Science of the Total Environment 627 (2018) 322–331

Contents lists available at ScienceDirect

Science of the Total Environment

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

Arsenic, cadmium and lead in fresh and processed tuna marketed in Galicia (NW Spain): Risk assessment of dietary exposure

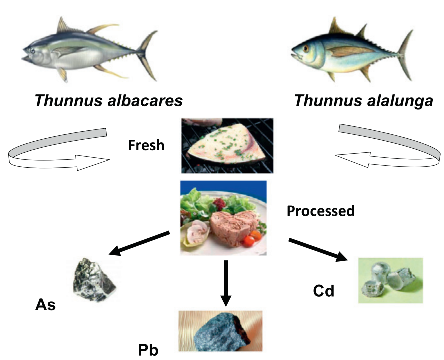
Ricardo Núñez, M. Ángeles García, Julián Alonso, M. Julia Melgar *

Department of Toxicology, Faculty of Veterinary. University of Santiago de Compostela, 27002 Lugo, Spain

HIGHLIGHTS

- In tuna, both fresh and processed, the lead content was negligible.
- Total As and Cd levels on processed tuna were lower than fresh tuna.
- The order of As and Cd concentrations was: olive oil > natural > pickled sauce
- These observed results do not pose a risk to the consumer (EU Legislation).
- THQs showed that there is no cancer risk for the tuna consuming population.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 3 October 2017
Received in revised form 24 January 2018
Accepted 24 January 2018
Available online xxxx

Editor: Yolanda Picó

Keywords:
Arsenic
Cadmium
Lead
Tuna
Risk assessment

ABSTRACT

Currently, metal bioaccumulation in fish is increasing and is a cause of concern due to toxicity. Total arsenic, cadmium and lead concentrations in fresh and processed tuna (110 samples) marketed in Galicia (NW Spain) were determined by ICP-MS spectrometry. The average concentrations of As and Cd, 3.78 and 0.024 mg kg^{-1} w.w., respectively, in fresh tuna were statistically significantly higher than those in processed tuna ($p < 0.001$). The contents in processed tuna were 0.295–7.85 mg kg^{-1} for As and ND-0.045 mg kg^{-1} for Cd. The Pb content was negligible in both types of tuna. In canned tuna, decreasing As and Cd concentrations were observed in different preparation-packaging media: olive oil > natural > pickled sauce. Of the two species studied in canned tuna, *Thunnus alalunga* showed statistically significant higher levels both for As 1.28 mg kg^{-1} ($p < 0.001$) and Pb 0.013 mg kg^{-1} ($p = 0.0496$) than *Thunnus albacares*. No samples surpassed the limits set by the EU for Cd and Pb. The limit for As in fish has not been established, but the arsenic contents in fresh tuna reported here are important, as they are among the highest reported in the literature. Considering public health in children and adults with respect to the investigated metals, the estimated daily intakes (EDIs) did not exceed the tolerable intakes. No chronic systemic risk was found since all the target hazard quotients (THQs-TTHQs) were far below 1 (critical value), and the carcinogenic risk (CR) for As did not exceed the acceptable value of 10^{-5}. Thus, tuna consumption in the Galician diet does not pose a risk for different population groups in terms of these studied metals/metalloids.

1. Introduction

Fish is considered essential to a healthy diet because it is a low-fat, high quality protein aliment, providing vitamins and a wide range of

* Corresponding author.
E-mail address: mj.melgar@usc.es (M.J. Melgar).

https://doi.org/10.1016/j.scitotenv.2018.01.253
0048-9697/© 2018 Published by Elsevier B.V.

other important nutrients (Andayesh et al., 2015; García et al., 2016; Olmedo et al., 2013; Storelli et al., 2010). Nutritionally, a relevant feature that distinguishes fish is the high content of omega-3 polyunsaturated fatty acids (PUFAs), which protect against coronary heart disease and contribute to satisfactory neurodevelopment in children (Olmedo et al., 2013; Ruelas-Inzunza et al., 2012). However, fish and seafood have been identified as the foodstuffs with the highest contribution to the total dietary uptake of chemical contaminants (among them Cd, As and Pb) in the general population (Bae et al., 2017; Bosch et al., 2016; Cano-Sancho et al., 2015; Hosseini et al., 2015; Massadeh et al., 2017; Perelló et al., 2014; Raber et al., 2012). These toxic elements cause variable pathological effects both when the daily intake exceeds permissible limits and under chronic low dose exposure; this circumstance is especially concerning for sensitive consumer groups (Andayesh et al., 2015; Varol et al., 2017).

Heavy metals are considered the most marked forms of pollution in aquatic environments, and since heavy metals are not biodegradable, they can accumulate at the top of the food chain (Çulha et al., 2016; Mol, 2011a, Milatou et al., 2015; Ugarte et al., 2012;Yabanli et al., 2016). Therefore, predator fish such as tuna can be used for biomonitoring marine contamination (Emami Khansari et al., 2005; Raimundo et al., 2017).

The natural input of metals in the marine environment due to natural sources (volcanic activity and erosion of the crust's earth) has been minimized by the severe impact of anthropogenic activities (industrial, fuel emissions, mining, agrochemicals, etc.) (Araújo and Cedeño-Macías, 2016).

Arsenic is a clear example of this pollution (EFSA, 2009), it is transported into the aquatic environment through atmospheric deposition and riverine input (Wu et al., 2015) and is another dangerous toxic element that may accumulate in marine organisms that grow in contaminated water (Çulha et al., 2016; Ugarte et al., 2012; Yabanli et al., 2016).

Exposure to dietary inorganic arsenic (iAs) causes a wide range of adverse effects on the human body. Currently, cancer risk assessments are being conducted for fish consumption due to their metal/metalloid contents, especially iAs (Copat et al., 2013; Çulha et al., 2016; Yabanli et al., 2016). According to Cano-Lamadrid et al. (2016) iAs has been classified as a Group 1 carcinogen by the International Agency for Research on Cancer (IARC). This classification is due to the induction of certain types of cancers, including skin, lung, and bladder (EFSA, 2009). In addition, iAs is considered the most toxic form for humans.

Cadmium, which is mainly derived from anthropogenic emissions of fuel combustion and its subsequent atmospheric deposition, is a major source of dissolved Cd in the oceans and bioaccumulates in fish (Araújo and Cedeño-Macías, 2016; Bosch et al., 2016; Quan et al., 2016).

Cadmium toxicity includes renal failure, as kidney is the primary target organ, as well as hypertension, neurological and digestive disorders, cancers, mutations, and skeletal weakness (Andayesh et al., 2015; EFSA, 2011; Torres et al., 2016).

Finally, with respect to lead, Torres et al. (2016) showed the evolution of human impact on the seawater environment; its pollution increased during the industrial age and rose significantly when Pb was added to gasoline fuel for vehicles. Regulations adopted to reduce the allowable gasoline Pb content have significantly contributed to a reduction in Pb concentrations in the marine environment (Bosch et al., 2016; Witt et al., 2010).

Lead toxic effects predominate in the central nervous system, especially in neurodevelopment periods. In addition, multiple organic systems (cardiovascular, digestive, immune, urinary, etc.) can be harmed (EFSA, 2010; Iwegbue, 2015).

In the Spanish population, seafood is the major contributor of Cd to the diet (Chiocchetti et al., 2017). Shellfish tends to accumulate more Cd than fish (Yusà et al., 2008). Thus, the Agencia Española de Consumo, Seguridad Alimentaria y Nutrición (AECOSAN, 2011) has urged consumers to reduce their intake of shellfish to limit dietary exposure to Cd. Furthermore, Galicia leads in consumption of these food items in Spain. Thus, seafood is an engine for the Galician economy and is also essential for its traditional cuisine.

In this context, it is interesting to consider tuna and specially processed tuna, which among fishery products are doubtless the most widely and most frequently consumed (Ruelas-Inzunza et al., 2012; Storelli et al., 2010). These foods merit study due to several factors of technical processing and storage, which might alter the concentrations of trace elements in preserved fish (Ganjavi et al., 2010; Hosseini et al., 2015).

The main objective of the present study was to perform human health risk assessment for three elements (As, Cd, and Pb) ingested via consumption of tuna. For this purpose, the concentrations of arsenic, cadmium and lead in different tuna presentations (fresh and processed) were determined and compared with those obtained in previous studies; likewise, EDIs and THQs for the three cited elements and the CRs for iAs were calculated to assess the potential health risk for Galician consumers based on their intake.

2. Materials and methods

2.1. Sampling

A total of 110 samples of fresh (raw) and commercial (10) and technologically processed (packaged: canned and glass) tuna fish products (100) were collected in 2013–2014. The origin of fresh samples (*Thunus albacares*) was large supermarkets and the central market of Lugo (Galicia – NW Spain), and all samples were stored in a freezer at −32 °C before the analysis, which was performed within 12 weeks. Processed tuna samples (*T. albacares* and *T. alalunga*) were purchased in some of the major supermarkets in the city. To select the three most popular brands, a random sampling was performed, using as the inclusion criteria: (a) frequent consumption in Spain (MERCASA, 2016) and (b) two type of packing tuna with different preparations: canned [natural (30), olive oil (30) and pickle sauce (30)], and glass [olive oil (10)]. The exclusion criteria (a) did not consider batch but (b) non-fresh samples or expired canned samples. The samples were transferred into clean plastic bags, coded with their brand, type of packaging and medium preparation, and frozen at −20 °C until analysis (García et al., 2016).

2.2. Sample preparation

To obtain three drained homogeneous samples of each biological sample for analysis, a portion of muscle tissue was taken from fresh tuna, washed with HNO_3 and rinsed with distilled and deionized water (Milli-Q® water). For the packaged samples, we drained the liquid medium for 24 h (García et al., 2016).

Sample mineralization consisted of a wet digestion with a suprapur acid mixture [conc. HCl and conc. HNO_3 (3:1)] using a variant of the EPA 3052 method (EPA, 1996) and according to the process described by García et al. (2016). Finally, the solutions were transferred to polyethylene bottles and refrigerated until analysis by ICP-MS. In parallel, blanks and certified reference material (*Reference Material 8414 Bovine Muscle Powder Agriculture Canada from National Institute of Standards and Technology*) samples were processed similarly to verify the accuracy and precision of this method.

2.3. Total arsenic, cadmium, and lead analyses and quality control method

Samples were analysed using a Varian 820 ICP-MS spectrometer. All samples were analysed in triplicate according to Melgar et al. (2014). In this work, to calculate the estimated intake for iAs from tuna, a 3% iAs content in tAs (Copat et al., 2013; Varol et al., 2017) was assumed.