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Assessment of human health hazards associated with the dietary exposure to organic and inorganic contaminants through the consumption of fishery products in Spain

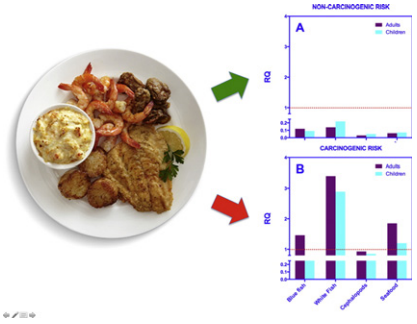
Ángel Rodríguez-Hernández [a,b,c], María Camacho [a,b,c], Luis A. Henríquez-Hernández [a,b,c], Luis D. Boada [a,b,c], Norberto Ruiz-Suárez [a,b,c], Pilar F. Valerón [a,b,c], Maira Almeida González [a,b,c], Annalisa Zaccaroni [d], Manuel Zumbado [a,b,c], Octavio P. Luzardo [a,b,c,*]

[a] Toxicology Unit, Research Institute of Biomedical and Health Sciences (IUIBS), University of Las Palmas de Gran Canaria, Plaza Dr. Pasteur s/n, 35016 Las Palmas de Gran Canaria, Spain
[b] Instituto Canario de Investigación del Cáncer (ICIC), Plaza Dr. Pasteur s/n, 35016 Las Palmas de Gran Canaria, Spain
[c] Spanish Biomedical Research Centre in Physiopathology of Obesity and Nutrition (CIBEROBn), Plaza Dr. Pasteur s/n, 35016 Las Palmas de Gran Canaria, Spain
[d] Large Pelagic Vertebrate Group, Veterinary Faculty, University of Bologna, Viale Vespucci 2, Cesenatico (FC) 47042, Italy

HIGHLIGHTS

- The daily intake of persistent pollutants through fish consumption is estimated
- Dietary intake of individual pollutants did not exceed the Tolerable Daily Intakes.
- Consumption of fishery products does not pose risk of acute toxicity for the Spaniards
- These results may be useful for the design of proper risk communication campaigns.

GRAPHICAL ABSTRACT



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ABSTRACT

In this work we have evaluated the potential carcinogenic and acutely toxic risks associated to the exposure to highly prevalent organic and inorganic contaminants through the consumption of fishery products by the Spanish population. The concentrations of 8 organochlorine pesticides (OCPs), 18 polychlorinated biphenils (PCBs), 7 polycyclic aromatic hydrocarbons (expressed as benzo[*a*]pyrene toxic equivalents ($B[a]P_{eq}$)), and three inorganic toxic elements [arsenic (As), cadmium (Cd), and mercury (Hg)] were determined in 93 samples of the most consumed species of white fish, blue fish, cephalopods and seafood species, which were acquired directly in markets and supermarkets in the Canary Islands, Spain. The chemical concentration data were combined with the pattern of consumption of these foodstuffs in order to calculate the daily intake of these contaminants, and on this basis the risk quotients for carcinogenicity and acute toxicity were determined for Spanish adults and children. Our results showed that the daily intake of OCPs, PCBs and $B[a]P_{eq}$, which is associated to blue fish consumption was the highest within the fish group. The estimated intake of pollutants can be considered low or very low for the individual contaminants, when compared to reference values, except in the case of HCB and As. All the estimated intakes were below the reported Tolerable Daily Intakes. Considering the additive effects of multiple contaminants, the risk of acute toxic effects can also be considered as low or very low. However, our results reflect

* Corresponding author at: Toxicology Unit, Department of Clinical Sciences, Universidad de Las Palmas de Gran Canaria, Plaza Dr. Pasteur s/n, 35016 Las Palmas de Gran Canaria, Spain.
E-mail address: octavio.perez@ulpgc.es (O.P. Luzardo).

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that the current consumption of white fish in adults and children, and also the blue fish in the case of adults, poses a moderate carcinogenic risk to Spanish consumers, mainly related to their concentrations of As. The conclusions of this research may be useful for the design of appropriate risk communication campaigns.

1. Introduction

Organic and inorganic contaminants, such as legacy pesticides, polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), mercury (Hg), arsenic (As), or cadmium (Cd) are commonly targeted contaminants for research and in monitoring programs. In the last decades, efforts have been made to raise knowledge about the adverse effects on humans and animals, worldwide distribution pattern, and new methods are developed to analyze these compounds in very different matrices and various environmental media (Luzardo et al., 2013b; Sharma et al., 2014). Thus, numerous studies have revealed that these toxic compounds, individually and in combination, may contribute to the development of severe health problems such as cancer, immune suppression or genotoxic effects in humans, even with long-term low-dose exposure (Bergman et al., 2012; Jarvis et al., 2014; WHO, 2003), and many of them have demonstrated endocrine disrupting effects in both animals and humans (Camacho et al., 2014; Kortenkamp et al., 2011). In fact, the use of organochlorine pesticides (OCPs) and PCBs is now banned in most developed countries, but they are still widespread in the environment (Almeida-González et al., 2012; Kakuschke et al., 2010; Luzardo et al., 2014).

Although there are different routes of exposure for humans to these pollutants, it has been established that ingestion of food contributes more than 90% of total human exposure, and that the fatty fraction of food represents the main entrance to the human body (Darnerud et al., 2006; Vazquez et al., 2015). In the last decade, studies on human dietary exposure to persistent pollutants have been carried out in various countries over the world and it has been reported that the dietary intakes vary considerably between countries. The dietary intakes are mainly influenced by the specific dietary habits of each country (Domingo and Bocio, 2007; Storelli et al., 2011). The daily intake of contaminants needs to be calculated on the basis of the typical food basket consumed in the country obtained from surveys on consumers. The dietary exposure to a wide range of persistent organic and inorganic pollutants of Spanish consumers has been investigated by several authors in the past years for different food groups, such as milk and cheese (Almeida-González et al., 2012; Luzardo et al., 2012), eggs (Luzardo et al., 2013a), yogurt (Rodríguez-Hernández et al., 2015c), meat and processed meat (Rodríguez-Hernández et al., 2015a; Rodríguez-Hernández et al., 2015b), and seafood (Bocio et al., 2007; Domingo and Bocio, 2007; Falcó et al., 2006). Also several basket market studies have been performed in Spain including the major food groups (Bocio and Domingo, 2005; Bocio et al., 2005; Falco et al., 2003; Llobet et al., 2003a; Llobet et al., 2003b; Llobet et al., 2003c), and even the consumption of foods of animal origin has been investigated as a determinant of contamination by OCPs and PCBs (Boada et al., 2014). However, to date only few studies have estimated the carcinogenic risk associated to the exposure to contaminants associated to certain food groups in the Spanish population (Rodríguez-Hernández et al., 2015a; Rodríguez-Hernández et al., 2015b), and to our knowledge none has been developed for the seafood group.

Fish is an important supplier of high quality nutrients such as omega 3 fatty acids, which have been proven reduce the risk of stroke, lower blood pressure and improve arterial integrity, and even decrease the risks of certain cancers (Kris-Etherton et al., 2002). However, fish is also one of the main contributors of the total dietary intake of environmental pollutants (Bocio et al., 2005; Falco et al., 2003; Llobet et al., 2003b; Llobet et al., 2003c). Thus, on the one hand, the health benefits of sea foodstuff consumption have been proven but on the other hand there also exist an increasing concern of the potential risk arising from exposure to toxic pollutants through the intake of fishery products. Because of the growing public concern about the health effects of food borne diseases related to chemical pollutants, there exists the need carrying out studies on particular food groups (such as fish), based on their current pattern of consumption by a given population. In some guidance documents for environmental risk assessment, a reference point from toxicity testing is divided by a default assessment factor and the result compared to the predicted exposure by computing their ratio, which is known as the risk quotient (RQ) (EFSA, 2015; USEPA, 2000). It has been proposed that RQ is a good method to estimate the risk to carcinogenic and acutely toxic effects associated to food contaminants in a population and that is useful to establish exposure limits to those chemicals.

As fish is a staple food of the Spanish diet, with an average consumption of 26.4 kg/person/year (MAGRAMA, 2015) we have designed this study in which we assess the toxic potential of the current pattern of consumption of this food group by the Spanish population. We have acquired seafood samples directly at points of sale to the consumer, and the sampling was designed to follow the Spanish consumers' preferences. We have assessed two types of health risks associated with the consumption of seafood: the carcinogenic risk, and the acute toxicity potential. In this research we have calculated the RQs considering multiple contaminants in fishery products for both carcinogenic and acutely toxic effects, and on this basis we calculated the number of healthy meals per month for a safe consumption in the Spanish population. Obviously, the results of this study need to be considered in the context of the proven health benefits of the nutrients of fish, but may serve for the design of appropriate risk communication campaigns in order to reduce the consumption of certain types of seafood with the aim of optimizing the risk-to-benefit balance.

2. Material and methods

2.1. Sampling

We selected for this study the most consumed species of seafood: fish (white fish and blue fish), cephalopods, crustaceans and bivalve mollusks in Spain, according to the data available (AECOSAN, 2006; AECOSAN, 2011). A total of 93 samples from the main commercial species (MAGRAMA, 2015; Martín Cerdeño, 2010) were randomly acquired from multinational retailers settled in the Canary Islands (Spain) between September and November of 2014. The samples purchased were transported to the Laboratory of Toxicology of the University of Las Palmas de Gran Canaria (ULPGC) and processed immediately upon arrival at the laboratory. We processed and analyzed only the edible parts of seafood (muscle + skin, depending on how the species are consumed). Each sample was constituted by five individual subsamples for each species of fish and cephalopods (fillets, small fishes, or parts of octopus and squids), and six subsamples of each species of crustaceans and mollusks to give pooled samples (using a stainless steel domestic food processor). Thus, 5 to 6 of these composites were used to obtain the data of each species. After that, all samples were frozen at −80 °C (until analysis).

The species of white fish included in this study were: wreckfish (*Polyprion americanus*), megrim (*Stephanolepis hispidus*), sole (*Solea vulgaris*), seabass (*Dicentrarchus labrax*), hake (*Merluccius merluccius*), toothed sparus (*Dentex dentex*), parrot fish (*Sparisoma cretense*), gilt head fish (*Sparus aurata*) and iridescent shark (*Pangasius hypophthalmus*). The selected species of blue fish were: tuna (*Thunnus thynnus*), salmon (*Salmo salar*), sardine (*Sardina pilchardus*), and trout (*Salmo trutta*). Additionally, we included those most consumed species of other seafood