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Individual methylmercury intake estimates from local seafood of the Mediterranean Sea, in Italy

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ABSTRACT

A Seafood Frequency Questionnaire (SFQ) broken down in more than 42 items with 8-week coverage was interview-administered to 278 adults aged 19–82 years (167 women, 98 in the reproductive age 19–45 years, and 111 men), resident on the Italian Mediterranean shore and frequent buyer at local fish markets. Methylmercury (MeHg) intake on individual basis was estimated for a selected occurrence equal to the median value + Median Absolute Deviation (MAD) in each seafood species reported (conservative scenario). MeHg occurrence was derived from an extensive seafood database referred to years 2009–2011. Accounting for an average body weight of 62.2 kg, 24.6% of women resulted overexposed with respect to the European Food Safety Authority (EFSA) Tolerable Weekly Intake (TWI) for MeHg of 1.3 μg/kg bw, with a mean of 0.92 μg/kg bw. In the vulnerable group aged 19–45 years, 29.6% exceeded the TWI. Rather than the amount of seafood consumed, the seafood choice appears to be the main determinant of the MeHg intake. Risk awareness was reported in the 49% of SFQs. Uncertainties related to such estimates from questionnaires are discussed, in order to give adequate health recommendations without compromising seafood consumption in the Mediterranean region.

1. Introduction

The Mediterranean Sea is worldwide acknowledged as a mercury (Hg) hotspot, due to the presence of relevant cinnabar settlements on the coastal shores and of the historical related mining and industrial activities discontinued in the last decades, only (UNEP, 2002). Methylmercury (MeHg) originates from inorganic Hg biomethylation in the marine sediments and bioaccumulates in the seafood. Mediterranean wild seafood species from FAO 37 fishing area show total Hg (Hg_{TOT}) and MeHg levels higher than those reported from other FAO zones (Brambilla et al., 2013; FAO and WHO, 2011). As environmental pollutant, MeHg exerts neurotoxic effects on the human central nervous system, particularly for exposures within the first months of the foetal development (Grandjean and Landrigan, 2006). Fish and seafood give the most relevant contribution in the dietary exposure to MeHg in high and frequent consumers and in general population, as well (EFSA, 2012).

Abbreviations: bw, body weight; EC, European Commission; EFSA, European Food Safety Authority; EU, European Union; FAO, Food and Agriculture Organization of the United Nations; FFQ, Food Frequency Questionnaire; Hg, mercury; Hg_{TOT}, total mercury; IQ, intelligence quotient; JECFA, Joint FAO/WHO Expert Committee on Food Additives; LC-PUFA, Long Chained Poly-Unsaturated Fatty Acid; MAD, Median Absolute Deviation; MeHg, methylmercury; MRL, Maximum Residue Limit; RDA, Recommended Daily Allowance; RfD, Reference Dose; SD, standard deviation; SFQ, Seafood Frequency Questionnaire; TWI, Tolerable Weekly Intake; UNEP, United Nations Environment Programme; US EPA, United States of America Environmental Protection Agency; WHO, World Health Organization.

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A Tolerable Weekly Intake (TWI) of 1.3 ng/kg body weight (bw) has been recently proposed for MeHg by the European Food Safety Authority (EFSA, 2012) on the basis of multiple epidemiological studies that observed adverse effects in children as consequence of maternal exposures. Other Agencies, in Japan, USA, New Zealand, Australia, UK, and Canada indicate MeHg Reference Doses (RfD) ranging from 0.10 to 0.47 ng/kg bw per day, based on multiple endpoints (Mahaffey et al., 2011). As complementary risk management measures, Maximum Residue Limits (MRLs) for Hg_{TOT} in seafood have been issued: for instance, the European Commission (EC) fixed in the 0.5–1.0 mg/kg range the MRL for low/top predatory fish (EC, 2006); moreover, the release of dietary recommendations to most vulnerable groups, on geo-referenced basis, has been suggested to European Union National Authorities by EFSA, as already done by other Agencies (Ser and Watanabe, 2012; Ström et al., 2011; US EPA, 2004).

In biomonitoring studies, a strong correlation was found between Hg_{TOT} concentration in human hair and fish consumption (Diez et al., 2008). Furthermore, according to Bellanger et al. (2013) human hair mercury concentrations are higher in Southern Europe and lower in Eastern Europe: a relevant part of susceptible individuals such as women in the reproductive age may be overexposed via seafood intake with potential consequences on pre-natal/perinatal brain development and a potential IQ reduction in the progeny. The association between prenatal mercury exposure and infant neurodevelopment in coastal areas of four Mediterranean Countries has been recently demonstrated by Valent et al. (2013). However, inconsistencies have been reported among MeHg daily intakes when computed starting from Food Frequency Questionnaires (FFQs) accounting for inventoried Hg_{TOT}/MeHg occurrence in seafood (external dose), or when derived from the one-compartment toxicokinetic model from human biomonitoring data of MeHg in blood and hair (internal dose) (Gosselin et al., 2006; JECFA, 2004). Noisel et al. (2011) found that on a group of 23 fishermen, the MeHg intake estimated with FFQs exceeded the US EPA RfD of 0.1 μg/kg bw per day in 21 subjects, against a smaller proportion (6/23) when computed on internal dose. Similarly, an overestimation of the intakes derived from FFQs compared to those recovered from the biomonitoring data has been reported by Sirot et al. (2008), Ström et al. (2011) and Valent et al. (2013).

Apart from the intrinsic uncertainties of the toxicokinetic modelling, as those generated from differences in MeHg concentrations between cord and maternal blood and/or from deviations from the steady-state, a possible source of bias in FFQ estimates may rely on the attribution of the proper MeHg occurrence to each seafood species, and on the seafood consumption habits when not broken down into species-specific details (EFSA, 2012; Miklavĉiĉ et al., 2013; Pouzaud et al., 2010). An overestimation of the intake of a contaminant through seafood consumption by means of FFQs might also stem from the fact that people generally tend to over report their life habits or activities that are known to be "healthy" or to have a positive effect (e.g., consumption of fish) (Männistö et al., 1996). Anyway, FFQ based estimates do not involve human blood or hair collection and analysis, with related ethical and financial issues, thus are a less expensive and less time-consuming methodology and should be ranked among the most cost-effective screening tools able to highlight a potential MeHg overexposure in individuals. In the present work we illustrate the outcomes of the MeHg intake assessment from a Seafood Frequency Questionnaire (SFQ) based on 45 seafood items administered to 278 adults living on the Italian Mediterranean shore. The aim is both to identify those habits able to reduce the risks associated to local seafood consumption thus maximizing the correlated benefits, and to identify, with a presumed reduced rate of false positivity, potentially overexposed individuals.

2. Materials and methods

2.1. Food consumption

A specific questionnaire targeted to record Seafood Frequency Consumption (SFQ) of Mediterranean coastal population has been developed for the evaluation of MeHg intake. The SFQ was adapted from guidelines of UNEP (2008) and of the US EPA-related website Fish-facts.org (2009). The following information was selected as the most relevant: age and gender; habitual residence; number of seafood servings per week and their average amount (over the last 8 weeks). Options to indicate portion of: (*a*) less than 100 g, (*b*) from 100 to 200 g, (*c*) from 200 to 300 g, and (*d*) more than 300 g were suggested; the orientation to consider as reference a 100 g fillet serving derived from a 350 g whole weight farmed sea bream of market size was also given. A positive list of 45 fish and seafood species ranked among wild, farmed, and preserved/canned/frozen categories, and open to other choices, was provided. For selected choices, it was asked to indicate which one were "often eaten" or "sometimes eaten" over an 8-week time. The detailed amount (in grams) and frequency of each seafood species was not requested. The questionnaires were interviewer-administered to allow the answer to potential open questions (i.e., proper identification of seafood species also according to local names, report of seafood species consumed other than those listed, estimates of the portion size when not framed among the suggested ones). The SFQ was pre-tested to ensure that the meanings of the seafood names, the portion-size descriptors and the other information requested were clear to the subjects in order to reduce potential ambiguity in the responses (Cade et al., 2004). The consumers' awareness about potential MeHg risk along with Long Chained Poly-Unsaturated Fatty Acid (LC-PUFA) benefits deriving from seafood consumption was also framed in the SFQ. Lastly, interviewed individuals might indicate their preference about the origin of the fish and seafood they used to buy among local, national, wild, farmed or preserved. The questionnaire was proposed to adults aged 18 and over at fish market sites during the spring/summer of 2013. Fig. 1 indicates the location of the coastal fish markets considered for the interviews, along with the codes of the FAO 37 fishing sub-areas.

Fish and seafood consumptions expressed on a body weight basis in the female and male groups (not normally distributed) were tested for significant differences, with the hypothesis that gender or age might influence consumption habits in terms of cumulative consumption (Mann–Whitney *U* test for non-parametric statistics). Statistical significance was defined as $p \leqslant 0.05$.

2.2. Mercury/methylmercury occurrence

Occurrence data referred to each species reported in the questionnaires were derived from a national database on Mediterranean seafood encompassing 69 different species (Brambilla et al., 2013). In some cases, MeHg concentrations were derived from Hg_{TOT} levels through the use of a correlation equation. For those species not inventoried in the database, such as canned tuna and eel, or for fish caught or farmed in FAO zones other than 37 (as in the case of salmon), data were recovered from the international literature, with the priority given to data referred to samples drawn on the national market (di Domenico et al., 2011; EFSA, 2012; Ginsberg and Toal, 2009; Storelli et al., 2010). In the case of a full lack of data (i.e. fish and shellfish soup) an average of the occurrence values of the species listed in the recipe was adopted.

As to MeHg occurrence for the single seafood species, different values were considered according to the following scenarios: (A) the median value (P50) reported for each one of the selected seafood species (for farmed species: mean value); (B) the median