Regulatory Toxicology and Pharmacology 65 (2013) 269–277

Contents lists available at SciVerse ScienceDirect

Regulatory Toxicology and Pharmacology

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

Mercury occurrence in Italian seafood from the Mediterranean Sea and possible intake scenarios of the Italian coastal population

Gianfranco Brambilla [a,*], Maria Cesarina Abete [b], Giovanni Binato [c], Eugenio Chiaravalle [d], Maurizio Cossu [e], Elena Dellatte [a], Roberto Miniero [a], Roberta Orletti [f], Pierluigi Piras [g], Alessandra Roncarati [h], Alessandro Ubaldi [i], Giannina Chessa [e]

[a] *Istituto Superiore di Sanità – Environment Dept., Rome, Italy*
[b] *Istituto Zooprofilattico Sperimentale del Piemonte, Liguria e Valle d'Aosta, Turin, Italy*
[c] *Istituto Zooprofilattico Sperimentale delle Venezie, Padua, Italy*
[d] *Istituto Zooprofilattico Sperimentale della Puglia e Basilicata, Foggia, Italy*
[e] *Istituto Zooprofilattico Sperimentale della Sardegna, Sassari, Italy*
[f] *Istituto Zooprofilattico Sperimentale dell'Umbria e delle Marche, Ancona, Italy*
[g] *Azienda Sanitaria of Carbonia (Sardinia), Italy*
[h] *Scuola di Scienze Mediche Veterinarie, Università degli Studi di Camerino, Italy*
[i] *Istituto Zooprofilattico Sperimentale del Lazio e della Toscana, Rome, Italy*

ARTICLE INFO

Article history:
Received 26 October 2012
Available online 10 January 2013

Keywords:
Mercury
Methyl mercury
Mediterranean seafood
Maximum residue limits
Tolerable intakes
Advisories

ABSTRACT

Mercury (Hg) settlements in the Mediterranean Sea determine a potential toxicological relevance of seafood intakes for coastal populations. To assess this possibility, fish, molluscs, and crustaceans of commercial size of 69 different species were sampled and analyzed for total mercury (Hg_{TOT}) from georeferenced areas and evaluated for their compliance with the European Union Maximum Residue Limits of 0.5 and 1.0 mg/kg wet weight (ww). Accounting for the weekly estimated seafood intake in the Italian coastal population (mean 469–626 g/person/week) it was then possible to recover threshold contamination values in seafood. Under a Tolerable Weekly Intake of 1.3 μg/kg/bw/week, a threshold seafood contamination <0.10 mg/kg ww has been derived for sensitive groups. A suitable algorithm based on the parallel MeHg and Hg_{TOT} analysis on the most consumed species, helped to refine the uncertainties related to the conservative assumption in seafood all the Hg_{TOT} present is in form of MeHg. This work aims to improve the link between the risk management and risk assessment strategies, with the identification of those fish and seafood species, that, when regularly consumed, could determine or prevent potential Hg_{TOT}/MeHg overintakes in sensitive groups.

1. Introduction

It is generally acknowledged that the Mediterranean Sea represents a geological hot spot for mercury (Hg): about half the world's Hg resources are located in this area, with abundant deposits in sites facing the Italian seas (Covelli et al., 2012; Grassi and Netti, 2000; Martinez-Frias et al., 1998; Moldenhauer et al., 2008). Because of the natural presence of mercury and the development of related industrial and anthropogenic activities since ancient times, the release of mercury into the marine environment is recognized as the main determinant of the bio-magnification of its predominant organic form – methyl mercury (MeHg) – in Mediterranean seafood from different trophic levels (TLs) (UNEP, 2002; FAO/WHO, 2011). A previous evaluation by the (US) National Research Council established an intake limit of 0.7 μg/kg body weight per week (NRC, 2000). Very recently, the European Food Safety

Abbreviations: AAS, Atomic Absorption Spectrophotometry; bw, body weight; DDA, Direct Automatic Analyzer; dw, dry weight; EFSA, European Food Safety Authority; EU, European Union; EC, European Commission; FAO, Food and Agriculture Organisation of the United Nations; Hg, Mercury; Hg_{TOT}, Total Mercury; ICP-MS, Inductively Coupled Plasma Mass Spectrometry; LCn3PUFAs, long chained omega-3 fatty acids; LOQ, Limit of Quantification; MAD, Median Absolute Dispersion; MeHg, methyl mercury; MRL, Maximum Residue Limit; NRC, National Research Council; PUFA, poly-unsaturated fatty acids; RDA, Recommended Daily Allowance; RfD, Reference Dose; Sd, standard deviation; Se, standard error; TL, Trophic Level; TWI, Tolerable Weekly Intake; UNEP, United Nations Environment Programmel; US-EPA, United States of America, Environmental Protection Agency; US-FDA, United States of America, Food and Drug Administration; TDA, Thermal Decomposition Amalgamation; WHO, World Health Organisation; ww, wet weight.

* Corresponding author. Address: Istituto Superiore di Sanità, Dipartimento Ambiente e connessa Prevenzione Primaria, Viale Regina Elena, 299 I-00161 Rome, Italy. Fax: +39 6 49902836.
E-mail address: gianfranco.brambilla@iss.it (G. Brambilla).

0273-2300/$ - see front matter © 2013 Elsevier Inc. All rights reserved.
http://dx.doi.org/10.1016/j.yrtph.2012.12.009

Authority revised its former opinion on mercury and methylmercury in food: A Tolerable Weekly Intake (TWI) of 1.3 μg/kg body weight (bw) for MeHg is now proposed lower than the former one of 1.6 (EFSA, 2012a), 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, have proposed MeHg Reference Doses (RfD) ranging from 0.1 to 0.47 μg/kg/day, based on multiple endpoints (Mahaffey et al., 2011). Recent papers have focused on biota sampled from Mediterranean industrial hot spots in Southern Italy, such as the gulf of Taranto (Di Leo et al., 2010; Spada et al., 2012) and the Augusta basin (Gabellini et al., 2011). The findings indicate that the presence of MeHg in local seafood of a low trophic level (TL) may represent a potential health concern for the population. A detailed biomonitoring study of fishermen and their families in a Northern Africa Mediterranean coastal village (Elhamri et al., 2007) found a correlation between the number of seafood meals based on anchovies and pilchards and methyl-mercury levels in hair. The same correlation with fish consumption was described by Dıez et al. (2008) in a study carried out on 237 subjects recruited in the urban population of Neaples (I); Freire et al. (2010) reported a possible association between fish consumption and cognitive development in a Spanish cohort of prescholar children. When extrapolated to dietary intakes, the Hg levels in hair indicated potential overexposure with respect to the oral reference dose. Plenty of data are available on Hg_{TOT} and MeHg levels for Italian marine species; however, some of the investigations were restricted to few species of seafood, or limited geo-referenced areas (Perugini et al., 2009; Storelli et al., 2002, 2003, 2005a; Copat et al., 2012). In some cases, the reported contamination data were biased by being expressed only on a dry basis (Ipolyi et al., 2004). In other studies, the sampling was affected by a strict environmental approach: this is the case for species that are not of commercial size and/or interest (Storelli et al., 2005b) or for fish as bioindicators of the environmental status in known highly polluted sites (Corsi et al., 2002). In this study, we aim to give a more comprehensive picture of the occurrence of Hg_{TOT} and MeHg in seafood recovered from fishing and aquaculture activities in areas of Italy presumed to be not impacted. Together with a better definition of MeHg levels on a geo-referenced basis, the final aim is to give a first ranking of the contributions of commercial fish and seafood species to MeHg exposure in the coastal populations, as already done in other countries (Groth, 2010; Lambert et al., 2012). The information will be then translated into intake advice for sensitive groups, without compromising the nutritional benefits of seafood-based diets (FAO/WHO, 2011).

2. Materials and methods

2.1. Sampling

Fish and seafood of market size belonging to 69 different seafood species were sampled in the period 2009–2011 from the following national fishing areas belonging to FAO Area 37: North Tyrrhenian (NT), Central Tyrrhenian (CT), South Tyrrhenian (ST), Ionian (IS), North Adriatic (NA), Central Adriatic (CA) and South Adriatic (SA) seas, as illustrated in Fig. 1. Registered offshore aquaculture farms (bivalve molluscs, seabass and seabream) were located in the corresponding fishing areas. Sampling activity was framed within both national monitoring plans for residue analysis and the AQUAFISH and FISR (2010) nationally/regionally-funded projects. Seafood of the more prevalent commercial size in the catch was considered for the sampling of edible parts. Each sample of the farmed seabass and seabream species was represented by 20–30 different individuals of commercial size, 350–400 g ww. Fish TLs for the Mediterranean Sea were taken from Stergiou and Karpouzi (2002) or from the specific database consulted (http://www.fishbase.org/search.php).

2.2. Analysis

Total Mercury determination was performed using different analytical techniques, including Inductively Coupled Plasma Mass Spectrometry (ICP-MS), Atomic Absorption Spectrophotometry (AAS) and Direct Automatic Analyzer based on Thermal Decomposition Amalgamation coupled with Atomic Absorption Spectrophotometry (TDA/AAS), according to the different equipment available in the official labs. In ICP-MS, Hg_{TOT} determination was performed by Agilent 7500CE ICP-MS. 209Bi was used as internal standard element to compensate matrix effect and signal drift. For each batch of samples processed, a method blank was carried throughout the entire sample preparation and analytical process. The calibration curve was verified at the end of each analytical batch using continuing calibration verification (CCV) at or near midrange. In AAS, the determination was performed by Perkin Elmer Flow Injection Mercury System FIMS 400. In TDA/AAS, a DMA-80 Direct Mercury Analyzer (Milestone Inc.) and an AMA254 Automatic Mercury Analyzer (FKV, Altec) were used. The quality control of the data produced was verified and controlled using Certified Reference Materials FAPAS T07139 – Canned fish (Certified Value: 0.539 mg/kg dw), BCR 278R – Mussel Tissue (Certified Value: 0,196 mg/kg dw), DORM-2 – Dogfish Muscle (Certified Value: 4.64 mg/kg dw), DORM-3 – Fish Protein (Certified Value: 0.382 mg/kg dw). All labs were intercalibrated through successful participation in internationally organised proficiency tests, with LOQs between 0.010 and 0.001 mg/kg ww, for Hg_{TOT}, according to the different analytical equipment. Within the FISR project activities (FISR, 2010), a panel of 36 fish market samples from 10 wild species caught from not impacted fishing areas (anchovies, $N = 6$; pilchards, $N = 2$; red mullets, $N = 6$; Atlantic mackerel, $N = 2$; bullet tuna, $N = 3$; European hake, $N = 6$; Atlantic horse mackerel, $N = 3$; Atlantic bonito, $N = 4$; bluefin tuna, $N = 2$; Swordfish, $N = 2$) were also analyzed for MeHg, using GC–MS (SIM) according to Ipolyi et al. (2004), adding Ethyl-Mercury (EtHg) as Internal Standard. The accredited method with a LOQ of 0.010 mg/kg ww, was under quality control using BCR-463 – tuna fish (certified value 3.04 mg/kg dw).

3. Theory/calculation

3.1. Conversion of HG_{TOT} into MeHg

The conversion of HG_{TOT} into MeHg for wild fish was performed using the following correlation equation taken from the parallel analysis of these contaminants $[MeHg] = -0.166\ (\pm 0.0827) + 1.14\ (\pm 0.0515) \log[Hg_{TOT}]$ $r = 0.967$, $r^2 = 93.5$, as described above (FISR, 2010). For seafood species, this ratio was derived from the scientific literature and set at 60% for octopus and crustaceans of the same trophic level (Raimundo et al., 2010) and at 40% for Mediterranean mussels (Di Leo et al., 2010). The MeHg values were taken directly from the specific analysis of farmed seabass and seabream of market size (350 g ww).

3.2. Fish and seafood consumption

The median, mean and P.95 of seafood consumption expressed as mg/kg bw per day were taken from the National Comprehensive Food Consumption Database (INRAN, 2012a), referred to seafood consumers only. The details concerning the most frequently consumed marine fish species are available on the same database