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Perfluoroalkylated substances (PFASs) and legacy persistent organic pollutants (POPs) in halibut and shrimp from coastal areas in the far north of Norway: Small survey of important dietary foodstuffs for coastal communities

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ABSTRACT

Halibut (*Hippoglossus hippoglossus*) and shrimps (*Pandalus borealis*) are regular foodstuffs for communities in northern Norway and important species for the coastal fishing industry. This is the first study to present a comprehensive overview of the contaminant status of these species, with emphasis on unregulated perfluoroalkylated substances (PFAS). The contaminant concentrations were low and within tolerable levels for human dietary exposure. Median Σpolychlorinated biphenyls (PCB) were 4.9 and 2.5 ng/g ww for halibut and unpeeled shrimps, respectively. Concentrations of perfluorooctane sulfonate (PFOS) – the most abundant PFASs – were 0.9 and 2.7 ng/g ww in halibut and shrimp, respectively. The halibut fillets were dominated by PCBs, which contributed to 50% of the total POPs load, followed by ΣDDTs; 26% and PFASs (18%), whereas shrimps were dominated by PFASs (74%). ΣPBDEs (polybrominated diphenyl ethers) contributed to 1–4% of the total POP load. Local sources are not contributing significantly to the contaminant burden in these species.

1. Introduction

Halibut (*Hippoglossus hippoglossus*) and shrimp (*Pandalus borealis*) are popular marine foods in Norway and are important commercial species present in coastal waters of northern Norway. Halibut are long-lived, benthic fish species that are piscivorous whereas shrimps are epibenthic and feed on detritus, as well as on pelagic lower trophic level organisms such as phytoplankton and zooplankton (IMR, 2014). The Norwegian fishing industry catches 5000 t of coastal shrimps every year, and 1400 t of halibut were caught in 2009 (IMR, 2014). The median fish dietary intake among the Norwegian population is 65 g fish/day, with high-consumers eating between 118 and 174 g fish/day (Bergsten, 2014; VKM, 2006, 2014a). However, it is not known how much of this comprises of shrimps and halibut. Marine foodstuffs are regularly scanned and analysed for nutrients, legacy and new pollutants by the National Institute of Nutrition and Seafood Research (NIFES) with data published in an open archive (NIFES, 2014). Persistent organic pollutants (POPs) reach the Arctic via long-range environmental transport (AMAP, 2003), although activities in coastal areas such as fisheries, shipping and the presence of harbours and associated coastal runoff from Arctic settlements may all serve to increase the levels of these contaminants. To date, however there have been relatively few surveys that have examined the levels of POPs such as polychlorinated biphenyls (PCBs), organochlorine pesticides (OCs) and polybrominated diphenyl ethers (PBDEs), in halibut and shrimps despite the fact that these chemicals are still cause for concern regarding their tendency to bioaccumulate with detrimental effects on both humans and wildlife (AMAP, 2011; Stockholm Convention, 2015). Furthermore, there are fewer data for newer contaminants such as the perfluoroalkylated substances (PFAS) and brominated flame retardants (BFRs), which, in some cases, may bioaccumulate and biomagnify in marine foodwebs and hence provide a dietary exposure pathway to humans (Carlsson et al., 2011; Haukås et al., 2007; Sørmo et al., 2009). Recent investigations of PFASs, PBDEs, PCBs and OC pesticides in marine food stuffs have been undertaken in Greenland and Iceland (Carlsson et al., 2014a, 2014b; Jörundsdóttir et al., 2012; Sturludottir et al., 2014), and PFAS in food stuffs from the Faroe Islands (Eriksson et al., 2013). Some of these data are comparable to the coastal species examined here.

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The aim of this study was to investigate contaminant concentrations in halibut and shrimps – species for which there are relatively few data – from coastal fishing regions in northern Norway and to compare levels to similar species from more remote parts of the Arctic. This provides insight into whether coastal fisheries have higher contaminant levels due to the proximity of additional sources of pollution. A further aim was to examine PFAS concentrations in relation to POPs to provide insight into their biological uptake and distribution within these two species. Given the health concern of emerging contaminants as well as legacy POPs in marine foodstuffs, this study puts these organisms into context as contributors to human dietary exposure to these chemicals. There are on-going long-term studies regarding human health in Tromsø (Berg et al., 2014; Jacobsen et al., 2012; Nost et al., 2013, 2014) and the results from this study provide important new data for the improvement of human exposure assessments for these chemicals.

2. Methods

Fresh fillets of halibut (*H. hippoglossus*) were purchased from local fishermen and fishmarkets and also caught from coastal waters close to Tromsø (PFAS: $n = 9$, other POPs: $n = 6$) over the period 2008–12. Shrimps (*P. borealis*) caught closeby from the Malangen and Kvænangen regions (unpeeled shrimp: $n = 9$, peeled shrimp: $n = 5$) were provided by a supplier in 2012. Length and weight of each halibut are reported in table S1. Tromsø is the largest city in northern Norway with ~70,000 inhabitants (Fig. 1). Sample handling, clean-up procedures and analyses were performed in accordance with well-established methods. The samples were analysed for a suit of PFAS, PCBs, PBDEs and OCs. All details, including abbreviations of PFASs can be found in the supplementary information (method description and table S2–4) and in earlier papers describing the methods for PFAS analyses (Carlsson et al., 2014a; Hanssen et al., 2013; Herzke et al., 2009) and PCB, PBDE, dichlorodiphenyltrichloroethane and its metabolites (DDTs), chlordanes and hexachlorobenzene (HCB) analyses (Crosse et al., 2012). PFASs were separated and quantified on a Thermo Scientific ultrahigh pressure liquid chromatography tandem mass-spectrometry (UHPLC–MS/MS). The PCBs, PBDEs and OCs were separated and quantified by a Thermo gas chromatograph (GC) connected to a mass spectrometer (MS) operated in electron ionisation mode (EI).

Basic statistics were performed with the Paleontological statistics software package for education and data analysis (PAST), e.g. Mann–Whitney's test or Kruskal–Wallis test (Hammer et al., 2001). A *p*-value of 0.05 was considered statistically significant if nothing else is stated. Samples below the limit of detection (LOD) are not included in the median or mean calculations (Table S5).

2.1. Quality control

The following 13C-labelled internal standards (IS) were used for quantification of the respective analysed compounds: For PFAS: PFBA, PFPA, PFHxA, PFHpA, PFOA, PFNA, PFDcA, PFUnA, PFDoA, PFHxS, PFOS and PFOSA (Wellington Laboratories Inc., Guelph, Ontario, Canada). The abbreviations are listed in Table S2. For PCBs, OCs and PBDEs, the following 13C-labelled IS were used: PCB-28, -52, -138, -153, and -180; PBDE-28, -47, -99, -100, -153, and -154, with quantification based on the recovery standards of PCB-30, 13C-labelled PCB-141 and 13C-labelled PCB-208 (PCBs/OCs) and PBDE-69 and 181 (PBDEs) purchased from Cambridge Isotope Laboratories, Andover, Massachusetts, US.

For chemical confirmation, quantifier and qualifier mass transitions were acquired for each analyte including the PFASs, except for PFBA and PFPA, where only a quantifier mass was acquired (Table S2). For PFAS analysis a laboratory blank and a standard reference material (SRM) were analysed every 10th sample PFAS 'ILS 2011' 'fish tissue' (developed during the PERFOOD project, KBBE; grant agreement no. 227525) was used as a reference material. The measured levels in these SRMs varied within an acceptable range (±20%) compared to

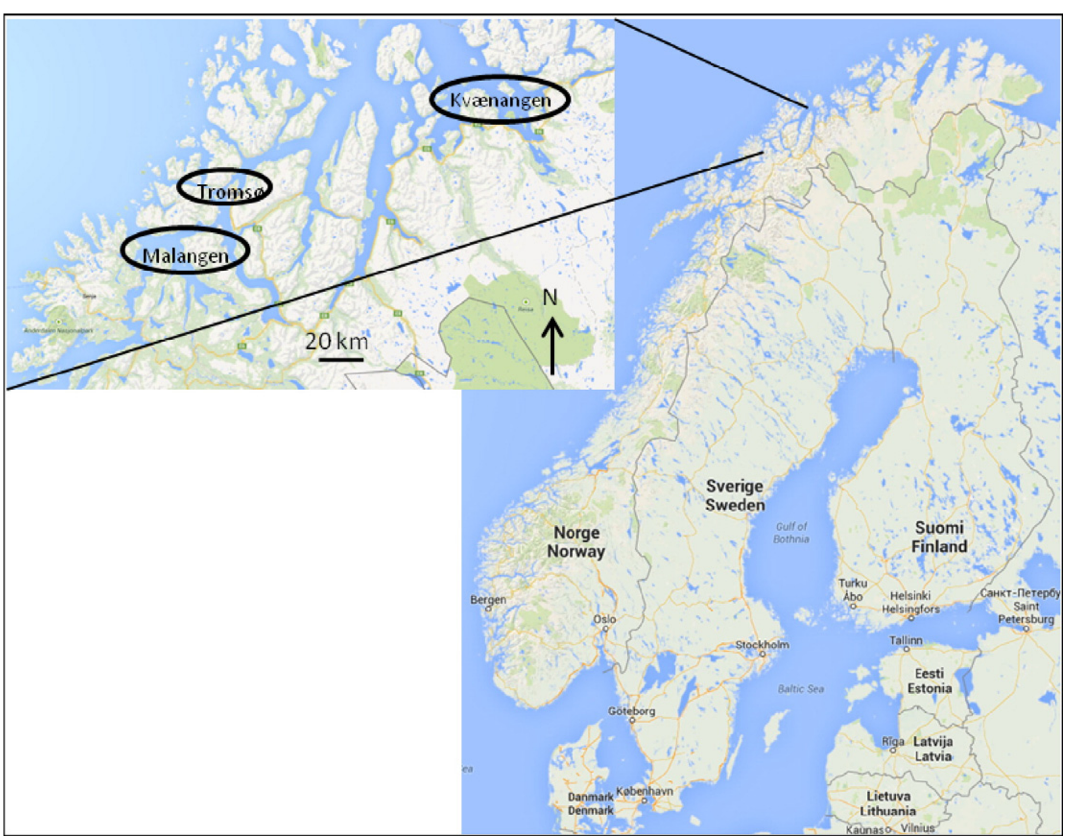

Fig. 1. Map showing Northern Norway with the sampling locations of Malangen (shrimps and halibut), Tromsø (halibut) and Kvænangen (shrimps) marked. Map from www.google.com.