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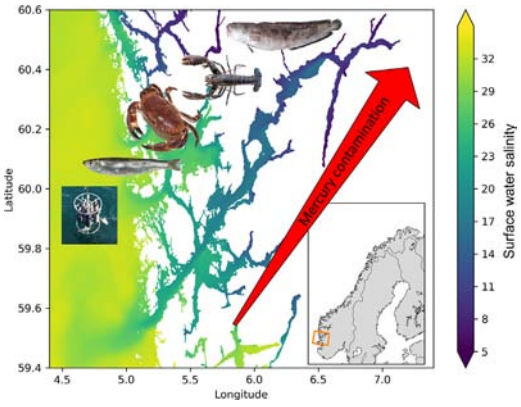
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HIGHLIGHTS

- Hardangerfjord is a mercury (Hg) contaminated ecosystem with a legacy point source.
- Hg species were analyzed in seawater, sediment and seafood.
- Hg concentrations in seawater, sediment and biota increased towards the inner fjord.
- Demersal fish from the entire fjord exceeded acceptable Hg limits for human consumption.

GRAPHICAL ABSTRACT



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ABSTRACT

Hardangerfjord is one of the longest fjords in the world and has historical mercury (Hg) contamination from a zinc plant in its inner sector. In order to investigate the extent of Hg transferred to abiotic and biotic ecosystem compartments, Hg and monomethylmercury (MeHg) concentrations were measured in seawater, sediment, and seafood commonly consumed by humans. Although total mercury in seawater has been described previously, this investigation reports novel MeHg data for seawater from Norwegian fjords. Total Hg and MeHg concentrations in seawater, sediment, and biota increased towards the point source of pollution (PSP) and multiple lines of evidence show a clear PSP effect in seawater and sediment concentrations. In fish, however, similar high concentrations were found in the inner part of another branch adjacent to the PSP. We postulate that, in addition to PSP, atmospheric Hg, terrestrial run-off and hydroelectric power stations are also important sources of Hg in this fjord ecosystem. Hg contamination gradually increased towards the inner part of the fjord for most fish species and crustaceans. Since the PSP and the atmospheric Hg pools were greater towards the inner part of the fjord, it is not entirely possible to discriminate the full extent of the PSP and the atmospheric Hg contribution to the fjord food web. The European Union (EU) Hg maximum level for consumption was exceeded in demersal fish species including tusk (*Brosme brosme*), blue ling (*Molva dypterygia*) and common ling (*Molva molva*) from

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the inner fjord (1.08 to 1.89 mg kg^{-1} ww) and from the outer fjord (0.49 to 1.07 mg kg^{-1} ww). Crustaceans were less contaminated and only European lobster (*Homarus gammarus*) from inner fjord exceeded the EU limit (0.62 mg kg^{-1} ww). Selenium (Se) concentrations were also measured in seafood species and Se-Hg co-exposure dynamics are also discussed.

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1. Introduction

Mercury (Hg) is a widespread global pollutant with significant implications for environmental and public health. Anthropogenic activities, such as emissions from coal-fired plants and mining have significantly increased the concentrations of Hg and monomethylmercury (MeHg) in the environment, including marine ecosystems and their inhabitants (Lamborg et al., 2014). Increased MeHg concentrations in some Arctic marine biota have been reported in comparison to pre-industrial times (Braune et al., 2005), however, the ocean is not uniformly polluted (Lamborg et al., 2014). For example, Vo et al. (2011) reported an increase in MeHg concentrations during a 120-year period in black-footed albatross museum specimens sampled from the Pacific Ocean, but recent studies have reported small-scale temporal declines in MeHg concentrations in coastal and pelagic fish species from the Atlantic Ocean (Cross et al., 2015; Lee et al., 2016). Although air-sea exchange, terrestrial inputs and atmospheric processes are recognized as important drivers of the Hg cycle, numerous important processes governing marine Hg biogeochemical cycling and bioaccumulation have a high degree of uncertainty and remain poorly understood (Strode et al., 2007; Black et al., 2012).

Inorganic Hg may exist in different forms such as elemental Hg, Hg^{2+} inorganic complexes, Hg^{2+} organic complexes, and Hg^{2+} with different degrees of bioavailability. However, inorganic Hg can be methylated by anaerobic, mainly sulfate reducing, bacteria in marine sediments (Compeau and Bartha, 1987) and also in the open water column (Topping and Davies, 1981). MeHg is highly neurotoxic and the most bioavailable form of mercury (Hong et al., 2012). Methylation dynamics and trophic transfer are critical processes involved in MeHg bioaccumulation in coastal and open ocean food webs (Bank et al., 2007; Senn et al., 2010). MeHg easily biomagnifies in the marine food web, and in top predator marine organisms 70 to 100% of the total Hg may be present in the MeHg form (Bloom, 1992; Magalhães et al., 2007; Hong et al., 2012). Fish may bioconcentrate MeHg as much as 10^6-fold compared to low seawater concentrations (Watras and Bloom, 1992).

Atmospheric deposition is considered an important source of Hg to the marine environment (Driscoll et al., 2013). Hg precipitated in terrestrial catchments and transported via run-off can be substantial for aquatic ecosystems including streams, rivers, ponds, lakes, and coastal zones. Although biotic methylation of inorganic Hg in the sediment and in the water column is the primary process governing MeHg, abiotic methylation may also occur, but at a far lower rate (Weber, 1993; Celo et al., 2006). Hg methylation in marine sediments has been shown to be enhanced by anaerobic conditions, increased temperature, decreased pH, and intermediate concentrations of organic carbon (Ullrich et al., 2001). Additionally, organic carbon composition and overall quality (i.e., humic substances content), sulfur availability, and fraction of Hg available for methylation have been shown to have important roles in controlling Hg methylation (Avramescu et al., 2011; Bełdowska et al., 2014; Schartup et al., 2014).

Seafood is the main contributor to MeHg exposure in humans (Batista et al., 1996; Al-Majed and Preston, 2000; Olivero et al., 2002) and the EU maximum level (EUML) of Hg (0.5 mg kg^{-1} ww) applies to most fish and fishery products for legal trade (EC, 2006). The interaction between MeHg and seafood nutrients, particularly selenium (Se), may influence the bioavailability and toxicity of MeHg (Ralston et al., 2008), and it is advantageous to measure and evaluate these elements simultaneously, across fish species, to make accurate decisions pertaining to food safety and human exposure.

The Hardangerfjord ecosystem is one of the longest fjords in western Norway (Fig. 1). The fjord is polluted by industry and other anthropogenic Hg pollution sources, including a zinc plant, hydroelectric power stations, and local mining and aquaculture facilities (deBruyn et al., 2006). The zinc plant has existed for ~100 years and produces zinc and aluminum fluoride at a site located 4 km north of Odda in the inner sector of Sørfjord, an arm of the Hardangerfjord (Fig. 1). Zinc ores typically contain Hg and zinc plants may emit high amounts of Hg to the atmosphere. For instance, it is estimated that approximately 107.7 tons of Hg was emitted to the atmosphere from zinc smelting activities in 2006 in China (Yin et al., 2012). Industrial wastes associated with zinc production with high concentrations of toxic metals were released to Sørfjord until 1986 (Julshamn and Grahl-Nielsen, 1996) even though a mercury removal system was introduced early in the 1970's. In the 1970's, it was estimated that an average of 1–3 kg of solid phase Hg per day was released into the local environment (Skei et al., 1972; Melhuus et al., 1978), most likely as metacinnabar (HgS). In 1986 the company initiated a waste treatment and processing program storing the main tailings and effluents from the zinc plant on land in mountain tunnels. However, the sediments in the inner part of the Sørfjord were already highly polluted with toxic trace metals including Hg, and today the Hardangerfjord ecosystem is still widely considered to be one of the most trace metal polluted fjords in the world (Skei et al., 1972; Everaert et al., 2017).

Early investigations on toxic trace metal contamination in the area focused on zinc (Zn), arsenic (As), cadmium (Cd), lead (Pb), and Hg in marine organisms such as brown algae (*Ascophyllum nodosum*), blue mussel (*Mytilus edulis*), flounder (*Platichthys flesus*) and saithe (*Gadus virens*). Hg received relatively little attention because Hg concentrations were not very high in the investigated species which were from low positions in the food web (Haug et al., 1974; Stenner and Nickless, 1974; Melhuus et al., 1978; Julshamn and Grahl-Nielsen, 1996). Julshamn et al. (2001) reported a significant decrease in toxic trace metals in Sørfjord following the termination of jarosite discharge in 1986, however, the degree of Hg contamination in demersal fish species was unknown. More recent investigations have reported Hg concentrations in fillets of tusk (*Brosme brosme*), inhabiting the demersal habitats of Sørfjord ~3 times greater than the EUML (Ruus and Green, 2007), and additional data (Kvangarsnes et al., 2012) led the Norwegian Food Safety Authority (NFSA) to issue extended consumption advisories for deep-water fish caught in the entire Hardangerfjord ecosystem, as well as for shellfish from the Sørfjord sector.

In this investigation, we focused on evaluating the spatial extent of Hg and MeHg concentrations in several Hardangerfjord ecosystem compartments including marine organisms consumed by humans, seawater, and sediment. We hypothesized that the zinc plant and surrounding highly polluted sediments, as a point source of pollution (PSP), would be an important driver of Hg contamination and spatial distribution in seawater, sediment, and biota. This would result in higher Hg and MeHg levels in the different ecosystem compartments sampled from the inner sector of the fjord compared to the outer sectors. Additionally, we compared our measurements in seafood to the EUML and discuss Se-Hg co-exposure dynamics.

2. Materials and methods

2.1. Study area

The Hardangerfjord ecosystem is the second longest fjord system in Norway, located in the western coastal region (59.4–60.6°N, 4.5–7.3°E;

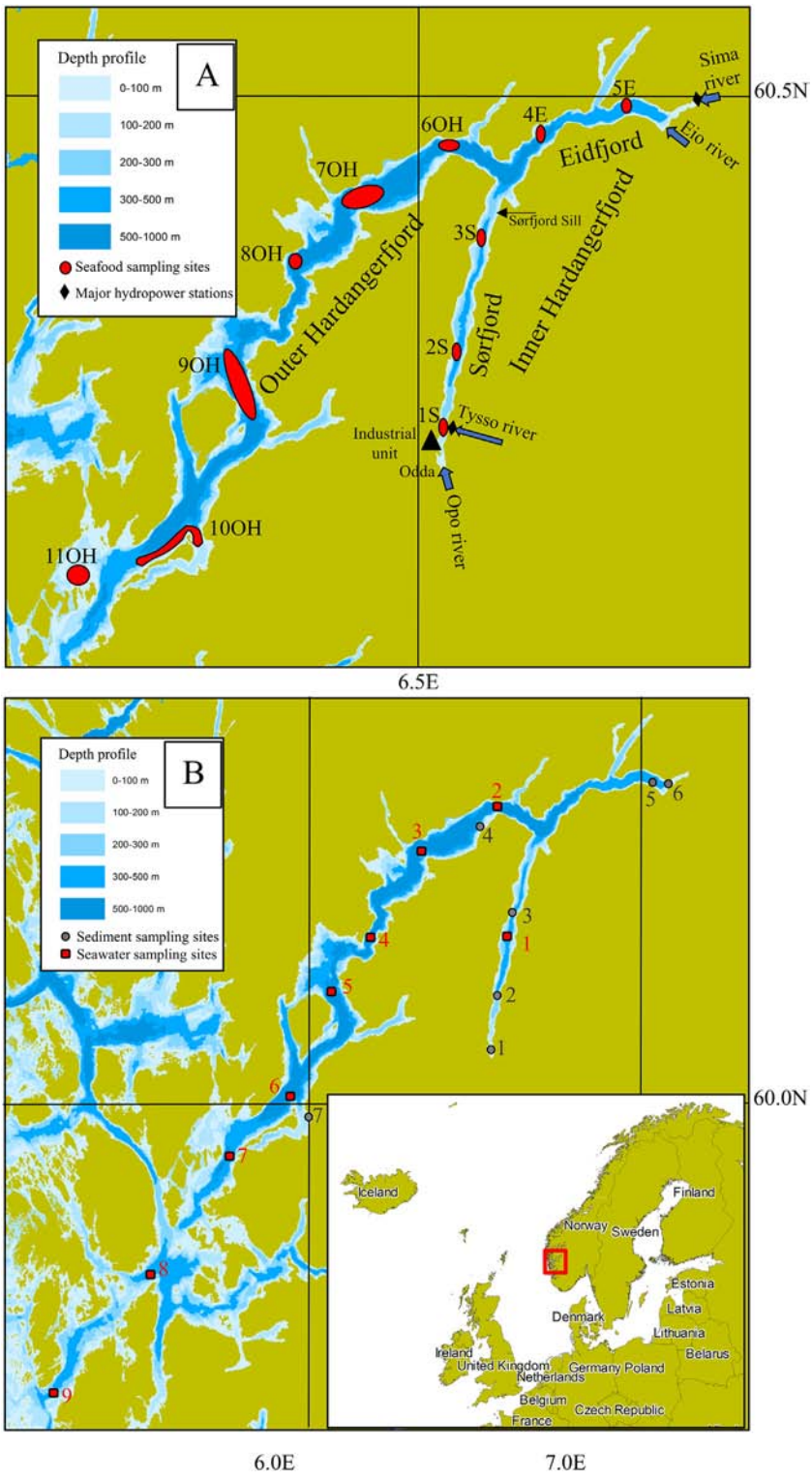


Fig. 1. Location of different sampling sites in Hardangerfjord. (A) Fish and crustacean species sampled in 2011 and (B) sediment and seawater sampled in 2015 and 2018. The letters after site numbers in map A represent the names of the fjords (S: Sørfjord; E: Eidfjord; OH: Outer Hardangerfjord). Details of biotic samples collected from each site are described in Table 1.

Fig. 1). The water depth ranges from 120 to 800 m and the fjord has several basins separated by shallower sills. The fjord ecosystem is connected to the ocean through one main fjord mouth and three narrower channels to the north. At the inner part, the fjord branches into Sørfjord to the south and Eidfjord to the northeast (Fig. 1). The Sørfjord is ~40 km long and up to 1 to 2 km wide and is substantially shallower than the main fjord, with depths of ~100 to 350 m and only ~50 m at the head of the fjord (Fig. 1). The Opo River is the main source of freshwater for Sørfjord. The Opo flows north at the head of the fjord within the Odda municipality (Fig. 1), and has a catchment area of 483 km^2 (Pettersson, 2008). River Tysso, with a catchment area of 390 km^2, is another large river which flows into the southern part of Sørfjord close to the PSP at Tyssedal that also houses a power station (Fig. 1). Eidfjord is the northwards fork extension of Hardangerfjord and is ~29 km long with depths reaching ~400–600 m. The Eio and Sima Rivers are both main sources of freshwater to the Eidfjord sector

of the Hardangerfjord ecosystem (Fig. 1), with catchment areas of 1173 km^2 and 146 km^2 respectively (Pettersson, 2008). Additionally, there is another hydroelectric power station located on the Sima River (Fig. 1). Apart from these four major rivers, the Glacier Folgefonna, consisting of three sub-glaciers with a total area of 200 km^2, is an important source of freshwater along with several other low-order and headwater streams within the catchment area of the fjord.

2.2. Sediment, seawater and seafood sampling and preparation

Fish were caught during cruises organized by the Institute of Marine Research (IMR) as part of a larger Hardangerfjord study. The demersal deep-water fishes blue ling (*Molva dypterygia*) (4 sites) and tusk (*Brosme brosme*) (8 sites) were caught using long line fishing. Common ling (*Molva molva*) (7 sites) and Atlantic wolffish (*Anarhichas lupus*) (2 sites) were sampled using a trammel net and European sprat (*Sprattus sprattus*) (5 sites) were sampled using purse seine nets. Crustacean species including brown crab (*Cancer pagurus*) (2 sites), European lobster (*Homarus gammarus*) (3 sites) and Norway lobster (*Nephrops norvegicus*) (1 site) were caught using lobster trap and trammel nets. All seafood sampling was conducted during 2011 (Table 1 and Fig. 1A). Due to a low number of samples, data for wolffish and Norway lobster were not included in the spatial distribution analyses.

All fish and crustacean specimens were shipped whole and frozen to the Institute of Marine Research, Bergen, Norway. Individual weights (g) and lengths (cm) of fish and crustaceans were measured and registered in the Laboratory Information Management System (LIMS). For all fish species except sprat, skin and bone free fish fillets were dissected. For tusk, we also analyzed liver tissue. For sprat, 25 whole fish were composited and homogenized. For European and Norway lobster, the tail meat was dissected, while for the brown crab, both claw meat (both claws) and brown meat (mixture of hepatopancreas, gonads and internal white meat), were sampled and analyzed. All biota samples were homogenized using a food processor, and all samples, except liver of tusk and brown meat of crab were subsequently lyophilized. After lyophilization to a constant mass, the water content (% moisture) of each sample was calculated and recorded prior to Hg and Se analyses.

Sediment samples (7 sites) were collected from the top 15 ± 2 cm of the bottom sediment using a van Veen grab or by diving. The sediment sampling was conducted during April – July 2015. The samples were frozen (−30 °C) before being sent to the laboratory for analyses.

Seawater samples (9 sites) were collected during May 28–31, 2018 on the *RV* Hans Brattstrøm (Fig. 1B). Seawater was collected using acid-washed Niskin-Type oceanographic general purpose, plastic water samplers (2.5 L model; Hydro-Bios Inc.) at depths of 15, 50, and 300 m. Trace metal clean sampling techniques (Bravo et al., 2018) were employed using acid-washed 120 mL and 250 mL Teflon bottles (Nalgene FEP). Teflon bottles and silicon tubing were acid washed in a Milestone acid-washer using 37% ultra-pure, trace metal grade HNO_3 and were rinsed five times using Milli-Q deionized water. The Niskin type plastic water samples were acid washed using two consecutive overnight, acid baths (1 HNO_3 and 1 HCL at 10% volume:volume prepared with milli-Q water). Teflon bottles and Niskin type bottles were dried in an EPA clean 100 room under a laminar flow hood. Bottles were stored in double plastic bags before and immediately after sampling seawater. Seawater was collected using a standard oceanographic rosette (Hydro-Bios, Inc.) and samples were transferred to individually labeled Teflon bottles using acid-washed silicon tubing that was rinsed between samples with deionized water and stored in a sterile and clean plastic bag. Seawater was then acidified using 0.5% ultrapure HCl (volume:volume) and placed in a dark refrigerator (4 °C) prior to laboratory analyses.

2.3. Total mercury and selenium measurements in biota

The concentrations of Hg and Se were determined using inductively coupled plasma-mass spectrometry (ICP-MS) after microwave digestion. First, weighed samples were digested using concentrated (65%) HNO_3 and 30% H_2O_2 in a microwave oven (Milestone Microwave digestion system: MLS-1200 MEGA Microwave Digestion Rotor - MDR 300/10). Hg and Se concentrations were determined using quantitative ICP-MS (Agilent 7500 with collision cell and ICP-ChemStation software). A standard curve was used to determine the concentration of Hg and Se. Germanium (Ge), thulium (Tm) and rhodium (Rh) were used either individually or in combination as internal standards, and gold (Au) was added to stabilize the Hg signals. The method is a CEN standard and Norway accredited laboratory method (ISO 17025) for these two elements (NMKL, 2007; CEN, 2009) and is described in detail elsewhere (Julshamn et al., 2007). Accuracy and precision of these methods have been tested by analyzing certified reference materials and the recoveries of both Hg and Se ranged from 80% to 120%. Certified reference material (CRM) 1566 (oyster tissue) from the National Institute of Standards and Technology (NIST, Gaithersburg, USA) and lobster hepatopancreas (TORT-2, TORT-3) from the National Research Council of Canada (Ottawa, Canada) were used for measurement quality control by including them in each sample run. The limits of quantification (LOQ) of this method were 0.005 and 0.01 mg kg^{-1} dry weight (dw) for Hg and Se, respectively.

2.4. Mercury speciation in sediment samples

Methylmercury concentrations in sediment samples were measured using EPA method 1630 (USEPA, 1998). Samples were prepared by leaching potassium bromide and copper sulfate solution to release the organic Hg species from inorganic complexes. MeHg was subsequently extracted by dichloromethane. An aliquot of the dichloromethane was then back-extracted into ultrapure deionized water by purging with argon. Samples were treated with sodium tetraethyl borate to form MeHg. Inorganic Hg was simultaneously converted to diethyl Hg. The ethylated Hg species are volatile and are stripped of the solution by purging with N_2 and then adsorbed onto Tenax traps. Hg-species were then thermally desorbed from the Tenax traps in a stream of helium

Table 1
Number of fish and crustacean samples collected from different sites in Hardangerfjord in 2011 (S: Sørfjord; E: Eidfjord; OH: Outer Hardangerfjord). Locations are shown on the map (Fig. 1).

Species	Scientific name	N	Sampling stations (N)										
			1S	2S	3S	4E	5E	6OH	7OH	8OH	9OH	10OH	11OH
Blue ling	*Molva dypterygia*	41			5		2		20	6	7		
Common ling	*Molva molva*	30	1		1			6	3	4	13		2
European Sprat[a]	*Sprattus sprattus*	5				1	1				1	2	
Tusk	*Brosme brosme*	138	2		8	7	24		30		13	32	22
Wolffish	*Anarhichas lupus*	4								3	1		
Brown crab	*Cancer pagurus*	20		10								10	
European lobster	*Homarus gammarus*	26		5						11		10	
Norway lobster	*Nephrops norvegicus*	10										10	

[a] Each sample is a composite of 25 whole specimens.

and separated by means of isothermal gas chromatography. Finally, the methyl/ethylated Hg species are decomposed to elemental Hg and detected using Cold Vapor-Atomic Fluorescence Spectroscopy (CV-AFS) by heating a pyrolysis column to 700–800 °C. The LOQ was 0.05 μg kg^{-1} dw. Total Hg in sediment was measured using laboratory accredited methods (EN ISO12846) and Cold Vapor-Atomic Absorption Spectrometry (CV-AAS) technique (ISO12846, 2012). The LOQ was 0.001 mg kg^{-1} dw and the measurement uncertainty was 20%. The sediment analyses were conducted by Eurofins Environment Testing Norway AS, Moss, Norway.

2.5. Mercury speciation in seawater

Inorganic Hg and MeHg concentrations in unfiltered seawater samples were simultaneously measured using the species-specific isotope dilution, and a GC-ICP-MS method developed for Hg speciation at ultra-trace levels in seawater (Monperrus et al., 2005; Cavalheiro et al., 2016; Bravo et al., 2018). The analyses were operated by a capillary gas chromatograph (Trace GC Ultra, Thermo Fisher, equipped with a TriPlus RSH auto-sampler) hyphenated to an inductively coupled plasma mass spectrometer (ICP-MS Thermo X Series 2). Briefly, an aliquot of 100 mL of unfiltered water sample was accurately weighed and spiked with known amounts and of isotopically enriched standards solutions Me(201)Hg and (199)inorganic Hg (ISC Science, Spain). Spiked samples were left overnight for equilibration in a laminar flow hood. The pH of the solution was then adjusted to 3.9 by adding 5 mL of sodium acetate-acetic acid 0.1 M buffer solution and about 1 mL of ultrapure ammonium hydroxide solution. At last, 250 μL of isooctane (HPLC grade) and 80 μL of sodium tetra-propyl borate solution (5% w/v, Merseburger Spezial Chemikalien, Germany) were added to achieve the derivatization of the Hg species and its subsequent extraction into the GC solvent. The vials were capped and shaken for 20 min at 400 rpm (orbital shaker); then the isooctane was recovered and analyzed in triplicate by GC-ICP-MS.

All materials were cleaned prior to use according to ultra-trace standard operating protocols (Bravo et al., 2018). In absence of any Certified Reference Material available for organomercury species, quality assurance and quality control (QA/QC) was based on reagent blank analyses, replicated assays and an extensive QA/QC procedure described elsewhere (Cavalheiro et al., 2016). Additionally, repeated participations in international inter-laboratory comparison exercises (GEOTRACES intercalibration cruises for Hg species in seawater) complement the QA/QC effort.

Inorganic Hg concentrations measured in the blanks averaged 0.016 ± 0.003 ng L^{-1}, whereas no MeHg was observed in the blanks. The MeHg blank equivalent concentration for the GC-ICP-MS instrument was estimated at 0.002 ± 0.001 ng L^{-1}. The detection limits of this method were 0.03 ng L^{-1} for inorganic Hg and 0.008 ng L^{-1} for MeHg, respectively. The measurement error (calculated by analyzing each sample three times) was <2.9% and 4.9% for inorganic Hg and MeHg concentrations, respectively. All seawater samples were analyzed at the IPREM laboratory (CNRS/University of Pau, France) within 28 days after sampling.

2.6. Salinity measurements and modeling

The salinity was observed in situ using a portable instrument (SAIV A/S SD 208) measuring the conductivity, temperature, and depth (CTD). The instrument was used in STD mode, and calculations of salinity from the conductivity were done automatically using the instrument's software. The accuracy of the salinity is ±0.003 with a range from 0 to 50. The instrument also measured dissolved oxygen (range: 0–20 mg L^{-1} accuracy: ±0.2 mg L^{-1}) supplied by SAIV A/S. The instrument was sampled with a time interval of 1 s and lowered with a speed of 0.2 ms^{-1}. Data was downloaded from the instrument for every 0.1 m in the upper 10 m and for every meter under 10 m depth. In addition to measuring the salinity in situ water was sampled using a multi water sampler slim line 6 with mounted plastic Niskin bottles supplied by Hydro-Bios. Water samples for salinity analyses were taken at a depth of 300 m at every site. The water samples were bottled and analyzed at the in-house salinity lab using a Guildline 8410A portasal (range: 0.004–76, Accuracy: ±0.003). By comparing the in-situ measurements to the salinity data from the seawater samples it became evident that the SAIV SD 208 instrument showed a deviation in its calibration (−0.12) and therefore we used a correction value of +0.12.

The salinity distribution of the fjord was modeled using the Regional Ocean Model System (ROMS) solving the hydrodynamic equations (Haidvogel et al., 2000; Shchepetkin and McWilliams, 2005). The model was set up with a horizontal resolution of 160 m × 160 m, with 35 terrain following coordinates in the vertical. 170 rivers were included with daily run-off from the Norwegian Water Resources and Energy Directorate (NVE) and atmospheric conditions were provided by 2.5 km resolved AROME model provided by the Norwegian Meteorological Institute (http://thredds.met.no). The model was run with an internal time-step of 6 s, writing environmental data as temperature, salinity and currents every hour. Further details of the model setup are described in Albretsen (2011). The model simulation was started 1st of April 2018, where the first month is considered spin-up time. The salinity distribution at the time of the cruise is illustrated as the mean salinity from May 28th to May 31st in 2018, for the sea surface.

2.7. Statistical analyses

Data were log transformed to meet the assumption of normal distribution and homogeneity of variances prior to statistical analyses. Analysis of covariance (ANCOVA) was used for comparison of Hg concentrations across sampling sites for seafood species with length as a covariate to remove the possible effect of length across sites. One-way analysis of variance (ANOVA) was used for crustacean species since length measurements and Hg concentrations were not correlated. In European lobster, the Hg concentrations increased with increasing weights, however since weight was not significantly different between sites, ANOVA was used for comparison across sampling sites. Independent Student's *t*-tests were used to compare length and Hg concentration between the inner and outer sections of Hardangerfjord. For post-hoc comparisons, unequal sample Tukey-HSD tests were used to evaluate the effects of unequal sampling efforts and unbalanced design. Only sites with two or more individuals were considered for spatial comparisons. Distance from PSP was calculated as distance from the industrial unit close to Odda and distance from the open ocean was calculated from the mouth of the Hardangerfjord at Kvinnsvika (Fig. 1). Statistical significance was accepted at $P < 0.05$ (Zar, 2010). All statistical analyses were performed using STATISTICA 13 (Statsoft Inc., Tulsa, USA) or GraphPad Prism 7.02 (GraphPad software Inc., San Diego, CA, USA).

2.8. Selenium health benefit value

Selenium health benefit value (HBV_{Se}) has been suggested as an evaluation index showing the Se amount provided in fish after sequestration of Hg and was calculated using the following formula (Ralston et al., 2016):

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

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

2.9. Bioconcentration Factors and Biota-Sediment Accumulation Factors

Bioconcentration Factors (BCF) and Biota-Sediment Accumulation Factors (BSAF) for tusk were calculated for total Hg and MeHg using the following formulas:

$$BCF = Log\left(\frac{Hg\ concentration\ in\ fillet}{Hg\ concentration\ in\ water}\right)$$

$$BSAF = Log\left(\frac{Hg\ concentration\ in\ fillet}{Hg\ concentration\ in\ sediment}\right)$$

BCF was calculated using average seawater concentration from 15, 50 and 300 m depths closest to the tusk sampling location and 100% of Hg in tusk fillet was assumed to be MeHg.

3. Results and discussion

3.1. Hg and Se concentrations in seafood

Tusk and blue ling fillet samples collected from the inner sector of Hardangerfjord had the highest mean Hg concentrations (1.87 and 1.44 mg kg^{-1} ww, respectively) and all individual fish were above the EUML of 0.5 mg kg^{-1} ww (Table 2). In comparison tusk and blue ling samples from outer Hardangerfjord had lower Hg concentrations, but the mean levels were still higher than EUML (mean = 0.84 and 1.07 mg kg^{-1} ww, respectively). Wolffish (0.14 mg kg^{-1} ww) and sprat (0.01 in outer and 0.03 mg kg^{-1} ww in the inner Hardangerfjord) had the lowest Hg concentrations. In a previous study, Azad et al. (2019) showed that Hg concentrations in blue ling and tusk from the Northeast Atlantic Ocean were similarly high, whereas common ling had lower concentrations and wolffish had the lowest of all demersal fish species analyzed in this study. The high concentrations of Hg in tusk, blue ling, and common ling were likely influenced by their high trophic position, and preference for deep-water, demersal habitats (Bergstad, 1991; Husebø et al., 2002; McMeans et al., 2010). Atlantic wolffish feed on molluscs, echinoderms, and other low trophic level prey species (Falk-Petersen et al., 2010), and this may explain their lower Hg concentrations.

Crustaceans had lower concentrations of Hg than demersal fish species (Table 2) likely as a result of their considerably lower trophic position and similar observations have been reported from Spain (Olmedo et al., 2013). European lobster tail meat sampled from inner Hardangerfjord had the highest mean Hg concentration of all sampled crustaceans (0.62 mg kg^{-1} ww). These values are higher than those previously reported in commercially caught European lobster from Scotland (Barrento et al., 2008; Noël et al., 2011). European lobster from outer Hardangerfjord had a mean Hg concentration of

Table 2
Mean, first and third quartiles, standard deviation and standard error of Hg and Se levels (mg kg^{-1} ww) in muscle tissue and length (cm) of demersal fish and crustacean species from the Hardangerfjord ecosystem, 2011. HBV_{Se} are calculated from mean values.

Species	Scientific name	Area	N	Hg (mg kg^{-1} ww)					Se (mg kg^{-1} ww)					Length (cm)					Percent with Hg ≥ 0.5 (mg kg^{-1} ww)	HBVSe
				Mean	Q25	Q75	SD	SE	Mean	Q25	Q75	SD	SE	Mean	Q25	Q75	SD	SE		
Blue ling	*Molva dypterygia*	Out. Hard.	33	1.07	0.64	1.19	0.83	0.15	0.43	0.38	0.49	0.08	0.01	90.73	80.00	97.00	13.65	2.38	93.9	0.4
		Inn. Hard.	8	1.44	1.07	1.85	0.66	0.23	0.50	0.49	0.53	0.04	0.01	92.33	86.00	100.00	8.41	3.43	100	−1.7
Common ling	*Molva molva*	Out. Hard.	28	0.49	0.19	0.59	0.44	0.08	0.47	0.42	0.51	0.08	0.01	73.93	61.50	83.50	18.63	3.52	35.7	5.0
		Inn. Hard.	2	1.08	0.40	1.76	0.97	0.68	0.65	0.43	0.87	0.32	0.22	78.00	72.00	84.00	8.49	6.00	50	4.7
Tusk	*Brosme brosme*	Out. Hard.	97	0.84	0.42	1.11	0.52	0.05	0.59	0.51	0.64	0.11	0.01	64.26	55.00	75.00	13.68	1.39	64.9	5.0
		Inn. Hard.	41	1.89	1.26	2.19	0.89	0.14	0.72	0.57	0.83	0.23	0.04	62.95	55.00	69.00	9.85	1.54	100	−0.6
Sprat[a]	*Sprattus sprattus*	Out. Hard.	3[a]	0.01					0.43					7.27						
		Inn. Hard.	2[a]	0.03					0.42					8.20						
Wolffish	*Anarhichas Lupus*	Out. Hard.	4	0.14	0.11	0.16	0.04	0.02	0.47	0.27	0.66	0.36	0.18	79.00	74.00	84.00	6.32	3.16	0	5.8
All fishes		Out. Hard.	162	0.63					0.49					76.98					48.6	4.1
		Inn. Hard.	51	1.47					0.62					77.76					83.3	0.8
Brown crab	*Cancer pagurus*	Out. Hard.	10	0.12	0.05	0.20	0.08	0.02	1.47	0.92	1.83	0.80	0.25	14.85	14.40	15.40	1.49	0.47	0	18.6
		Inn. Hard.	10	0.22	0.13	0.30	0.14	0.05	0.76	0.60	0.74	0.33	0.10	15.37	13.40	17.60	2.45	0.77	10	9.5
European lobster	*Homarus gammarus*	Out. Hard.	21	0.19	0.16	0.23	0.07	0.01	0.61	0.49	0.65	0.19	0.04	26.55	25.50	27.00	1.48	0.32	0	7.6
		Inn. Hard.	5	0.62	0.63	0.70	0.13	0.06	0.55	0.49	0.65	0.14	0.06	27.80	27.00	30.00	2.77	1.24	80	5.5
Norway lobster	*Nephrops Norvegicus*	Out. Hard.	10	0.20	0.19	0.22	0.03	0.01	0.99	0.87	1.05	0.21	0.07	18.27	16.90	19.30	1.63	0.51	0	12.4
All Crustaceans		Out. Hard.	41	0.17					1.02					19.89					0	12.9
		Inn. Hard.	15	0.42					0.65					21.59					45	7.5
All species		Out. Hard.	203	0.44					0.72					52.51					27.8	7.8
		Inn. Hard.	67	1.05					0.64					55.29					68	3.5

[a] Each sample is a composite of 25 whole specimens and thus, percent exceeding EUML and HBV_{Se} are not calculated.

0.19 mg kg^{-1} ww that is consistent with their reported range. Claw meat of brown crab had lower Hg concentrations than European lobster with mean values of 0.22 and 0.12 mg kg^{-1} ww in samples from inner and outer Hardangerfjord, respectively (Table 2). The Hg concentrations in brown crab samples from outer Hardangerfjord were similar to the mean reported for this species from the Norwegian coast (0.1 mg kg^{-1} ww) (IMR, 2018), whereas Hg concentrations in samples from the inner fjord were ~2-fold higher. Norway lobster was only sampled from outer Hardangerfjord. The mean Hg concentration in tail meat of Norway lobster was 0.20 mg kg^{-1} ww, similar to European lobster and within the range reported for Norway lobsters caught in other regions of Norway (IMR, 2018). The Hg levels in Norway lobster measured in this investigation were lower than the reported levels in samples from the Mediterranean (Cresson et al., 2014). All crustaceans in this study are benthic carnivores (Cristo and Cartes, 1998; Meeren, 2007; IMR, 2008), and the observed variation in Hg concentrations is likely driven by several factors including body size, toxicokinetics, growth dilution, prey type, ecosystem methylation potential, and species migration patterns.

Overall, Se concentrations in all sampled taxa were less variable than Hg concentrations. Se concentrations in fish species from outer Hardangerfjord were ~50% lower compared to crustaceans analyzed from the same area (mean = 0.49 vs 1.02 mg kg^{-1} ww; Table 2). However, Se concentrations in fish and crustaceans sampled from the inner part of Hardangerfjord, where Hg contamination in sediment and seawater was substantially higher, were similar (mean = 0.62 in fishes vs mean = 0.65 mg kg^{-1} ww in crustaceans). Fish Se concentrations were greater in the inner sector of Hardangerfjord compared to the less contaminated areas of the fjord, whereas crustacean Se concentrations were lower in the inner sector (Table 2; Fig. 1).

The liver in fishes and the hepatopancreas in crustaceans both play significant roles in the distribution of toxic trace metals and high concentrations have often been reported (Engel, 1983; Romeo et al., 1999). Tusk liver contained higher concentrations of both Hg (6.39 vs 1.37 mg kg^{-1} ww) and Se (9.95 vs 0.66 mg kg^{-1} ww) in comparison to fillet tissue (Fig. S1; Table 2). Tusk sampled from the inner sector of Hardangerfjord had greater Hg and Se concentrations in liver compared to the outer sector (Hg: 8.14 vs 4.63 mg kg^{-1} ww; Se: 10.42 vs 9.48 mg kg^{-1} ww). However, brown meat of crab (a mixture of hepatopancreas, gonad and internal connective tissue) sampled from the inner sector had higher Hg concentrations (0.16 vs 0.06 mg kg^{-1} ww), whereas Se concentrations were 42% lower compared to the outer fjord area (0.83 vs 1.42 mg kg^{-1} ww).

Se concentrations increased concomitantly with Hg concentrations in all fish species and Pearson's correlation coefficient ranged from $r = 0.36$ in common ling to $r = 0.49$ in tusk. Similar findings have been reported in several fish species from the Northeast Atlantic Ocean (Azad et al., 2019). Crustacean Se concentrations in muscle varied in the opposite direction of Hg and decreased slightly with increasing Hg concentrations and no significant correlation was observed (Fig. 3). Similarly, hepatic Hg and Se concentrations in tusk increased concomitantly ($r = 0.73$; $P < 0.0001$). However, no correlation was found between Hg and Se concentrations for brown crab hepatopancreas (Fig. S2). Collectively, these findings suggest an organ specific distribution pattern in fish and crustacean species that may be driven by differential uptake mechanisms and toxicokinetics of Hg and Se.

3.2. Seafood Hg concentrations and body size

Hg concentrations increased with both length and weight in all sampled fish species (Fig. 2; Table S1). Length explained a larger part of the variation in fillet Hg concentrations (r^2 between 0.18 and 0.60; $P < 0.01$) than weight (r^2 between 0.20 and 0.40; $P < 0.01$). Over time, Hg bioaccumulation leads to increasing concentration with fish age (Power et al., 2002). Our data shows that fish length is a better proxy for age than weight, as weight can be affected by seasonal variation and food availability, body condition and rates of gonad maturation (Table S1). Hg concentrations were not correlated to length in crustaceans except for Norway lobster, where a negative linear relationship was observed ($r^2 = 0.42$; $P < 0.05$). However, Hg concentrations increased with weight in both European lobster ($r^2 = 0.19$; $P < 0.05$) and Norway lobster ($r^2 = 0.68$; $P < 0.01$), but not in brown crab. Since crustaceans molt, their length increases incrementally and during the periods in between molting steps, the weight may be a better predictor of growth than length (Cameron, 1989).

In many crustaceans, clear differences in Hg concentrations between sexes and interactions with length have been reported. For example, female Norway lobster from the Ligurian Sea (Minganti et al., 1990) and outside Scotland (Canli and Furness, 1993) showed steeper increases in Hg concentrations with length than males. This is likely due to slower growth rates of females in comparison to males and as a result of more energy investment related to reproduction. In this study, crustaceans were not sexed and consequently it was not possible to make comparisons among sexes. Analyzing individuals without information on sex could also mask effects of size on Hg concentrations in crustaceans.

3.3. Spatial variation of Hg in seafood and sediments

In most of the studied species, Hg concentrations were higher in samples of marine organisms collected towards the inner fjord and PSP at Odda than in samples taken in the outer fjord (Figs. 1; 4). This spatial variation was consistent across crustacean and fish species including European lobster, crab, tusk and sprat, but not for blue ling or common ling.

The highest mean concentration of Hg in tusk were observed in the two Eidfjord sites 4E and 5E (2.88 and 1.78 mg kg^{-1} ww), and not in the inner part of Sørfjord where sites 1S and 2S also showed high Hg values (1.90 and 1.36 mg kg^{-1} ww). The differences between sites 4E, 5E and 1S were, however, not significant (Fig. 4) and considering the limited number of tusk collected from 4E (n = 7) and 1S (n = 2), tusk from both branches appear to be contaminated at similar levels. However, the observed high Hg concentrations in tusk from Eidfjord, 47 and 59 km from the PSP, indicate that PSP may not be the only source of Hg to the biota in Hardangerfjord. The tusk from Eidfjord may hence have been influenced by the freshwater inputs from two large rivers and the hydroelectric power station located upstream on the Sima River (Fig. 1). Moreover, substantial transport of Hg from PSP in Odda to Eidfjord does not seem very likely, based on the sediment concentrations of Hg in Sørfjord. Measured concentrations decreased rapidly from 2.26 mg kg^{-1} dw at site 1 to 0.72 mg kg^{-1} dw at site 2 and 0.03 mg kg^{-1} dw at site 3. At site 6 in Eidfjord the sediment concentration was again a bit higher, with 0.17 mg kg^{-1} dw, but still more than an order of magnitude lower than at site 1. The combination of depth (350 m) and the Sørfjord sill may also prevent the movement of contaminants. However, the run-off from Opo River and fjord/estuarine water circulation driven by local tidal conditions may also redistribute and resuspend the contaminants to outside Sørfjord, but the majority of Hg from PSP stays within the Sørfjord sector. If transport of Hg should take place from PSP to Eidfjord, resuspended Hg would have to be transported in higher water layers, over the Sørfjord sill and to the right-hand side due to the Coriolis force effect before being deposited in Eidfjord. Tusk from site 7OH close to Steinstø, had significantly lower Hg concentrations than tusk from the Eidfjord sites, but significantly higher Hg concentrations than tusk from the three outermost sites.

Hg concentrations in sediment increased from the outer Hardangerfjord towards the inner fjord and PSP at Odda (Fig. 5; Table S3) and were in good accordance with the tusk Hg data. Sediments were sampled from the top and intermediate layers (15 ± 2 cm) resulting in an integrated sample which limits our resolution of the interpretation. However, our spatial results are consistent with other studies and show an increasing gradient of mercury from offshore

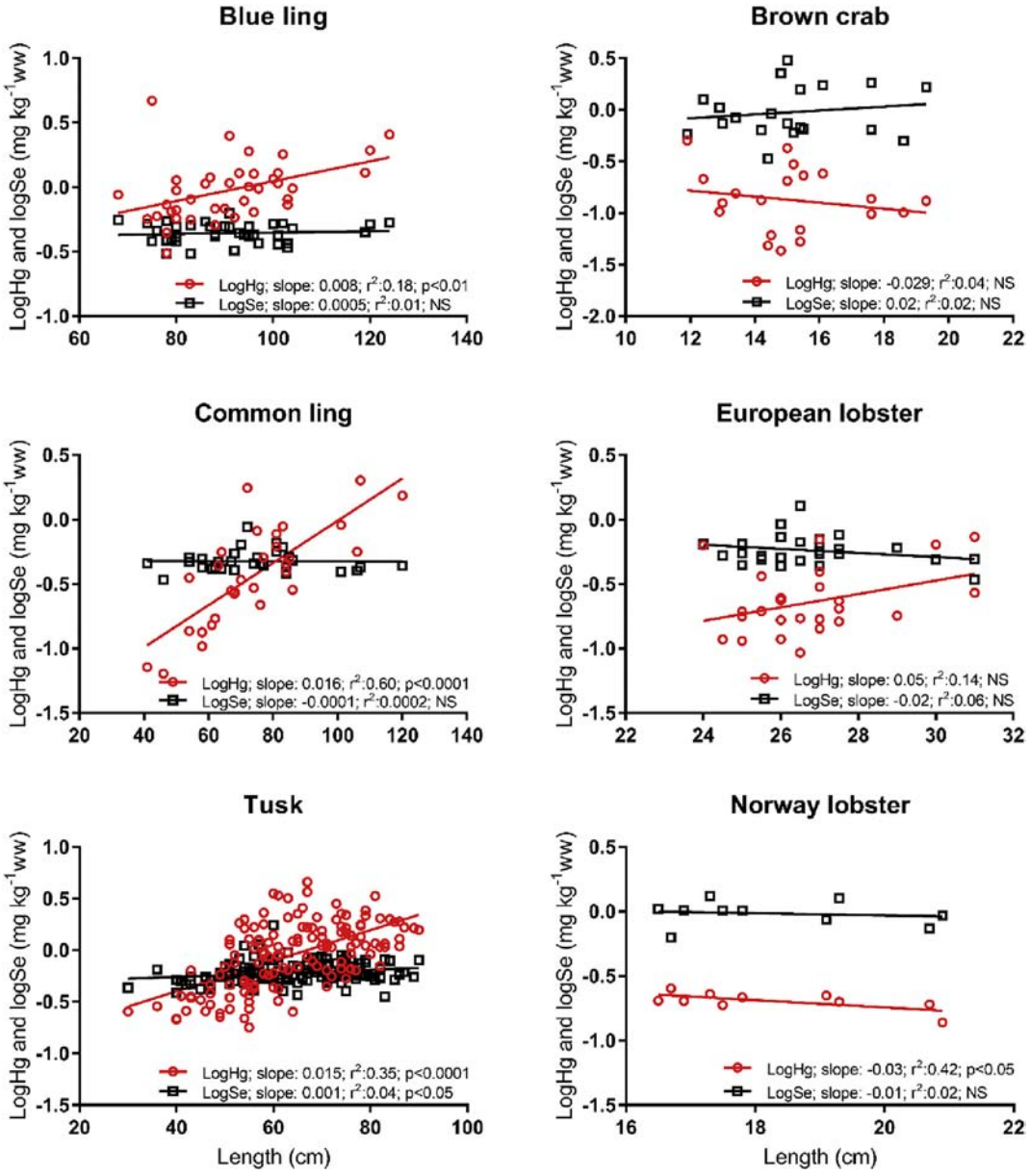


Fig. 2. Linear regression between length and log Hg (red circles) and length and log Se (black squares) in fish and crustacean species from Hardangerfjord sampled in 2011. Slope, r^2 and P are presented. NS = not significant.

to the interior of the fjord. Using a meta-analysis, Everaert et al. (2017) reported Hg concentrations in sediment samples of 0.13 mg kg^{-1} dw in Norwegian inner fjord areas and 0.02–0.03 mg kg^{-1} dw in offshore areas. Their reported levels from inner fjord areas were comparable to Hg concentrations measured at sites 5 and 6 in Eidfjord (0.072 and 0.173 mg kg^{-1}dw), whereas the reported levels in offshore areas were comparable to the Hg concentrations in sediment from the less impacted areas of outer Hardangerfjord (0.015–0.050 mg kg^{-1} dw, sites 3, 4 and 7).

Concentrations of Hg in sediments were not significantly correlated with distance from PSP (Fig. 5). On the other hand, distance from open ocean that takes into account both input from catchment and PSP in the same direction, showed a significant correlation with Hg concentrations in sediment (Kendall tau 0.90; $P < 0.05$) (Table S4). For tusk, there was a significant correlation between Hg concentration and distances from both ocean and PSP, but the correlation with distance from open ocean was stronger (Fig. 5; $r^2 = 0.59$ and $r^2 = 0.76$, respectively) and overall Hg concentrations in both tusk and sediment were in good accordance.

A recently published study, which included tusk specimens from sites 1S and 7OH as well as tusk from other areas on the Norwegian coast, showed that the Hg stable isotope values were different in Hardangerfjord, particularly Sørfjord, compared to the open coast of Norway (Rua-Ibarz et al., 2019). The isotopic composition changed somewhat from Sørfjord to the outer Hardangerfjord, to a profile more similar to that of the open coast. This indicated that in the outer Hardangerfjord there was an influence from the zinc plant in Sørfjord, but also from atmospheric sources.

In areas not impacted by specific sources of pollution, atmospheric deposition of Hg is considered a major source of Hg to the ecosystems (Mason et al., 1994), and in coastal ecosystems Hg mostly originates from freshwater input, organic matter decomposition and erosion (Bełdowska et al., 2014). Fjords naturally have large river inputs often at the ends and these often drain large catchment areas. This freshwater run-off contains Hg deposited over the entire catchment area, including throughfall (Kahl et al., 2007). In the Hardangerfjord, there are two large rivers at the end of Sørfjord located close to PSP and two large rivers at the end of Eidfjord (Fig. 1). The River Eio at the end of Eidfjord has the

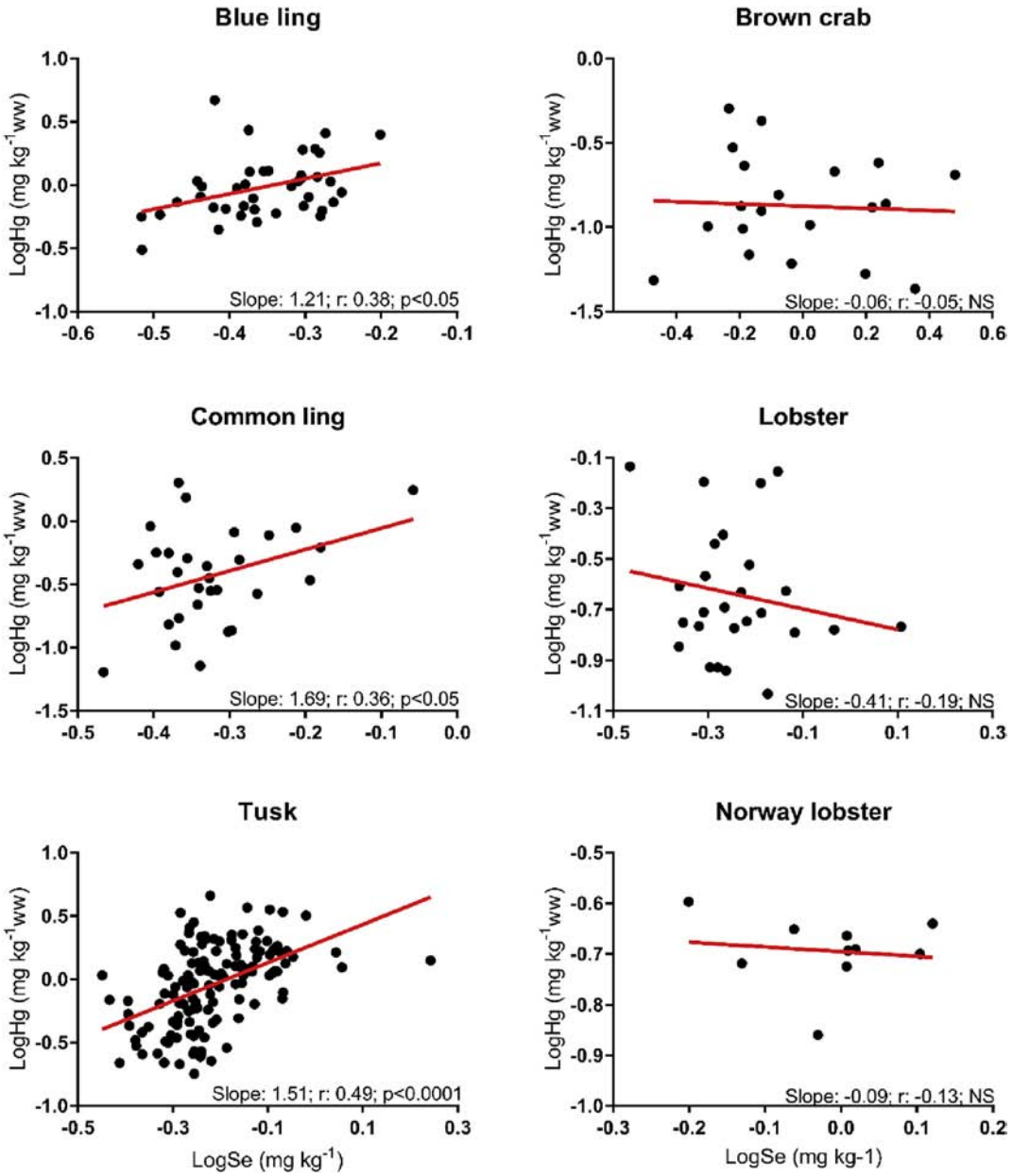


Fig. 3. Relationship between log Hg and log Se (mg kg^{-1} ww) in fish and crustacean species from Hardangerfjord, 2011. Slope, r and *P* are presented. NS = not significant.

largest catchment area in the inner part of Hardangerfjord and is likely to transport larger amounts of atmospherically deposited Hg than the other rivers. Sima, the other main river in Eidfjord sector, also has a hydroelectric power plant that may impact the Hg load as well as methylation (Schartup et al., 2015). In hydroelectric stations, water usually comes from the hypolimnion layer of the reservoir which often has favorable conditions for Hg methylation. Additionally, wetting and drying from periodic flooding of the adjacent soils can also increase MeHg production and bioavailability. These increases in MeHg are largely driven by the timing, frequency and severity of the reservoir flooding. Water released from the reservoir to the fjord is often enriched in MeHg (Pestana et al., 2018) and several studies have reported increased Hg levels in water, plankton and fish from downstream of hydroelectric dams (Hylander et al., 2006; Kasper et al., 2014). Also, in Sørfjord there are several hydroelectric power plants. Freshwater inputs from the rivers is reflected in the salinity measurements and modeling that showed a decreasing trend in surface water salinity from the outer part of Hardangerfjord towards both Sørfjord and Eidfjord (Fig. 6; Fig. S3). The rivers also deliver significant amounts of terrestrial organic matter (Jassby and Cloern, 2000) that may influence Hg methylation and bioavailability dynamics (Lambertsson and Nilsson, 2006).

3.4. Mercury methylation in sediments

Concentrations of MeHg in sediment varied from 0.12 μg kg^{-1} dw at site 4 to 8.4 μg kg^{-1} dw at site 1, closest to PSP (Fig. 5). Atmospheric deposition and terrestrial run-off have been suggested as significant sources of MeHg and inorganic Hg that can be methylated, particularly in estuarine and coastal areas (Mason et al., 2012; Schartup et al., 2015). However, close to the PSP, a relatively high concentration of MeHg indicates that methylation of inorganic Hg originating from the zinc plant is taking place to some degree. High concentrations of both total Hg and MeHg were found close to the PSP (site 1). Comparing the Hg concentrations in sediment at the end of Sørfjord, close to PSP, with Eidfjord (2.26 vs 0.17 mg kg^{-1} dw) and the MeHg concentrations in these sites (8.4 vs 0.82 μg kg^{-1} dw) shows that methylation efficiency (i.e., % MeHg) from PSP is similar (0.37% vs 0.47%) (Table S3). In general, MeHg concentrations in sediment increased towards the inner part of the fjord (Figs. 1; 5) and

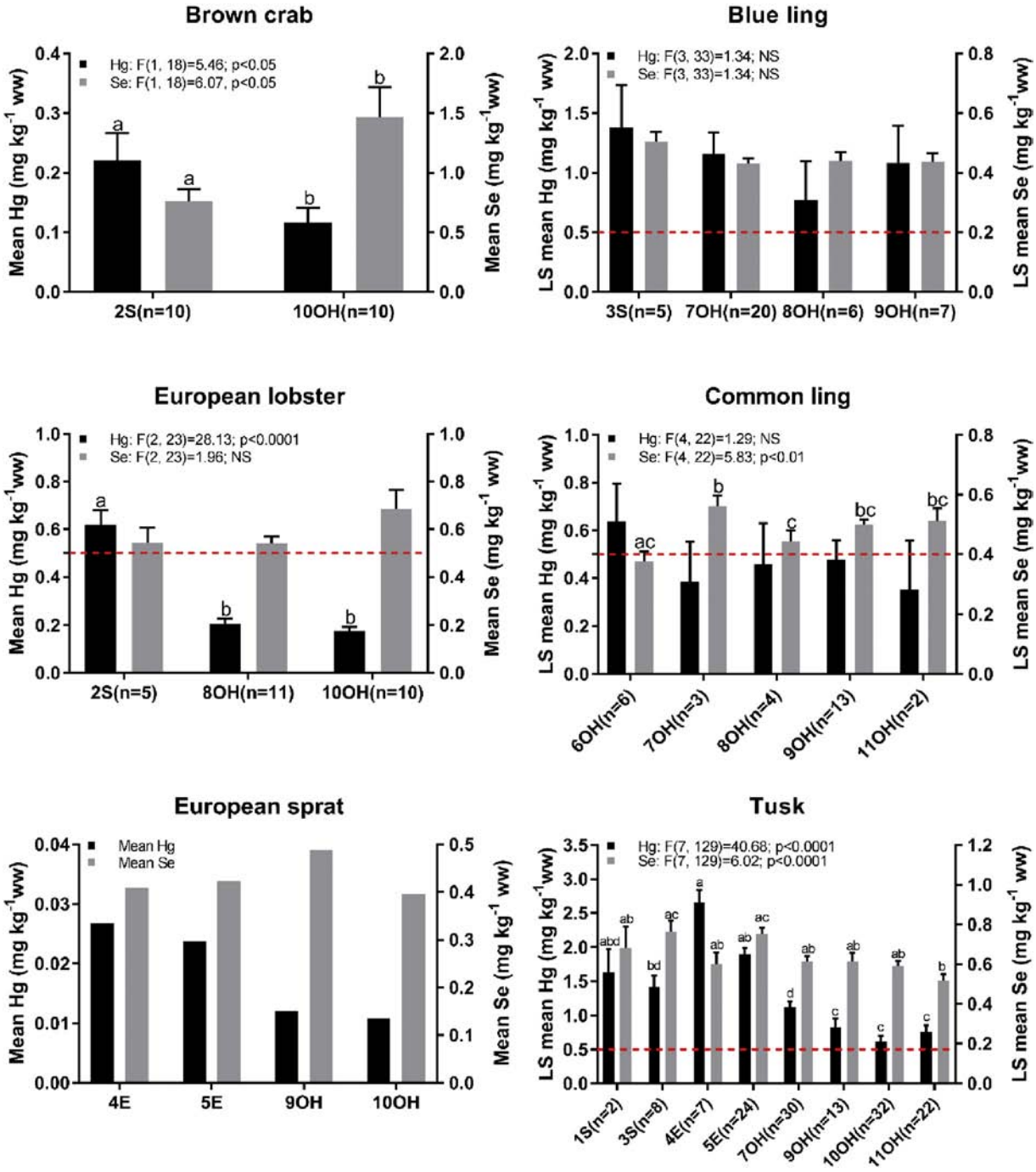


Fig. 4. Least squares means (adjusted for mean length) + standard error of Hg and Se concentrations in fish and crustacean species collected from different sites in Hardangerfjord, 2011. Hg and Se concentrations are presented on the left and right Y axes, respectively. ANCOVA/ANOVA test results are presented and letters were used to show significant differences when applicable. For lobster and brown crab ANOVA was used for comparisons between areas and arithmetic means are presented. Only composite samples of sprat and their arithmetic means are shown. Stations are sorted according to the distance from point source of pollution (PSP) at Odda. Letters after each station number represent the location in detail; S=Sørfjord, E = Eidfjord and OH=Outer Hardangerfjord. The dashed red lines show the EU maximum level of Hg (0.5 mg kg^{-1} ww).

were well correlated with total Hg concentrations (Kendall tau 0.71; $P < 0.05$), and we can conclude that Hg concentrations likely had an important influence on MeHg production in sediments in Hardangerfjord. In a study from Öre River estuary in Sweden, organic matter was shown to be a primary factor controlling MeHg formation in estuarine sediments, while total Hg had little or no effect on net MeHg production (Lambertsson and Nilsson, 2006). The main difference between the Hardangerfjord system and the Öre River Estuary studied by Lambertsson and Nilsson (2006) was the absence of local anthropogenic pollution in the Öre River Estuary, and consequently much lower concentrations of THg (ca. 18 times) in sediment samples than what we observed in inner Sørfjord. In another study, from the estuarine environment of the Penobscot River, Maine, USA, with high concentrations of Hg in sediment originating from industrial sources, a clear positive linear relationship was observed between Hg and MeHg concentrations (Rudd et al., 2018).

Distance from the open ocean was the best predictor for MeHg variation between the sites (Kendall tau 0.81; $P < 0.05$), while no correlation between MeHg and distance from PSP was detected since MeHg levels were relatively high in the inner sectors of both Sørfjord and Eidfjord (Fig. 1). Methylmercury concentrations in the sediments are

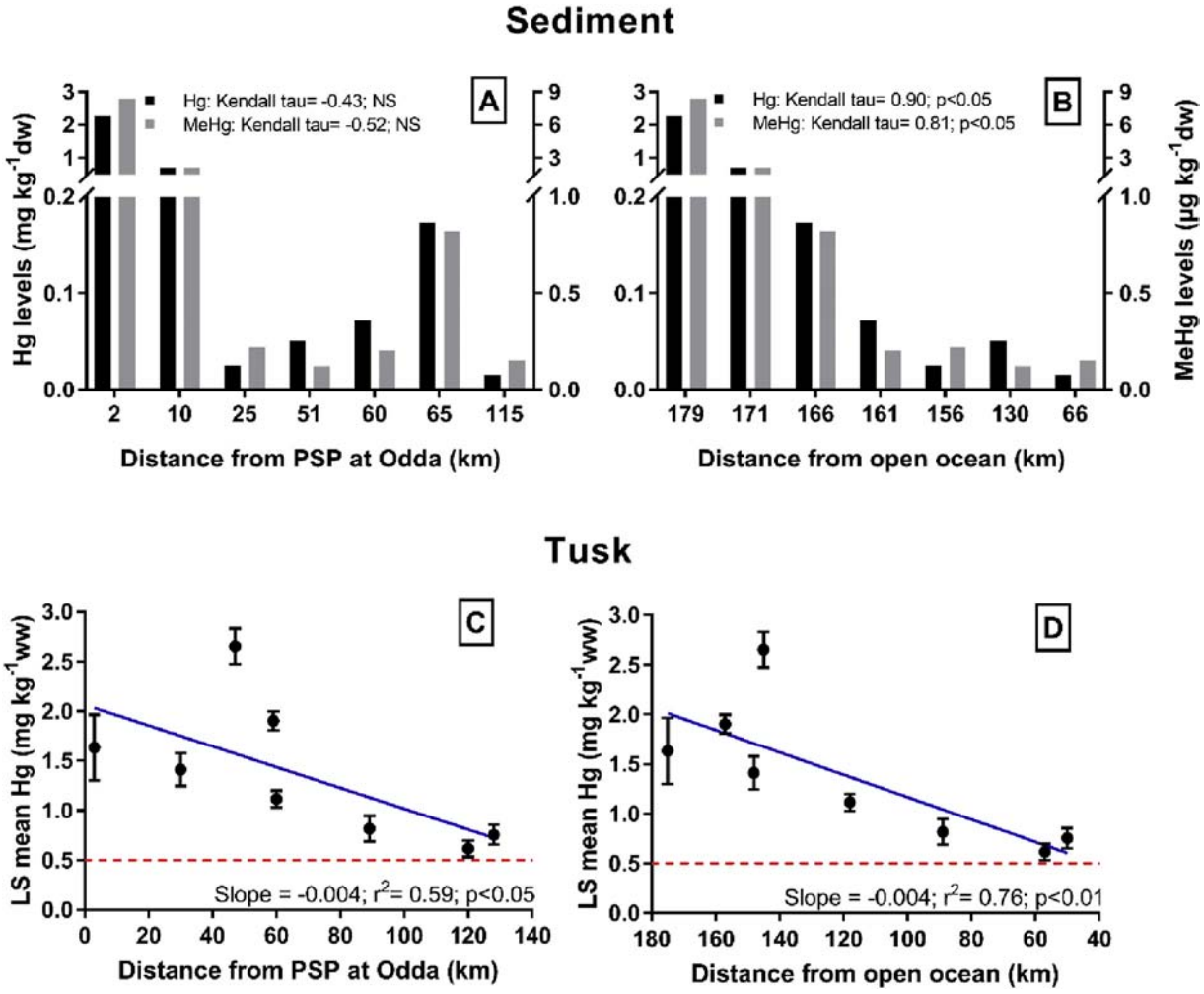


Fig. 5. Mercury pollution in sediment and tusk fillet sampled from different sites in the Hardangerfjord ecosystem. A and B: Total Hg and MeHg concentrations in sediment samples collected from different sites sorted by distance from point source of pollution (PSP) at Odda and distance from the open ocean. Nonparametric Kendall tau correlation coefficients are presented. NS = not significant. C and D: Least squares means Hg ± standard error of tusk fillet (adjusted for mean length) collected from different sites with varying distance from the point source of pollution (PSP) at Odda and distance from the open ocean. Dashed red lines show the EU maximum level of Hg (0.5 mg kg^{-1} ww).

likely governed by Hg concentrations, anaerobic microbial activity mainly driven by sulfate reducing bacteria in the inner sector of the fjord, and/or by organic matter quantity and composition.

3.5. Mercury speciation in seawater

Hg species and physiochemical parameters were measured in seawater samples taken from nine sites and three depths including 15, 50, and 300 m (Table 3). Salinity and temperature measurements showed that the three sampling depths belong to different hydrographic layers. Brackish layers were restricted to the upper 7 m of the fjord at the time of measurement. Water samples taken at 15 and 50 m depths were both within the intermediate layer while samples from 300 m depths were under the sill level in the fjord basin water. Total Hg concentrations increased with depth (mean of all sites 0.25 ng L^{-1} at 15 m; 0.43 ng L^{-1} at 50 m and 0.52 ng L^{-1} at 300 m; Fig. S4), whereas the MeHg concentrations were highest at 50 m and lowest at 15 m depth (0.02 ng L^{-1} at 15 m; 0.09 ng L^{-1} at 50 m and 0.04 ng L^{-1} at 300 m; Table 3; Fig. S4). The lower total Hg concentrations observed in the shallower layers may be related to the physical properties involved with water residence time in fjord ecosystems. Internal waves generated by wind conditions creating up- and down-welling at the coast are an important forcing mechanism for the renewal of the fjord water above the sill (Asplin et al., 1999). These internal waves are shown to occur irregularly 1 to 2 times a month and are restricted to the upper 30 m in May and June in the Hardangerfjord ecosystem (Asplin et al., 2014). Therefore, the water at 15 m depth will be exchanged more frequently than the water at 50 m depth, despite both depths being intermediate layers. The lower concentration of Hg found at 15 m depth compared to 50 m depth can be explained as a mixed effect of both different water residence times and that the deeper layers receive Hg deposited from the upper layers. The overall highest concentration of Hg was found at the 300 m depth level in sites 4 and 1 (1.65 and 1.55 ng L^{-1}) and also at 50 m at site 9 (1.2 ng L^{-1}). MeHg concentrations at all depths were highest at site 1 close to the PSP (0.04, 0.25 and 0.11 ng L^{-1} at 15, 50 and 300 m depths, respectively).

MeHg concentration at 50 and 300 m depths in seawater, as well as total Hg and MeHg concentrations in sediment, increased gradually towards the PSP indicating a possible interaction between Hg pools in surface sediments and deep layers of seawater. At deep parts of the Hardangerfjord ecosystem, below the sill, water exchange and mixing are very limited. MeHg produced in sediments as well as biological production of MeHg under the mixed layer that sinks as particles to deeper water are probably the main sources of MeHg in deep-water environments (Blum et al., 2013). MeHg concentrations in seawater at 50 and 300 m depths increased from the outer fjord towards PSP and the inner part of Hardangerfjord (Kendall tau −0.94 and −0.93 respectively; Table S5). In the inner part, a higher effect of the PSP, anaerobic conditions (i.e. lower oxygen conditions at the fjord's interior) and

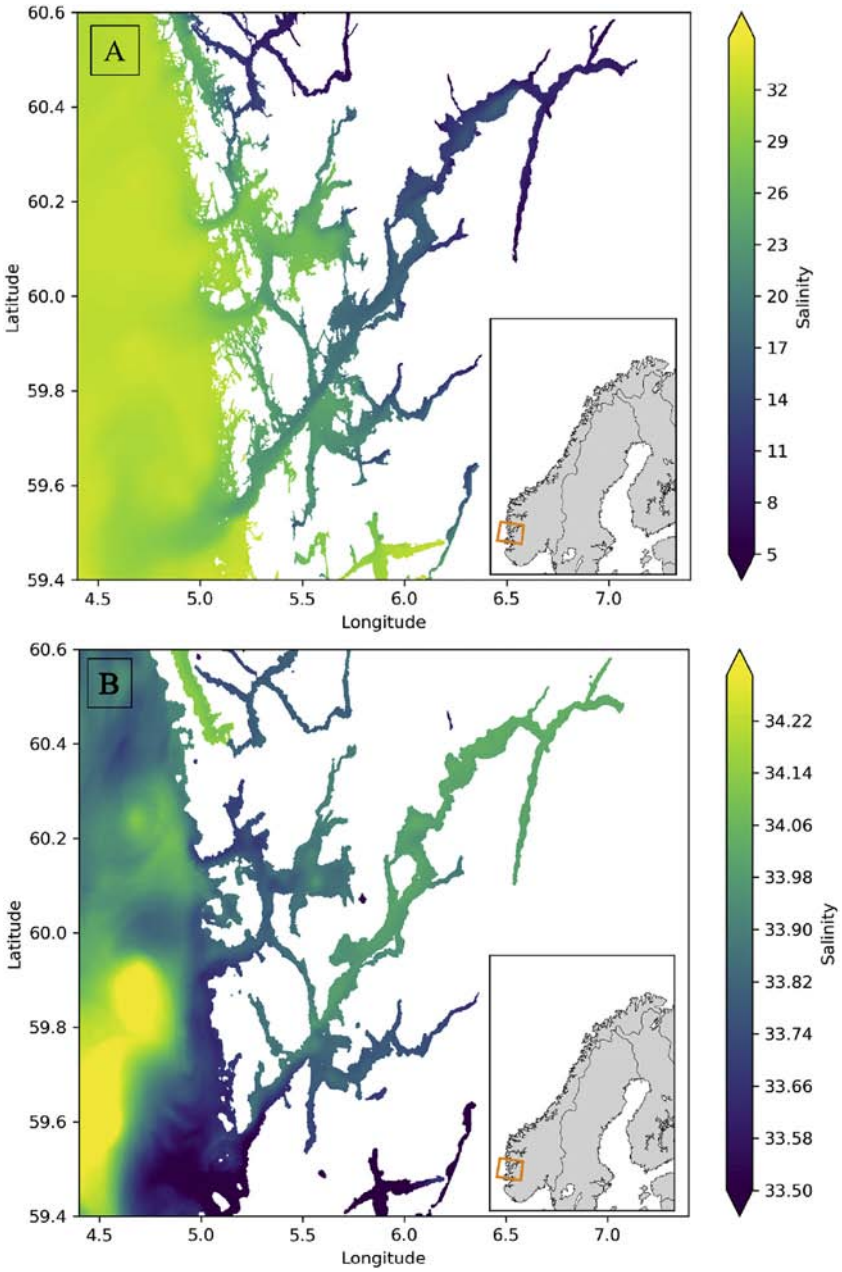


Fig. 6. Salinity level modeled for surface (A) and 50 m depth (B) in Hardangerfjord seawater sampled in May 2018. Note the different salinity scales in each map.

terrestrial run-off are expected. There was no such significant trend at 15 m depth (Table S5) where MeHg concentrations were generally low at all sites (up to 0.04 ng L^{-1}).

Percent MeHg increased significantly towards PSP for the 50 m depth samples but not at 15 m nor 300 m (Table S5). Oxygen concentrations at both 50 and 300 m depths decreased towards the inner part of the fjord, in the opposite trend of MeHg concentration and percent MeHg at 50 m depths. Lower oxygen concentrations in deep layers are typical of fjords due to lower rates of water exchange inside fjord sills. A combination of low oxygen concentrations and higher organic matter bound Hg^{2+} within fjords likely provided ideal conditions for biotic methylation and higher MeHg concentrations (Soerensen et al., 2018).

3.6. Bioconcentration Factors and Biota Sediment Accumulation Factors

For each site in the Hardangerfjord, Bioconcentration Factor (BCF) and Biota Sediment Accumulation Factor (BSAF) were calculated for total Hg and MeHg in tusk fillet tissue (Fig. S5). These are indicators of how much THg and MeHg are transferred to tusk fillet from water and sediment, respectively. Tusk was chosen for this purpose as it is a benthic feeder and a deep-water fish species with low vagility (Cohen et al., 1990). Tusk samples were collected across a broad area and in both the inner and outer sectors of the Hardangerfjord ecosystem. BCF values varied from 6.2 to 7.0 for total Hg and from 7.0 to 7.5 for MeHg (Fig. S5). Tusk BSAF values for total Hg was 0.2 at site 1S, and between 1.4 and 1.8 for the other sites and BSAF values for MeHg varied between 2.6 and 4.2. Site 1S closest to PSP had lower BSAF than the other sites due to very high Hg concentration in the sediment close to the PSP that was not reflected in the tusk fillets. Both BCF and BSAF were higher for MeHg than total Hg at all sites due to lower MeHg concentration in seawater and sediment compared to total Hg and the more efficient trophic transfer and bioavailability of MeHg. Lower BCF values close to the PSP at sites 1S and 3S and much lower BSAF values for both MeHg and Hg at site 1S compared to other parts of Hardangerfjord (Fig. 5S) may indicate that Hg and MeHg originating from PSP is less bioavailable compared to the Hg pool in other parts of the Hardangerfjord ecosystem.

Table 3
Mercury speciation and physiochemical properties of seawater from the different sampling sites in Hardangerfjord, May 2018.

Site	Sampling depth (m)	Max depth (m)	MeHg (ng L^{-1})	SD	iHg (ng L^{-1})	SD	THg (ng L^{-1})	% MeHg	Temperature (°C)	Salinity CTD[a]	Salinity (Water sample)	Oxygen (%)	Oxygen (mg L^{-1})	Latitude	Longitude
1	15	380	0.04	0.0011	0.40	0.01	0.45	9.33	7.25	31.762		97.62	10.10	60°	6° 35,747
	50		0.25	0.0146	0.53	0.01	0.78	32.17	8.22	34.65		50.47	5.01	14,656	
	300		0.11	0.0102	1.45	0.05	1.55	6.98	7.65	34.963	34.95	49.08	4.93		
2	15	780	0.02	0.0011	0.24	0.01	0.25	7.17	7.00	31.656		96.48	10.05	60° 26,58	6° 34,33
	50		0.15	0.0059	0.25	0.01	0.39	37.18	8.28	34.713		56.54	5.61		
	300		0.07	0.0014	0.17	0.01	0.24	29.45	7.68	34.98	34.98	61.30	6.15		
3	15	800	0.01	0.0002	0.13	0.01	0.14	7.68	7.25	31.762		97.62	10.10	60° 23.40	6° 20,43
	50		0.13	0.0030	0.31	0.02	0.45	29.39	8.22	34.65		50.47	5.01		
	300		0.06	0.0003	0.11	0.01	0.17	33.01	7.71	35.012	34.99	61.15	6.13		
	770		0.10	0.0015	0.20	0.01	0.30	32.40	7424	35.058	35.05	53.27	5.37		
4	15	500	0.01	0.0001	0.13	0.00	0.15	9.46	7.04	31.630		99.43	10.35	60° 15.55	6° 11,43
	50		0.07	0.0010	0.21	0.02	0.28	25.31	8.39	34.721		61.23	6.06		
	300		0.03	0.0011	1.63	0.04	1.65	1.53	7.75	34.999	34.99	68.17	6.83		
5	15	650	0.01	0.0004	0.20	0.01	0.21	6.25	7.09	31.580		100.50	10.45	60° 09.12	6° 04,73
	50		0.06	0.0046	0.13	0.01	0.19	32.46	8.47	34.684		64.91	6.41		
	300		0.04	0.0013	0.22	0.01	0.26	16.54	7.72	34.996	34.99	67.76	6.79		
6	15	500	0.04	0.0003	0.20	0.01	0.24	16.63	7.01	31.568		98.46	10.26	60° 00.47	5° 56,15
	50		0.05	0.0016	0.16	0.01	0.21	23.82	8.53	34.693		67.93	6.70		
	300		0.02	0.0006	0.15	0.01	0.17	9.31	7.65	34.974	34.97	75.50	7.58		
7	15	500	0.04	0.0005	0.24	0.01	0.27	13.30	7.06	31.644		99.14	10.31	59° 55.07	5° 45,15
	50		0.04	0.0013	0.27	0.02	0.31	12.93	8.51	34.713		68.57	6.77		
	300		< LOD	< LOD	0.21	0.01	0.21	<LOD	7.50	34.951	34.95	79.60	8.02		
8	15	330	0.02	0.0005	0.21	0.01	0.23	7.53	7.97	32.802		95.16	9.62	59° 44.45	5° 30,38
	50		0.02	0.0003	0.09	0.01	0.10	14.69	7.86	34.726		77.32	7.74		
	300		0.01	0.0001	0.15	0.01	0.16	5.23	7.22	35.079	35.08	81.81	8.29		
9	15	330	0.01	0.0002	0.25	0.01	0.26	4.21	8.74	31.792		99.73	9.98	59° 35.72	5° 15,72
	50		0.04	0.0003	1.16	0.02	1.20	3.26	7.69	34.619		79.00	7.94		
	300		0.01	0.0002	0.28	0.01	0.29	2.44	7.24	34.9	34.91	79.81	8.09		

LOD: limit of detection.
[a] An offset of 0.12 was added.

3.7. Does point source of pollution drive the spatial distribution of Hg in Hardangerfjord?

To investigate the effect of the point source, length adjusted Hg concentrations in tusk from different sites were analyzed as a function of distance from the PSP at Odda and the distance from the open ocean. The distance from PSP explained 59% of the variation of mean Hg concentrations in tusk fillet, but distance from the open ocean improved the model to 76% variance explained (Fig. 5C and D). The same trend was observed when Hg concentrations in individual tusk were used (Fig. S6). An explanation for this may be that an increased distance from the open ocean not only increases the effect of the PSP, but also water residence time, freshwater run-off and terrestrial organic matter.

Hg concentrations in sediment were very high near the PSP at Odda and decreased sharply towards the outer parts of Sørfjord (Fig. 5), indicating that Hg pollution from PSP is likely quite local. Probably a limited amount of Hg is transported from PSP, and Hg found in the outer Hardangerfjord and Eidfjord may originate largely from terrestrial run-off (Rua-Ibarz et al., 2019). A study of local soils showed that Hg concentrations around the zinc plant are very high compared with background concentrations, but within a 10–20 km distance from the source, Hg concentrations are comparable to background conditions (Svendsen et al., 2007). Thus, air emissions and atmospheric deposition of Hg from this point source will likely remain mostly inside the catchment area of the Sørfjord, and the major effect of Hg emissions is concentrated at the head of the fjord near Odda as well as the southern half of Sørfjord. However, Hg can also be distributed long distances to outside catchment areas via atmospheric transport (Fitzgerald et al., 1998).

The average MeHg concentration in the sediment in Sørfjord closest to the PSP was approximately 10 times higher than sediments in the inner Eidfjord sector. Even so, the mean concentration of Hg in tusk fillet (length adjusted) in Sørfjord was approximately 30% lower than in tusk from Eidfjord. This suggests that MeHg from different sites may not be equally bioavailable in these branches or that other factors such as trophic position varies across sites. In a mesocosm experiment, using Hg isotope tracers in both inorganic and organic forms, Jonsson et al. (2014) showed that MeHg from terrestrial and atmospheric sources have higher bioavailability compared to MeHg formed in the sediment and that MeHg from terrestrial run-off has a significant effect on MeHg burdens in estuarine biota. These findings could explain the trend in our results, where MeHg produced in sediments from inorganic Hg originating from the PSP appear to be less bioavailable than MeHg in Eidfjord that likely mainly originates from terrestrial run-off and atmospheric deposition, although the effects from local hydropower stations may also be substantial. High Hg concentrations in tusk have been reported from inner Nordfjord (another fjord in western Norway) compared to open ocean habitats (Berg et al., 2000), but further investigations in a fjord ecosystem without a point source are required to fully evaluate this hypothesis. However, it seems likely that the fjord ecosystems favor high Hg accumulation in deep-water, demersal fish compared to pelagic species, with some exceptions. Moreover, life history characteristics, and spatial and temporal variation in trophic complexity in these ecosystems, must also be considered important drivers of Hg in seafood species inhabiting fjords and other coastal environments, especially considering that subtle differences in diet and food web position may lead to substantial differences in Hg bioaccumulation (Bank et al., 2007). The high freshwater input from large catchment areas are believed to deliver highly bioavailable terrestrial MeHg to the fjord. When run-off reaches the fjord, the increase in salinity increases the partitioning of contaminants bound to organic matter in the particulate phase and thus the contaminants sedimentation can be enhanced from suspended particulate matter entering the sediments (Turner and Millward, 2002). MeHg will be retained in the fjord due to limited exchange of bottom water and the presence of a shallow sill. Additionally, Wang et al. (2018) reported the importance of Hg methylation in subsurface water in predicting Hg in marine biota from the Arctic. Future research should evaluate the role of subsurface methylation in relation to Hg dynamics in fjord food webs.

Using a linear model, distance from the open ocean explained 76% of the variation in Hg concentrations in tusk fillets. The linear model can be used to estimate the range of Hg contamination in deep-water species such as tusk. Therefore, tusk can be considered an important bioindicator for Hg contamination in high trophic species inhabiting fjord ecosystems especially since they have a very wide distribution and low vagility.

3.8. Comparison of Hardangerfjord seafood with the EU maximum level

Hg concentrations in fish and fishery products, including claw and tail meat of crustaceans, are regulated by the EU in different categories and should be below the maximum level (EUML) of 0.5 mg kg^{-1} ww for all species investigated in this study (EC, 2006). Tusk and blue ling collected from all sites in both inner and outer Hardangerfjord had 2–3 times higher average Hg concentrations than the EUML and all individual measurements in samples collected from inner Hardangerfjord exceeded the EUML (Table 3). Mean Hg concentrations in common ling from the inner part of the fjord exceeded the EUML by ~2-fold, and mean Hg concentrations in samples from the outer part were close to EUML (0.49 mg kg^{-1} ww). Hg levels in wolffish and sprat were well below the EUML. The sampled crustacean species had average Hg concentrations below the EUML, except for European lobster caught in Sørfjord that had 0.62 mg Hg kg^{-1} ww and four out of five specimens had concentrations above EUML. The EUML regulates commercial fishery in this area, as it is illegal to sell food exceeding EUMLs. To protect local recreational fishers and their families from MeHg exposure, the Norwegian Food Safety Authority (NFSA) has issued a consumption advisory to avoid blue ling and tusk from the whole Hardangerfjord and common ling from Sørfjord. Further, pregnant and nursing women are advised by NFSA to avoid consumption of crab, European lobster and sentinel fish species from Sørfjord (www.miljostatus.no).

Recently, Selenium Health Benefit Value (HBV_{Se}), was suggested as a comprehensive human health index considering the Se co-exposure that potentially reduces bioavailability, exposure and toxicity of MeHg (Ralston et al., 2016). Negative HBV_{Se} values imply higher molar concentration of Hg than Se, and consumption of seafood with negative values may be more detrimental for human health than consumption of seafood with positive values. In this investigation only blue ling and tusk from inner Hardangerfjord had HBV_{Se} with negative values of −1.7 and −0.6, respectively.

In general, crustaceans had higher HBV_{Se} values than fish species (~3 times higher in the outer part of the fjord and ~9 times higher in the inner sectors) since they contain less Hg and more Se (Table 2). Although Hg and Se concentrations were correlated in both tusk and blue ling from the inner part of Hardangerfjord, tusk with higher Hg concentrations had higher HBV_{Se} values than blue ling. This may be due to differences in bioaccumulation mechanisms and toxicokinetics of Se and Hg across taxa which have important implications for seafood safety and overall food security.

4. Conclusions

Hardangerfjord is a Hg impacted fjord with a pollution source at the end of its inner sector and provides a unique opportunity to investigate Hg bioavailability in seafood species commonly consumed by humans. Although the direct release of jarosite containing contaminants from the zinc plant into Hardangerfjord was stopped in 1986, legacy Hg is still present in the environment and concentrations in seawater and sediment were highest close to this point source at the inner most part of Sørfjord (Fig. 1). Tusk, blue ling and common ling from the entire Hardangerfjord area and European lobster from the inner part of the Hardangerfjord are highly polluted by Hg and well above the EUML. Concentrations of Hg in both seafood, sediment, and seawater increased from the open ocean to the inner part of the fjord. Although sediment concentrations were ten times higher in the inner fjord branch with a PSP (Sørfjord) compared to an adjacent fjord branch that may have been influenced by freshwater inputs, Hg concentrations in the demersal fish species tusk sampled from each branch were similar. Although Hg originating from the point source was methylated in sediments and Hg contamination in both fish and crustacean species increased towards the PSP, atmospheric Hg transferred by run-off and hydroelectric power stations cannot be ruled out as important sources of Hg to biota.

The effects of the PSP, run-off and organic matter input from the catchment, anaerobic conditions, and residence time gradually increased in the same direction (towards inner parts) and therefore it is difficult to separate the effect of these different Hg pools on biota. Adding a study in another fjord with similar conditions, but without a pollution point source or conducting Hg stable isotope analysis on terrestrial and marine ecosystem compartments from Hardangerfjord will likely help to better understand the relationship between different sources of Hg, local biogeochemistry patterns and overall bioavailability, fate, and transport of MeHg.

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Appendix A. Supplementary data

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