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Effects of industrial processing on essential elements and regulated and emerging contaminant levels in seafood

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Mitigation of contaminants in industrial processing was studied for prawns (cooked and peeled), Greenland halibut (cold smoked) and Atlantic salmon (cold smoked and trimmed). Raw prawns had significantly higher cadmium, chromium, iron, selenium and zinc content in autumn than in spring, while summer levels typically were intermediate. Peeling raw prawns increased mercury concentration but reduced the concentration of all other elements including inorganic arsenic, total arsenic, chromium, zinc, selenium but especially cadmium, copper and iron ($p < 0.05$), however interaction between seasons and processing was observed.

Non-toxic organic arsenic in raw Greenland halibut ($N = 10$) and salmon ($N = 4$) did not transform to carcinogenic inorganic arsenic during industrial cold smoking. Hence inorganic arsenic was low (<0.003 mg/kg wet weight) in both raw and smoked fillets rich in organic arsenic (up to 9.0 mg/kg for farmed salmon and 0.7 mg/kg for wild caught Greenland halibut per wet weight). Processing salmon did not significantly change any levels (calculated both per wet weight, dry weight or lipid content). Cold smoking decreased total arsenic (17%) and increased PCB congeners (10–22%) in Greenland halibut (wet weight). However PFOS, PCB and PBDE congeners were not different in processed Greenland halibut when corrected for water loss or lipid content.

1. Introduction

Seafood is one of the most important food commodities consumed worldwide and recognized as a high-quality and healthy food item. Although marine fish supplies have been declining during recent years, farmed aquaculture products continue to increase globally (constituting around 50% of the total seafood market) due to the technological advances in production and relatively lower production costs (compared to non-farmed fisheries; FAO, 2014). Aquaculture has contributed to a rise in fish consumption in Europe that has now stabilised at 22 kg per capita per year, with Iceland and Portugal being the populations with the highest intake. Seafood is an important source of several beneficial components (e.g. ω3 polyunsaturated fatty acids (PUFAs), vitamin D, selenium and iodine) and the health benefits of a diet, which is rich in seafood have been extensively recognized. The World Health Organization (WHO), the Food and Agriculture Organization (FAO) as well as several national food authorities recommend a regular fish consumption of 1–2 servings per week in order to provide an adequate level of ω3-PUFA (FAO/WHO, 2003). On the other hand, seafood, like other types of food, may also be a source of exposure for humans to environmental contaminants like persistent organic pollutants (POPs) and toxic elements. Currently, the contaminant level in seafood is in most countries being monitored by national surveillance programmes, which form the basis for conduction of risk assessment and risk management with regards to seafood consumption. However, at present, only relatively few data is available for contaminants where no maximum limits have been set by authorities, i.e. the so-called emerging contaminants (Vandermersch et al., 2015), which include compounds such as toxic elements, endocrine disruptors, brominated flame retardants, pharmaceuticals and personal care products, polycyclic aromatic hydrocarbons and derivatives, microplastics and marine toxins. The Marine Strategy Framework Directive emphasizes the need for further monitoring of these compounds in seafood for human

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Abbreviations

α-HBCD	α-hexabromocyclododecane
As	arsenic
BFR	brominated flame retardants
Cd	cadmium
Cr	chromium
Cu	cobber
Fe	iron
GC-HRMS	gas chromatography high-resolution mass spectrometry
Hg	mercury
iAs	inorganic arsenic
LC-MS/MS	liquid chromatography tandem mass spectrometry
Pb	lead
PBDE	polybrominated diphenyl ether
PCB	polychlorinated biphenyls
PFC	perfluorinated compound
PFNA	perfluorononanoate
PFOSA	perfluorooctane sulfonamide
PFOS	perfluorooctane sulfonate
PFOA	perfluorooctanoate
Se	selenium
Sn	tin
TBBPA	tetrabromobisphenol A
ww	wet weight
Zn	zinc
2,4,6-TBP	2,4,6-tribromophenol

consumption for determination of Good Environmental Status of the marine environment (MSFD, 2010). For non dioxin-like PCB maximum levels have been established by European Commission Regulation 1259/2011 (ECR, 2011).

Trace elements occur widely in all types of foodstuffs, either because of their natural occurrence in the environment or from contamination during food production, processing and storage. Essential trace elements (e.g. iron, zinc, copper, selenium, chromium and iodine) are crucial for human health whereas others are potentially dangerous (De la Guardia and Garrigues, 2015). Seafood has been identified as one of the food items which may contain relatively high concentrations of a number of the toxic trace elements (Llobet et al., 2003; Bocio et al., 2005), typically identified as lead, cadmium and mercury, for which maximum limit in seafood are regulated by the European Commission regulation1881/2006 (ECR, 2006). For other (toxic) trace elements, so far no maximum levels have been laid down in the EU legislation, partly due to lack of knowledge on their occurrence in seafood. For some trace elements, the chemical form (i.e. the elemental speciation) in which the element is ingested may play a significant role from a toxicological point of view. For example, methylmercury is known to be much more toxic than inorganic mercury compounds, while inorganic arsenic is known to be more toxic than the organic species of arsenic (EFSA, 2009; 2012).

Halogenated contaminants *i.e.* chlorinated contaminants such as polychlorinated biphenyls (PCBs), brominated flame retardants (BFR) as e.g. polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecanes (HBCDs) and polyfluorinated compunds (PFC), are all anthropogenic persistent chemicals ubiquitous in the environment, even in remote places like the Arctic (Glasius et al., 2005). The chlorinated and brominated compounds are lipophilic and bioaccumulate and biomagnify in the lipid phases of the food chain, while the perfluorinated compounds have surfactant properties and more tend to occur in muscles tissue, liver and blood (Stahl et al., 2011). The applications of brominated flame retardants are or have been as flame retardants in many materials including also plastics and the perfluorinated compounds are typically used in industrial and consumer products (impregnating agent for paper, textiles etc.) (Stahl et al., 2011). While PCB is a well-known contaminant, the occurrence of the other compound groups mentioned is not very frequently reported. Recent reports by e.g. Van Leeuwen et al. (2009) on halogenated contaminants in farmed seafood and Glasius et al. (2005) and Carlsson et al. (2014, 2016) who conducted surveys of halogenated contaminants in seafood from remote areas of Greenland and Northern Norway, respectively, emphasise the increasing interest in these types of compounds.

The majority of data on contaminant level in seafood have been achieved from the analysis of raw seafood. However, most seafood is consumed processed and prepared and hence risk evaluations based on raw seafood may be biased when not taking the effect of processing on the contaminant level into account. Therefore, in order to conduct improved risk assessments, seafood processing and preparation procedures should also be taken into account. There is currently a lack in our knowledge on how industrial processing may influence the levels of contaminants in seafood. In order to improve the risk assessment further information on processed seafood is needed, so that the evaluation can be done on products being consumed.

The aim of the present study was to assess whether selected industrial processing procedures (smoking, fat trimming, peeling, cooking) affect the content of a range of emerging contaminants in seafood products. The results obtained in the study furthermore contribute with novel data on emerging contaminants in some commercially relevant seafood products, that is Atlantic salmon, Greenland halibut and prawns. In addition the seasonal influence on contamination levels in raw prawns and further processing mitigation was evaluated. It is expected that the results obtained here will be of value for future risk assessments of these contaminants in seafood and contribute to the optimization of industrial processing of seafood and hereby contribute to improving the food safety of these products.

2. Materials and methods

2.1. Prawns (Pandalus borealis)

Prawns (3–7 years old) were caught by net near the west coast of Greenland (Fig. 1) in the FAO 21 Major fishing area, more precise in the Northwest Atlantic Fisheries Organization (NAFO) subarea 1A and 1B (grid LE23, LH25, LH24, LK25, LL25,LS14, LJ27, LT15, KG16, KR17 and KT17 in Fig. S1). They were caught in different seasons; spring (25.3–1.4 2014), summer (15.-17.8 2014) and autumn (28.-29.11 2014). Each shipping of raw prawns was industrially processed at the Royal Greenland A/S factory in Sisimiut and Ilulisat into cooked prawns with shell/head/guts and cooked and peeled prawns without shell/head/guts. Raw and cooked prawns were sampled immediately before the cooker (CoolSteam-Cooker, Laitram Machinery, New Orleans, Lousiana, USA), peeled prawns were taken after the digester (Laitram Machinery) before glazing. Surface water from the city lake was applied in the processing. Samples within the season came from different areas and inshore vessels, but from the same maturation tank. Prawns (500 g) were packed in freezer bags, and pressed flat without air before freezing and were