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Factors influencing risk assessments of brominated flame-retardants; evidence based on seafood from the North East Atlantic Ocean

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ABSTRACT

Brominated flame-retardants (BFRs) such as polybrominated diphenyl ethers (PBDE) and hexabromocyclododecane (HBCD) are considered hazardous to human health. Due to their persistence, they are still present in the environment and in biota and seafood is major contributor of BFRs to human exposure. Here, we used data from > 9700 samples of wild and farmed fish, fish feed and fish feed ingredients collected from the North Atlantic between 2006 and 2016 aiming to investigate factors influencing the risk assessments of BFRs.

Due to most representative number of analyses, PBDEs were the main focus of investigation. Mean ΣPBDE in fillet samples ranged from below quantification in Atlantic cod fillet to 2.0 μg kg^{-1} in Atlantic halibut. The main congener contributing to the ΣPBDE in all species was BDE 47. Factors affecting the level of BFR in seafood were multifaceted, and the levels were within species mainly determined by fish age, geographical origin and time of sampling. BDE 47, 99, 153 and HBCD were selected for margin of exposure (MOE) evaluation. When other sources of BFR than seafood were excluded, our risk assessment showed low risk at the current dietary intake of seafood. However, the dietary intake of BDE 99 may be of concern for toddlers when all sources are considered. The choice of fish species, dietary studies, choice of statistics, as well as exposure from other sources than seafood, were all factors that influenced the final MOE of BFRs. We propose the use of regression on order statistics as a tool for risk assessment, to illustrate means and spreads in large surveillance datasets to avoid the issue of measurements below the limit of quantification. A harmonized, updated evaluation of the risk associated with exposure to BFRs from diet, air and dust is warranted, where the fish species most commonly consumed also is taken into consideration.

1. Introduction

Brominated flame retardants (BFRs) constitute a diverse group of compounds used in several commercial commodities to prevent or restrain fire. For instance, the legacy BFRs such as polybrominated diphenyl ethers (PBDE), hexabromocyclododecanes (HBCD) and tetrabromobisphenol A (TBBPA) have been used in electrical components, furniture and insulation-foam. The European Union (EU) has taken precautionary measures regarding these specific BFRs, and has issued bans or restrictions on their production and use (EC, 2003; EC, 2008; EFSA, 2012; Koch et al., 2015). However, due to their persistence they are still present in the environment and consequently in biota (Danon-Schaffer et al., 2013), particularly in aquatic organisms. Hence, the legacy BFRs, are still relevant BFR classes for monitoring (EC, 2014; EFSA, 2006).

Food of animal origin, particularly fat-rich seafood, is traditionally

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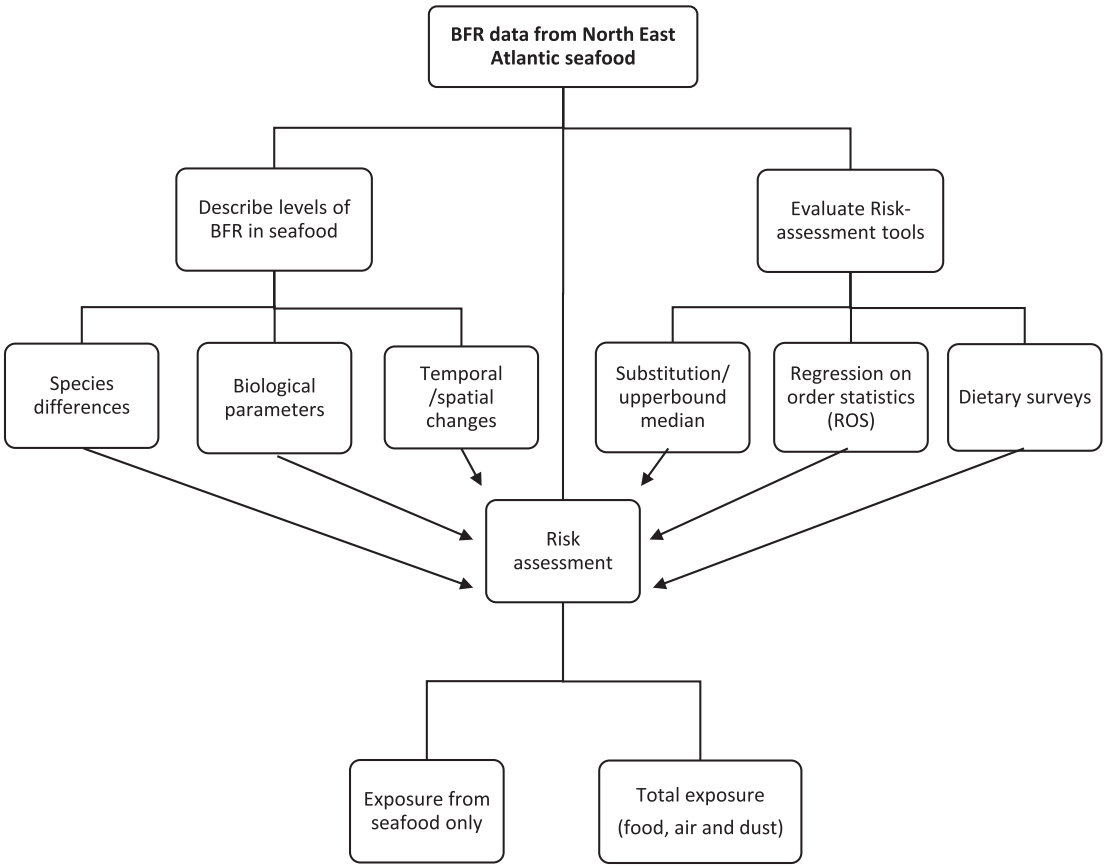

Fig. 1. Flowchart describing the objective and aim of the study. This study has evaluated both levels of BFR in seafood, and the factors affecting these, and different risk assessment tools. These two aspects have been assessed according to their effect on risk assessment from both multiple sources of exposure and from seafood exposure alone.

regarded as a major contributor of BFRs to human exposure (Cruz et al., 2015; EFSA, 2011a; EFSA, 2011b; Koch et al., 2015), although the impact of non-food sources should also be considered (Koch et al., 2015; Martellini et al., 2016). The contribution of Norwegian seafood for human BFR exposure is of interest not only for the Norwegian population who traditionally have a high seafood intake, but also for the population of countries which import seafood from Norwegian waters. Norway is the world's second largest exporter of fish and fishery products including both farmed and wild fish (FAO, 2016). Whereas food exposure assessments generally use “fish” or “seafood” as general food intake categories, commercial Norwegian seafood consist of several different species with a large variation in fat content, age, position in the marine food chain, harvest location and season of capture, which all affect BFR levels and congener composition and thus cause different exposure. In this paper, we highlight factors that may cause variation in risk assessments of BFRs in seafood. We assessed the impact of exposure from other sources than seafood, and how choice of statistics related to reporting limit of quantification (LOQ) in surveillance data, affects risk assessment (Fig. 1). Based on an extensive dataset, we highlight species-specific risk of seafood consumption in terms of the legacy BFRs. The levels of PBDEs, HBCD and TBBPA in the main commercial fish species harvested in and near Norwegian waters are also described. Further, we evaluate factors affecting the level of the different BFRs in seafood species, such as age, fat content, geographical origin, time of sampling and feed.

2. Materials and methods

2.1. Sample material

The data presented in the current study comprise results from analyses of 9764 marine samples including both wild and farmed fish, fish feed and fish feed ingredients collected between 2006 and 2016. A total of 9211 samples were analyzed for ΣPBDE, here defined as sum of BDE 28, 47, 99, 100, 153, 154 and 183, 1453 for HBCD and 352 for TBBPA (Table 1). Additionally 383 samples of fish feed or fish feed ingredients were analyzed; 383 were analyzed for ΣPBDE, 275 for HBCD, and 69 for TBBPA. An overview of all analyses and results are given in [dataset] Appendix data. Sampling was done primarily on commercial fish species used as food, with the exceptions of certain forage fish (capelin and polar cod). Sampling locations for wild fish represent Norwegian fishing grounds including areas beyond the Norwegian territorial boundaries (Fig. 1). Fish were mainly sampled in seasons when commercial fishing occurs for the different species. Farmed Atlantic salmon (*Salmo salar*), were collected from all regions along the Norwegian coast with aquaculture activity. Twelve of the wild Atlantic salmon caught at sea were found to originate from fish farms, using methodology described elsewhere (Fiske et al., 2005; Lund et al., 1991), and are treated as a separate group hereafter called escapees. Fish feed and fish feed ingredients were sampled from Norwegian feed producers or at fish farms, representative of fish-feed production in Norway.

All samples were analyzed at the Institute of Marine Research (IMR) or Eurofins Gfa GmbH (Hamburg, Germany). The farmed fish, feed ingredients and fish feed were sampled by the Norwegian Food Safety Authority, while the wild fish were sampled by the IMR. The current study includes data on the legacy brominated flame-retardants PBDEs (28, 47, 66, 99, 100, 119, 138, 153, 154 and 183), HBCD and TBBPA.

2.2. Sample preparation

Fish length, weight and sex were recorded for each fish sampled