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Elemental content of brown crab (*Cancer pagurus*) – Is it safe for human consumption? A recent case study from Mausund, Norway

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HIGHLIGHTS

- Brown crabs (N = 65) demonstrated high Cd concentrations in brown meat.
- The Cd intake dose from 100 gr of brown crab media (dry) reached 20.2 mg.
- Fe, Cu, Ni, Se, Ca, Mg and P can exceed the tolerable upper intake levels.
- The Se:Cd ratio was proposed as an essential criterion of risk from Cd exposure.
- Brown meat consumption must be addressed by regulatory bodies.

GRAPHICAL ABSTRACT

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ABSTRACT

The brown crab (*Cancer pagurus*; the edible crab) is consumed worldwide and greatly appreciated for the essential elements (e.g., Zn, Se) that it contains. However, alongside these, it contains toxic elements that can pose serious risks for human health. For the brown meat parts of the brown crab, which contain considerable Cd concentrations, official legal limits of exposure are still lacking by regulatory bodies, rendering its consumption a potential food safety threat. In this study, a survey was conducted during 2016–2018 in a major Mid-Norway commercial crab fishing area of Mausund in Frøya municipality, Norway, where brown crab media samples were collected to assess the occurrence and profile of select elements, including toxic elements (Cd, Pb, Hg, Sr, As). A yearly median concentration ranging from 6.75 (2016) to 14.0 (2017) mg Cd/kg dry weight (est.: 2.11 (2016) to 4.37 (2017) mg Cd/kg wet weight) indicated high Cd concentrations, which were alarming when compared to the maximum allowed concentration of 0.5 mg/kg wet weight set by the E.U. for raw white crab meat. Human exposures were assessed by estimating intakes of elements per 100 g serving portion of dry edible media (white/brown meat), and the tolerable upper intake levels (ULs) (as specified by the *United States National Academy of Sciences*) were exceeded for Fe, Cu, Ni, Se, Ca, Mg and P. The median Se and Cd estimated human dietary intakes (EDIs) (from brown crab) were 8.21–8.59 and 9.64–20.0 μg/kg body weight, respectively. The human intake dose (ID) of Cd (from brown crab) reached the order of a few tens of milligrams (mg). In addition, 33% of brown crab samples were found to be suitable for human consumption when the Se:Cd concentration ratio was applied and a stoichiometric excess of Se over Cd was indicated.

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1. Introduction

The human consumption of brown crab (*Cancer pagurus*; the edible crab) is well-established worldwide. In the E.U. (28 countries) alone, the catch of brown crab was estimated to be approximately 45.000 tons (t) in 2017 (EuroStat, 2019). In the same year, the European countries with the highest volume of catch were in decreasing order: The United Kingdom (33,800 t) > Ireland (6600 t) > Norway (4 900 t) > France (4 300 t) (EuroStat, 2019). These catches denote the abundancy of brown crab in the Norwegian and North Sea. In Norway, most of the crab catches are obtained from Mid-Norway (Søvik et al., 2017; Woll et al., 2006). White (muscle meat; mainly derived from claws and legs) and brown meat (mainly derived from hepatopancreas and gonads) are either consumed individually or in combination. In Norway, whole brown crabs are consumed regularly (Bergsten, 2004), while in other countries, such as Portugal, it is documented that the brown meat part of the crab is consumed the most (Maulvault et al., 2013).

Assessment of the risks and benefits of brown crab human consumption remains challenging and controversial. Brown crabs are established as an important source of essential elements (e.g., Se and Zn), but unfortunately, they are also a significant source of toxic elements (e.g., Cd and Hg) (Barrento et al., 2009a,b,c; Maulvault et al., 2011, 2012, 2013). Most marine animals, including the brown crab species, demonstrate variability in elemental concentrations, attributed mainly to differences among the marine environments (e.g., location, pollution pressures) to which they are exposed (Barrento et al., 2009c; Kannan et al., 1995; Tsangaris et al., 2016). Thus far, several norwegian and international reports (Barrento et al., 2009b; Jensen and Wasmuth, 2010; Julshamn et al., 2012a, b) raised concerns over the high concentrations of Cd found in brown crab meat. The Cd concentrations found in white meat (from brown crab) represent an issue for many countries worldwide, including Norway and the E.U., where legal limits are already in place (E.U., 2004, 2006, 2011; VKM, 2015). However, for the brown meat (from brown crab), which generally contains even higher Cd concentrations, official legal limits of exposure are still lacking, rendering its consumption a potential food safety concern. In 2015, the Norwegian Food Safety Authority performed a risk assessment on the risk of dietary Cd exposure in the norwegian population with a special focus on foodstuffs that contain high Cd concentrations, including brown meat-based products (VKM, 2015). The risk assessment concluded that the consumption of brown meat (from brown crab) products is indeed of high concern, in contrast to the consumption of white meat, which does not pose an actual human risk.

In the present study, brown crab, local wild-caught predatory fish and deep-sea sediment matrices were collected during 2016–2018 from the marine environment of Mausund, which is a major norwegian fishing and aquaculture location (island complex) surrounded by the Norwegian Sea and located off the coast of Mid-Norway (belonging to Frøya municipality, Trøndelag County) (Ervik et al., 2018), with the main aims of: (i) assessing potential recent elemental influences in brown crab from relevant marine sources (fish and sediments); (ii) investigating the occurrence of toxic elements in brown crab meat; and (iii) evaluating human exposures and risks from the consumption of brown crab media (dry). The elemental concentrations measured in local wild-caught predatory fish were compared to those in brown crab since both are natural co-habitants in the Mausund area.

2. Materials and methods

2.1. Chemicals and materials

Multielement standard working solutions that contained 70 elements were prepared from PS-70 stock solution (*Elemental Scientific, Omaha, NE, U.S.*). Concentrated nitric acid (UltraPure grade) was obtained by distillation with *Milestone SubPur (Sorisole, BG, Italy)*. Water was purified with a Milli-Q grade water purification system (*Q-option, Elga Labwater, Veolia Water Systems LTD, U. K.*). Two sets of calibration solutions (CS) were purchased from *Elemental Scientific* (*Omaha, NE, U.S.*) that were obtained from two independent producers.

2.2. Sample collection

2.2.1. Biological matrices

Three sampling campaigns were performed in Mausund (in a 50 km^2 radius area; Fig. S1) during the month of September between 2016 and 2018 (one campaign per year). The crabs (*Cancer pagurus*; $N = 65$ in total) were collected using crab pots in three locations in 2016 ($N = 15$); five locations in 2017 ($N = 27$); and four locations in 2018 ($N = 23$) (Fig. 1). In total, 65 brown crab biological samples were collected (one sample per crab), including 3 gonad (raw brown crab meat), 4 claw (raw white brown crab meat) and 58 hepatopancreas (raw brown crab meat) samples.

The Atlantic halibut (*Hippoglossus hippoglossus*; $N = 3$) and cod (*Gadus morhua*; $N = 7$) fish were caught in 2017 and 2018, respectively. Other fish species were caught in 2018, namely porbeagle shark (*Lamna nasus*; $N = 1$), rabbit fish (*Chimaera monstrosa*: ghost shark; $N = 1$), haddock (*Melanogrammus aeglefinus*; $N = 1$), common ling (*Molva molva*; $N = 3$), pollock (*Theragra finnmarchica*; $N = 1$), lumpfish (*Cyclopterus lumpus*; $N = 1$) and skate (*Amblyraja hyperborean*; $N = 2$). All these fish species can either act as predator or prey for the brown crab (Woll, 2006), thus demonstrating a predator-prey interrelationship. Cod were caught using specific cod traps, while the other fish species were caught using regular fishing gear (e.g., fishing line).

Fig. 1. Locations of brown crab sampling in 2016 (1–3), 2017 (4–8) and 2018 (9–12); geographical coordinates shown in Table S1.

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Fish fillet samples were obtained from Atlantic halibut (k = 3), cod (k = 1), porbeagle shark (k = 1), rabbit fish (k = 1), common ling (k = 1), and pollock (k = 1) by obtaining the tissue between the gut and the main dorsal fin. For the Atlantic halibut, the fillet samples were obtained from the stomach area (in front of the gut opening), which is richer in fat. In addition to the fish fillet samples, a liver and stomach sample were obtained from the porbeagle shark, two liver samples and one stomach sample were obtained from cod, and an intestine sample was obtained from the pollock. Moreover, concerning the common ling samples, an additional liver and intestine sample were obtained from a single fish.

From the lumpfish, haddock and skate species, no fish fillet samples were obtained. A liver and stomach sample were obtained from the haddock, a gonads sample (raw eggs) was obtained from the lumpfish, and a stool, liver and stomach sample were obtained from the skate.

The fish and crab species were kept in sea water after collection and were transferred alive to Mausund Fieldstation, where they were euthanized prior to sampling of the tissues. Thereafter, the samples were kept frozen at −20 °C. The sampling coordinates for all species are provided in supporting information (Tables S1 and S2; Figure S2).

2.2.2. Deep-sea sediment matrix

Sediment samples (N = 10) were collected with a Van Veen grab sampler in 2017 (N = 5) and 2018 (N = 5; deepest point-158 m); ~10 gr was obtained per sediment sample. The sampling coordinates are provided in supporting information (Table S3; Figure S3). The samples were kept intact and transported refrigerated (10 °C) to the laboratory after collection. The sediment samples were directly stored at − 20 °C.

2.3. Sample preparation

All sample matrices were freeze-dried (Alpha 1–2 LDplus; *Christ, Harz, Germany*) and homogenized before further sample preparation; thus, concentrations corresponded to dry weight (d.w.), which were consequently not influenced by the water content. When necessary (for comparison purposes), concentrations from brown crab and fish biological media were converted from d.w. to wet weight (w.w.) dividing by a factor of 3.2 and 5, respectively (Cresson et al., 2017; Knutsen et al., 2018). The water content in brown crab and fish biological media was estimated at 69 and 80%, respectively (Cresson et al., 2017; Knutsen et al., 2018). A portion of 100–400 mg of the dried environmental or biological sample was transferred to a polytetrafluoroethylene (PTFE) vessel and 9 mL of aqueous nitric acid (50% v/v) was added. Digestion of the samples was carried out in a high-pressure microwave system, and after the digested samples cooled down to room temperature, they were diluted with Milli-Q water and transferred for HR-ICP-MS analysis.

2.4. HR-ICP-MS analysis

Elemental analyses were performed using an *Element 2* instrument *(Thermo Finnigan model; Bremen, Germany)*. The samples were introduced using a combination of SC2 DX autosampler (with ULPA filter dust cover) and PrepFAST flow injection analysis system *(Elemental Scientific, Inc. Omaha, NE, U.S.)* with a total flow of 200 μL/min. The instrument was equipped with a PFA-ST nebulizer, spray chamber (PFA Barrel 35 mm), demountable torch, quarts standard injector, sample cone and skimmer cones. Methane (CH_4) was introduced into argon (Ar) gas to avoid the formation of oxides and provide enhanced sensitivity, especially for Se and As. The cool, auxiliary, nebulizer and T-connection gas flow were set at 15.5, 1.10, 0.75, and 0.55 L/min, respectively. The LOQ of every element was set at a concentration where repeatability (RSD%) was 25% (supporting information).

2.5. Classification of essential and non-essential (toxic) elements

The essential inorganic chemicals for maintaining biological activities within an ecosystem/organism are established as essential minerals (or elements), which can be further classified into micro- (trace) and macro- (major) minerals (Zoroddu et al., 2019). Trace elements that are essential for biological activities include Fe, Cu, B, Mo, Ni, Zn, Mn, Se and V (Vincent, 2010; Zoroddu et al., 2019). Toxic elements that are mainly established as non-essential include Pb, Cd and Hg (Sobolev et al., 2019). The macro-minerals (essential) include K, Ca, Mg and P (Zoroddu et al., 2019). Sr, As and Cr are highly controversial elements (e.g., Sr replacing Ca in human media, organic vs. inorganic As toxicity, Cr(III) vs. Cr(VI)) in terms of essentiality; nonetheless, here Sr and As were considered non-essential (toxic) elements, while Cr was considered an essential element (Nielsen, 2004; Vincent, 2010; Zoroddu et al., 2019). It must be noted that the essential elements can demonstrate toxicity for an ecosystem/organism above specific thresholds (Voulgaris et al., 2019; Zoroddu et al., 2019).

2.6. Quality assurance and quality control (QA/QC)

Accuracy and reproducibility were ensured by measuring replicates of certified calibration solutions, and through frequent analysis of the customized reference soil material GBW 07408 (GSS-8) (Chinese National Center for Standard Materials). One of the purchased calibration sets was used as the calibration solution (CS) and the other for quality assurance/quality control (QA/QC). Instrumental repeatability of the measurements was established for every individual sample by scanning it three consecutive times. For each batch of 20 samples analyzed, one procedural blank was analyzed, and a calibration check standard was injected as a check for drift in instrumental sensitivity.

2.7. Data analysis

HR-ICP-MS data treatment was performed with the built-in software of the *Element 2* instrument *(Thermo Finnigan model; Bremen, Germany)*. Statistical treatment was performed with STATGRAPHICS Centurion XV software package (Stat Point, Inc., Version 2002) and Excel (Microsoft, 2010). Data analysis did not include censored data (i.e., nondetects; NDs). The statistical significance was set at $p < 0.05$.

3. Results and discussion

3.1. Elemental profile in deep-sea sediments

The elemental concentrations in the surface sediments (mg/kg d.w.) collected in 2017 and 2018 are given in Tables S4 and S5, respectively, and the logarithmic transformed median values are depicted in Fig. S4. In deep-sea sediments, Ca (med. 106569–201 758 mg/kg d.w.), Na (4246–14301), Fe (1561–5578), Al (1627–5526), S (1094–3813), Mg (1441–2719), K (752–2458), Sr (549–1277) and P (167–515) were found in abundance, followed (in decreasing order; median) by: Ti (77.6–234 mg/kg d.w.) > Mn (57.6–130) > Ba (11.2–45.2) > Zn (8.57–30.4) > Ce (6.03–14.2) > Pb (4.16–10.9) > Cu (2.14–9.13). The high concentrations of Fe, Ca, Al, P, K, Mg, S, Sr and Ti were attributed to the underlying occurring geological minerals of the location (Faust et al., 2014; Nordgulen et al., 1990). These elements derive mainly from natural occurring Fe-Mg minerals (e.g., amphibole and dolomite), potassium feldspar

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(e.g., $K(AlSi_3O_8]$), illite, calcite and aragonite (e.g., limestone, which is a type of sedimentary rock consisting mainly of $CaCO_3$), fluorapatite $Ca_5(PO_4)_3F$, apatite and hydroxyapatite (e.g., $Ca_5(PO_4)_3OH$), TiO_2 containing minerals, and sulphide minerals (e.g., pyrite (FeS_2)) that are found in abundance in the seabed around Mausund (Faust et al., 2014, 2017; Nordgulen et al., 1990). Elements found in minor concentrations including Se, V, Y, Rb, Zn, and Ce are also released from the seabed minerals (Tabelin et al. 2018). Other elements of interest, namely Cr, Ni, As, Se, Sn, Cd and Hg, demonstrated median concentrations of 4.14–13.3, 2.41–9.37, 0.84–2.41, 0.37–0.82, 0.15–0.51, 0.06–0.16 and 0.01–0.07 mg/kg d.w., respectively. As far as Pb, Sb, As, Bi, W and Cd were concerned, these elements are documented preferentially attached to smaller-size aerosols available for long-range atmospheric transport (Gladney et al., 1976; Greenberg et al., 1978; Steinnes et al., 2011), indicating that the inputs of these elements in the marine environment are not derived solely from local sources. On the contrary, Cr, Ni and Cu concentrations derive mainly from local point sources (Christensen et al., 2018). Comparing to selected elements analyzed in sediments during a detailed 2015 survey in Mausund, no significant differences were observed (Ervik et al., 2018) that would further indicate any recent anthropogenic pollution inputs, which in turn would currently affect the brown crab populations.

3.2. Elements in biological media of fish species: Focusing on Pb, Cd, Hg and As

The concentrations of elements determined in fish species are given in Tables S6–S15. For the Atlantic halibut, the median concentrations of As and Hg were 69.7 and 0.37 mg/kg d.w., respectively (est.: 13.9 and 0.07 mg/kg w.w., respectively), while Cd was not detected, and Pb demonstrated a median concentration of 0.01 mg/kg d.w (est.: 0.002 mg/kg w.w.). The concentrations of As determined in Atlantic halibut were higher than those reported in samples during 2012–2015 (mean: 11.3 mg/kg d.w.; est. mean: 2.26 mg/kg w.w.) from Mausund (Ervik et al., 2018), but in most instances they were lower than those reported from Norwegian waters during 2006–2010, which reached up to 48 and 15 mg/kg w.w. in Greenland and Atlantic halibut, respectively (Julshamn et al., 2012a, 2013a). The Cd, Pb and Hg concentrations did not exceed the upper limits set (for muscle meat) by the E.U. of 0.05, 0.30, 1 (specific for Atlantic halibut) mg/kg w.w., respectively (E.U., 2006, 2011).

For cod, the concentrations of Cd (0.10–0.13 mg/kg d.w.), Hg (0.50–0.56 mg/kg d.w.), Pb (0.008–0.07 mg/kg d.w.) and As (6.07–14.4 mg/kg d.w.) were found to be similar to those reported during 2012–2015, where the concentrations for Cd, Hg, Pb and As were 0.12 (mean), 0.56 (mean), 0.02 (median) and 9.45 (median) mg/kg d.w., respectively (Ervik et al., 2018). Here, the Cd concentration in one fillet sample (0.43 mg/kg d.w.; est.: 0.09 mg/kg w.w.) exceeded the upper Cd limit (for muscle meat; 0.05 mg/kg w.w.) (E.U., 2006, 2011). The median Hg concentrations were below the upper limit (for muscle meat: 0.05 mg/kg w.w.) (E.U., 2006, 2011). Individually, one stomach (0.83 mg/kg d.w.; est.: 0.17 mg/kg w.w.) and four fillet samples (1.70, 1.04, 0.76 and 0.59 mg/kg d.w.; est.: 0.34, 0.21, 0.15 and 0.12 mg/kg w.w.) demonstrated concentrations that exceeded this Hg limit. All Pb concentrations were found below the upper Pb limit (for muscle meat: 0.30 mg/kg w.w.) (E.U., 2006, 2011).

The porbeagle shark demonstrated Cd, Hg and Pb median concentrations of 0.49, 0.28 and 0.69 mg/kg d.w., respectively. Cd concentrations exceeded the upper limit (for muscle meat: 0.05 mg/kg w.w.) (E.U., 2006, 2011) in the liver (0.32 mg/kg d.w.; est. 0.06 mg/kg w.w.) and stomach (0.67 mg/kg d.w.; est.: 0.13 mg/kg w.w.) sample; however, in the fillet sample, Cd was not detected. The Hg concentration determined in porbeagle shark fillet was 1.87 mg/kg d.w. (est. 0.37 mg/kg w.w.), which was lower than the upper limit (for muscle meat) of 1 (Hg; specific for shark species) mg/kg w.w. (E.U., 2006, 2011). The Pb concentration determined in porbeagle shark fillet was 2.68 mg/kg d.w. (est. 0.54 mg/kg w.w.), which was higher than the upper limit (for muscle meat) of 0.30 mg/kg w.w. (E.U., 2006, 2011). The concentrations of As found in porbeagle shark (median: 21.4 mg/kg d.w.) were lower than those determined in Atlantic halibut, but higher than those determined in cod (Table S6–S8).

The rabbit fish demonstrated As, Hg, Pb and Cd fillet concentrations of 93.8, 2.47, 0.42 and 0.02 mg/kg d.w., respectively. The common ling showed As, Cd, Hg and Pb median concentrations of 18.0, 0.02, 0.26 and 0.05 mg/kg d.w., respectively, while the pollock showed As, Cd, Hg and Pb median concentrations of 10.1, 0.05, 0.26 and 0.04 mg/kg d.w., respectively. In addition, in the pollock fillet sample, Cd was not detected, while As, Hg and Pb showed concentrations of 18.0, 0.47 and 0.01 mg/kg d.w., respectively.

The stomach sample from the haddock demonstrated a relatively high Pb concentration (8.98 mg/kg d.w.), while the stool and liver sample from the skate contained Cd concentrations of 1.89 and 4.29 mg/kg d.w., respectively. Additionally, the stool and liver sample from the skate contained Pb and Hg concentrations of 1.33 and 1.80 mg/kg d.w., respectively. The lumpfish eggs showed minimal concentrations of As, Cd and Pb compared to those determined in biological media from other fish species, while Hg was not detected (Table S15). Overall, As and Hg were found in higher concentrations in fish media from Mausund compared to Pb and Cd.

3.3. Elevated Cd concentrations in brown meat from brown crab

The analysis of the biological media from brown crab (61 out of the 65 samples were brown meat samples; Tables S16 - S18) indicated extremely high Cd concentrations when compared to the maximum allowed concentration of 0.5 mg/kg w.w. set by the E.U. for raw white crab meat (Fig. 2). Cd was found higher than the LOQ (0.02 mg/kg) in 63 out of 65 samples with a yearly median concentration ranging from 6.75 (2016) to 14.0 (2017) mg/kg d.w. (est.: 2.11 (2016) to 4.37 (2017) mg Cd/kg w.w.) (Fig. 2). The 4 claw samples (raw white crab meat) were measured < 0.1 mg/kg d.w., while the 3 gonad (raw brown crab meat) samples demonstrated concentrations of 45.0, 6.20 and 9.32 mg/kg d.w. (est. 14.0, 1.93 and 2.91 mg/kg w.w., respectively). Overall, only 5 samples (1 brown and all 4 white meat samples) demonstrated Cd concentrations < 0.5 mg/kg w.w.. Two concentration points are not depicted in Fig. 2, 122 (2017) and 203 mg/kg d.w. (2018) (est. 38.1 and 63.4 mg/kg w.w., respectively), which were the maximum concentrations measured; previous reports on Cd concentrations in brown crab are depicted in Table 1.

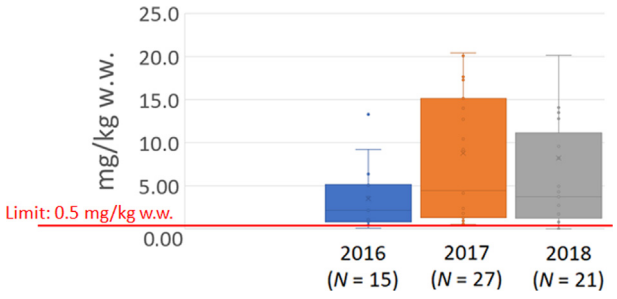

Fig. 2. Cd concentrations measured in brown crab media (61 out of the 65 samples are brown meat samples) compared to the maximum allowed concentration of 0.5 mg/kg w.w. set by the E.U. for raw white crab meat.

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Table 1
Studies reporting Cd concentrations (mg/kg wet weight) in brown crab media.

Location	Year	Number of samples	Brown meat Mean or median/range	White meat Mean or median/ range	References
Borough of Birsay, Scotland	1978	35 (brown meat)/33 (claw meat)	7.30/1.12–49.4	–/0.08–0.27	Davies et al., 1981
Eyemouth, Scotland	1978	60	2.60/0.48-8.17	–/–	
Lock Ewe, Scotland	1982	20	10.8/0.83-32.2	–/–	Falconer et al., 1986
English Channel	2008	20	18.0/–	0.10/–	Barrento et al., 2009b
Scottish coast		20	25.0/–	0.10/–	
Norway (along the coast)	2013–2014	197	8.70/0.24–43.0	0.62/0.02–4.50	Frantzen et al., 2015
Senja, Norway	2015	20	9.30/1.60–29.0	0.53/0.03–3.20	Frantzen and Maage, 2016
Kvaløya, Norway		19	30.0/7.30–58.0	0.25/0.06–0.74	
Hadselfjorden, Norway	2012	10	10.0/5.60–14.0	0.69/0.10–2.50	Julshamn et al., 2013b,c
Nykvåg, Norway		10	8.70/3.20–21.0	0.58/0.18–1.20	
Rollnesboen, Norway		9	7.90/4.70–12.0	0.38/0.07–1.30	
Akkarnes, Norway		10	10.0/3.20–15.0	0.81/0.24–2.20	
Floholm nord, Norway		10	13.0/1.50–28.0	1.30/0.34–3.00	
Stø, Norway		10	8.80/0.90–16.0	0.60/0.13–1.20	
Eidsfjorden/Ramberg/Straumsnes, Norway		10	7.40/2.60–13.0	0.73/0.10–1.80	

The results strongly indicate that it is of paramount importance to have maximum Cd concentrations regulated in brown meat from brown crab, acknowledging also that while brown meat samples can contain Cd concentrations even higher than 50 mg/kg w.w. (Table 1), the upper Cd concentration limit set for safe consumption of white crab meat is significantly lower, 0.5 mg/kg w.w. It is noteworthy that no recent Cd pollution pressures were identified regarding brown crab in this specific Mid-Norway marine habitat: the Cd concentrations (in d.w.) measured in deep-sea sediments (Section 3.1.) and in biological media from predatory fish (Section 3.2.) were ~ 2–3 orders of magnitude lower than those measured in the brown meat from brown crabs. The findings here further suggest that the Cd concentrations in brown meat from this specific Mid-Norway marine habitat of brown crab are likely to be not a result of pollution, but rather a result of natural bioaccumulation, as recently indicated in the literature (Wiech et al., 2018). Nevertheless, it is a fact that there is no full understanding yet of what actually affects the edible crab throughout the seasons. It has been documented though, that the brown crab can migrate down to 400 m and more during the winter season (Bakke et al., 2019). In brown meat samples (from brown crab), which were obtained during 2012–2015 from Mausund, Cd mean concentrations were reported to be significantly lower (mean: 10.3 mg/kg d.w.) (Ervik et al., 2018). Here, where the samples were collected during 2016–2018, the yearly mean Cd concentrations ranged 11.1 (2016)-28.0 (2017) mg/kg d.w., indicating an increasing trend in Cd concentrations in brown meat from Mausund (in contrast to deep-sea sediments; see Section 3.1.).

3.4. Hg, Pb and As concentrations in brown crab media

For all samples, Hg concentrations were significantly lower than the maximum allowed concentration of 0.5 mg/kg w.w. for raw white crab meat (E.U., 2006, 2011) (Fig. S5). In contrast to the Cd findings (Section 3.3.), the Hg median concentrations were not statistically different (in d.w.) between brown crab and other miscellaneous fish media (Section 3.2) (p = 0.20 > 0.05; Kruskal-Wallis test). Compared to brown meat concentrations measured during 2012–2015 from Mausund (Ervik et al., 2018), Hg concentrations (in d.w.) reported here were found to be similar to 2016, but ~ 2 fold higher in 2017 and 2018.

Pb concentrations for all samples were also significantly lower than the maximum allowed concentration of 0.5 mg/kg w.w. for raw white crab meat (Fig. S6). Comparing our results with a previous Norwegian survey conducted in 2011 (Julshamn et al., 2012b), some crab samples (claw and brown meat) from the municipalities of Frøya and Hitra, which are in close geographical proximity to Mausund, showed concentrations that reached even up to 0.30–0.40 mg/kg w.w. The Pb concentrations reported in brown meat during 2012–2015 (mean: 0.16 mg/kg d.w.) from Mausund (Ervik et al., 2018) were similar to those reported here.

The concentrations of As determined in this study were ~ 2–3 fold higher than those reported in samples during 2012–2015 (mean: 44.2 mg/kg d.w.) from Mausund (Ervik et al., 2018). Furthermore, As and Pb median concentrations (in d.w.) were ~ 1 order of magnitude and ~ 3-fold higher, respectively, in the brown crab than in the fish media (Section 3.2.), denoting higher bioaccumulation potential in crab biological media for these two elements.

3.5. Human exposure / consumer risks assessment

Human exposure was assessed by estimating intakes of elements per 100 g dry serving portion of edible media (white/brown meat) from brown crab. Estimated dietary intake (EDI; μg /kg body weight (b.w.)) and intake dose (ID; mg or μg) were calculated for select elements based on simple steady-state toxico-kinetic equations (Eqs. (1) and (2)):

$$\text{Estimated daily intake (EDI; } \mu g/\text{kg b.w.}) = \frac{\text{Elemental concentration in crab media } \left(\frac{\mu g}{\text{kg,d.w.}}\right) \text{x 0.1 kg d.w.}}{\text{70 kg b.w.}} \quad (1)$$

$$\text{Intake dose (ID; mg or } \mu g) = \text{Elemental concentration in crab media} \left(\frac{\text{mg or } \mu g}{\text{kg d.w.}}\right) \times \text{0.1 kg d.w.} \quad (2)$$

The EDIs were calculated for an adult individual older than 19 years of age and weighing 70 kg (Barrento et al., 2009d; USNAS, 2019). However, it should be noted that several uncertainties (e.g., way of consumption/cooking and bioavailability) exist in the assessment of exposure based on the measured concentrations in brown crab media. Currently, adequate intakes (AIs) and tolerable upper intake levels (ULs) for essential elements are specified by the Food and Nutrition Board, Institute of Medicine, United States National Academy of Sciences (USNAS, 2019), and these values

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were used here for assessing the consumption risk (USNAS, 2019). The EDI (μg/kg b.w.) and ID (mg or μg) are presented in Tables 2 and 3, respectively.

3.5.1. Intake doses for essential elements

The median ID values indicated that indeed the edible media from brown crab is an excellent source of essential elements (Barrento et al., 2009d). However, due to their abundance, by consuming just 100 gr of dry edible media, ULs can be exceeded for specific elements; here, ULs were exceeded for Fe, Cu, Ni, Se, Ca, Mg and P. Se has an important anti-oxidative stress role against toxic metals, including Cd and Hg (Sobolev et al., 2019). However, in higher concentrations it can be toxic (Bansal and Asthana, 2018). In addition, it is noteworthy that currently intake values are not specified for K (USNAS, 2019).

3.5.2. Intake doses for toxic elements

The median ID values indicated that 100 gr of dry edible media can expose humans to high concentrations of toxic elements; Cd and As in particular, reached the order of a few tens to thousands of mg, respectively, as observed here. As is regularly found higher than 50 mg/kg w.w. in seafood, which according to the literature is considered a high concentration (Francesconi, 2007). However, Maulvault et al. (2011) reported an upper limit of 76 mg/kg w.w. for As in crustaceans [which was established by the *United States Food and Drug Administration* (FDA) in 2010]. The highest median concentrations reported here for As were 107 mg/kg d.w. (est. 33.4 mg/kg w.w.) in 2017 and 117 mg/kg d.w. (est. 36.5 mg/kg w.w.) in 2018, while the overall maximum concentration reached 3287 mg/kg d.w. (est. 1027 mg/kg w.w.).

Cd and Hg are classified as human carcinogens, and there is evidence suggesting that Se interacts with those in vivo, reducing their toxicity (Calatayud et al., 2012; Schöpfer et al., 2010). A molar Se:Cd or Hg ratio < 1 appears to increase the toxic potential of Cd or Hg, while those which approach or exceed (≥)1, appear to increasingly protect against Hg or Cd toxicity (Calatayud et al., 2012; Schöpfer et al., 2010). Here brown crab media showed a median Se:Cd and Se:Hg ratio of 0.56 and 29.0, respectively. Se:Cd ratio values > 1 were shown in only 22 out of the 65 brown crab samples, while Se:Hg ratio values > 1 were shown in all 65 samples. For comparison purposes, fish media demonstrated for the same ratios, values > 1 in ~90% of samples with a median Se:Cd and Se:Hg ratio of 97.5 and 6.75, respectively. Based on these ratios (and not on the concentrations) and by accepting that Se protects against Cd toxicity, it can be suggested that potentially 33% of these brown crab media samples can be safely consumed. Nonetheless, this suggestion must be further confirmed in future studies. Moreover, alongside with Se, other essential elements (e.g., Zn) should be further evaluated in terms of their protective effects (as individual elements or in combination with other elements) against toxic elements in order to obtain a clear understanding of actual exposures from brown crab media consumption and toxic effects.

4. Conclusions

In summary, compared to local wild-caught predatory fish, brown crab media demonstrated significantly higher concentrations of Cd, As and Pb, while Hg concentrations were not statistically different. Cd was the most abundant toxic element in brown crab media, which was also indicated as a great risk for human health, and consequently needs to be further addressed by regulatory bodies. The human exposure assessment of brown crab indicated that the essential elements, namely Fe, Cu, Ni, Se, Ca, Mg and P, can potentially exceed the established ULs. Moreover,

Table 2
Estimated daily intake (μg/kg b.w.) of select essential and toxic elements from brown crab media consumption.

Sampling Year	As			Cd			Hg			Pb			Se			Sr		
	Min.	Median	Max.	Min.	Median	Max.	Min.	Median	Max.	Min.	Median	Max.	Min.	Median	Max.	Min.	Median	Max.
2016	51.0	98.7	4695	0.17	9.64	60.7	0.03	0.14	0.39	0.03	0.13	0.33	3.89	8.21	25.8	9.73	377	611
2017	47.2	152	454	2.36	20.0	172	0.11	0.46	1.11	0.04	0.19	0.83	2.64	8.59	36.0	145	478	1118
2018	47.2	167	302	0.04	17.0	288	0.07	0.36	0.80	0.03	0.16	0.43	3.41	8.23	27.8	18.2	458	1150
Sampling Year	Ca			Cu			Zn			Mg			Mn			K		
	Min.	Median	Max.	Min.	Median	Max.	Min.	Median	Max.	Min.	Median	Max.	Min.	Median	Max.	Min.	Median	Max.
2016	660	35426	67500	11.3	31.0	504	55.6	95.9	310	818	3110	4084	5.80	11.4	14.1	4673	10740	22552
2017	7521	35227	80151	17.1	105.3	1334	45.1	96.3	229	2357	3294	6478	6.25	12.9	28.7	8064	12281	34960
2018	1350	34118	85752	18.0	57.0	804	47.7	99.9	541	1714	3403	5198	1.00	10.0	28.6	6760	11861	23243
Sampling Year	B			Fe			Cr			Mo			Ni			P		
	Min.	Median	Max.	Min.	Median	Max.	Min.	Median	Max.	Min.	Median	Max.	Min.	Median	Max.	Min.	Median	Max.
2016	1.70	8.90	12.1	18.6	214	571	0.21	0.47	1.76	0.17	0.86	2.46	0.04	2.13	21.5	12847	33516	50294
2017	4.91	7.27	15.0	177	425	1134	0.30	0.80	6.24	0.28	1.04	3.18	0.24	3.30	15.4	18588	33372	58304
2018	3.07	7.06	13.6	23.8	328	890	0.03	0.64	5.77	0.06	0.77	2.29	0.17	2.24	16.0	30580	12325	64245
Sampling Year	V																	
	Min.	Median	Max.															
2016	0.24	0.79	16.5															
2017	0.30	1.40	16.2															
2018	0.04	1.07	12.1															

Table 3
Intake Dose (ID; mg or μg) for select essential and toxic elements from 100 gr (dry) brown crab media consumption.

As (mg)			Cd (mg)			Hg (μg)			Pb (μg)			Se (μg)			Sr (mg)		
Medians	Max.	UL	Medians	Max.	UL	Medians	Max.	UL	Medians	Max.	UL	Medians	Max.	UL	Medians	Max.	UL
6.91-11.7	329	-	0.68-1.40	20.2	-	10.0-32.0	78	-	9.1-13.3	58.0	-	575-601	2520	400	26.4-33.4	80.5	-
*Ca (mg)			Cu (mg)			Zn (mg)			Mg (mg)			Mn (mg)			K (mg)		
Medians	Max.	UL	Medians	Max.	UL	Medians	Max.	UL	Medians	Max.	UL	Medians	Max.	UL	Medians	Max.	UL
2388-2479	6002	2500	2.17-7.37	93.4	10	6.71-6.99	37.9	40	217-238	453	350	0.70-0.91	2.01	11	752-859	2447	-
B(mg)			Fe(mg)			Cr (μg)			Mo (μg)			Ni (μg)			P (mg)		
Medians	Max.	UL	Medians	Max.	UL	Medians	Max.	UL	Medians	Max.	UL	Medians	Max.	UL	Medians	Max.	UL
0.49-0.62	1.05	20	15.0-29.8	79.4	45	33.0-56.0	437	20	54.0-73.0	223	2000	149-231	1510	1000	2140-2346	4497	3000
V (μg)																	
Medians	Max.	UL															
55.0-98.0	1160	1800															

*Ca: The median IDs were below the daily UL of 2500 mg for individuals between 19 and 50 years of age but exceeded the daily UL of 2000 mg for individuals older than 50 years of age.

based on the molar ratio of Se:Cd in brown crab media, it was suggested that 33% of those samples could be potentially consumed, further proposing this ratio as an essential criterion of risk from Cd exposure, rather than the sole Cd content.

Declaration of Competing Interest

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

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Appendix A. Supplementary data

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