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Title:

The influence of a submerged meadow on uptake and trophic transfer of legacy mercury from contaminated sediment in the food web in a brackish Norwegian fjord

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Abstract

There are contradicting results on the importance of legacy mercury (Hg) contaminated sediments to Hg fish tissue concentrations. Still, sediment remediation actions often aim at minimizing ecosystem exposure and human risk caused by the consumption of fish and seafood. The aim of this study was to investigate the possible influence of a permanently submerged meadow on the availability and transfer of Hg from sediment to biota, three decades after the Hg discharges was halted and the previous biota survey was carried out, in the severely contaminated brackish fjord Gunneklevfjorden in southern Norway. We examined total Hg (Tot-Hg) and methyl-Hg (MeHg) concentrations and stable isotopes of nitrogen (N) and carbon (C) in zooplankton, benthic invertebrates, and fish to map the food web and to investigate the trophic transfer of Hg. Sediment and water data were available from a previous study. Overlap in δ^{13}C in benthos and fish reveals that benthos is a preferred prey to fish, though despite elevated Tot-Hg concentrations in benthos, fish predator (perch, pike and eel) Hg concentrations are comparable to concentrations reported in nearby lakes without contaminated sediments (mean 1.6 ± 1.3 mg Tot-Hg kg^{-1} dw). We propose that habitat reliance is an important factor controlling the uptake of Hg from sediments, as both benthos and fish prefer to forage within the meadow where sediment concentrations of Tot-Hg are lower than outside the macrophyte meadow, though %MeHg is higher than outside. Further, we propose that sediment remediation actions performed outside the meadow may have limited effect on the Hg concentrations in fish.

Keywords

Methyl mercury, Bioaccumulation, Stable isotopes, Habitat reliance, European perch, Sediment remediation

1 Introduction

Mercury (Hg) contaminated sediments are typically found in rivers and estuaries influenced by historic industrial discharges, and are believed to be a source to uptake of Hg into aquatic food webs (Driscoll et al., 2013). Exposure of Hg to aquatic food webs is thought to be strongly influenced by the production of bioavailable and bioaccumulating organic methyl mercury (MeHg) often constitute 95 - 99 % of total Hg (Tot-Hg) in fish (Wiener and Spry, 1996), though inorganic Hg (IHg) is often a significant part of total Hg (Tot-Hg) at lower trophic levels. It is commonly acknowledged that microbial production of MeHg from IHg is stimulated within freshwater wetlands, estuarine and coastal marine environments (AMAP, 2011, Compeau and Bartha, 1985, Donaldson et al., 2010, Hollweg et al., 2009, King et al., 2000, Lehnherr et al., 2011, Lehnherr, 2014), and contaminated sediment in these environments has been shown to represent a pool of IHg available for MeHg production (Merritt and Amirbahman, 2009, Jaglal, 2011). Further, MeHg production has been shown to primarily occur in the surface, or oxic-anoxic interface of sediments (Skyllberg et al., 2007, Kim et al., 2008), potentially exposing sediment dwelling organisms to pockets of MeHg. Given that benthic secondary production plays a central role in supporting higher aquatic trophic level production (Vander Zanden and Vadeboncoeur, 2002), uptake of MeHg in benthos may be an important entrance of Hg into the aquatic food webs. However, there are contradicting results on the importance of legacy Hg contaminated sediments to Hg fish tissue concentrations (Hodson et al., 2014, Lehnherr et al., 2011), reflecting the complexity of factors controlling methylation, exposure, uptake and bioaccumulation of Hg. Studies have demonstrated similar Hg concentrations in fish from highly contaminated waters and waters without direct industrial discharges (Schaefer et al., 2004). This may be explained by limited MeHg accumulation in Hg contaminated surface waters (Bloom, 1992, Watras and Bloom, 1992), which has been suggested to be due to enhanced reductive demethylation of MeHg by Hg-resistant bacteria at high Hg concentrations (Schaefer et al., 2004). However, the direct uptake of MeHg into biota from sediment may be influenced by other mechanisms controlling the availability of MeHg in sediments. Recently, attention has been drawn towards effects of bioturbation as well as the impact from submerged macrophytes on Hg methylation in sediments, revealing higher %MeHg within vegetated habitats (Regier et al., 2012, Bravo et al., 2014, Cosio et al., 2014, Olsen et al., 2018). As sediment remediation actions often aim for minimizing ecosystem exposure and human risk caused by the consumption of Hg contaminated fish and seafood, enhanced knowledge of the spatial factors influencing exposure and uptake of Hg is crucial to target measures that may resolve issues of high Hg concentrations in fish.

The aim of this study was to investigate the possible influence of a permanently submerged meadow on the transfer of Hg from sediment to biota, in the severely contaminated brackish fjord Gunneklevfjorden in southern Norway. Biota Hg concentrations has not been reported since 1989 (Berge and Knutzen, 1989). Sediment Hg concentrations in the fjord is high due to substantial industrial Hg discharges during 1947-1987, though the Tot-Hg concentrations are lower within the submerged meadow than in non-vegetated areas (Olsen et al., 2018). We hypothesize that habitat reliance (within and outside the meadow) is an important factor controlling uptake of Hg into aquatic food webs. To test our hypotheses, we examined the food web by analyses of stable isotope of nitrogen (N) and carbon (C) in zooplankton, benthic invertebrates, and fish, and we analyzed Hg concentrations in biota within and outside the meadow. δ^{15}N values are commonly used to estimate trophic levels (*TL*) in aquatic food webs (Cabana and Rasmussen, 1996), whereas δ^{13}C values are used to distinguish between pelagic and littoral derived organic carbon in the diet of aquatic consumers (France, 1995). Water data was available from Olsen et al. (2016), and sediment data and pore water was available through a recently published paper by Olsen et al. (2018).

2 Materials and methods

2.1 Study area

Our study site, Gunneklevfjorden, is a 0.7 km^2, shallow (max depth: 11 m), brackish fjord, located in southern Norway (**Figure 1**). The fjord connects to the urban impacted river Skienselva in the north and to the industrialized and also severely contaminated fjord Frierfjorden, in the south. The fjord is strongly influenced by the river and the tidal influence is weak, though the surface water salinity varies between 1–5 PSU and bottom waters may reach 20 PSU (Molvær, 1979, Molvær, 1989). Salinity during our study period (August 2013) was observed to be 1.1–2.5 PSU in surface waters and 2.7-3.5 PSU in bottom waters. The permanently submerged meadow covers 10% of the fjord area (70 000 m^2) down to approximately 3 m depth and is dominated by the vascular plant curly-leaf pondweed (*Potamogeton crispus*), whereas the rest of the fjord consists of very soft non-vegetated sediments (Olsen et al., 2016). Between 1947 and 1987, approximately 80 tonnes of Hg was released from a chlor-alkali plant situated at the nearby industrial site Herøya (Skei, 1989). The entire fjord system outside of Gunneklevfjorden (approximately 70 km^2) is influenced by various industrial activities at Herøya, though Gunneklevfjorden has been the main recipient for Hg discharges. The discharges are evident through high Tot-Hg and MeHg concentrations (492 mg kg^{-1} and 269 µg kg^{-1}, respectively) in intermediate sediment depths (10-20 cm), whereas natural recovery has resulted in reduced surface concentrations (0-5 cm) of 1.9-5.5 mg Tot-Hg kg^{-1} and 1.3-3.2 µg MeHg kg^{-1} (Olsen et al., 2018) with the lowest concentrations found within the meadow (**Table SI-A**). Water Hg concentrations are generally low (**Table SI-A**) and sediment is recognized as the primary Hg source to the water column, as the concentration of Hg in the influencing river water is lower than in the Gunneklevfjorden (Olsen et al., 2015). The Norwegian Environment Agency has prioritized Gunneklevfjorden as well as the adjacent Frierfjorden for sediment remediation measures (www.miljodirektoratet.no).

Uptake of Hg in biota in the fjord was first reported in algae (*Cladophora* sp.) in 1975 (Rygg, 1979). Later, Berge and Knutzen (1989) revealed Hg concentrations in fish muscle tissue up to 1.55 mg kg^{-1} ww in eel, *Anguilla anguilla*, and reported mean Hg concentrations for eel 0.92 mg kg^{-1} ww (n=20); perch (*Perca fluviatilis*) 1.34 mg kg^{-1} ww (n=9); pike (*Esox lucius*) 1.26 mg kg^{-1} ww (n=1) and flounder (*Platichthys flesus*) 0.55 mg kg^{-1} ww (n=11). Dietary restrictions and prohibition of fishing has been implemented by the Norwegian Food Safety Authority since the 1960s. No further studies or monitoring of Hg in biota have been conducted during the natural recovery period, i.e. from 1987 to present.

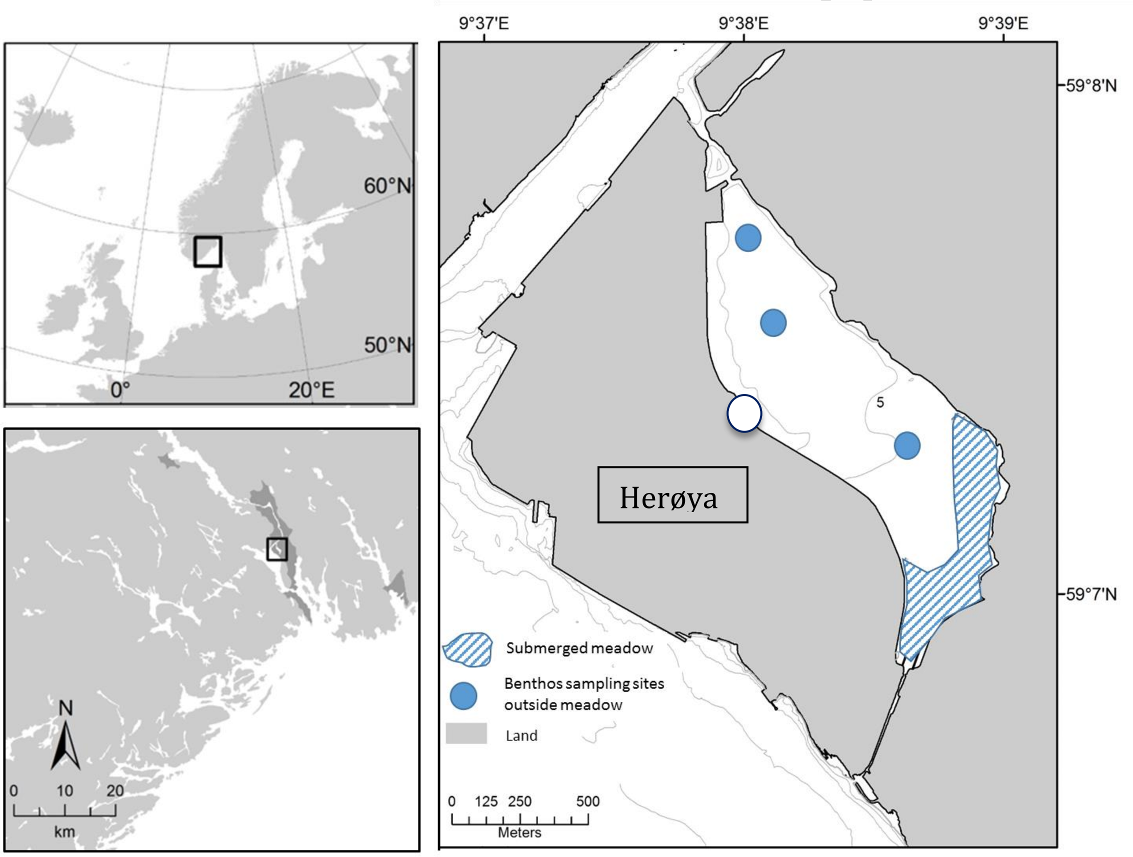

Figure 1*. Gunneklevfjorden, Norway. The previous discharge point at Herøya is marked with a white circle. Benthos were collected at multiple sampling sites within the submerged meadow and at three sampling sites outside the meadow. Fish were mainly caught within or close to the meadow. Zooplankton were collected by net hauling across the entire fjord.*

2.2 Sampling

2.2.1 Zooplankton

Zooplankton was collected by seine nets (mesh size: 45 µm) in August 2013, by two vertical net hauls from just above the sea bed (5 m depth) to the surface at a central point in the fjord, and two surface horizontal hauls (1-2 m). The four samples were kept cool (~4°C) in 2 L plastic bottles. Within 24 hours, each sample was filtered again through a 45 µm seine net into a small glass bottle and fixed with Lugol's solution (I_3K solution). Zooplankton in each of the four samples were then identified in a microscope to the lowest appropriate taxonomic level, and the approximate proportion (by number) of each taxonomic group was estimated (**Table SI-B**). The four zooplankton samples were then stored frozen (~ -18°C) and thereafter freeze dried prior to homogenization and analysis of Tot-Hg and stable isotopes (C and N).

2.2.2 Benthic invertebrates

Benthic organisms were collected within and outside the meadow in August 2013 (**Figure 1**) by use of Ekman grab, invertebrate traps, and by "washing" of approximately 500 mL submerged macrophytes (displacement volume). The traps consisted of three chained bundles of hemp rope left for 72 hours on the sea bottom. Previous studies have revealed that these traps act as artificial substrate, attracting an intermediate size fraction of benthic organisms commonly associated with underwater vegetation (Kraufvelin et al., 2002). All macrophyte samples and ropes were well washed in a bucket of local water before water was sieved through a 0.5 mm sieve. Samples from within and outside the meadow were kept separate. Our observations regardless of sampling technique, were that benthic organisms were 10 - to - 100 -fold more abundant in samples collected within the meadow compared to outside. All benthic organisms were kept alive in site water and stored cold (~ 4°C) for maximum

24 hrs before identified to lowest possible taxonomic levels in a microscope and then distributed into 10 samples representing six different taxonomic groups, keeping samples from within and outside the meadow separate (**Table SI-C**). All six taxonomic groups were present inside the meadow and three groups were present outside the meadow. Amphipods from outside the meadow were distributed into two samples. Amphipod individuals dominated by numbers followed by molluscs (mainly gastropods) and chironomids. Other mosquito larvae were grouped together in one sample. Only soft parts of molluscs (gastropods) were kept for chemical analysis. All 10 samples were frozen (~18°C) and freeze dried (composite sample weight ranged 0.002 - 0.8 g dw) prior to analysis of Hg and stable isotopes of C and N.

2.2.3 Fish

A total of 548 fish from five species (**Table SI-D**) were collected by gillnets, fish traps and beach seine at several sites within and nearby the meadow at two occasions in August 2013 (**Figure 1**). The fish was not assigned a sampling site (within or outside the meadow), due to their mobility. Perch predominated in the catch (n = 523), and a random selection of approximately 20 perch within five length intervals were incorporated in further analyses (n=111) together with the rest of the fish (n=25; **Table SI-D**). All fish (n=136) were transported to the laboratory and stored cool (~ 4 °C), and processed within 48 hours. Standard length and weight of all individuals were measured, before sampling of dorsal muscle tissue. The tissue samples were frozen (~ -18 °C) and thereafter freeze dried prior to analysis of Tot-Hg and stable isotopes of C and N of all samples. All samples were weighed before and after freeze drying to determine dry matter content. A random selection of six samples were analyzed for MeHg. Age determination was conducted on burnt and transversally sectioned otoliths under a light microscope. In most individuals, also operculum was sampled as a supplementary support for age determination.

2.3 Chemical analyses

2.3.1 Mercury analyses in biota

All freeze dried biota samples were analyzed in triplicates for Tot-Hg by thermal decomposition and subsequent direct atomic absorption spectrophotometry (Lumex Mercury Analyzer RA915). Quality control samples included reference material (DORM-3; fish protein) for every 10th analysis and sample triplicates (20 mg dw). Relative standard deviation (RSD) of sample and reference material triplicates was always <10 % and recovered amount of reference material always 90 % – 110 %. MeHg was determined in only one mixed sample of benthos (molluscs) and in five randomly selected fishes (**Table 3**), due to insufficient mass of other benthic taxonomic groups as well as zooplankton. Prior to MeHg analyses, the freeze dried material was dissolved in nitric acid (HNO_3, 30 %, 1–10 mL depending on available sample) under heating (16 hours, 65 °C), following method described in Braaten et al. (2014), before chemical analysis was carried out following USEPA Method 1630 (USEPA, 1998). The analysis of MeHg is based on ethylation, purge and trap, and quantification by cold vapor atomic fluorescence spectrophotometer (CVAFS). Quality assurance/quality control samples included method blanks (n=4); sample duplicates (n=3); matrix spikes (n=3); and certified reference material (n=2). The certified MeHg concentrations of the reference materials used were 0.355 ± 0.056 mg/kg for DORM-3 (fish protein; National Research Council of Canada). Samples that were analyzed in duplicate were also used for matrix spike. Samples chosen for matrix spiking were added 10 000 pg MeHg (as MeHgOH, 1.0 mL of 10.0 ng/mL).

For each analyzing batch, quality control is performed, included blank samples, blank spiked samples, sample duplicates and matrix spiked samples. The RSD of the sample duplicates was always <20% for MeHg. The amount found in blank and matrix spiked samples was 80 % – 120 %. Wet weight (ww) concentrations of Tot-Hg and MeHg in fish were calculated from the sample specific dry matter content (average of all fish samples 20.5%).

2.3.2 Bioconcentration factor

Bioconcentration factor (*BCF*) is the ratio between concentration in biota and in the environment. *BCF* for Tot-Hg and MeHg was calculated for the lower trophic levels: i.e. for zooplankton relative to water concentration, and for benthos (only molluscs) relative to pore water as well as to water concentration (**Table SI-E**), to investigate the uptake of Hg into biota.

2.3.3 Stable isotopes analyses and calculation of biomagnification factor

All organisms analyzed for Tot-Hg were also analyzed for the stable isotopes ^{14}N, ^{15}N, ^{12}C and ^{13}C at the Norwegian Institute for Energy Technology, to calculate δ^{15}N and δ^{13}C. In aquatic food chains, δ^{15}N is observed to increase by approximately 3.4 ‰ per TL (Post, 2002). Hence, *TL* was calculated according to the following equation:

$$TL = (\delta^{15}N_{consumer} - \delta^{15}N_{baseline})/3.4 + TL_{baseline} \qquad (Eq.\ 1)$$

Normally, primary consumers are used for baseline corrections because their larger body size and greater longevity result in less seasonality in the δ^{15}N signatures than primary producers and, thus, level out much of the large temporal variations typically present in short living primary producers such as algae (Cabana and Rasmussen, 1996). Common primary consumers typically used for baseline correction of pelagic food webs are shallow zooplankton and mussels, while molluscs such as gastropods, amphipods and trichopterans are often used for baseline corrections in littoral food webs. Chironomids and deep zooplankton are often used in profundal food webs (Vander Zanden and Rasmussen, 1999). We used the lowest δ^{15}N value for molluscs as baseline (5.74 ‰) for the littoral food web (benthos and fish), whereas for the pelagic (zooplankton) we used the lowest zooplankton δ^{15}N value (2.97).

The Trophic Magnification Slope (*TMS*) is routinely used as an indicator of biomagnifying potential of Hg in food webs (Lavoie et al., 2013), expressed as the slope (*b*) of the linear relationship between Hg concentration and δ^{15}N within a food web:

$$\mathrm{Log}_{10}[Hg] = a + b(\delta^{15}\mathrm{N}) \qquad (Eq.\ 2)$$

A significant and positive slope indicates biomagnification up the food web.

Accordingly, the Trophic Magnification Factor (*TMF*) can be estimated by the following equation:

$$TMF = 10^{(b \cdot 3.4)} \qquad (Eq.\ 3)$$

where *b* is the slope in *Eq 2.*, while 3.4 is the empirically increase in δ^{15}N (in ‰) per trophic level (Post (2002). Thus, *TMF* estimates the average increase per trophic level.

2.4 Data analyses

In the statistical models, Hg concentrations were log-transformed to improve normality and avoid heteroscadicity and influence of outliers. A cluster analysis was performed on the data to identify different functional groups in the food web. We used a hierarchical clustering procedure (Ward’s minimum variance) in the statistical program JMP (SAS Institute) on data on Tot-Hg (log-transformed), δ^{15}N and δ^{13}C. ANOVA was used to recognize differences between groups (zooplankton, benthos and fish), between taxonomic groups or species, and within and outside the meadow, followed by pairwise comparison using *t*-tests with pooled SD to identify which groups or species differed significantly from others (post hoc tests). Pearson's correlation coefficients was used to identify potential predictors for Hg concentrations.

To reveal the extent of biomagnification of Tot-Hg in the food web, we made a statistical model (ANCOVA) explaining Tot-Hg (log-transformed) in the different functional groups (zooplankton, benthos and fish) as a function of individual trophic levels (δ^{15}N). Further, we tested the different functional groups for differences in regression slopes by introducing an interaction effects between δ^{15}N and the group factor. The n was low for all taxonomic groups and fish species except perch and perch was the only species with enough individuals to investigate bioaccumulation within one single species as well as possible biomagnification of Hg by the development of a linear regression model (GLM). The R (SIAR) approach for prediction of proportions of food sources to consumers was considered. However, the sample size of benthos and zooplankton was found to be too small for this analyses to be relevant. Further, supporting data from stomach analyses was not available.

3 Results

Results from chemical analysis of stable isotopes of C and N, and Tot-Hg and MeHg in biota and calculation of *BCF* for zooplankton and benthos, is presented as Supplementary Information; **Table SI-E**.

3.1 Food web structure

A three-group pattern with zooplankton, benthos and fish as functional groups was confirmed by a hierarchical clustering with average linkage and a two-way clustering based on the variables Tot-Hg, δ^{13}C and δ^{15}N (**Figure SI-A**). Zooplankton and benthos represents lower trophic levels, benthos and fish overlap in intermediate trophic levels, and fish represents the top predators at the highest trophic levels (**Figure 2**). The δ^{15}N and δ^{13}C pattern indicates that benthic invertebrates comprise a larger portion of fish diets in Gunneklevfjorden than zooplankton. With the exception of young perch, none of the fish species is typically planktonic feeders are rather strongly linked to vegetation areas, with a possible exception to flounder. They are all carnivorous or omnivorous with potentially overlapping diets (Pethon, 1989). There are no significant differences in δ^{15}N between the benthic groups (p = 0.84) nor between the fish species (*p* = 0.65). Differences between within and outside the meadow is presented in 3.2.3.

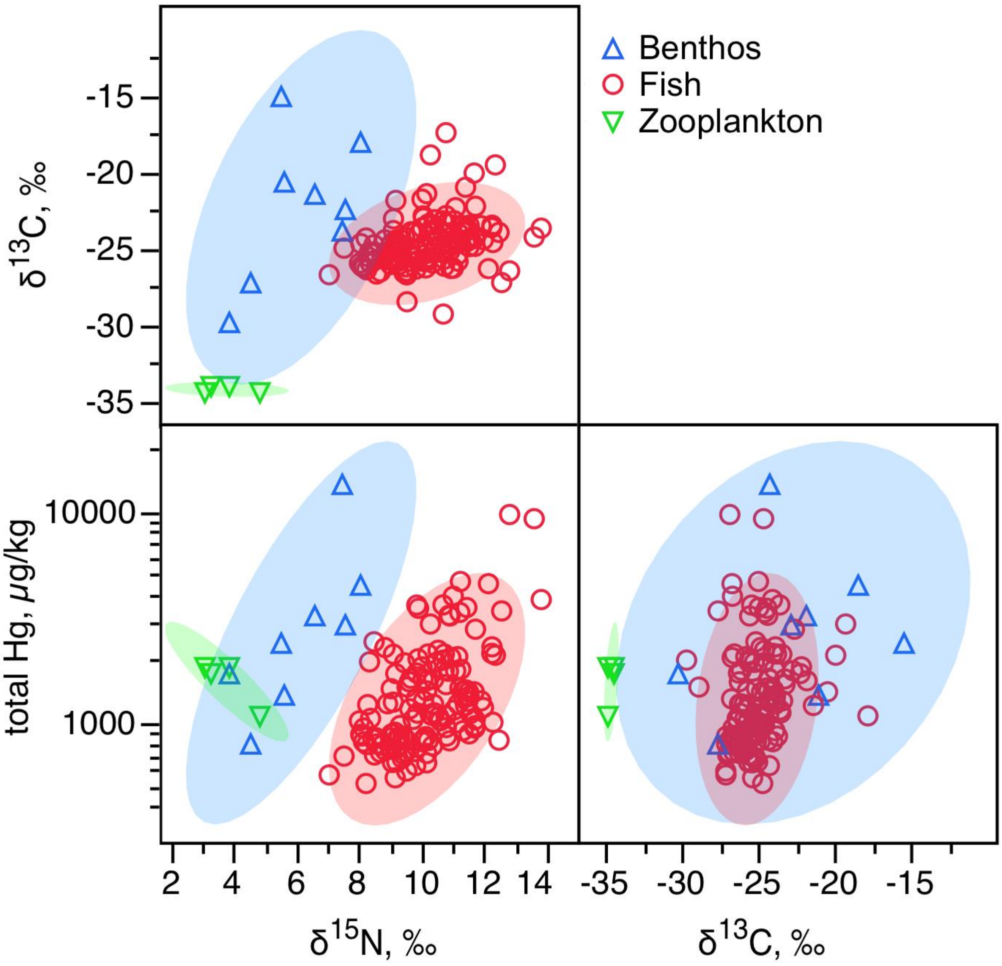

***Figure 2**. Bivariate scatterplots of log Tot-Hg, $\delta^{15}N$ and $\delta^{13}C$ (with 90% confidence ellipses) including the three functional groups zooplankton, benthos and fish in Gunneklevfjorden identified in by a hierarchical clustering (Figure SI-A). Trombidiformes is not included in the plot due to lack of Hg analysis and Helopdella is not included due to carnivore dietary preferences.*

The weak dietary link between zooplankton and fish is supported by the significant difference in δ^{13}C ($p < 2e^{-16}$); Zooplankton has a very depleted and typical pelagic δ^{13}C-signature, while fish and benthos reveals relatively equal and less depleted, more littoral δ^{13}C-signatures (**Figure 2**). Further statistical testing (ANOVA) excluded other benthic groups than the three numerously dominating amphipods, molluscs and chironomids that we found both within and outside the meadow, and reveals significantly different δ^{13}C -signatures ($p = 0.03$) with a less depleted and thus more littoral δ^{13}C-signature in molluscs than in the two other groups (**Figure 3**). There is no significant correlation between δ^{13}C and δ^{15}N in benthos.

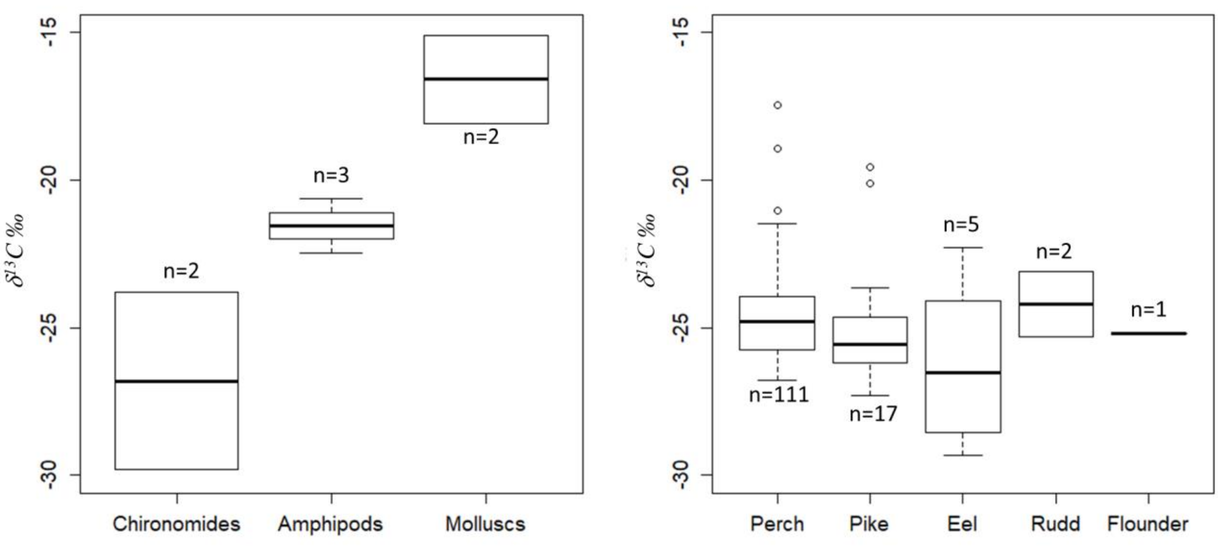

***Figure 3**. $\delta^{13}C$-signatures in benthos (left) and fish (right) from Gunneklevfjorden in 2013.*

There is no significant difference in δ^{13}C-signatures between the fish species (*p* = 0.37: **Figure 3**), and all species has a depleted δ^{13}C-signatures that reflects that of chironomids and amphipods ($\delta^{13}C < -20$) more than molluscs ($\delta^{13}C > -20$). In the most numerous species perch (n = 111), there is a positive correlation (Pearson's) for δ^{15}N with δ^{13}C ($r = 0.35$, $p = 0.0001$), age ($r = 0.43$, $p < 0.0001$) and length ($r = 0.43$, $p < 0.0001$), supporting a gradual ontogentic dietary shift (Persson, 1986) towards a more littoral based diet as the fish grow older and reach higher trophic levels. There is no significant correlation between δ^{13}C and δ^{15}N for pike (n = 17, r= 0.46, p=0.06) and eel (n=5, r=0.27, p=0.65).

3.2 Hg in biota

3.2.1 Exposure and uptake of Hg

The concentration of Tot-Hg in the three functional groups overlapped (**Figure 2**), ranging 0.2 – 13.9 mg Tot-Hg kg^{-1} dw with the lowest concentration found in one perch and the highest in a mixed sample of chironomids (**Table SI-E**). Mean concentrations for all fish were 1.6±1.3 mg Tot-Hg kg^{-1} dw (calculated to 0.3±0.3 mg Hg kg^{-1} ww), that is below the mean values reported in 1989, see chapter 2.1 (Berge and Knutzen, 1989). The % MeHg is low in benthos (2.3 % MeHg) compared to fish (64 – 83 % MeHg), revealing that high concentrations of Tot-Hg in benthos most likely are caused by IHg or Hg-complexes other than MeHg. MeHg was not measured in zooplankton. The Tot-Hg concentrations in benthos do not correlate significantly with δ^{13}C, and there is no significant difference in δ^{13}C in benthos within and outside the meadow, thus the differences in diet do not seem to explain different Tot-Hg concentrations. However, a significant positive correlation between δ^{13}C and Tot-Hg (log-transformed) in fish (*n* = 136, *r* = 0.2, *p* = 0.02), indicated higher concentrations of Tot-Hg in fish with less depleted (more littoral) δ^{13}C-signature.

The exposure and uptake of Tot-Hg as indicated by the calculated $BCF_{Tot\text{-}Hg}$ is higher in zooplankton ($BCF_{Tot\text{-}Hg}$ = 454 000) than in benthos ($BCF_{Tot\text{-}Hg}$ = 3 000), based on water concentrations for zooplankton and pore water concentration for molluscs (**Table SI-E**). However, when we base the calculation of $BCF_{Tot\text{-}Hg}$ for molluscs on bottom water concentrations (5 m depth) that might be a more relevant exposure pathway than pore water for gastropods, we get a $BCF_{Tot\text{-}Hg}$ of 412 000 which is almost similar to zooplankton. For MeHg the calculated BCF_{MeHg} for molluscs is larger when based on pore water (BCF_{MeHg} = 10 300 000) than on bottom water (BCF_{MeHg} = 3 512 000), indicating that MeHg may be more available for uptake from pore water than from bottom water.

3.2.2 Biomagnification and trophic transfer of Hg

Biomagnification explored by an ANCOVA model of Tot-Hg in the three functional groups shows the interaction term (δ^{15}N x group) to be statistically insignificant, indicating parallel slopes between Tot-Hg and δ^{15}N for the three groups (**Figure 4**). A *post hoc* test (contrasts) could not prove any statistical difference between the adjusted means of Tot-Hg for zooplankton and benthos (*p* = 0.49) whereas the adjusted mean for fish was significantly lower compared to these (*p* = 0.001). This supports previous findings of a decoupling in the biomagnification of Tot-Hg between the groups (fish vs. benthos and zooplankton), that reflects the significant larger fraction of IHg in benthos and zooplankton and low trophic transfer to fish.

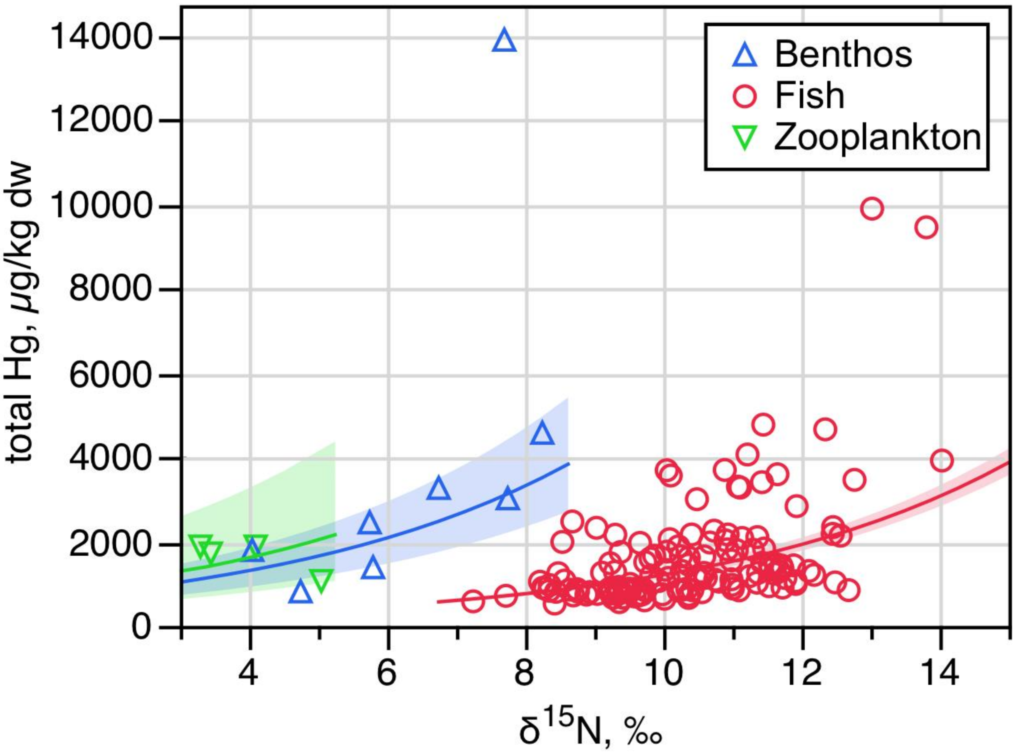

Figure 4. *Relationship between Tot-Hg (µg kg^{-1} dw) and δ^{15}N in zooplankton, benthos and fish in Gunneklevfjord. The curves and their 95 % confidence intervals are based on an analysis of covariance model giving identical regression coefficients (parallel regression lines on a semi-log scale).*

Overall trophic transfer of Tot-Hg in the food web was explored by calculating *TMS* and *TMF* for Tot-Hg in fish following *eq. 2* and *eq. 3*. Invertebrates were excluded to avoid bias due to the high proportion of IHg concentrations in benthos (**Table SI-E**). All fish were included in the calculation which allowed for measures of δ^{15}N ranging approximately 3 trophic levels according to *eq.1*, thus in compliance with recommendations in estimates of contaminant biomagnification (Borgå et al., 2012):

TMS: 0.106 (d.f.=135, p=1.06e^{-12})

TMF: 2.3

In addition, *TMS* = 0.09 (*d.f.* =109, *p* < 0.001) and *TMF* = 2.1 was calculated for perch solely, for direct comparison with the *TMF* = 4.6 reported from studies of perch in nearby lakes not influenced by direct industrial discharges (Økelsrud et al., 2016). The relatively low *TMF* in Gunneklevfjorden explains why Tot-Hg concentrations in fish doesn't reach higher than 9.9 mg kg^{-1} dw (2.5 mg kg^{-1} ww, **Table SI-E**) despite the severely contaminated sediments.

3.2.3 Habitat reliance influencing bioaccumulation of Hg in benthos

Investigation of bioaccumulation in benthos by an ANOVA based on a pooled dataset of the three most numerous benthic groups (amphipods, molluscs and chironomids) (**Figure 5**), revealed that trophic levels (δ^{15}N) explained 72% (multiple $R^2 = 0.72$, $p = 0.015$) of the variance of Tot-Hg. Further, there was a significant difference ($p = 0.05$) in δ^{15}N between benthos collected within and outside the meadow. This may reflect that benthos (amphipods) on high trophic levels are more mobile and less stationary within the meadow. Influence of habitat reliance on exposure and uptake of Tot-Hg in benthos is indicated by significant lower values of Log Tot-Hg ($p = 0.01$) within the meadow (**Figure 5**) where sediment Tot-Hg concentrations are lower than outside.

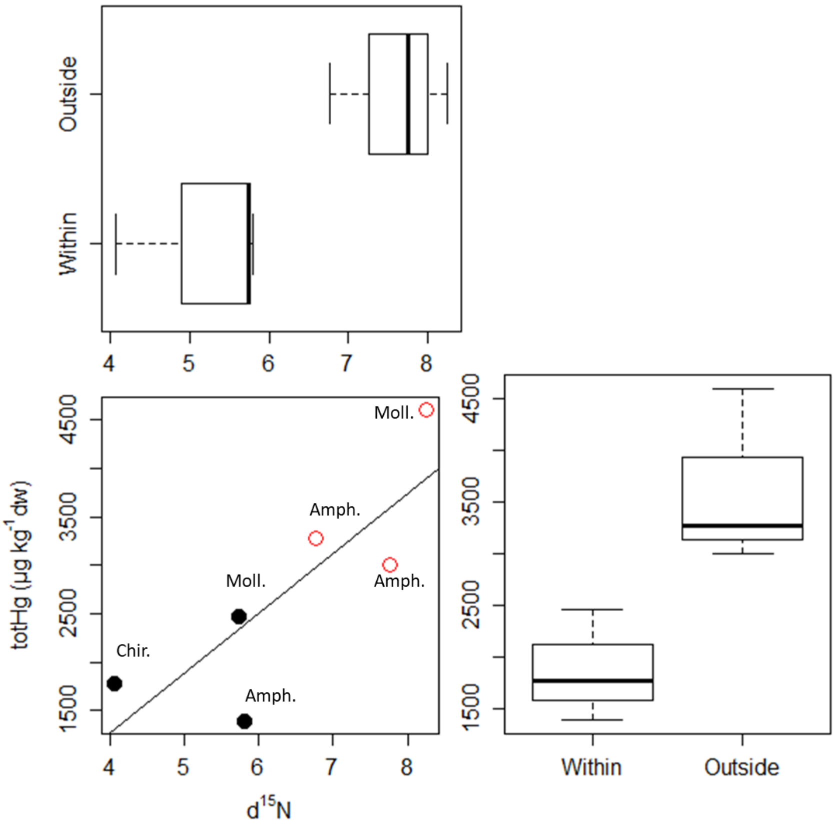

Figure 5. Pooled data on δ^{15}N and Tot-Hg (µg kg^{-1} dw)within (dark dots ●) and outside (open dots ○) the meadow. Down left: linear model for δ^{15}N and Tot-Hg (p=0.015). One sample with very high Tot-Hg concentration (14 000 µg kg^{-1} dw) is not included in the figure due to graphical scale issues. Benthos groups are denoted. Top left: δ^{15}N. Down right: Tot-Hg (µg kg^{-1} dw). The differences between within and outside the meadow were significant on a 95% level.

3.2.4 Bioaccumulation and trophic transfer of Hg based on perch only

Results indicate a moderate to strong positive correlation (Pearson’s, $p < 0.001$) for concentrations of Tot-Hg in perch (µg kg^{-1} dw) with δ^{15}N ($r = 0.42$), age ($r = 0.76$), weight ($r = 0.84$,) and length ($r = 0.76$), and a somewhat weaker correlation with δ^{13}C ($r = 0.19$, $p = 0 .037$) A GLM for concentrations of Tot-Hg $[C_1]$ in perch solely based on δ^{15}N, δ^{13}C and an interaction between age and length as predictors seems to give the best fit to the dataset, explaining 76 % of the variance ($R^2 = 0.76$, *d.f.* = 105, $p << 0.001$). However, a more simplified model for LogTot-Hg $[C_2]$, with δ^{15}N and length as predictors explains 63 % of the variance in the perch dataset ($R^2=0.63$, *d.f.* = 108, $p << 0.001$):

$[C_2]=a+b_1(\delta^{15}\mathrm{N})+b_2(Length)$

This simple model for Tot-Hg in perch supports bioaccumulation as well as trophic transfer and biomagnification of Hg.

4 Discussion

4.1 Food web structure

Biota samples collected for this study spans approximately five trophic levels from zooplankton and benthos representing primary consumers to fish. Trophic structure (based on δ^{15}N) and δ^{13}C signatures suggest that the connection between benthic and pelagic food chains in the Gunneklevfjord is weak. The δ^{13}C signatures in fish are predominantly littoral, suggesting a benthic dominated diet, and the δ^{13}C signatures mainly reflect amphipods, chironomids and other fish. This fits well with the general knowledge of vegetated habitat preference for the species caught. However, it is known that perch feeds on plankton in its first year (Pethon, 1989), before the diet shifts towards benthic invertebrates and small fish and our data on correlation for δ^{15}N with δ^{13}C, age and length for perch support a gradual ontogentic dietary shift towards a more littoral based diet (Persson, 1986). From length 15-20 cm perch mainly feed on fish, including its own species (Pethon, 1989). The perch collected is in the length range of 6 - 46 cm, with median length 16 cm. Hence, approximately half of the individuals of perch in our dataset probably had a diet dominated by other fish, which is reflected in the high trophic position of perch in the food web.

4.2 Trophic transfer of Hg in biota

Our results indicates that fish Tot-Hg concentrations is reduced since the last survey (Berge and Knutzen, 1989). Bioaccumulation of Tot-Hg in the food chain has been detected with a *TMF*> 1 for Tot-Hg, based on fish across several trophic levels. This mainly reflects the high proportion of the bioaccumulating MeHg in fish (63.4-82.5%), though %MeHg has been reported higher in other studies (Jones et al., 2014). The Tot-Hg concentrations in perch in Gunneklevfjord overlapped with concentrations reported from two nearby lakes within the same catchment area where there are no direct Hg sources or contaminated sediment, only atmospheric deposition in the catchment area (Økelsrud et al., 2016), questioning the importance of the sediment as a source. In these lakes, the mean Tot-Hg concentrations in perch for three sampling sites were 0.46, 0.65 and 1.68 mg Tot-Hg kg^{-1} dw, whereas mean for perch from Gunneklevfjorden is 1.4 mg Tot-Hg kg^{-1} dw, supporting that there are site specific factors influencing the availability and uptake of MeHg. Further, *TMF* = 2.1 for perch in Gunneklevfjorden is lower than the *TMF* = 4.6 recently reported from the two non-industrialized lakes within the same catchment area (Økelsrud et al., 2016). It is worth noting that the calculation of *TMF* based on Tot-Hg and not MeHg, probably affects *TMF* in the direction of a lower value (a less steep slope) due to the high proportion of inorganic Tot-Hg at the lowest trophic level. We have sought to reduce this impact by calculating *TMF* based on fish only and excluding benthos, while *TMF* in the referenced study (Økelsrud et al., 2016) included benthos. The *TMS*=0.106 calculated in Gunneklevfjorden is also lower than mean *TMS* reported by Lavoie et al. (2013), compiling 69 studies reporting on 205 aquatic food webs worldwide (*TMS* 0.16), suggesting that biomagnification of Hg in Gunneklevfjorden is generally low. Our findings are in accordance with Hodson et al. (2014) who found that Tot-Hg and

MeHg concentrations in sediment and pore water were unrelated to concentrations in amphipods and yellow perch (*Perca flavescens*) in a study in the contaminated Lake Saint Francis in Canada. Possible explanations for the somewhat limited transfer of Hg through the food chain in Gunneklevfjorden is discussed in the following chapters.

4.2.1 Low uptake of MeHg in benthos

Low uptake rates of MeHg from the Gunneklevfjorden sediment to lower trophic levels in the food chain may be due to low MeHg concentrations in sediment and pore water (**Table SI-A**) or limited exposure of important surface dwelling benthic groups (molluscs and amphipods) to contaminated sediment and pore water. The concentrations of Tot-Hg in zooplankton and benthos were almost similar. The pore water concentrations of Tot-Hg are 100-200x the concentrations measured in the water (**Table SI-A**). However, concentrations of MeHg in pore water is only 0.3-3x the concentrations in water, and %MeHg is significantly higher in water than in pore water. This reveals relatively low bioavailability of MeHg in the sediments, which can explain a significantly higher $BCF_{Tot\text{-}Hg}$ in zooplankton than in benthos when BCF is calculated on basis of water concentrations for zooplankton and pore water concentration for molluscs. Low bioavailability of Hg in the Gunneklevfjord sediment is in accordance with other studies reporting that recent deposited, transported or mineralized Hg tends to be more reactive towards methylation and bioaccumulation than Hg that has aged in place in sediments and soil (Hsu-Kim et al., 2018)

4.2.2 Length of food chain

Results indicating an increase in Hg with relative trophic level in fish is in accordance with the well-established phenomenon of Hg biomagnification in food webs (McIntyre and Beauchamp, 2007, Garcia and Carignan, 2005, Cabana et al., 1994, Vander Zanden and Rasmussen, 1996). Consequently, the length of the food chain will influence on the concentrations accumulated in top predators. The number of trophic levels revealed in our dataset are approximately two from secondary littoral consumers to fish, whereas the most numerous fish species perch covers approximately two-three trophic levels. This is similar to the food webs investigated in nearby lakes (Økelsrud et al., 2016), and therefore not a plausible explanation for the somewhat low *TMF* in Gunneklevfjorden.

4.2.3 Possible effects of species specific migration patterns and seasonality in prey abundance

Seasonal migration patterns, residence time and dietary preferences may affect Tot-Hg uptake in predator fish. It is known that perch can migrate out of brackish areas and into rivers for wintering, pike has been observed in the adjacent Frierfjorden and can probably migrate as long as the salinity is below 10 ‰ (Pethon, 1989), flounder is a marine fish with a wide salinity tolerance and typically migrates into the sea and into deeper waters in the winter, and the eel's life cycle involves long migrations and several years in the ocean before it returns to freshwater localities. Rudd is the species most likely to stay in Gunneklevfjord all year to forage on vegetation, molluscs and insect larvae. The two individuals of rudd in our data set had the highest %MeHg (74.6 and 82.5 %MeHg), which may reflect their stationary living and full-time exposure to the Hg sources in the Gunneklevfjord. Further, seasonality in prey abundance may affect the Hg concentrations in fish. In particular, the chironomids have a life cycle that indicates variation in prevalence throughout the year. In addition, previous studies have shown that Hg methylation rate in the sediment is not constant throughout the year, and MeHg pore water concentrations may be low at some times of the year (Razavi et al., 2013).

4.2.4 Habitat reliance

Benthos that do not live permanently buried in the sediment, such as molluscs and amphipods, are likely to be exposed to bottom water as well as to sediment pore water, and probably also to Hg in the plant material (at least the molluscs) or in the periphyton they might be grazing. Sources like sediment organic matter (SOM), particulate organic matter (POM) and macroalgae were not taken into account in our study. We find that including SOM as a food source might not be correct in this case as the benthic invertebrates found are more likely to feed on detritus, periphyton and bacteria on the sediment, rocks and vegetation. Even though POM may be a source of C for zooplankton, the dominating species *Bosmina longirostris* may be much more selective than *Daphnia* sp. (DeMott, 1982). *B. longirostris* have both a raptorial feeding mode (grabbing particles) as well as a non-selective filtration mode of smaller particles (DeMott, 1982, Bleiwas and Stokes, 1985). Including the very diverse and unspecified POM may not be representative for the diet of *B. longirostris*, and not for the fish preferring *B. longirostris* as their main prey. The similarity in $BCF_{Tot\text{-}Hg}$ for zooplankton and benthos when $BCF_{Tot\text{-}Hg}$ for molluscs is calculated on basis of bottom waters (5 m) rather than pore water (**Table SI-A**), indicates that the availability of Hg and the uptake rates in molluscs and zooplankton in Gunneklevfjorden may be quite similar. In a recent study, we reported Hg concentrations in the macrophytes in Gunneklevfjorden on 50 - 1321 ng Tot-Hg kg^{-1} dw and 1 - 52 ng MeHg kg^{-1} dw (Olsen et al., 2016), that is much lower than the sediment concentrations (**Table SI-A**) though the %MeHg in the macrophytes was higher (1.4-17.3 %MeHg). Hence, macrophytes should be considered a potential Hg source to benthic or vegetation based food webs, as suggested by Beauvais-Fluck et al. (2017), though the exposure to the various sources (sediment, water, macrophytes) may vary with both benthic and predator (fish, birds, mammals) habitat reliance and prey preferences. Our data does not reveal whether the fish in Gunneklevfjorden feed mainly within or outside the vegetation belt. However, several observations supports that the meadow is an important feeding habitat for the fish in Gunneklevfjorden: i) the species found generally have a strong connection to vegetation; ii) we observed a high prevalence of prey in the vegetation area compared to outside; and iii) there was a littoral dominance of δ^{13}C-signatures in the fish. Feeding within the meadow means that the benthic prey is exposed to lower Hg concentrations than prey outside the meadow, which was demonstrated by the significant difference in Tot-Hg in benthos within and outside the meadow. The indication that benthos (amphipods) on higher trophic levels are more mobile and move farther away from the meadow than benthos on lower trophic levels (**Figure 5**), is complimentary to earlier studies reporting on the importance of mobility to search for high quality food (Jørgensen and Christie, 2003, Kraufvelin et al., 2006) and emigration to limit population growth and stabilize the community (Christie and Kraufvelin, 2004). Thus, although the Tot-Hg concentrations in sediments and the benthos outside the meadow are higher than within the meadow, these areas may have less significance for the trophic transfer of Hg into fish. Accordingly, because of lower Tot-Hg concentrations in benthos inside the meadow, the dietary exposure of fish to Tot-Hg might be lower when feeding on benthos inside the meadow, though the MeHg exposure may be higher due to a higher %MeHg in sediments. Hence, the correlation between δ^{13}C and Tot-Hg (log-transformed) in fish may be related to enhanced exposure to MeHg when feeding within the shallow meadow despite lower Tot-Hg concentrations in sediments, given the higher fraction of MeHg within the vegetated habitat (**Table SI-A**).

4.2.5 Implications for sediment remediation

Based on our results, the sediment Tot-Hg concentrations outside the meadow seem to be of minor importance for transfer of Hg from sediment to the food web. Given that the purpose of sediment remediation actions are to reduce the Hg concentrations in biota and minimize human risk caused by the consumption of Hg contaminated fish and seafood, we propose that remediation actions within the meadow will have more effect on Hg concentrations in fish than outside the meadow, despite higher Hg concentrations outside.

5 Conclusion

The aquatic food chain in Gunneklevfjorden is based on benthos, primarily amphipods and chironomids, and the submerged meadow is the preferred habitat for both benthos and fish. Molluscs do not seem to represent an important prey to fish, though they may play a role in the remobilization and transfer of Hg from sediment through macrophytes and/or periphyton into food webs. The Tot-Hg concentrations in fish seem to have decreased since the last survey reported in 1989, in the same period as surface sediment concentrations have declined. Despite still high Tot-Hg concentrations in sediment, especially outside the meadow, transfer of MeHg from sediment to the food chain are generally low. We propose that the meadow is the most important exposure site for sediment MeHg, due to habitat reliance and higher %MeHg in sediment, though the importance of other non-sediment sources to Hg in fish should be explored following the reduction in sediment Tot-Hg concentrations. Also, the possible influence of seasonality in biotic as well as abiotic factors on availability and uptake of MeHg into food webs should be investigated. Following these results, we propose that sediment remediation actions outside of the meadow may have limited effect on Tot-Hg concentrations in fish.

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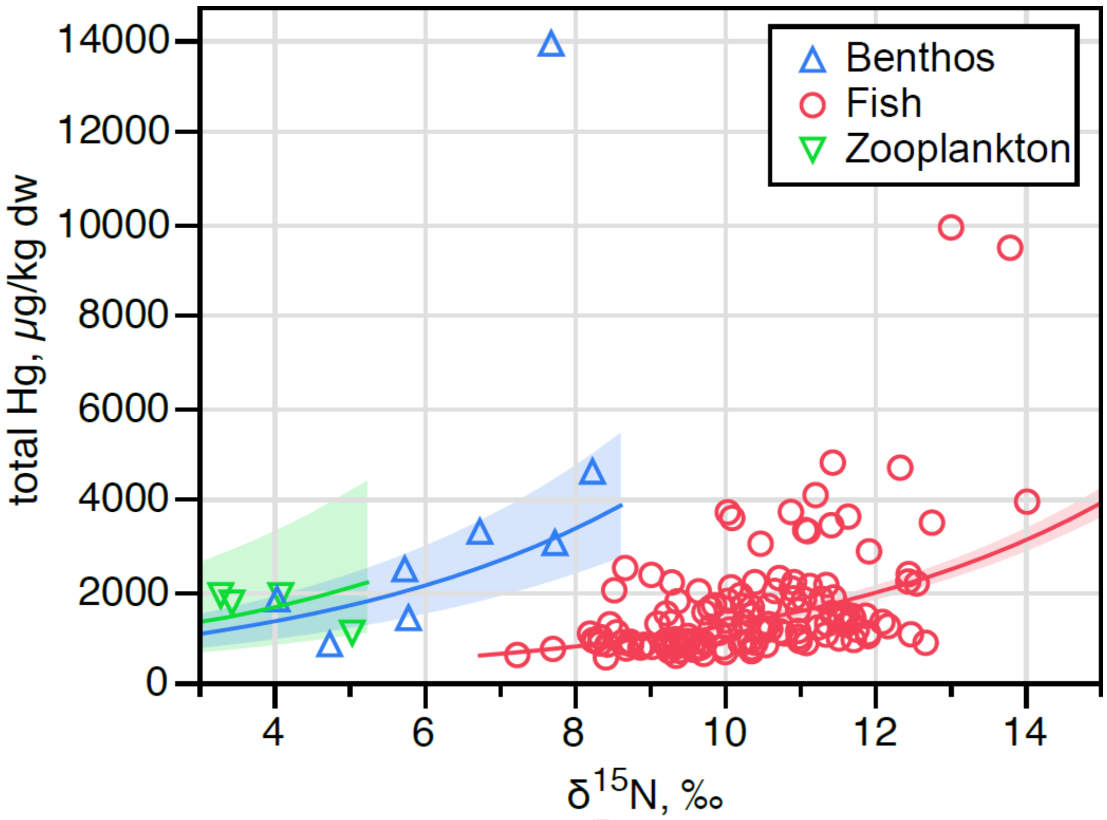
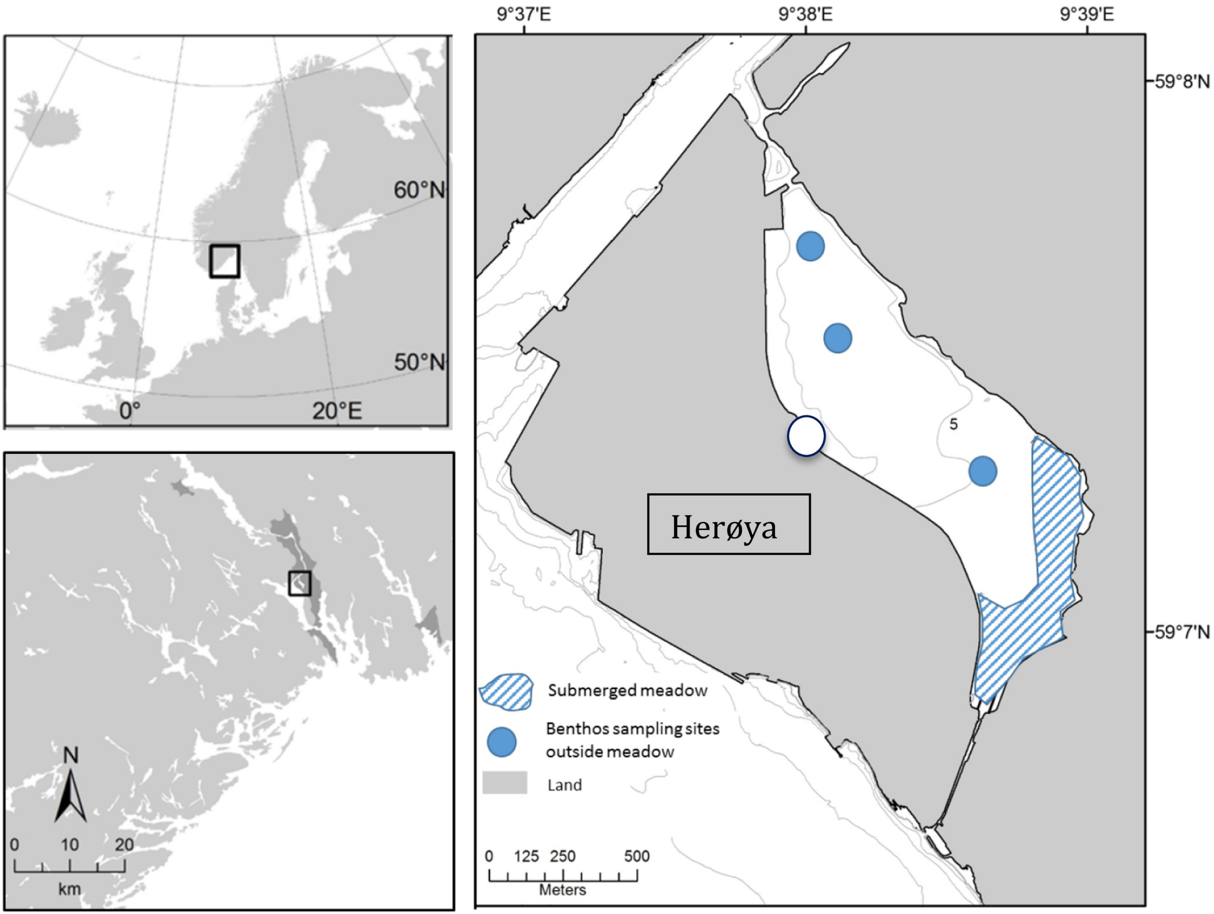


Graphical abstract

Highlights

- The possible influence of macrophytes to bioavailability of Hg studied
- Invertebrates comprise a larger portion of fish diets than zooplankton
- The concentrations of Tot-Hg in benthos, zooplankton and fish are overlapping
- Biomagnification of Hg in fish is low, with TMF = 2.3
- Transfer of sediment MeHg to the food chain is limited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km
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5
Herøya
Submerged meadow
Benthos sampling sites outside meadow
Land
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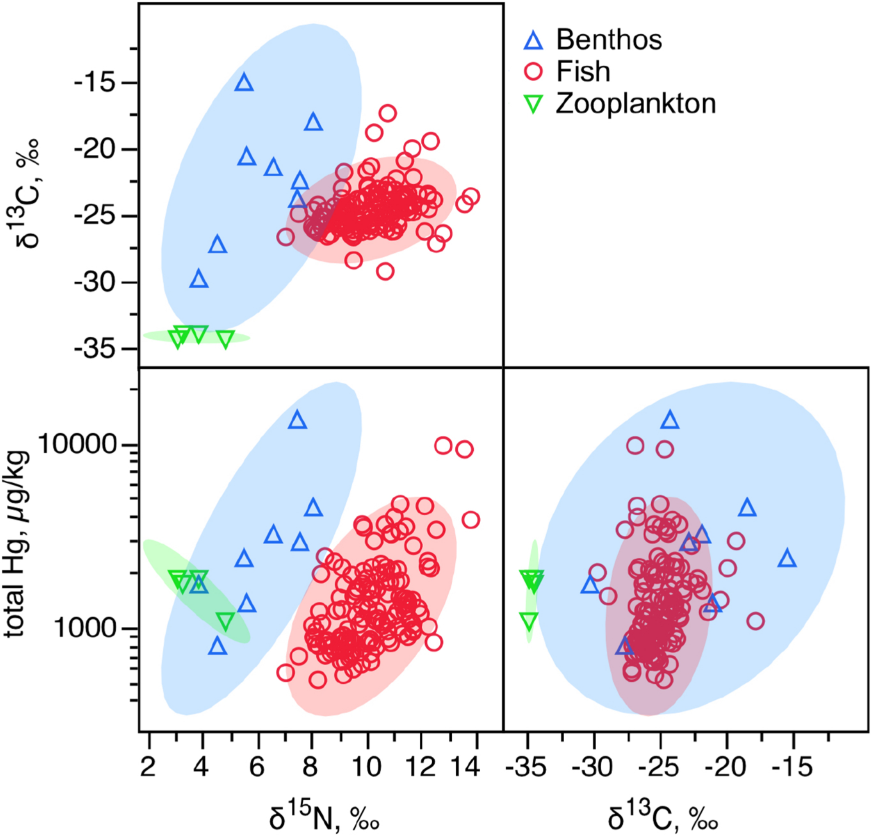
250
500
Meters

Figure 1


△ Benthos
○ Fish
▽ Zooplankton
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-20
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-35
δ13C, ‰
10000
1000
total Hg, µg/kg
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14
δ15N, ‰
-35
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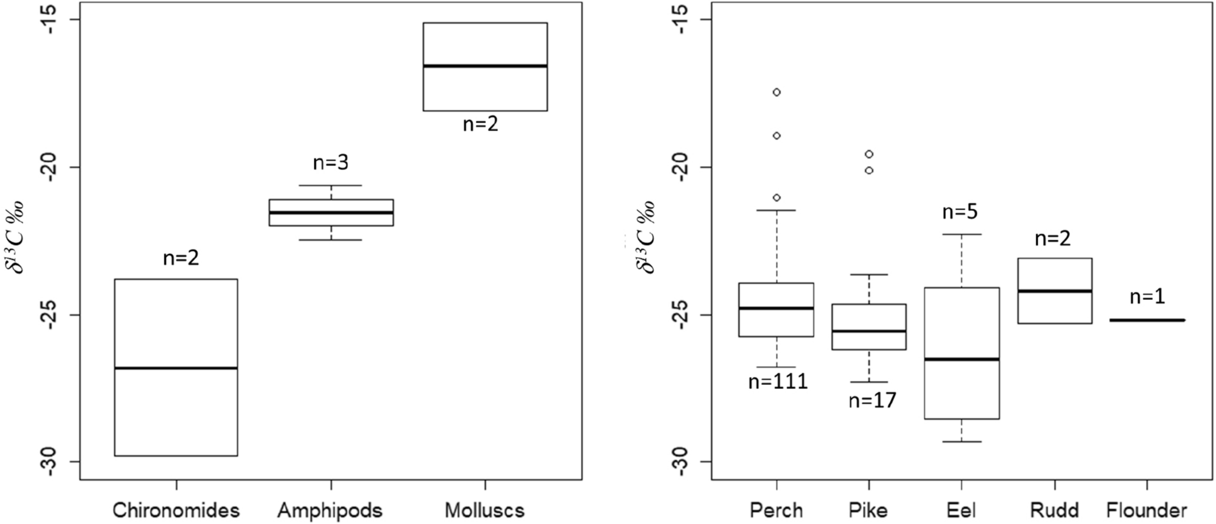
δ13C, ‰

Figure 2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$\delta^{13}C$ ‰
n=2
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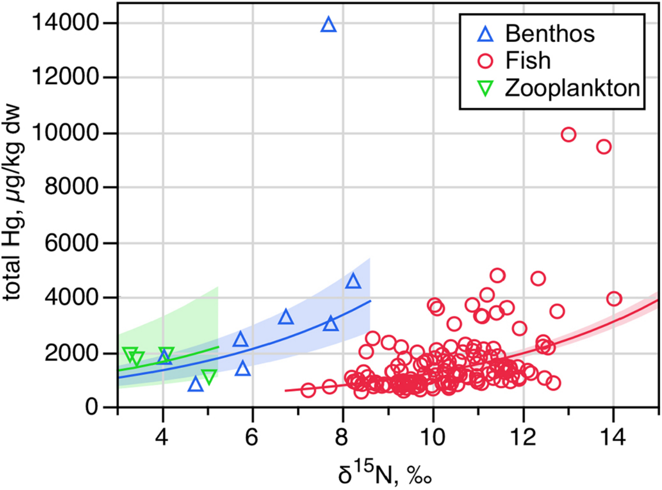
n=2
Chironomides
Amphipods
Molluscs
-15
-20
-25
-30
$\delta^{13}C$ ‰
n=111
n=17
n=5
n=2
n=1
Perch
Pike
Eel
Rudd
Flounder

Figure 3



14000
12000
10000
8000
6000
4000
2000
0
total Hg, μg/kg dw
4
6
8
10
12
14
δ^{15}N, ‰
Benthos
Fish
Zooplankton

Figure 4

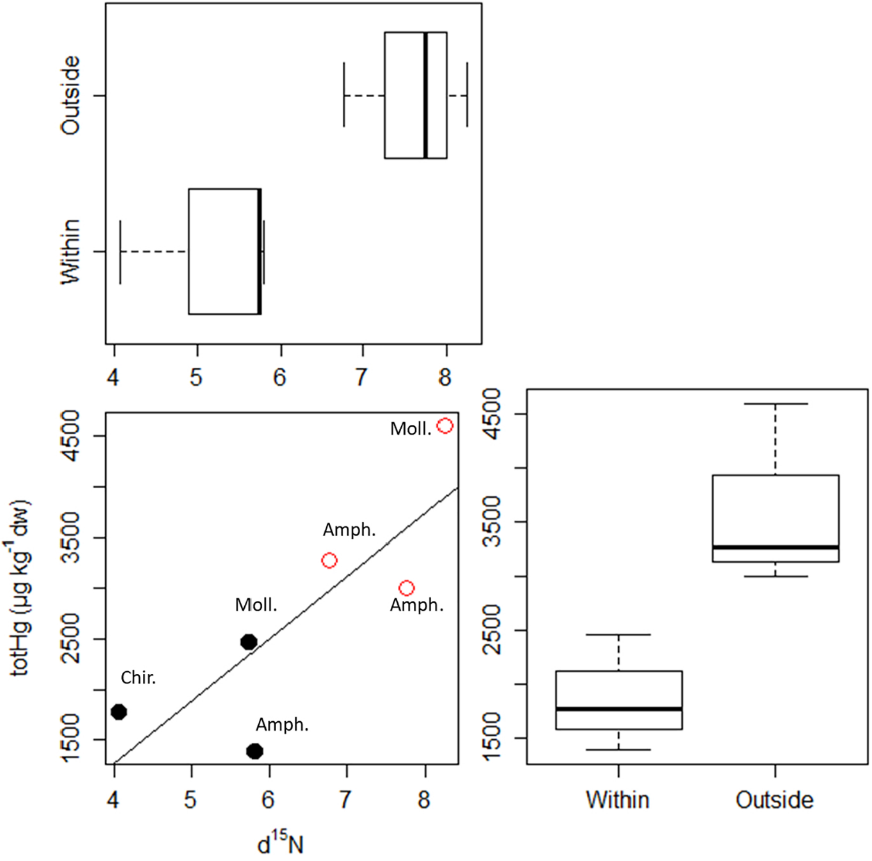

Outside
Within
4
5
6
7
8
totHg (µg kg-1 dw)
4500
3500
2500
1500
Moll.
Amph.
Moll.
Amph.
Chir.
Amph.
d15N
4500
3500
2500
1500
Within
Outside

Figure 5