (Gearing et al., 1977). In our study the innermost part of Sognefjord had similar values of $\delta^{13}C$ to terrigenous sediment, while the outer part had much higher $\delta^{13}C$ than typical marine sediment (~7‰). Since we did not sample the sediment in the coastal and offshore areas, it is not possible to extend our interpretation to these habitats.

Conversely, $\delta^{13}C$ in tusk fillet tissue samples increased from offshore (−18.7‰) towards inner Sognefjord (−17.2‰), indicating that the source of carbon at the base of the tusk's food web changes with fjord position. Opposing trends of the $\delta^{13}C$ signature in sediment and tusk fillet tissue indicate that the tusk's food web is not sediment based and highlights the potential importance and role of pelagic energy sources. The measured $\delta^{13}C$ values in the inner part were very similar to values previously found in oceanic tusk from Icelandic waters (−16.7‰; McMeans et al., 2010), while the values found in the outer part were slightly lower.

In many estuarine ecosystems the phytoplankton production is insufficient to support the heterotrophic energy demands and therefore allochthonous carbon originating from terrestrial plants can play a substantial role in the pelagic food web dynamics (Harfmann et al., 2019; Vargas et al., 2011). Similar process in Sognefjord may be occurring, particularly during autumn and winter when phytoplanktonic production is low.

Phytoplankton may have depleted $\delta^{13}C$ compared to dissolved and particulate organic matter with high levels of allochthonous carbon originating from terrestrial plant detritus (Jones et al., 1998; Grey et al., 2000; Rautio et al., 2011; Van den Meersche et al., 2009). When zooplankton replace their typical diet (phytoplankton) with organic carbon originating from terrestrial plants, $\delta^{13}C$ values will be enriched (Karlsson et al., 2003; Pulido-Villena et al., 2005; Rautio et al., 2011) and this can be efficiently transferred across trophic levels to the top of the food web. These processes vary substantially across geographical areas with different catchment history, watershed characteristics (e.g. vegetation species in the catchment, hydrology and biogeochemistry) and food web dynamics. These carbon based processes are well documented in freshwater ecosystems however information on marine ecosystems and fjords is currently still quite limited. Further investigations are required in order to further elucidate the distribution and carbon source apportionment patterns in the Sognefjord food web.

4.3. Mercury accumulation pathways in tusk

Mercury exposure in tusk, as measured by Hg concentrations in fillets and liver, showed that Hg increased from offshore and coast towards the inner sectors of Sognefjord and was strongly correlated with $\delta^{13}C$ values.

We observed a spatial trend of $\delta^{13}C$ in tusk fillets that was likely indicative of more terrestrial energy sources from freshwater runoff in samples collected from the fjord's interior. Thus, increasing $\delta^{13}C$ in tusk from the open ocean towards the inner Sognefjord possibly reflects the increasing influence of freshwater runoff on Hg bioaccumulation. Other studies have also shown a gradual increase in $\delta^{13}C$ from offshore towards inshore and used this parameter to explain the variation in Hg accumulation in marine fish (Le Croizier et al., 2019; Taylor et al., 2019). It is likely that Hg originating from the terrestrial landscape and subsequent water column methylation could be driving the overall bioaccumulation regime of tusk in fjord ecosystems (Wang et al., 2018) however further research is needed to verify this hypothesis.

The observed higher Hg concentrations in fillets and liver samples of tusk from the inner part of Sognefjord compared to the outer part may thus reflect increased bioavailability of Hg caused by increased input of terrestrial organic carbon, or that higher input of freshwater from rivers and streams is followed by more input of Hg from the catchment area, or possibly both. Additionally, the degree to which differences between the food webs of the inner fjord areas and coastal and offshore habitats might also be governing Hg bioaccumulation in tusk is ultimately unknown. MeHg originating from the atmosphere and terrestrial runoff has higher bioavailability compared to MeHg formed *in situ* in sediments (Jonsson et al., 2014). Also, Soerensen et al. (2017) found that in the northern Baltic Sea allochthonous organic matter and labile dissolved organic carbon were important drivers for seasonal and spatial variation of MeHg in seawater in an ecosystem with generally slow turnover of water masses. Organic carbon content has also been shown to control MeHg levels in seawater and estuarine marine food webs in coastal ecosystems along the Northeast coast of USA (Taylor et al., 2019). Although fjords are distinctly different from the above mentioned ecosystems, our investigation is in good accordance with the findings from estuarine and freshwater investigations which reported that higher terrestrial input of organic matter may lead to increased Hg methylation rates and more efficient subsequent trophic transfer to the base of the food web and ultimately to apex predators.

In this study we did not find any spatially increasing gradients in sediment Hg towards the inner part of the fjord. Traditionally, Hg methylation in sediments from saltwater environments has been considered the main source of Hg in food webs of coastal areas (Compeau and Bartha, 1987). However, recent studies have indicated that Hg methylation in fjords and estuarine ecosystems often occurs within the water column (Schartup et al., 2015; Wang et al., 2018). Water column methylation or MeHg originating from land and a food web of pelagic origin via benthic-pelagic coupling may be a plausible explanation for why tusk fillet and sediment data did not follow similar spatial trends with regard to either Hg or $\delta^{13}C$. Mercury, entering the fjord from land together with terrestrial organic carbon, may quickly methylate in the water column and subsequently enter the base of the food web, thus exposing tusk to more Hg contaminated prey however further research is needed to verify these hypotheses.

Tusk from the different sites had mean $\delta^{15}N$ values between 14.8 and 16.1‰, which is indicative of a high trophic position in marine food webs as expected as tusk mainly feed on other fish and large crustaceans (Bergstad, 1991). An average $\delta^{15}N$ of only 12.7‰ was however found in tusk from Icelandic waters (McMeans et al., 2010). These differences may be related to geographical variation in $\delta^{15}N$ at the base of the food web (i.e., different baselines of $\delta^{15}N$). Trophic level, as estimated by $\delta^{15}N$, was similar across all habitats except offshore which had a relatively lower trophic level than other habitats. Larger individuals are physically able to ingest larger prey and trophic level and fish body size are often well correlated (Romanuk et al., 2011). It is possible that tusk from offshore areas may have slightly lower trophic positions than individuals inhabiting the fjord due to prey distribution and abundance patterns. One limitation in using bulk $\delta^{15}N$ to estimate trophic position in top predators is that baseline variation in $\delta^{15}N$ could be confounding the interpretation (Cabana and Rasmussen, 1996), and trophic level may not relate similarly to $\delta^{15}N$ in all areas. Future studies on tusk should consider the use of compound specific stable isotope analysis of amino acids (CSIA-AA), since this method is less sensitive to changes in baseline nitrogen values in contrast to the bulk stable isotope analyses (Ishikawa, 2018).

Length adjusted LS mean Hg concentrations in tusk fillets from different sites were not correlated with mean $\delta^{15}N$. Thus, while trophic level played an important role in the variation between individuals, the variation in Hg concentrations from different sites and habitats appears to have been independent of their trophic level and differences in their food items for the size class of individuals measured in this study. Kenji et al. (2020) investigated the marine food web in Minamata bay and showed that variation in Hg is mostly linked to food sources and $\delta^{13}C$ values rather than estimates of $\delta^{15}N$.

Hydroelectric reservoirs influence the water cycle in fjord ecosystems and could affect the local Hg biogeochemical cycle. Episodic or seasonal water level fluctuation and flooding of marginal areas of the reservoir (often covered with vegetation, oxidized soils, organic matter and leaf litter) can enhance methylation and facilitate MeHg transport into the reservoir. Many studies have shown elevated levels of MeHg in both water and biota downstream of these reservoirs (Kasper et al., 2014; Calder et al., 2016; Pestana et al., 2018). Since the Sognefjord area has many hydroelectric plants, particularly within its inner parts (www.nve.no), they can potentially influence local Hg bioaccumulation regimes.

5. Conclusions

Tusk from a long marine fjord system (Sognefjord, Norway) showed elevated Hg concentrations with mean values above the EU's maximum level at most sites. Sognefjord has no major point source of Hg pollution, and fjord characteristics, particularly high amount of runoff containing Hg species and terrestrial carbon from the catchment, likely have significant effects on Hg concentrations at the top of the food web and are important drivers of Hg bioaccumulation in tusk. Measured $\delta^{13}C$ increased along a linear gradient from offshore towards inner Sognefjord and was well correlated with Hg concentrations in tusk fillets, indicating that the influence of terrestrial carbon and freshwater input may have a significant effect on mercury bioaccumulation in predatory fish from fjord ecosystems. Although previous studies have shown that organic matter is the main driver of MeHg in the sub-arctic coastal areas in seawater, sediment and plankton (Lambertsson and Nilsson, 2006; Schartup et al., 2015; Soerensen et al., 2017), this study clarifies the strong linkage between organic carbon sources and Hg bioaccumulation in a demersal, long-lived, predatory fish species inhabiting fjord ecosystems.

Author Statement

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Methodology, Writing - review & editing, Project administration. Michael S. Bank, Methodology, Writing - review & editing. Lise Madsen, Methodology, Writing - review & editing. Amund Maage, Writing - review & editing, Funding acquisition, Resources.

Summary of main findings

Hg concentrations in tusk gradually increased towards the inner fjord while Hg levels in sediments were at background concentrations. Carbon source was the best predictor of spatial Hg variation in tusk.

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

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