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Drivers of the accumulation of mercury and organochlorine pollutants in Mediterranean lean fish and dietary significance

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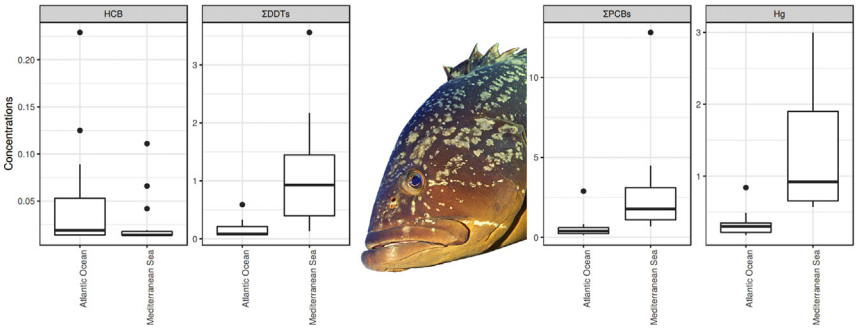
HIGHLIGHTS

- 40% of Mediterranean lean fish had Hg concentrations above EU recommended values.
- The levels of organochlorine compounds in all specimens were below the EU threshold.
- Hg levels were correlated with specimen weight but the trend was species dependent.
- The organochlorine compound concentrations were not correlated with specimen weight.
- Mediterranean populations feeding on fish have higher EWI for Hg than the EFSA PTWIs.

GRAPHICAL ABSTRACT



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ABSTRACT

An integrated assessment of lean fish of commercial value as Hg and organochlorine compound source into the population of the Balearic Islands were reported. Dependences between pollutant concentrations, trophic level, fish species, specimen weight and physical-chemical properties were evaluated.

Hg and total DDTs showed highest variability between fish species whereas PCBs and HCB displayed more constant median values. The organochlorine compounds found in highest concentrations were those with highest hydrophobicity, consistently with their higher bioaccumulation potential. These pollutant concentrations were higher in Mediterranean than Atlantic fish. Higher median total DDT and PCBs concentrations were also observed in the third than the second trophic level species. The observed concentrations were below the threshold recommended by the EU for human consumption (75 ng/g wet weight).

The Hg concentrations were higher in Mediterranean than Atlantic fish, with average values of 1.5 μg/g ww and 0.43 μg/g ww, respectively. Forty-one percent of the specimens from the Mediterranean and 25% of dusky grouper specimens from the Atlantic Ocean showed Hg concentrations above the EU recommended limits for human consumption, either 0.5 μg/g ww or 1 μg/g ww.

In the third trophic level, a significant dependence between median Hg concentrations and weight of each studied species was observed, which remained significant in specimen weight correlations. Independent species correlations of Hg concentrations vs individual weight generally showed higher concentrations at higher weight. Weight/size of the individuals was therefore an important factor for Hg accumulation but the trend was modulated by a species effect.

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Extrapolation of the observed Hg concentrations in Mediterranean fish to Provisional Tolerable Weekly Intakes (PTWIs) showed higher intakes than the thresholds recommended by EFSA for adults and children, 110% and 140%, respectively. The estimated PTWIs for MeHg corresponded to 310% and 400% of the recommended threshold values. The PTWI values for organochlorine compounds were lower than those recommended.

1. Introduction

Fish consumption has been associated to human accumulation of mercury (Hg) and organochlorine pesticides (Llop et al., 2010; Gascon et al., 2012, 2013; Garí et al., 2013; Vizcaino et al., 2014; Costa et al., 2016; Bravo et al., 2017). Despite their different origin and structure, OCs and Hg share common properties such as (I) strong chemical stability and environmental persistence, (II) bioconcentration in living organisms and biomagnification through the food chain due to their hydrophobic character and (III) toxicity for humans and wild animals. Methylmercury is a more toxic form than the original metal whose human intake is also attributed to fish and seafood (Calatayud et al., 2012; Garí et al., 2013; Perello et al., 2014; Cano-Sancho et al., 2015; Obeid et al., 2017).

Human exposure to environmental concentrations of organochlorine compounds with high number of chlorine substituents has been associated to diverse deleterious effects, e.g. hexachlorobenzene (HCB) to attention deficit hyperactivity syndrome (Ribas-Fitó et al., 2007), alteration of thyroid hormones (Álvarez-Pedrerol et al., 2008; Sala et al., 2001), overweight (Smink et al., 2008) and thyroid cancer (Grimalt et al., 1994), DDE to increases of asthma (Ribas-Fito et al., 2006; Sunyer et al., 2006), alteration of thyroid hormones (Álvarez-Pedrerol et al., 2008) and urinary coproporphyrins (Sunyer et al., 2008), DDT to decreases of cognitive skills (Ribas-Fito et al., 2006; Morales et al., 2008), oncogene mutation (Porta et al., 1999), and polychlorobiphenyls (PCBs) to overweight (Valvi et al., 2012), oncogene mutation (Porta et al., 1999; Howsam et al., 2004), impaired liver metabolism (Sala et al., 2001) and neuropsychological development (Forns et al., 2012; Gascon et al., 2013) among others.

On the other hand, methylmercury (MeHg) targets the nervous system, especially during the children developmental stage (Grandjean et al., 1997; World Health Organization - International Programme on Chemical Safety (WHO-IPCS), 1990). Contamination episodes in Japan showed irreversible neurological damage upon exposure to this compound (Harada, 1995) and neurotoxicity and neurodevelopmental risk among children exposed to low or moderate Hg levels have been investigated (Karagas et al., 2012).

Accordingly, the production and use of OCs and Hg has been restricted and/or banned in many countries. However, these contaminants are still found in the environmental compartments (Arellano et al., 2011; Lamborg et al., 2014), foodstuff (Martí-Cid et al., 2010; Olmedo et al., 2013) and human tissues (Vizcaino et al., 2014; Garí et al., 2013).

Even though there are many sources of human exposure to these pollutants, it is well-known that ingestion is the main route (representing >90% of the total exposure) and, especially, through consumption of fatty food such as fish (Xu et al., 2017 and Vázquez et al., 2015). Until very recently, fish consumption recommendations for vulnerable population groups, such as infants and pregnant women, have focussed on certain big, migratory and oily fish species (EFSA, 2012, 2015; AESA, 2006). However, since Hg is primarily associated with muscle tissue rather than fat, predatory but non-migratory fish species, e.g. lean fish, may also accumulate this compound. This is particularly relevant for carnivorous species feeding at the top of the food chain which increase the concentrations of these pollutants by bioaccumulation. To date, few studies have assessed the potential role of predatory but non-migratory fish species regularly consumed by general and infant populations as sources of organochlorine compounds and Hg.

This concern is high in island populations for being typically high fish consumers, particularly from local markets. These populations are prone to accumulate high levels of these pollutants (Grandjean et al., 1997; Myers et al., 2000; Murata et al., 2002). Previous studies on newborns and preschool children from Mediterranean populations have shown high Hg concentrations in blood and hair (Freire et al., 2010; Garí et al., 2013; Llop et al., 2014). They evidence the need for assessment of the role of different fish species in human exposure to these pollutants.

In this sense, we performed two preliminary studies on the intake of these compounds, one on Hg in Mediterranean lean fish and seafood (Llull et al., 2017) and another on a diet evaluation of the population from Menorca (Junqué et al., 2017). Now, an integrated assessment of lean fish species of commercial value as sources of both Hg and organochlorine compounds in islander populations is reported. The Balearic Islands are adequate reference sites for their location in the western Mediterranean. Thus, most of the fish specimens were obtained from fishermen of these islands. The study has been extended to fish specimens from Tunisia and Egypt. Samples from the Atlantic Ocean, in front of Senegal and Mauritania, two big fishery areas due to upwelling, have also been collected and examined for comparison.

To acquire further data on the possible human health risks of fish consumption, this study examined the weekly intakes of both children and adults and comparison with the Provisional Tolerable Weekly Intakes (PTWIs) established by the European Food and Safety Authority (EFSA, 2012). The study is also aimed to ascertain whether the concentrations of Total Hg are compliant with the maximum values in fishery products established by the European Legislation. Taking into account the role of biotic and abiotic factors in the accumulation of these pollutants in fish (Verhaert et al., 2017 and Storelli et al., 2002), the dependences between pollutant concentrations, fish species and specimen weight have been also evaluated.

The results of the present study need to be considered to design guidelines for decreasing human incorporation of these compounds through diet which is consistent with the EFSA (2015) recommendations.

2. Materials and methods

2.1. Sampling

Between March 2016 and August 2017, 104 commercial fish samples from the Western Mediterranean Sea were collected (Fig. 1). Most of them ($n = 102$) in waters nearby the Balearic Islands (Majorca, $n = 66$; Menorca, $n = 17$; Ibiza, $n = 18$) and the rest of the samples were from Tunisia ($n = 2$) and Egypt ($n = 1$). Additional fish samples ($n = 14$) from the Atlantic Ocean, in front of the Senegal ($n = 4$) and Mauritania ($n = 10$) coasts, were also collected.

Twenty-one lean fish species from the Balearic Islands were selected considering those most consumed by the population (Table 1; SMAP, 2015). They encompassed angler (*Lophius piscatorius*), European hake (*Merluccius merluccius*), common dentex (*Dentex dentex*), common pandora (*Pagellus erythrinus*), dusky grouper (*Epinephelus marginatus*), Mediterranean moray (*Muraena helena*), red scorpionfish (*Scorpaena scrofa*), small-spotted catshark (*Scyliorhinus canicula*), conger (*Conger conger*), Mediterranean rainbow grasse (*Coris julis*), four-spot megrim (*Lepidorhombus boscii*), annular seabream (*Diplodus annularis*), common two-banded seabream (*Diplodus vulgaris*), John dory (*Zeus faber*),