Environmental Research 158 (2017) 16–23

Contents lists available at ScienceDirect

Environmental Research

journal homepage: www.elsevier.com/locate/envres

Mercury concentrations in lean fish from the Western Mediterranean Sea: Dietary exposure and risk assessment in the population of the Balearic Islands

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ARTICLE INFO

Keywords:
Estimated weekly intake
Lean fish
Mercury
Methylmercury
Western Mediterranean Sea

ABSTRACT

The present study reports total mercury (THg) and methylmercury (MeHg) concentrations in 32 different lean fish species from the Western Mediterranean Sea, with a special focus on the Balearic Islands. The concentrations of THg ranged between 0.05 mg/kg ww and 3.1 mg/kg ww (mean 0.41 mg/kg ww). A considerable number of the most frequently fish species consumed by the Spanish population exceed the maximum levels proposed by the European legislation when they originate from the Mediterranean Sea, such as dusky grouper (100% of the examined specimens), common dentex (65%), conger (45%), common sole (38%), hake (26%) and angler (15%), among others. The estimated weekly intakes (EWI) in children (7–12 years of age) and adults from the Spanish population (2.7 μg/kg bw and 2.1 μg/kg bw, respectively) for population only consuming Mediterranean fish were below the provisional tolerable weekly intake (PTWI) of THg established by EFSA in 2012, 4 μg/kg bw. However, the equivalent estimations for methylmercury, involving PTWI of 1.3 μg/kg bw, were two times higher in children and above 50% in adults. For hake, sole, angler and dusky grouper, the most frequently consumed fish, the estimated weekly intakes in both children and adults were below the maximum levels accepted. These intakes correspond to maximum potential estimations because fish from non-Mediterranean origin is often consumed by the Spanish population including the one from the Balearic Islands.

1. Introduction

Mercury is widely distributed throughout the environment as consequence of natural and anthropogenic processes. The most common sources of mercury releases include industrial activities, such as artisanal and small scale gold mining, energy production, and chlor-alkali plants, as well as waste sites (UNEP and WHO, 2008). Anthropogenic effects are responsible for the increases of the global inventory of mercury in the oceans. Mercury emissions to the atmosphere due to mining and fossil fuel burning (Hg_0) are deposited into seawaters after oxidation to Hg^{2+}. There, Hg may undergo bioaccumulation and scavenging by organic-rich particles, be eventually transported from surface to deep waters and reduced back to Hg_0 and to methylmercury (Lamborg et al., 2014).

Methylmercury is a more toxic form than the original metal. This compound targets the nervous system, especially during the children developmental stage (Grandjean et al., 1997; UNEP and WHO, 2008). The major source of methylmercury intake in humans is fish and seafood products (Calatayud et al., 2012; Garí et al., 2013; Perelló et al., 2014; Cano-Sancho et al., 2015a; Obeid et al., 2017). Food sources other than fishery products may also contribute to mercury body burdens, but mainly in the form of inorganic mercury, which is less toxic than the methylated form (Perelló et al., 2014). This compound in fish is bound to tissue protein rather than to fatty deposits. It biomagnifies through the food web and apical predators, which are carnivorous species feeding at the top of the food chain and tend to increase the concentrations.

Until very recently, fish consumption recommendations' to vulnerable population groups, such as infants and pregnant women, have focussed on certain big, migratory and oily fish species (EFSA, 2012, 2015a; AESA, 2006). However, since mercury is associated primarily with muscle tissue rather than to fat deposits, predatory but non-migratory fish species, *e.g.* lean fish, may also accumulate this compound.

Island populations are typically high fish consumers, particularly from local markets. These populations are more prone to accumulate high mercury levels (Grandjean et al., 1997; Myers et al., 2000; Murata

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http://dx.doi.org/10.1016/j.envres.2017.05.033
Received 21 March 2017; Received in revised form 24 May 2017; Accepted 27 May 2017

et al., 2002). Previous studies on newborns and preschool children from Mediterranean populations have shown high mercury concentrations in blood and hair (Freire et al., 2010; Garí et al., 2013; Llop et al., 2014).

To date, few studies have assessed the potential role of predatory but non-migratory fish species regularly consumed by general and infant populations as mercury sources. The present study is devoted to determine the concentrations of total mercury and methylmercury in a great variety of lean fish species from the Western Mediterranean Sea, with a special focus on the Balearic Islands. This study has been extended to fish specimens from the Portmán Bay, a highly polluted area in Cartagena (Murcia, South Spain), Tunisia and Egypt. Samples from the Atlantic Ocean, in front of Senegal and Mauritania, have also been collected and examined for comparison. The study is also aimed to ascertain whether the concentrations of THg are compliant with the maximum values in fishery products established by the European Legislation (EC, 2011).

Possible human health risks of fish consumption are assessed by examination of 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).

2. Materials and methods

2.1. Sampling

Between February 2014 and August 2016, 406 commercial seafood samples from the Western Mediterranean Sea were collected (Fig. 1). Most of them (n = 374) in waters nearby the Balearic Islands (Majorca, n = 173; Menorca, n = 122; Ibiza, n = 79) and the rest of the samples were from Portmán (Múrcia, South of Spain, n = 26), Tunisia (n = 2) and Egypt (n = 4). Additional fish samples from the Atlantic Ocean, in front of the Senegal and Mauritania coasts, were also collected (n = 14).

Thirty-one fish species from the Balearic Islands were selected considering the most consumed by the population (SMAP, 2015). These species were obtained by both commercial and recreational fishing. Samples were collected in action halls and local fish markets. Those from Tunisia, Egypt and the Atlantic Ocean, were obtained from importer facilities in the Balearic Islands.

The specimens from Portmán Bay, *Pagrus pagrus* and *Mullus barbatus*, were collected onboard the R/V Ángeles Alvariño (IEO) in August 2014, in the context of the MIDAS Project.

Information on length, weight, date of sampling and catch location was obtained whenever possible. Some samples were analysed in pools, *e.g.* anchovies (mean specimens in each pool, n = 77), sardines (n = 32), picarel (n = 14), pearly razorfish (n = 6), Atlantic horse mackerel (n = 6), comber and painted comber (n = 5), blackspot seabream (n = 4), black scorpionfish (n = 2) and common sole (n = 2).

2.2. Mercury and methylmercury analyses

Total Mercury (THg) was determined by cold vapor atomic absorption spectrometry, inductively coupled plasma mass spectrometry (ICP-MS, Agilent 7900) and automatic mercury analysis (AMA-254).

Methylmercury was analysed by high performance liquid chromatography coupled to ICP-MS with isotopic dilution (Agilent 7900/Agilent 1200LC).

Mercury and methylmercury concentrations were expressed as Hg (mg/kg) by reference to sample wet weight (ww). Concentration means and ranges were used for data reporting. Statistical analyses and graphs were performed using the R software (R Core Team, 2015).

The limits of quantification (LQs) were 0.1 mg/kg ww in both methods. Concentrations below LQs were assumed to be ½ of the LQs, 0.05 mg/kg ww. Similar figures were obtained from the calculation of these values with a reverse Kaplan-Meier estimator (Gillespie et al., 2010).

2.3. Estimated dietary intakes and threshold values

A first estimate of the dietary weekly THg intakes (mg Hg) through fish consumption were calculated by multiplying the median fish concentrations of all analysed samples (mg/kg ww) by the weekly average fish consumptions of the Spanish population, 46.4 g/day and 71.1 g/day for 7–12 year old children and adults older than 17 years, respectively (AESA, 2006). Comparison of these results with those reported by EFSA for Spain give similar figures, *e.g.* 36.3 g/day and 63.6 g/day for children and adults, respectively (EFSA, 2015b). Estimated Weekly Intakes (EWIs; μg/kg bw) of THg were obtained from the dietary weekly intakes after normalization to the mean body weights of each population group, *e.g.* 34.48 kg for children and 68.48 kg for adults.

The EWIs were compared to the PTWIs of both THg and methylmercury, 4 μg/kg bw and 1.3 μg/kg bw, respectively (EFSA, 2012). The PTWI percentages (%PTWI) were calculated as 100·EWI/PTWI.

In addition to the calculation of EWIs and %PTWIs for total fish consumption, specific calculations for consumption of European hake, angler, common sole and dusky grouper were also performed because these species are the most frequently consumed by the Spanish population (Table 4).

3. Results

The present study reports the concentrations of mercury in 420 fish specimens from 32 species collected at several locations of the Western Mediterranean Sea and the Atlantic coast. Mediterranean morey, red scorpionfish, angler, conger, European hake and dusky grouper were the most studied species, with a total of 27–38 specimens for each

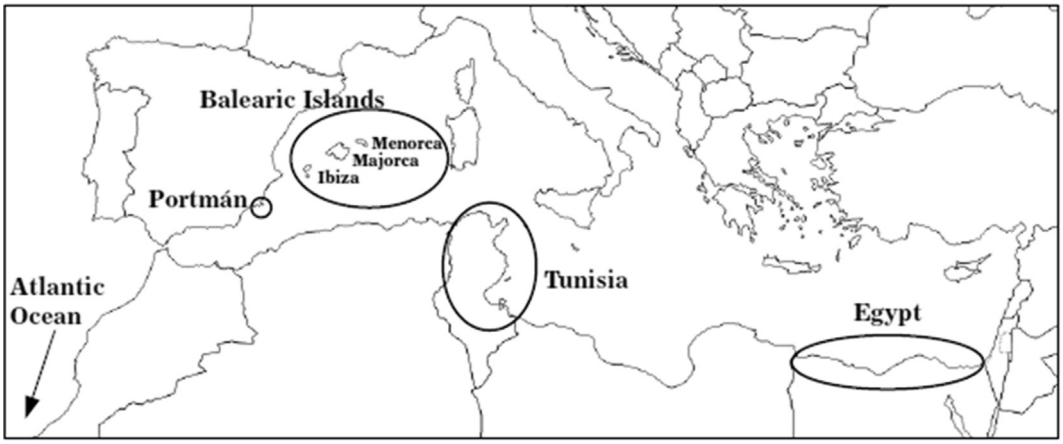

Fig. 1. Map of the fish sampling sites in the Mediterranean Sea and Atlantic Ocean considered in the present study.