RESEARCH ARTICLE



Toxic and essential elements in seafood from Mausund, Norway

Hilde Ervik¹ · Tor Erik Finne² · Bjørn Munro Jenssen³

Received: 23 August 2017 / Accepted: 10 December 2017 / Published online: 26 December 2017 © Springer-Verlag GmbH Germany, part of Springer Nature 2017

Abstract

In annual surveys conducted during the period 2012–2015, concentrations of the toxic or essential elements B, Se, Cd, Sn, Cs, Hg, Pb, Al, Cr, Mn, Fe, Ni, Cu, Zn, and As were analyzed in brown meat of edible crab (*Cancer pagurus*), and filets of cod (*Gadus morhua*) and halibut (*Hippoglossus hippoglossus*) in one of the most important commercial crab fishing areas in Norway, at Mausund in Frøya municipality in Sør-Trøndelag, Norway. Concentrations of the elements were analyzed in sediments in 2015. Several salmon farms are located in this area. Samples were extracted by HNO₃ and analyzed using inductively coupled plasma mass spectroscopy (ICP-MS). Crab, cod, and halibut were caught in a total of five locations. One of these locations was in the proximity of a salmon farm. In edible crabs, the mean ranks were significantly different between two locations only for Sn (p = 0.034). When all data were pooled, the mean ranks statistics showed significant difference between all years for the elements Se (p = < 0.001), Cs (p = 0.005), Mn (p = 0.002), Zn (p = 0.006), and As (p = 0.001) in edible crab. The study showed elevated levels of Cd in edible crabs in 2012, 2013, 2014, and 2015, with the highest levels in 2015. In cod, there were significant differences between locations for the elements B (p = 0.003), and Pb (p = 0.04), as well as between the years for the elements B, Sn, Cs, Hg, Cr, and As (p = < 0.01). The study showed elevated level of Pb in cod in 2013. Halibut showed no significant differences between years or locations for any elements. The Cd, Hg, and Pb values of the sediments in this study indicate that local geogenic sources cannot be considered a major cause of high values in local biota.

Keywords Pollution · Ecotoxicology · Bioaccumulation · Toxic elements · Essential elements · Monitoring · Seafood · Sediment

Introduction

Marine biological resources are becoming increasingly important to meet the demand for food for the increasing human population. Uptake and/or accumulation of toxic elements, such as mercury (Hg), lead (Pb), cadmium (Cd), arsenic

Responsible editor: Philippe Garrigues

Electronic supplementary material The online version of this article (https://doi.org/10.1007/s11356-017-1000-4) contains supplementary material, which is available to authorized users.

Hilde Ervik hilde.ervik@ntnu.no

- ¹ Faculty of Social and Educational Sciences, Norwegian University of Science and Technology, NO-7491 Trondheim, Norway
- ² Geochemistry and Hydrogeology, Geological Survey of Norway, P.O. Box 6315 Torgard, NO-7491 Trondheim, Norway
- ³ Department of Biology, Norwegian University of Science and Technology, NO-7491 Trondheim, Norway

(As), and aluminum (Al), and essential elements, such as copper (Cu), zinc (Zn), and iron (Fe), in seafood may, however, compromise human food safety (Pedersen et al. 2014). Factors that could affect concentrations of elements in the marine ecosystem, include season, climate variability, food web dynamics, local geochemical formations, and local human pollution and long-distance pollution, such as Cd compounds from production processes, and Zn, Cu, and Pb from combustion processes (coal and oil) (European Commission DG Environment 2000). In recent years, there has been particular focus on Cd, because of concerns about high concentrations of Cd reported in brown meat in edible crab (Cancer pagurus), and questions have been raised on the causes for these high concentrations of Cd (Falk 2014). There have also been concerns about high concentrations of Cu in sediments near salmon farming cages, where the local pollution can be caused by the use of Cu as an antifouling biocide (Guardiola et al. 2012).

Results from studies conducted in the vicinity of salmon (*Salmo salar*) farms on whether the salmon aquaculture are a source of toxic elements or increasing concentrations of, e.g., Cu, in local marine environments, are inconsistent, indicating that there is a knowledge gap in relation to contaminant

exposure relevant for human health. Results from studies in Canada have shown that salmon aquaculture can result in increased levels of several elements in the sediments (Chou et al. 2002, 2004). On the other hand, Bustnes et al. (2011) presented results showing that samples of liver from cod (*Gadus morhua*) and saithe (*Pollachius virens*) caught near Norwegian salmon farms, did not provide evidence that farm-associated feral fish had higher levels of elements compared to the controls that were caught at distances > 4000 m from these farms. Arechavala-Lopez et al. (2015) supported the findings of Bustnes et al. (2011) by documenting higher levels of Fe, As, Se, Zn, and B in the livers, and As, B, Li, Hg, and Sr in muscles of wild saithe (*Pollachius virens*) caught in distances > 2500 m from a salmon farm.

Several environmental studies have reported the presence of toxic elements in the tissue of crustaceans and have evaluated the risks of these concentrations in relation to human consumption. Cadmium concentrations in brown meat of edible crabs (Cancer pagurus) harvested along the Scottish coast and the English Channel (Barrento et al. 2009), in France (Noël et al. 2011), and in Norway (Falk 2014) have been reported to be above the concentrations set by the European Food Safety Authority as the current limit for white meat, which is 0.50 mg kg $^{-1}$ wet weight (w.w.). In brown meat and claw meat from edible crabs sampled at three fish farms located in the counties of Sogn og Fjordane, Møre og Romsdal, and Nord-Trøndelag in Norway, the average concentrations in brown meat were reported to be from 0.86 to 7.82 mg Cd/kg w.w., with the highest concentrations in Sogn og Fjordane and Nord-Trøndelag. The results showed significant variation between individuals within each of the fishfarms (Falk 2014). The authors argued that this indicated that Cd in the fish feed is not likely to be a significant source. However, it is also possible that this variation is due to that the individuals consumed different amounts of fish feed, or that local currents affected the sediment concentrations of Cd originating from the fish farm.

The aim of this study was to examine whether there were variations in the element concentrations of B, Se, Cd, Sn, Cs, Hg, Pb, Al, Cr, Mn, Fe, Ni, Cu, Zn, and As in edible crab, cod, and halibut *(Hippoglossus hippoglossus)* in the Mausund region between the years 2012–2015, and whether there were variations in element concentrations in crab, fish, and sediment between locations, of which one location was in the close proximation from a salmon fish farm.

The specific objectives of the study were (1) to compare the concentrations of toxic or essential elements in edible crab and halibut, caught at two different locations within the region, cod caught at five locations within the region, and sediment sampled at three locations within the region; (2) to investigate

if the concentrations of any of the elements in the location close to the fish farm differed from that at the other locations; and (3) to establish whether there were between year (2012–2015) variations in the concentrations of toxic or essential elements in edible crab, cod, and halibut in the region,

Materials and methods

During four consecutive years (2012–2015) four classes of students taking the course "Technology and Theory of Research 2" at Byåsen Upper Secondary School, Trondheim, conducted an annual research project (Marine Environmental Monitoring), designed to address the previously described aims. The samplings were conducted in the first half of September each year. The sampling region in the inshore Mausund area, covered approximately 50 km² (Table 1). Edible crab, cod, and halibut were caught by the students under supervision in a total of five locations (each covering approximately 1 km²). In addition, edible crabs and sediment were collected by teachers participating in a postgraduate course at the Norwegian University of Science (NTNU) in mid-October 2015.

Halibut and cod were caught using lines and cod traps, and crab using crab pots in the selected locations (Table 1). Cod, halibut, and crab were kept in sea water and brought alive to Mausund fieldstation, and euthanized at the field station, prior to sampling of the tissues. The students followed standardized procedures to register sampling positions, and weight and length of the individual animals.

One gram (g) of fish fillet, obtained by a cut from the tissue between the gat and the dorsal fin, was sampled from each fish of cod as described for salmon, by Johnsen et al. (2011) and by a cut from the stomach area in front of the gat from each fish of halibut. From edible crab, 1 g of brown meat (hepatopancreas or hard roe) from each of the individuals was sampled. The samples were immediately frozen in 25 mL polystyrene cups. The samples were analyzed using inductively coupled plasma mass spectrometry (ICP-MS) at the Norwegian University of Science and Technology (NTNU).

Sixty-six edible crabs, fifty cods, and nine halibuts were caught at Mausund during 2012–2015 and samples of sediments were collected at locations 1, 2, and 3 in 2015 (Table 1). Location 1 was close to a salmon farm and this issue will be discussed later.

At location 1, crab, cod, and halibut were caught each year during the period 2012–2015. At location 2, crab was caught in 2014 and 2015, and cod was caught in 2013 and 2014. At location 3, cod were caught in 2012. At location 4, cod were caught in 2013, while at location 5, cod and halibut were caught in 2014 (Table 1). To establish if there were between-year variations in the concentrations of toxic or essensial

 Table 1
 Locations 1–5 and sampled species in the survey area at Mausund

7411

| Location | Species | 2012 | 2013 | 2014 | 2015 |
|----------|---------|---------------------------------|----------------------------------|---------------------------------|---------------------------------|
| 1 | Crab | N 63° 52′ 16″ E 008° 41′ 34″ | N 63° 52′ 11″ E 008° 40′ 59″ | N 63° 51′ 53″ E 008° 41′ 04″ | N 63° 52′ 08″ E 008° 40′ 34″ |
| 1 | Cod | N 63° 51′ 56″ E 008° 41′ 02″ | N 63° 52′ 16″ E 008° 41′04″ | N 63° 51′ 31″ E 008° 40′ 43″ | N 63° 52′ 12″ E 008° 41′ 19″ |
| 1 | Halibut | N 63° 52′ 34″ E 008° 41′03″ | N 63° 52′30″ E 008° 41′ 17″ | N 63° 52′ 06″ E 008° 41′ 47″ | N 63° 52′ 21″ E 008° 40′ 25″ |
| 2 | Crab | | | N 63° 50′ 38″ E 008° 36′ 08″ | N 63° 50′ 20″ E 008° 35′ 35″ |
| 2 | Cod | | N 63° 50′ 26″ E 008° 36′ 46″ | N 63° 50′ 36″ E 008° 36′ 58″ | |
| 3 | Cod | N 63° 53′ 07″ E 008° 39′ 18″ | | | |
| 4 | Cod | | N 63° 51′ 51″ E 008° 38′, 30″ | | |
| 5 | Cod | | | N 63° 52′ 04″ E 008° 34′ 03″ | |
| 5 | Halibut | | | N 63° 52′ 06″ E 008° 34′ 08″ | |

elements in the animals in the region (Fig. 1), the results from all the five locations were pooled.

Sediments

Samples of seabed sediments were collected in 2015 in locations 1–3 (Table 1), using a small (0.1 m^2) van Veen grab. After retrieving the grab and emptying the contents carefully into a large polyethylene box, the sediments were inspected for the integrity of the surface material and the stratigraphy of the sediment. One of the sample materials, location 3, was sufficiently cohesive to be sliced into 1-cm-thick slices after inserting a transparent polyethylene tube with an internal diameter of 110 mm and cutting a cylinder through the entire thickness of the sediment sample. By pushing a piston upwards against the sample, slices of 1-cm thickness were cut using polyethylene skewers as the sediment cylinder emerged at the top of the tube. Each of the slices was put in separate zip-lock polyethylene bags, tagged, and stored at -18 °C. The samples from the two other locations were treated similarly, except for the slicing, since the samples from the other stations were bulk samples of the uppermost 6-7 cm of the sediments.

Following freeze drying of the eight samples, a slice of each was taken for Ultra Clave digestion of 300 mg using 50% HNO₃ (50/50 HNO₃/water), and the solution was analyzed using ICP-MS for 59 elements, including the 15 elements B, Se, Cd, Sn, Cs, Hg, Pb, Al, Cr, Mn, Fe, Ni, Cu, Zn, and As. The principle of analysis used by inductively coupled plasma mass spectrometry (ICP-MS), element 2, as described by Bustnes et al. (2011). The concentrations of the elements are presented on as microgram per gram dry weight (d.w.).

Statistical treatment

The data were skewed and not normally distributed (IBM SPSS Statistics, version 21). Thus, differences in element concentrations were tested between the locations for all 15 elements using the non-parametric analysis of variance (Kruskal-Wallis test). For crab, cod, and halibut differences were tested using IBM SPSS Statistics, version 21, whereas for the sediment differences were tested using STATISTICA 6.0. For the between-year comparison of element concentrations in the animals, the results from all locations were pooled for every year, and ANOVA Kruskal-Wallis non-parametric test was used to test for between-year differences. Spearman's rank-order correlation was used to test if there were correlation between variables.

Results

Crab

Crabs were collected at locations 1 and 2 in Mausund, and the concentrations of the analyzed elements in edible crabs caught at these two locations during 2012–2015, is presented in Table 2 and Supporting Information (SI 1). The results from the pooled material, from all years, are listed in Table 3.

The results showed that only the concentrations of Sn differed among the two locations 1 and 2, where location 1 was close to a salmon farm. As shown in Table 3, there were significant differences in the concentrations of As, Cs, Mn, Se, and Zn in the pooled samples of the crabs between the years. Further tests showed that the As, Se, and Mn



Fig. 1 Mausund (63° N, 008° E), Frøya municipality in Sør-Trøndelag, Norway

concentrations were significantly higher in 2014 and 2015 than in 2012 and 2013, and that the Cs and Zn concentrations were significantly higher in 2014 and 2015 than in 2012 (Table 3). Thus, this indicates that there may have been a temporal increase in the concentrations of As, Cs, Mn, Se, and Zn during the 4-year period.

Cod

The concentrations of the analyzed elements in cod caught at location1, location 2, location 3, location 4, and location 5 in Mausund, 2012–2015, are presented in SI 2 and SI 3. The results on the pooled material from all years, are listed in Table 4.

The results showed that only the concentrations of B and Pb differed among the five locations (SI 3). The concentrations of B and Pb in the location close to the fish farm were not elevated as compared some of the other locations (SI 3). As shown in Table 4, there were significant differences for the concentrations of As, B, Cr, Cs, Hg, and Sn in the pooled samples between the years. Further tests showed that the As concentrations were significantly higher in 2014 than in 2015 and significantly higher in 2015 and 2014 than in 2012 and 2013; that the B concentrations were significantly higher in 2013, 2014, and 2015 than in 2012; and that the Cr concentrations were significantly higher in 2015 than in 2012, 2013, and 2014, and significantly lower in 2014 than in 2013 and 2012. The Cs concentrations were significantly higher in 2013, 2014, and 2015 than in 2012 and higher in 2013 than in 2014, whereas the Hg concentrations were significantly higher in 2013, 2014, and 2015 than in 2012, and the Sn concentrations were significantly lower in 2015 than in 2013 and 2014 (Table 4). Correlation analysis showed that there were no temporal trends in the element concentrations from 2012 to 2015.

Halibut

A total of nine samples of halibut, from location 1 and location 5 (Table 1), were analyzed during the 4-year period. The concentrations of the elements B, Se, Cd, Sn, Cs, Hg, Pb, Al, Cr, Mn, Fe, Ni, Cu, Zn, and As in fish filets of halibut are presented in Table 5. The results showed that the mean ranks were not statistically different between the two locations for the elements B, Se, Cd, Sn, Cs, Hg, Pb, Al, Cr, Mn, Fe, Ni, Cu, Zn, and As (p < 0.05).

Sediments

A total of eight samples of sediment, from location 1, location 2, and location 3, were sampled in 2015.

The mean ranks were not statistically different between these three locations for any of the elements B, Se, Cd, Sn, Cs, Hg, Pb, Al, Cr, Mn, Fe, Ni, Cu, Zn, and As (Table 6).

Discussion

In countries where much seafood, such as edible crab and fish, is consumed, and where there is high salmon farming activity,

Table 2Analyze of the elements B, Se, Cd, Sn, Cs, Hg, Pb, Al, Cr, Mn,Fe, Ni, Cu, Zn, and As in brown meat in edible crab (*Cancer pagurus*)Kruskal-Wallis H test between two locations; χ^2 and p values of means

caught at location 1 and location 2 in Mausund, 2012–2015. Median and mean concentrations ($\mu g g^{-1} d. w.$) and standard deviation (SD). Results of

| Elements | | Al | As | В | Cd | Cr | Cs | Cu | Fe | Hg | Mn | Ni | Pb | Se | Sn | Zn |
|---------------------|----------------------------------|-------------------------------|-------------------------------|----------------------------|------------------------------|-------------------------------|-------------------------------|-------------------------------|-------------------------------|-------------------------------|----------------------------|------------------------------|-------------------------------|----------------------------|-------------------------------|-------------------------------|
| Location 1 | Median Mean SD <i>n</i> | 1.13 1.73 2.76 43 | 42.26 43.94 23.46 43 | 2.00 2.74 2.35 43 | 1.68 3.38 4.80 43 | 0.099 0.192 0.276 43 | 0.011 0.012 0.005 43 | 18.01 32.39 36.04 43 | 84.48 99.47 76.65 43 | 0.079 0.094 0.058 43 | 5.22 5.76 3.31 43 | 0.292 0.714 1.02 43 | 0.044 0.052 0.034 43 | 4.29 4.41 2.59 43 | 0.003 0.004 0.003 43 | 57.80 98.40 90.15 43 |
| Location 2 | Median Mean SD <i>n</i> | 0.810 0.816 0.739 23 | 42.20 44.74 16.27 23 | 3.30 3.19 2.27 23 | 2.29 23.19 64.18 23 | 0.040 0.161 0.202 23 | 0.012 0.013 0.005 23 | 19.88 44.45 63.75 23 | 93.50 89.54 54.32 23 | 0.033 0.157 0.162 23 | 6.49 6.23 1.64 23 | 0.192 0.934 1.84 23 | 0.044 0.549 1.07 23 | 4.39 5.60 5.73 23 | 0.002 0.002 0.002 23 | 92.40 109.8 64.97 23 |
| Between 2 locations | χ^2 p | | | | | | | | | | | | | | 4.48 0.03 | |

studies has been conducted to monitor the environment of these species to investigate if they are exposed to contaminants, including toxic elements, and accumulate these to levels exceeding regulatory guidelines. High concentrations of Cd have been documented in brown meat in edible crabs in Norway (Falk 2014) and high levels of some trace metals have been reported in liver in wild saithe in the vicinity of Hitra Island, Norway, which is relatively close to the study area Mausund (Arechavala-Lopez et al. 2015). The causes of these high concentrations have not been established.

In the present study, the toxic or essential elements, B, Se, Cd, Sn, Cs, Hg, Pb, Al, Cr, Mn, Fe, Ni, Cu, Zn, and As was analyzed in edible crab, cod, halibut, and sediment and the only element that differed between the location close to a fish farm (location 1) and the other locations was Sn, which was significantly higher in brown meat of the crabs from location 1 as compared to crabs from location 2. When considering that

the between-location differences were tested for 15 elements, it is possible that this is a false positive finding. Nevertheless, it is possible that Sn may originate from organotin compounds, such as tributyltin (TBT). TBT has been used as a biocide in anti-fouling paint in marine vessels and constructions, including in fish farm installations (Berge et al. 2006). However, in Norway a ban of the use of TBT in fish farm constructions was introduced in 1989 (Berge et al. 2006), and an international ban of the use of TBT on vessels were implemented in 2008 (Gippert 2009). Thus, since TBT has not been used in the fish farm industry since 1989, and we do not know the age or the history of the specific fish farm, it is not possible to conclude that the Sn originates from chemicals used in the fish farm industry. Furthermore, it should be noted that the concentrations of Sn were very low, and the actual concentration of Sn was only 0.001 μ g g⁻¹ d. w. higher in the crabs caught at the location close to the fish farm as compared

Table 3Median and mean concentrations ($\mu g g^{-1}$ dry weight), standarddeviation (SD), and minimum and maximum concentrations ($\mu g g^{-1}$ dryweight). of the elements B, Se, Cd, Sn, Cs, Hg, Pb, Al, Cr, Mn, Fe, Ni, Cu,

Zn, and As in brown meat in edible crab (*Cancer pagurus*). Results of Kruskal-Wallis *H* test; χ^2 and *p* values of means of all years pooled, and *p* values for any combination of annual means are given

| n = 66 | | Al | As | В | Cd | Cr | Cs | Cu | Fe | Hg | Mn | Ni | Pb | Se | Sn | Zn |
|-----------|----------|--------|---------|------|------|--------|---------|------|------|------|---------|------|------|---------|---------|--------|
| Median | | 0.90 | 42.2 | 2.11 | 2.03 | 0.09 | 0.011 | 18.6 | 88.6 | 0.09 | 6.00 | 0.26 | 0.05 | 4.30 | 0.003 | 62.4 |
| Mean | | 1.41 | 44.2 | 2.89 | 10.3 | 0.18 | 0.01 | 36.6 | 96 | 0.12 | 5.92 | 0.79 | 0.16 | 4.83 | 0.003 | 102 |
| SD | | 2.3 | 21.1 | 2.31 | 38.7 | 0.25 | 0.01 | 47.4 | 69.4 | 0.11 | 2.84 | 1.35 | 0.64 | 3.06 | < 0.001 | 81.9 |
| Min | | < 0.01 | 12.6 | 0.42 | 0.01 | < 0.01 | < 0.01 | 2.78 | 0.26 | 0.02 | 0.56 | 0.01 | 0.01 | 0.91 | < 0.001 | 20.9 |
| Max | | 16.6 | 105 | 12.3 | 306 | 1.00 | 0.03 | 274 | 317 | 0.76 | 16.9 | 7.38 | 5.17 | 18.8 | 0.02 | 540 |
| All years | χ^2 | | 17.1 | | | | 14.6 | | | | 17.5 | | | 20.1 | | 13.9 |
| | р | | 0.002 | | | | 0.01 | | | | 0.002 | | | < 0.001 | | 0.01 |
| 2012-2013 | | | | | | | | | | | | | | | | |
| 2012-2014 | | | 0.01 | | | 0.03 | < 0.001 | | | | < 0.001 | | | 0.01 | 0.02 | 0.01 |
| 2012-2015 | | | < 0.001 | | | | < 0.001 | | | | < 0.001 | | | < 0.001 | | 0.05 |
| 2013-2014 | | 0.02 | 0.03 | | 0.04 | 0.01 | | | | 0.05 | 0.01 | 0.02 | | 0.01 | | < 0.01 |
| 2013-2015 | | 0.01 | 0.04 | | | | 0.05 | | | | 0.04 | | | < 0.001 | | |
| 2014-2015 | | | | | | | | | | 0.02 | | | | | | 0.01 |

| | <i>n</i> = 50 | Al | As | В | Cd | Cr | Cs | Cu | Fe | Hg | Mn | Ni | Pb | Se | Sn | Zn |
|-----------|---------------|-------|--------|--------|---------|---------|--------|-------|-------|--------|-------|-------|-------|-------|--------|-------|
| Median | | 0.69 | 9.45 | 0.54 | 0.002 | 0.008 | 0.120 | 0.65 | 4.00 | 0.306 | 0.40 | 0.024 | 0.024 | 1.032 | 0.008 | 15.05 |
| Mean | | 5.58 | 14.7 | 0.666 | 0.122 | 0.934 | 0.138 | 1.45 | 11.3 | 0.565 | 1.01 | 0.050 | 0.109 | 1.09 | 0.156 | 16.7 |
| SD | | 16.2 | 12.4 | 0.447 | 0.333 | 0.249 | 0.890 | 3.76 | 21.4 | 0.723 | 3.22 | 0.083 | 0.340 | 0.365 | 0.254 | 8.7 |
| Min | | 0.130 | 1.85 | 0.070 | < 0.001 | < 0.001 | 0.006 | 0.180 | 1.380 | 0.034 | 0.080 | 0.005 | 0.003 | 0.200 | 0.001 | 1.50 |
| Max | | 74.9 | 51.5 | 2.50 | 0.200 | 1.65 | 0.370 | 25.8 | 109.8 | 3.15 | 23.0 | 0.538 | 2.19 | 2.46 | 0.160 | 52.5 |
| All years | χ^2 | | 16.2 | 17.7 | | 28.7 | 20.2 | | | 14.2 | | | | | 9.5 | |
| | р | | < 0.01 | < 0.01 | | < 0.01 | < 0.01 | | | < 0.01 | | | | | < 0.01 | |
| 2012-2013 | | | | < 0.01 | | | < 0.01 | | | < 0.01 | | | | | | |
| 2012-2014 | | | 0.01 | < 0.01 | | < 0.01 | < 0.01 | | | 0.01 | | | | | | |
| 2012-2015 | | | 0.03 | < 0.01 | | < 0.01 | < 0.01 | | | < 0.01 | | | | | | |
| 2013-2014 | | | 0.02 | | | < 0.01 | 0.01 | | | | | | | | | |
| 2013-2015 | | | 0.02 | | | 0.01 | | | | | | | | | 0.01 | |
| 2014-2015 | | | 0.01 | | | < 0.01 | | | | | | | | | 0.02 | |
| | | | | | | | | | | | | | | | | |

Table 4 Concentrations ($\mu g g^{-1}$ dry weight) of B, Se, Cd, Sn, Cs, Hg, Pb, Al, Cr, Mn, Fe, Ni, Cu, Zn, and As in fish filets of cod (*Gadus morhua*). Results of Kruskal-Wallis *H* test; χ^2 and *p* values of means for all years pooled, and *p* values for any combination of annual means

to the other location. Since there were no differences among the other elements in the crabs, nor in cod, halibut or in the sediments, it is concluded that the proximity to the fish farm most likely did not affected the concentrations of the investigated elements in the investigated marine species or in the sediments.

In the region Mausund, there were between year (2012–2015) variations in some concentrations of toxic or essential elements in edible crab and cod, but no variation in halibut. There were significant differences between the years for the elements Se, Cs, Mn, Zn, and As in brown meat in crab and for the elements B, Sn, Cs, Hg, Cr, and As in cod and there may have been a temporal increase in the concentrations of As, Cs, Mn, Se, and Zn in the brown meat in crab during the 4-year period between years 2012–2015.

Crab

The results from the present study showed a significant difference between location 1 and location 2 (Table 2) for the element Sn. In crabs, there were significant differences between 2012 and 2015 in the pooled samples from these locations for the elements As, Cs, Se, and Mn (Table 3). There were high concentrations of the elements As, Cd, Cu, Fe, and Zn in some

individuals, with an extremely high Cd concentration in brown meat at location 2 (Table 3).

Due to the health risks associated with high concentrations (Norwegian Scientific Committee for Food Safety 2015), special attention devoted to the element Cd and to explain the high levels of Cd found in brown meat in some crab individuals (Falk 2014). Juhlsham et al. (2012) conducted Norway's first (near) nationwide survey of heavy metal concentrations in edible crab. The study was spurred by the findings in 2010 and 2011 of widespread occurrence in claw meat of levels of Cd that exceeded the EU limit of 0.5 μ g g⁻¹ w.w. (Falk 2012; Finne 2013). In that particular study, ten crabs were caught and analyzed individually from each of 47 locations along the coast. Within one area, the crabs in the majority of investigated locations had higher levels of Cd than the EU limit, namely Salten, a district in Nordland county in Norway (Juhlsham et al. 2012). The variation of the Cd concentration within locations in the high-Cd region in Salten, was remarkably stable, indicating that the Cd-input to the region was most likely waterborne (dissolved or by microorganisms); whereas it could be argued that greater variability between individuals from the same location would indicate a source of more local character (Juhlsham et al. 2012). Since the results in the present study are given on a dry weight basis, it is not possible to assess whether or not the

Table 5Concentrations ($\mu g g^{-1} d.w.$) of elements in halibut (*Hippoglossus hippoglossus*); samples from location 1 (2012–2015) and location 5, 2014,in Mausund, Norway

| <i>n</i> = 9 | Al | As | В | Cd | Cr | Cs | Cu | Fe | Hg | Mn | Ni | Pb | Se | Sn | Zn |
|--------------|-------|------|-------|-------|-------|-------|-------|------|-------|-------|-------|-------|-------|-------|------|
| Mean | 0.994 | 11.3 | 0.27 | 0.012 | 0.011 | 0.086 | 0.911 | 8.16 | 0.270 | 0.471 | 0.013 | 0.011 | 1.65 | 0.008 | 13.8 |
| SD | 1.26 | 10.9 | 0.79 | 0.033 | 0.014 | 0.046 | 1.27 | 12.1 | 0.169 | 0.321 | 0.007 | 0.008 | 0.68 | 0.008 | 6.3 |
| Min | 0.130 | 3.69 | 0.110 | 0.000 | 0.000 | 0.013 | 0.340 | 1.66 | 0.038 | 0.190 | 0.003 | 0.001 | 0.400 | 0.002 | 4.60 |
| Max | 3.71 | 37.9 | 0.390 | 0.100 | 0.036 | 0.153 | 4.29 | 39.6 | 0.601 | 1.14 | 0.025 | 0.028 | 2.59 | 0.029 | 27.9 |

Table 6 Concentrations ($\mu g g^{-1} d. w.$) of the elements B, Se, Cd, Sn, Cs, Hg, Pb, Al, Cr, Mn, Fe, Ni, Cu, Zn, and as in sediments in sediments; samples collected in the survey area in 2015, Mausund, Norway

| n = 8 | Al | As | В | Cd | Cr | Cs | Cu | Fe | Hg | Mn | Ni | Pb | Se | Sn | Zn |
|-------|-------|------|------|-------|------|-------|------|-------|-------|------|------|------|------|-------|------|
| Mean | 1755 | 1.29 | 34 | 0.138 | 4.56 | 0.141 | 2.68 | 1484 | 0.013 | 46.6 | 3.08 | 3.77 | 0.39 | 0.226 | 11.9 |
| SD | 197.3 | 0.24 | 6.04 | 0.034 | 0.49 | 0.026 | 0.40 | 194.8 | 0.003 | 14.1 | 0.54 | 0.75 | 0.11 | 0.085 | 1.41 |
| Min | 1557 | 0.92 | 24.8 | 0.092 | 3.87 | 0.113 | 2.07 | 1239 | 0.008 | 32 | 2.21 | 2.91 | 0.23 | 0.164 | 10.3 |
| Max | 2073 | 1.68 | 43.9 | 0.191 | 5.18 | 0.180 | 3.33 | 1769 | 0.018 | 75 | 3.82 | 5.30 | 0.54 | 0.371 | 13.5 |

concentrations of Cd in the brown meat exceeded the EU limit of 0.5 μ g g⁻¹ w.w. Cd in claw meat of edible crabs.

The results from the present study show relatively large variations in the element concentrations between individuals from the same geographic region (i.e., Mausund). This may be related to the nomadic behavior of edible crabs, Bennett and Brown (1983) found that inshore, 90% of males and 76% of females were recaptured within 18 km. Thus, it is possible the uptake and accumulation of the elements in the crabs does not represent background levels in their diet at the locations where they were caught. It is possible that the knowledge of the local fishermen on the seasonal nomadic movement and pattern of the crabs may assist in understanding the variations of the element concentrations among the individual crabs in the region (Rosa et al. 2014).

Although it is possible that the cause of the relatively high concentrations of the elements As, Cd, Cu, Fe, and Zn (Table 3) could originate from long-distance transport of pollutants (European Commission DG Environment 2000), natural sources, or local pollution, could also be the origin of these relatively high concentrations in the crabs.

The Norwegian Food Safety Authority has not set a limit for As, Cu, Fe, Zn, or Cd in brown meat, and consumers are recommended to follow the consumption advice given by the Norwegian Scientific Committee for Food Safety (2015), related to Cd exposure from crabs. This recommendation is that adults can eat approximately one whole crab or two filled crab shells per month. Based on the high concentrations of Cd reported in brown meat of several individual edible crabs caught at location 2, we recommend that further investigations on Cd concentrations in sediments and biota should be conducted in a specific region close to location 2 that have depth of 100 m. It is possible that crabs may have seasonal migrations to this deep-water region and thus being exposed to high Cd levels in the prey in this particular location.

Since the crab fisheries in the region, according to the local fishermen, appear to be stable and thus most likely sustainable, the concentrations of the elements in brown meat in edible crabs in the present study, most reflect normal physiological concentrations for the essential elements Cu, Fe, and Zn.

Cod

In cod, there were significant differences between 2012 and 2015 for the elements As, B, Cr, Cs, Hg, and Sn and there were significant differences between locations for the elements B and Pb (Table 4).

The Norwegian Food Safety Authority has set a limit for Hg, Cd, and Pb in fish-fillet. The limit for Hg is 0.5 mg/kg (w.w.), the limit for Cd is 0.05 mg/kg (w.w), and the limit for Pb is 0.3 mg/kg (w.w). However, since the results in the present study are given on a dry weight basis, it is not possible to assess whether or not the concentrations of Cd and Pb in the cod filets exceeded these limits. The concentrations of some other elements in the present study (Table 5) may reflect the variation in normal physiology for the essential elements Cu, Fe, and Zn.

Halibut

There were no significant differences between location 1 and location 5, or between years, for toxic or essential elements in halibut. The Norwegian Food Safety Authority has set a limit for Hg, Cd, and Pb in fish-fillet. The limit for Hg is 0.5 mg/kg (w.w.), the limit for Cd is 0.05 mg/kg (w.w), and the limit for Pb is 0.3 mg/kg (w.w).

Sediments

In 2008 and 2014, NGU sampled sediments from 222 points within an area of 429 km², including the approximately 50 km² studied in the present study. The bedrock in Frøya municipality has been mapped as granite or granodioritic gneiss, rock types that mainly consist of quartz and feldspar minerals containing 12–30 μ g g⁻¹ Pb (Nordgulen et al. 1990). Chemical analysis of five rock cores by XRF yielded values for the total content of Pb similar to those reported by Nordgulen et al. (1990), whereas analysis of HNO₃-extracts of the cores showed concentrations of Pb between < 5 and 7 μ g g⁻¹(Slagstad 2014). This indicates that most of the lead is located in the silicate lattice of the feldspar minerals, and that it is not bioavailable. The Cd concentration in the same rock cores was below 1 μ g g⁻¹. Neither Nordgulen et al. (1990) nor Slagstad (2014) reported concentrations of Hg in

the sediments. Surficial deposits are scarce and often of high organic content. The thickness of sediments on the seabed is not known in detail, but the grain size of the sediments is generally quite coarse, mostly from sand and up. Comparing the Cd, Hg, and Pb values of the sediments in the present study to those of the bedrock and of the oil gas sector Environmental Monitoring Database (MOD 2012), indicates that local geogenic sources cannot be considered a major cause of high values of these elements in local biota.

Conclusion

Comparing the concentrations of toxic or essential elements in edible crab and halibut, caught at two different locations within the region, cod caught at five locations within the region and sediment sampled at three locations within the region, showed that except for the concentrations of Sn in brown meat in crab and B and Pb in cod, there appeared to be no difference between the location close to a fish farm and the other locations.

The study showed elevated levels of Cd in edible crabs in 2012, 2013, 2014, and 2015, with the highest levels in 2015, but there were relatively large variations in the Cd concentrations between individuals in the same geographic area. When all data were pooled, the mean ranks statistics showed significant difference between all years and there may also have been a temporal increase in the concentrations for the elements Se, Cs, Mn, Zn, and As in brown meat in edible crab. There should be a special attention to the toxic element As. There is significant difference between years and high values. As, Cd, and Pb concentrations in the sediments in this study indicate that local geogenic sources cannot be considered a major cause of high concentrations in local biota. Other sources could be local human pollution or long-distance pollution following the ocean currents.

Acknowledgements Special thanks to Odd Arne Arnesen for the invaluable help with coordinating the fieldwork, to Arvid Støen for the help with catching crabs, cod, and halibut, and to both for explaining the conditions in the geographical area. Thanks also to Harald Aursøy for the use of the field station, and to Syverin Lierhagen for analyzing the material. We also thank the enthusiastic students at Byåsen Upper Secondary School in Trondheim and the students in the postgraduate participating in the course Technology and theory of research SKOLE6625 at the Norwegian University of Science and Technology (NTNU).

References

wild saithe Pollachius virens. Mar Coast Fish: Dyn Manage Ecosyst Sci 7(1):59-67

- Barrento S, Marques A, Teixeira B, Carvalho ML, Vas-Pires P, Nunes ML (2009) Accumulation of elements (S, As, Br, Sr, Cd, Hg, Pb) in two populations of Cancer pagurus: ecological implications to human consumption. Food Chem Toxicol 47(1):150-156. https://doi.org/ 10.1016/j.fct.2008.10.021
- Bennett DB, Brown CG (1983) Crab (Canser pagurus) migrations in the English Channel. J Mar Biol Assoc U K 63(2):371-398. https://doi. org/10.1017/S0025315400070740
- Berge, J., Amundsen, C.E., Eggen, T., Hylland, K. and Bøe E (2006). Naturlig nedbryting og biotilgjengelighet av tinnorganiske forbindelser i marine sedimenter. Degradation and bioavailability of organotin compounds in marine sediments. NIVA-rapport 4996. Norwegian Institute of Water Research, Oslo. (In Norwegian, English summary). http://hdl.handle.net/11250/212772
- Bustnes JO, Nygård T, Dempster T, Ciesielski T, Jenssen BM, Bjørn PA, Uglem I (2011) Do salmon farms increase the concentrations of mercury and other elements in wild fish? J Environ Monit 13(6): 1687-1694. https://doi.org/10.1039/c1em10083a
- Chou CL, Haya K, Paon LA, Burridge L, Moffatt JD (2002) Aquaculturerelated trace metals in sediments and lobsters and relevance to environmental monitoring program ratings for near-field effects. Mar Pollut Bull 44(11):1259-1268. https://doi.org/10.1016/S0025-326X(02)00219-9
- Chou CL, Haya K, Paon LA, Moffatt JD (2004) A regression model using sediment chemistry for the evaluation of marine environmental impacts associated with salmon aquaculture cage wastes. Mar Pollut Bull 49(5-6):465-472. https://doi.org/10.1016/j.marpolbul.2004.02. 039
- European Commission DG Environment (2000) Working group on arsenic, cadmium and nickel compounds. Ambient air pollution by AS, CD and NI compounds
- Falk AH (2012) Kartlegging av kadmium i sediment, Saltenområdet i Nordland. Akvaplan-niva Rapport 6063.01, 44 pp. (In Norwegian)
- Falk AH (2014) Akvaplan niva. Rapport: 6676-01. Kadmium rundt oppdrettsanlegg. Er fiskefôr en potensiell kilde til kadmium i marint miljø? Report, Akvaplan Niva
- Finne TE (2013) Kadmium i løsmasser, overflatevann og grunnvann fra Salten som lokalt naturlig bidrag til forhøyet Cd- nivå i taskekrabbe langs kysten. (In Norwegian). NGU report 2013.056
- Gippert L (2009) The legal design of the International and European Union ban on tributyltin antifouling paint: direct and indirect effects. J Environ Manag 90(Suppl. 1):S86-S95. https://doi.org/10.1016/j. jenvman.2008.08.013
- Guardiola FA, Cuesta A, Meseguer J, Esteban MA (2012) Risks of using antifouling biocides in aquaculture. Int J Mol Sci 13(2):1541-1560. https://doi.org/10.3390/ijms13021541
- Johnsen CA, Hagen Ø, Adler M, Jönsson E, Kling P, Bickerdike R, Solberg S, Björnsson BT, Bendiksen EÅ (2011) Effects of feed, feeding regime and growth rate on flesh quality, connective tissue and plasma hormones in farmed Atlantic salmon (Salmon salar L.) Aquaculture 318(3-4):343-354. https://doi.org/10.1016/j. aquaculture.2011.05.040
- Juhlsham K, Nilsen B, Valdersnes S, Frantzen S (2012) Årsrapport 2011. Mattilsynets program Frememdstoffer i villfiskmed vekt på kystnære farvann. Delrapport I: Undersøkelser av miljøgifter i taskekrabbe. (In Norwegian, English summary) NIFES, Bergen
- MOD (2012) MiljøOvervåkingsDatabasen (environMental mOnitoring Database), https://projects.dnvgl. com/MOD/Default.aspx?TOOL=HJEM. Accessed March 2012
- Noël L, Chafey C, Testu C, Pinte J, Velge P, Guérin T (2011) Contamination levels of lead, cadmium and mercury in imported and domestic lobsters and large crab species consumed in France. Differences between white and brown meat. J Food Compos Anal 24(3):368-375. https://doi.org/10.1016/j.jfca.2010.08.011
- Arechavala-Lopez P, Sæther B-S, Marhuenda-Egea F, Sanchez-Jerez P, Uglem I (2015) Assessing the influence of salmon farming through total lipids, fatty acids, and trace elements in the liver and muscle of

- Nordgulen Ø, Solli A, Bøe R, Sundvoll B (1990) Geologiske undersøkingar på Frøya og Froøyane. (In Norwegian), NGU report 90.073
- Norwegian Scientific Committee for Food Safety (2015) Risk assessment of dietary cadmium exposure in the Norwegian population. VKM Report 2015:12
- Pedersen KL, Bach LT, Bjerregaard P (2014) Amount and metal composition of midgut gland metallothionein in shore crabs (Carcinus maenas) after exposure to cadmium in the food. Aquat Toxicol 150:182–188. https://doi.org/10.1016/j.aquatox.2014.03.009
- Rosa R, Carvalho AR, Angelini R (2014) Integrating fisherman knowledge and scientific analysis to assess changes in fish diversity and food web structure. Ocean Coast Manag 102:258–268. https://doi. org/10.1016/j.ocecoaman.2014.10.004
- Slagstad T (2014) Personal communication on NGU's ongoing LITO project, aiming at sampling all of Norway's bedrock in a 9×9 km grid for determination of chemistry and other geoscientific parameters