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Stable isotope analyses revealed the influence of foraging habitat on mercury accumulation in tropical coastal marine fish

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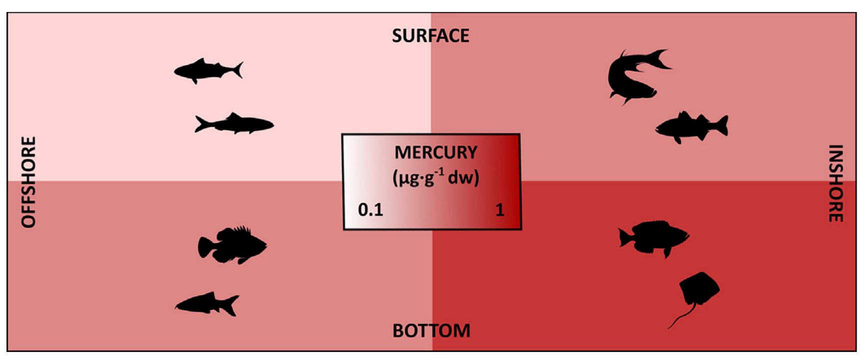
HIGHLIGHTS

- THg, $\delta^{13}C$, $\delta^{15}N$ were determined in 132 marine fish from a tropical coastal region.
- Coastal and demersal species were more contaminated than oceanic and pelagic ones.
- SIA suggest that Hg exposure derived from bottom feeding and/or in estuarine waters.
- Conspecific fish fed on different coastal food webs and Hg increased with fish size.
- Hg levels in fish from Senegal were not of concern for human consumption.

GRAPHICAL ABSTRACT



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ABSTRACT

Bioaccumulation of toxic metal elements including mercury (Hg) can be highly variable in marine fish species. Metal concentration is influenced by various species-specific physiological and ecological traits, including individual diet composition and foraging habitat. The impact of trophic ecology and habitat preference on Hg accumulation was analyzed through total Hg concentration and stable isotope ratios of carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$) in the muscle of 132 fish belonging to 23 different species from the Senegalese coast (West Africa), where the marine ecosystem is submitted to nutrient inputs from various sources such as upwelling or rivers. Species-specific ecological traits were first investigated and results showed that vertical (*i.e.* water column distribution) and horizontal habitat (*i.e.* distance from the coast) led to differential Hg accumulation among species. Coastal and demersal fish were more contaminated than offshore and pelagic species. Individual characteristics therefore revealed an increase of Hg concentration in muscle that paralleled trophic level for some locations. Considering all individuals, the main carbon source was significantly correlated with Hg concentration, again revealing a higher accumulation for fish foraging in nearshore and benthic habitats. The large intraspecific variability observed in stable isotope signatures highlights the need to conduct ecotoxicological studies at the individual level to ensure a thorough understanding of mechanisms driving metal accumulation in marine fish. For individuals from a same species and site, Hg variation was mainly explained by fish length, in accordance with the

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bioaccumulation of Hg over time. Finally, Hg concentrations in fish muscle are discussed regarding their human health impact. No individual exceeded the current maximum acceptable limit for seafood consumption set by both the European Union and the Food and Agriculture Organization of the United Nations. However, overconsumption of some coastal demersal species analyzed here could be of concern regarding human exposure to mercury.

1. Introduction

Coastal areas provide many ecosystem services such as fish production, ensuring the supply of marine products to local populations and supporting the economies of coastal countries, particularly through the export (Interwies and Görlitz, 2013). Among the many disturbances to which these ecosystems are subjected, mercury (Hg) contamination is a major environmental concern, due to its deleterious effects on marine organisms and human health (Streets et al., 2011). Hg originates from both natural and anthropogenic processes, including geological weathering, gold mining, combustion and industrial discharges. Although Hg is volatile and can be transported through the atmosphere over long distance away from emission sources, coastal environments are particularly subjected to Hg exposure since they are located at the interface between three main Hg sources: atmosphere, rivers and ocean (Cossa et al., 1996).

After being released into seawater, Hg can be methylated by microorganisms such as sulfate and iron-reducing bacteria or methanogens (Correia and Guimarães, 2017; Gilmour et al., 2013; Paranjape and Hall, 2017; Podar et al., 2015) in coastal and shelf sediments as well as in sub-thermocline oceanic waters, where oxygen concentration is low and organic matter is intensively remineralized (Blum et al., 2013; Cossa et al., 2017; Fitzgerald et al., 2007). Once in the organic methylmercury form (MeHg), this metal is easily taken up by phytoplankton and biomagnifies along trophic chains. Biomagnification (*i.e.* increase in MeHg content with trophic level) is responsible for high Hg concentration in the tissues of top predators such as tunas and sharks (McKinney et al., 2016). As trophic level is closely correlated with nitrogen stable isotope ratios (δ^{15}N) (Post, 2002), this marker has been extensively used to explain quantities of Hg in biota (Kiszka et al., 2015; McMeans et al., 2010; Pethybridge et al., 2012).

Species with similar trophic levels can, however, display different patterns of Hg accumulation (Bank et al., 2007; Le Croizier et al., 2016) and other factors have been linked to Hg concentrations. First, like other metal elements, Hg strongly binds to sulfhydryl groups of proteins in marine fish leading to a very slow excretion of the bioaccumulated Hg over time, particularly in muscle tissues (Kidd and Batchelar, 2011; Peng et al., 2016). As a consequence, Hg concentration has been extensively shown to increase with fish age, being positively correlated with size or weight (Bosch et al., 2016a; Chouvelon et al., 2014; Pethybridge et al., 2010). Second, Hg concentrations in fish tissues are also known to increase with depth of occurrence in the water column, corresponding to concentrations of dissolved organic mercury in seawater and/or proximity to sediments (Chouvelon et al., 2012; Choy et al., 2009). Third, ecosystem characteristics such as human activities, density and composition of phytoplankton communities can influence Hg bioaccumulation and subsequent biomagnification (Chen and Folt, 2005; Chouvelon et al., 2018; Condini et al., 2017; Heimbürger et al., 2010).

Finally, some studies focusing on Hg sources found differences in Hg concentrations between offshore and nearshore species or between pelagic and benthic species, depending on whether Hg methylation takes place in pelagic deep waters or in coastal sediments (Cresson et al., 2014; Sackett et al., 2015; Senn et al., 2010). Carbon stable isotope values (δ^{13}C) have commonly been used to discriminate oceanic/pelagic from coastal/demersal habits in marine fish in order to investigate the role of foraging habitat in Hg bioaccumulation (Cresson et al., 2014; Goutte et al., 2015; Pethybridge et al., 2012; Signa et al., 2017). Coastal fish species have a wide range of prey and habitat use, however, which leads to large intraspecific variability in metal accumulation (Bird et al., 2018; Le Croizier et al., 2016; Sackett et al., 2015). It therefore seems essential to conduct ecotoxicological studies at the individual level to better determine the origin of the accumulated Hg in fish tissues.

In this context, the general goal of the present study was to investigate the trophic origin of mercury exposure to coastal fish from the Canary Current Large Marine Ecosystem (CCLME) in Western Africa. This ecosystem is of particular interest since it is one of the world's major cold-water upwelling currents and ranks first in the world in terms of primary productivity (Messié and Chavez, 2015). It supports one of the largest fisheries among African large marine ecosystems and provides food to local populations but also to foreign countries through the attribution of fishing licenses and export of marine products. This marine ecosystem is prone to metal contamination due to urban effluents and industrial activities (Auger et al., 2015; Diop et al., 2015) and Hg has been recently identified as one of the toxic elements reported at significant concentrations in sediments and marine organisms from this coastal zone (Bodin et al., 2013; Diop and Amara, 2016; Net et al., 2015). Hence, as Hg can cause serious pathologies including neurological impairments (Bosch et al., 2016b) and fish consumption represents the major pathway for human exposure to Hg (Driscoll et al., 2013), a proper evaluation of Hg sources and concentrations was needed in fish from this area.

To fulfill this objective, total mercury concentrations were determined in a wide range of fish belonging to 23 different species. Total mercury was used as a proxy of methylmercury, since MeHg generally represents >80% of the total mercury found in fish muscle, regardless of their ecological characteristics (Andersen and Depledge, 1997; Bloom, 1992; Magalhães et al., 2007; Storelli et al., 2003). The main origin for Hg contamination was investigated through the reported habitat distribution of the species. This method allowed to discriminate Hg exposure between oceanic *versus* coastal and pelagic *versus* demersal sources, but did not take into account the individual specialization in terms of foraging habitat. Thus, a second complementary approach was undertaken using stable isotope analyses, particularly δ^{13}C that gives information at the individual level but only discriminate between oceanic/pelagic *versus* coastal/demersal sources. Finally, the potential health impact associated with fish consumption in the area was discussed in relation to the tolerable Hg intake limit established by the European Union and the Food and Agriculture Organization of the United Nations.

2. Materials and methods

2.1. Sampling

A total of 132 fish belonging to 23 different species were sampled at four different locations along the Senegalese coast in West Africa (from north to south): Saint-Louis, Dakar, Saloum and Casamance (Fig. 1). Saint-Louis, Saloum and Casamance are located off estuaries, while Dakar is located along a rocky coast. Samples used in this study were collected during a scientific cruise (doi,