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Mercury species in dab (*Limanda limanda*) from the North Sea, Baltic Sea and Icelandic waters in relation to host-specific variables

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ABSTRACT

In the framework of the ICON project (Integrated Assessment of Contaminant Impacts on the North Sea), muscle tissue from a total of 135 common dab (*Limanda limanda*) (20–28 cm total length) was collected in seven offshore sampling areas in the North Sea, at Iceland and in the Baltic Sea during Aug/Sept and December 2008 for a chemical mercury speciation analysis by means of gas chromatography and detection by cold vapour atomic fluorescence spectroscopy (GC-CVAFS). There was a highly significant correlation between concentrations of methylmercury ($MeHg^+$) and inorganic mercury (Hg^{2+}) in individual fish, and the mean ratio of $MeHg^+$ compared to Σ Hg ($MeHg^+ + Hg^{2+}$) was 94.0%. The results revealed statistically significant differences in concentrations of $MeHg^+$ and Hg^{2+}, respectively, between sampling areas. Mean concentrations in the German Bight (North Sea), in Icelandic waters and in Mecklenburg Bight (Baltic Sea) were low ($MeHg^+$: 0.023–0.036; Hg^{2+}: 0.001–0.002 mg/kg wet weight), while concentrations in dab from the Dogger Bank, Firth of Forth and the vicinity of the Ekofisk oil field (all North Sea) were significantly higher ($MeHg^+$: 0.059–0.101; Hg^{2+}: 0.003–0.004 mg/kg wet weight). Statistical correlation analysis on effects of host-specific factors revealed that neither length, weight, age, sex nor condition factor showed a significant relationship with Hg concentrations. However, Hg concentrations were significantly correlated with the Fish Disease Index (FDI), indicating a relationship between Hg concentrations and the health status of dab. Multiple linear regression analysis aiming to find factors affecting Hg concentrations revealed that only the sampling area had a highly significant main effect on Hg concentrations, and in some cases, additionally the condition factor contributed significantly to the final model. From the results, it cannot be excluded that elevated Hg concentration recorded in dab were linked to discharges from offshore oil and gas installations and that Hg affected the health status of dab.

1. Introduction

Mercury (Hg) is a naturally occurring element and is regarded as a global pollutant that affects human and ecosystem health (EU, 2005; Driscoll et al., 2013; UNEP, 2013). Human activity has increased the mobilization of Hg in the environment mainly since the industrial revolution in the 19th century, raising the amounts in the atmosphere, soils, fresh waters and oceans by a factor of 2–3 (Macdonald et al., 2005; UNEP, 2013). There is indication that, overall, anthropogenic emissions from fuel combustion and the industrial sector have increased since 2005, after a period of stable levels in the period 1990–2005 (UNEP, 2013). Since Hg is a highly toxic heavy metal found in many, including aquatic, ecosystems at partly high concentrations, it is still regarded as a global threat to human and environmental health, thus, having motivated national and international initiatives (US EPA, 2001; 2010; EU, 2005; Driscoll et al., 2013; UNEP, 2013).

According to Mason et al. (2012) and UNEP (2013), releases of Hg to water have doubled the amount in the top 100 m of the world's oceans in the last 100 years, and Zhang et al. (2015) estimated that the surface ocean Hg concentrations have increased fourfold over

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the last 600 years. For open sea systems, direct atmospheric deposition of Hg is regarded as major source of contamination (Mason et al., 2012; Driscoll et al., 2013), and half of the emitted anthropogenic Hg has accumulated in the oceans and marine sediments (Zhang et al., 2015). During atmospheric transport, chemical reactions convert elemental Hg to divalent forms that can bind to particulate matter. Both divalent Hg and particulate Hg are deposited from the atmosphere onto water bodies where microorganisms and chemical processes can convert a small proportion of the Hg to its organometallic form methylmercury ($MeHg^+$), the toxic Hg species of main concern regarding environmental and health effects in humans and wildlife (Swain et al., 2007; Roman et al., 2011; Driscoll et al., 2013). Due to its higher assimilation efficiency and lower elimination rate in aquatic animals compared to the inorganic mercury (Hg^{2+}), $MeHg^+$ is largely responsible for the accumulation of Hg in organisms (bioaccumulation) and its transfer from one trophic level to another (biomagnification) in the marine food web (Mason et al., 1996; Driscoll et al., 2013). The accumulation of $MeHg^+$ in aquatic organisms is largely related to their trophic level (Hammerschmidt and Fitzgerald, 2006; Driscoll et al., 2013; Hammerschmidt et al., 2013; Zhang et al., 2013). For instance, in phytoplankton, only 1–10% of the total Hg is present as $MeHg^+$, while up to 100% of the total Hg consists of $MeHg^+$ in fish and marine mammals (Bloom, 1992; Suedel et al., 1994; Hill et al., 1996; Bunke, 2007). Fish and other seafood are important pathways to human Hg exposure, and the most common route of exposure is the consumption of marine and freshwater fish (Sunderland, 2007; Swain et al., 2007; Roman et al., 2011; Dijkstra et al., 2013; Driscoll et al., 2013).

The toxicity of Hg is complex (Clarkson and Magos, 2006; Grandjean et al., 2010), partly because it may exist in different chemical forms in the environment all of which may exert toxic effects (McElwee et al., 2013). $MeHg^+$ has long been recognized as a potent neurotoxin in humans, affecting the central nervous system leading to brain damage and has been associated with cardiovascular and immunological effects (US EPA, 2001; 2010; Kaur, 2008; Grandjean et al., 2010; Roman et al., 2011; Karagas et al., 2012; Dijkstra et al., 2013). Fatal effects in humans related to consumption of highly $MeHg^+$ contaminated fish have been documented, e.g., during the Minamata and Niigata epidemics in Japan in the 1950s and 1960s (Ekino et al., 2007; Kaur, 2008).

In fish, mercury is concentrated in the muscle, liver and kidney (Dixon and Jones, 1994), and toxic effects such as sublethal tissue damage, depressed reproduction, effects on hormonal and endocrine systems, oxidative stress, genotoxicity and increased apoptosis have been recorded and attributed to $MeHg^+$ exposure, partly at environmentally realistic concentrations (Wiener and Spry, 1996; Oliveira Ribeiro et al., 2002; Gonzales et al., 2005; Mela et al., 2007; Drevnick et al., 2008; Klaper et al., 2008; Machado Da Rocha et al., 2009; Tan et al., 2009; Sandheinrich and Wiener, 2011; Depew et al., 2012).

In the Northeast Atlantic, including the North Sea, Hg and other inorganic and organic contaminants in fish are monitored on a regular basis as part of the OSPAR Coordinated Environmental Monitoring Programme (CEMP) and the Joint Assessment and Monitoring Programme (JAMP) (OSPAR, 2010a, 2011). According to the JAMP Technical Guidelines for monitoring of contaminants in biota, dab is the first choice flatfish species for chemical monitoring (OSPAR, 2011). Thus, data are available and have been used for environmental assessments (OSPAR, 2010b, 2014). However, since $MeHg^+$ is not among the contaminants measured on a routine basis, we applied speciation analysis in order to quantify $MeHg^+$ as Hg species of predominant toxicity.

The aims of the present study were to compare concentrations of the two Hg species $MeHg^+$ and Hg^{2+} in dab (*Limanda limanda*) from a geographically large area, encompassing stations in the North Sea and in areas in the Baltic Sea and Icelandic waters, and to analyse and assess the relationship with the host-specific factors sex, length, age, weight, condition factor and disease status as well as their effects on regional patterns in concentrations of mercury species. The results constitute a contribution to the overall aim of the ICON project (Integrated Assessment of Contaminant Impacts on the North Sea) to assess the status of selected European marine areas with regards to anthropogenic contaminants and their biological effects (Hylland et al., 2017). Within ICON, a large suite of chemical and biological indicators were measured in fish, mussels and sediments in an integrated way and the results of these studies are published in other contributions to the same volume.

2. Material and methods

Dab were collected during cruises no. 315 (29.08.–19.09.2008) and no. 317 (28.11.–16.12.2008) of RV Walther Herwig III by means of bottom trawling, using either a 140 ft (Baltic Sea and Iceland) or a GOV (North Sea) bottom trawl with standard configuration for stock assessment. Towing time was 30–60 min, towing speed 3–4 knots. The location of the seven sampling areas in the North Sea and off Iceland (both visited during cruise no. 315) and in the Baltic Sea (visited during cruise no. 317) is shown in Fig. 1, geographical coordinates are given in Table 1.

Live dab were randomly sorted from the catches and kept alive in tanks with running seawater of ambient water temperature prior to dissection. Following euthanasia, muscle tissue (without skin) was removed from the upper body side and individually deep frozen (−20 °C) in plastic bags. Fish size was measured to the nearest cm below and fish weight as total wet weight (g). Otoliths for subsequent age determination were removed and stored individually. Sex was determined by external and internal inspection.

Condition factors (CF) were calculated as Fulton's K = wet weight * 100/total length^3. Data characterizing the fish used for analysis are provided in Table 1.

The health status of dab was recorded based on external examination for gross diseases and parasites, following standard methodologies developed by the International Council for the Exploration of the Sea (ICES) (Bucke et al., 1996) and through the Biological Effects Quality Assurance in Monitoring (BEQUALM) programme (www.bequalm.org). A Fish Disease Index (FDI) was calculated for individual fish, based on the presence/absence of the nine disease conditions lymphocystis, epidermal hyperplasia/papilloma, acute/healing skin ulcerations, acute/healing fin rot/erosion, x-cell gill disease and infestation with *Stephanostomum baccatum* (Digenea), *Acanthochondria cornuta* and *Lepeophtheirus pectoralis* (both Copepoda), their intensity (3 severity grades), their suspected impact on the host (weighting by expert judgement) as well as on adjustment factors for effects of sex, length and sampling season on the disease prevalence (Lang and Wosniok, 2008; ICES, 2012; Lang et al., 2017; this issue).

Chemical speciation analysis of $MeHg^+$ and Hg^{2+} was carried out using samples from a total of 135 individual dab (99 females, 36 males), size range 20–28 cm total length (Table 1). The method for the determination of both mercury species was elaborated as part of a research project, performed 2005–2008 in the Institute for Fish and Fishery Products Cuxhaven of the Lower Saxony State Office for Consumer Protection and Food Safety. It was published as part of the final project report by Kruse and Bartelt (2008). In different aspects, the applied method is a development of the procedure of Harms and Bunke (2002). A complete description of the applied analytical procedure can be submitted by the performing institute (E-mail: Poststelle.IFF-Cux@Laves.Niedersachsen.de).

The subsequent overview shows the central operation steps, as