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Assessment of human health risk associated with methylmercury in the imported fish marketed in the Caribbean

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ABSTRACT

The decline in marine and freshwaters catches in recent years in Colombia has led to a change in dietary habits, with an increase in the purchase and consumption of imported fish. This is of particular concern as fish are sometimes caught in mercury-contaminated waters, and are subsequently sold canned or uncanned. In addition, canned tuna has received little attention as it is widely assumed that concentrations are low. In this study, total mercury (THg) and methylmercury (MeHg) concentrations were evaluated in three imported fish species marketed in Colombia, *Prochilodus lineatus, Prochilodus reticulatus,* and *Pangasianodon hypophthalmus,* plus four brands of canned tuna and one of sardines. One brand of tuna showed the highest mean concentrations of THg (0.543 ± 0.237 µg/g, wet weight, ww) and MeHg (0.518 ± 0.337 µg/g ww), while concentrations in *P. hypophthalmus* were approximately 30 times lower (≈0.02 µg/g ww). The estimated weekly intake (EWI) in children was above the provisional tolerable weekly intake (PTWI) of MeHg established by the Joint FAO/World Health Organization (WHO) Expert Committee on Food Additives (JECFA) in 2007, 1.6 µg/kg body weight (bw) per week, for all the canned tuna brands. Values for adults were below PTWI, whereas for women of childbearing age, values were above PTWI only for brand D of canned tuna. The estimate of the potential risk indicated that MeHg levels in canned tuna can generate negative effects in vulnerable groups, while the EWI of fresh fish did not pose a threat to the general population. Therefore, establishing strategies to address the high consumption of canned tuna, and continuous monitoring to control commercial food, are recommended to decrease Hg exposure.

1. Introduction

Fish have been recognized as an integral component of the diet, providing a healthy source of energy, high-quality protein, vitamins and a broad range of other important nutrients (Pieniak et al., 2010). In addition, fish are an important source of omega-3 polyunsaturated fatty acids that are known to reduce the risk of coronary heart disease and contribute to the normal neurological development of children (Mozaffarian and Wu, 2011; Swanson et al., 2012).

Despite the potential benefits of fish intake on health, chemical contaminants contained in these products have become a topic of concern, especially for regular fish consumers (Hightower and Moore, 2003; Burger et al., 2008; von Stackelberg et al., 2017). Trace metal contaminants are a risk to public health as they can accumulate in aquatic organisms (Dórea, 2008; Fuentes-Gandara et al., 2016), and diet is the main route of exposure for the general population (Kim and Lee, 2010; Bosch et al., 2015).

Of the trace metals, mercury (Hg) causes the most concern due to its wide distribution, persistence and mobility in the environment, bioaccumulation, toxicity and trophic transfer to human beings, mainly in its organic forms such as methylmercury (MeHg) (Díez et al., 2007; Marrugo-Negrete et al., 2008a; Liao et al., 2016; Salazar-Camacho et al., 2017). MeHg is the most toxic form of Hg, as it is easily bioaccumulated and biomagnified in the food chain (Carrasco et al., 2011; Gewurtz et al., 2011). Since a high percentage of Hg exists as MeHg in aquatic biota (Lacerda et al., 1994; Morel et al., 1998; Carrasco et al., 2009), the main source of human exposure to MeHg is through fish consumption (Rice et al., 2014), and is able to become a neurotoxic potential agent (UNEP, 2010).

In Colombia, the *per capita* consumption of fishery products is 4.73 kg/year, which is low compared with countries such as Spain (38 kg/year), Japan (54 kg/year), and the average of consumption in

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Latin America (18 kg/year) (MADR, 2013). The National Nutrition Situation Survey (ENSIN, 2010) concludes that 61% of Colombians consume fish or shellfish once a month, and only 27% consume them weekly.

According to the Organization for Economic Co-operation and Development (OECD), production from capture fisheries has decreased significantly (about 50%) over the last ten years, mainly due to overexploitation of the principally harvested species (OECD, 2016).

The decline in marine and freshwaters catches in recent years in Colombia has led to an increase in the purchase of imported fish. Total imports of fish products increased almost sevenfold over the period 2004–13. Imports are mainly from Vietnam, Chile, Ecuador, Senegal, China, South Africa and, since 2013, the USA (FAO, 2015). Recently, the National Administrative Department of Statistics of Colombia (DANE) reported that during 2016, around 96,000 Mg of fishery products arrived in the country, representing an increase of 2% compared to 2015. The most imported products are the basa fish from Vietnam, bocachico from Argentina and Venezuela and canned fish (e.g. tuna and sardine) from Ecuador, Chile, USA, China and Vietnam. The most notable trend is the importation of basa fish and canned tuna, products that are in high demand as they are competitive, low import cost aquaculture products sourced in Asia with a low market price.

The objectives of our study were to determine the THg and MeHg content in imported fish products, i.e fresh and canned fish, that are the most common and popular in Colombia, and subsequently estimate the weekly tolerable intake of Hg with reference to the recommendations from the international agencies.

2. Materials and methods

2.1. Collection and analysis of samples

Three fish species, and canned tuna and sardine samples, were purchased in local stores and supermarkets located in the centre and north of Barranquilla (northern Colombia) from September 2016 to March 2017. The samples were selected based on their presence in fisheries throughout the year and because they are the most common imported fish products consumed. The samples included bocachico argentino (*Prochilodus lineatus*), bocachico venezolano (*Prochilodus reticulatus*), basa fillet (*Pangasianodon hypophthalmus*) from Vietnam and four different brands of canned tuna (CT) from China (A), Ecuador (B and C) and Colombia (D), and one brand of canned sardines (CS) from Ecuador (E).

For each fish species and canned fish sample, 20 specimens and 20 cans of each brand were obtained, totalling 160 samples. Special care was taken to maintain the physical appearance and condition of the fresh fish samples. For each brand, we collected ten cans of tuna shredded in vegetable oil, ten cans of tuna in water and 20 cans of sardines in tomato sauce.

Fresh fish samples were individually packaged in labelled polyethylene bags, refrigerated and transported to the laboratory for examination. Similarly, canned samples were transported unopened to prevent contamination of samples. Dorsal muscle samples (3 cm, weighing approximately 10 g) were dissected, and immediately frozen and stored at −20 °C until THg and MeHg were determined. The canned fish samples were selected according to the brands of tuna and sardines that are certified by The National Institute of Food and Drug Monitoring (INVIMA), with existing health registration. For each canned fish sample, cans were opened and the water, vegetable oil or tomato sauce was drained. The meat was homogenised using a food blender with a stainless-steel blade.

For THg analysis, samples (0.5 g wet weight, ww) were digested with H_2SO_4/HNO_3, (2:1) for 2 h at 100 °C (Sadiq et al., 1991; Marrugo-Negrete et al., 2008a), and Hg concentration was determined using cold vapour atomic absorption spectrophotometry (CV-AAS) (Thermo Scientific model iCE series 3500). Quality control was performed using CRM IAEA-407 (THg $0.222 \pm 0.024\ \mu g\ g^{-1}$ dry weight, dw), and the recovery percentage for THg was $97.0 \pm 0.2\%$. The detection limit for THg was $0.014\ \mu g\ g^{-1}$ dw, calculated as three times the standard deviation (SD) of the blank (n = 7).

MeHg was analysed using gas chromatography with electron-capture detection (Marrugo-Negrete et al., 2008b) using a MeHg calibration curve (methylmercury chloride; Sigma-Aldrich, St Louis, MO, USA). The fish samples (0.2–0.3 g) were digested with HCl, NaBr and toluene. After centrifugation and several extraction steps with cysteine, an aliquot of the organic phase was injected into the gas chromatograph (Model PE Autosystem XL). The detection limit for MeHg was $0.009\ \mu g\ g^{-1}$, calculated from the SD of ten blanks. Analytical quality control of the method was performed in triplicate using dogfish muscle CRM DORM-2 ($4.47 \pm 0.32\ \mu g\ g^{-1}$), and the recovery percentage was $105 \pm 5\%$.

2.2. Human health risk assessment

To estimate the potential risk, the study was based on data from a survey on food consumption in different locations in Barranquilla. The surveys were carried out in three local fish stores located in the centre of the city of Barranquilla, adjacent to the public market, and also in two national chain supermarkets located in the north. Individuals (n = 150) from the general population that were shopping in the local stores and supermarkets were asked for their frequency of fish consumption, which species they consumed most frequently and the number of cans and fresh fish per day. The gender, age, educational level, social class and educational status of each subject were also recorded. 56% of the surveyed population was male, and around 6% of the women surveyed were pregnant (8–24 weeks). In addition, 58% of the population that bought fish in the local stores were at least at secondary level in school, while in supermarkets, 63% of the buyers were professionals. Finally, the people surveyed were a clear example of the inhabitants of the Caribbean region as they shared certain cultural traits, particularly the tradition of fish consumption.

Potential risk was evaluated from the estimated weekly intake (EWI) of Hg per kg of bodyweight of the person exposed (μg/kg body weight (bw)/week) using the equation described by UNEP (2008):

$$\mathrm{EWI} = \frac{\mathrm{WI} \times [\mathrm{Hg}]}{\mathrm{BW}} \tag{1}$$

where WI is the weekly intake of fish (kg/week), [Hg] is the concentration of Hg in fish ($\mu g\,/kg^{-1}$) and BW is the bodyweight of the person in kg. In addition, the permissible concentration of MeHg in fish for human consumption was determined. The concentration of MeHg that the consumed fish species should contain to avoid exceeding the EWI concentration established by FAO/WHO (2007) (e.g. the PTWI) was estimated by considering the EWI and the amount of fish eaten per week. Thus, MeHg content was calculated by the following equation:

$$[\mathrm{MeHg}]_{\mathrm{permissible}} = \frac{[\mathrm{MeHg}] \times \mathrm{PTWI}}{\mathrm{EWI}} \tag{2}$$

where [MeHg] is the concentration of MeHg in fish ($\mu g\,/kg^{-1}$) and PTWI is the provisional tolerable weekly intake, a reference value established by the Joint FAO/World Health Organization (WHO) Expert Committee on Food Additives (FAO/WHO, 2017). In 2006, the JECFA (FAO/WHO, 2007) established a PTWI for MeHg at 1.6 μg/kg bw/week for sensitive groups such as women of childbearing age (WCA) and young children (CHD). A higher ingestion rate (3.2 μg/kg bw/week) was allowed for adults from the general population (AGP). The maximum amount of fish that can be consumed weekly per person (MFW), without harmful effects to health, was calculated using the following equation:

$$\mathrm{MFW} = \frac{\mathrm{PTWI} \times \mathrm{WI}}{\mathrm{EWI}} \tag{3}$$