

Persistent organic pollutants in edible fish: a human and environmental health problem

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Abstract

Many persistent organic pollutants (POPs) pose serious health hazards to both the environment and human. Among these, polychlorobiphenyls (PCBs) are probable human carcinogens and can also pose non-cancer health hazards to intellectual functions and the nervous, immune and reproductive systems. The risks and hazards associated with POP residues in tissues are a function of the dioxin-like compound toxicity and an individual's exposure. Fish consumption might become a serious problem because of bioaccumulation as revealed in many studies worldwide.

We report data concerning the accumulation and pattern of hexachlorobenzene (HCB), *p,p'*-DDE and PCBs in edible tissues of commercial fish species (bluefin tuna *Thunnus thynnus*, swordfish *Xiphias gladius*, Atlantic mackerel *Scomber scombrus*) from Italian Seas and of the Antarctic toothfish *Dissostichus mawsoni* from the Ross Sea (Antarctica). The species analyzed are part of the human diet. 2,3,7,8-TCDD toxic equivalents (TEQs) and tolerable weekly intake (TWI) were also calculated to evaluate the toxic hazard for the population that include them in their diet.

Gaschromatography revealed 0.16 ± 0.24 and 0.4 ± 0.2 ng/g wet wt of HCB in Antarctic toothfish and bluefin tuna, respectively. *p,p'*-DDE concentrations were 38 ± 29 and 31 ± 38 ng/g wet wt in swordfish and bluefin tuna muscle, respectively, and 0.66 ± 0.57 ng/g wet wt in the Antarctic toothfish. PCBs showed higher concentrations and they were 89 ± 82 , 80 ± 86 and 5.2 ± 4.0 ng/g wet wt in the muscle of swordfish, bluefin tuna and Antarctic toothfish, respectively. In Mediterranean fish, the most abundant congeners were the most persistent PCB numbers 153, 138, 180, 118 and 170, which accounted for 51% and 47% of the total PCB residue in tuna fish and swordfish, respectively, and 18% in the Antarctic toothfish. TEQs were 1.97 and 4.65 pg/g wet wt in bluefin tuna muscle and gonads, respectively, and 0.11 pg/g wet wt in Antarctic toothfish. The TEQ weekly intake was calculated and values ranged 197–465 pg_{TEQ}/week when consuming 100 g of Mediterranean fish and therefore lower than the recommended TWI. Those values were higher (788–1860 pg_{TEQ}/week) than the recommended TWI, if 400 g of fish/week was consumed (with the exception of Antarctic fish).

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1. Introduction

Persistent organic pollutants (POPs) are carbon-based compounds and mixtures that include industrial chemicals such as polychlorobiphenyls, pesticides such as DDT, and

combustion by-products including dioxins. They are primarily products and by-products of industrial processes of relatively recent origin. POPs released to the environment can travel through air and water to regions far distant from their primary sources [1]. They can bioaccumulate, bioconcentrate and biomagnify because they are hydrophobic, persistent, toxic and have affinity for fat. The most toxic POPs are polychlorodibenzodioxins (PCDDs) and polychlorodibenzofurans (PCDFs). 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (2,3,7,8-TCDD) is

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known to be the most toxic compound for organisms [2]. The Environmental Protection Agency (EPA) has confirmed that dioxin is a cancer hazard to people. The International Agency for Research on Cancer (IARC) announced on February 14, 1997, that the most potent dioxin, the 2,3,7,8-TCDD, is now considered a Class 1 carcinogen, meaning a “known human carcinogen” [3]. Polychlorobiphenyls (PCBs) are considered dioxin-like compounds since the two benzene rings are able to rotate around a carbon–carbon bond and the molecule can thus assume a planar structure resembling dioxins. Depending on the number of chlorine atoms that are bonded to the *ortho* positions, a PCB congener may spend a shorter or longer time in that configuration, and this affects its toxicity [4]. Among the 209 PCBs, a hundred are used in the technical mixtures and can be detected in all the environmental compartments. The congeners 3,3',4,4' (PCB77), 3,3',4,4',5 (PCB126) and 3,3',4,4',5,5' (PCB169) are considered to be the most toxic among PCBs [4,5].

Once in the organisms, the model proposed for the mechanism of action of the 2,3,7,8-TCDD and related toxic halogenated hydrocarbons provides the following steps: (a) the initial formation of a cytosolic receptor complex, followed by (b) an activation step; (c) the formation of nuclear receptor complexes, and (d) their interaction with specific nuclear-binding site. This interaction contributes to an increase or induction of a 2,3,7,8-TCDD inducible gene, such as the CYP1A1. There is a decrease in receptor binding affinities with the decreasing lateral substitutions [4]. Some toxic effects due to POPs are: cancer, reproductive and developmental problems (for example, low birth weight, hormone alterations, lower IQ, emotional problems), alterations of the immune system (such as decrease in ability to fight cancer and infections), endocrine disruption (affecting the thyroid, the sex hormones), central nervous system defects, effects on the nervous system, liver damage, skin and eye disease and death [6,7].

Human exposure occurs to mixtures of compounds. It is widely known that dioxin-like compounds cause toxic effects in similar ways [4,8]. The EPA and other authorities use Toxicity Equivalency Factors (TEFs) to compare the potential toxicity of each of the individual dioxin-like compounds to the relative toxicity of TCDD. With such factors, the toxicity of a mixture can be expressed in terms of its Toxicity Equivalents or 2,3,7,8-TCDD toxic equivalents (TEQs), which is the amount of TCDD it would take to equal the combined toxic effect of all the dioxin-like compounds found in that mixture [4,8]. The TEQs of each chemical are then summed in order to obtain a single TCDD TEQ value for the complex mixtures of dioxin-like compounds found in the sample. EPA adopted the International approach in 1989 [9]. This procedure assigns I-TEFs for a total of 17 compounds. The World Health Organ-

ization (WHO) has established another approach for TEFs, that includes a total of 17 compounds [2]. The WHO approach for developing TEF values differs from the International approach for three compounds, two of which would not alone significantly change any TCDD TEQ value.

Due to the fact that dioxin-like compounds have been found in most food and drinking water, a Tolerable Daily Intake (TDI) has been defined in order to assess the eventual risk for humans through diet. TDI is an estimated quantity of a chemical contaminant in food or water that can be ingested daily over a lifetime without posing a significant risk to health. Currently, the TDI of TCDD and related compounds, suggested by the WHO, is 1 to 4 pg/kg of body weight per day [2,10] and the EPA risk specific dose is 0.01 pg/kg of body weight per day [9].

We analyzed edible tissues of three species of Mediterranean fish (bluefin tuna, swordfish, Atlantic mackerel), to evaluate the possible chemical intake through the diet. Tuna and swordfish are both top predators which feed on mackerel [11]. These species are very interesting because of their peculiar ecology and their widespread use in the human diet. We report on the residue levels of HCB, *p,p'*-DDE and PCBs in the fish edible tissues and the TEQ and TDI values in order to assess the possible risk to humans through the diet. POP concentrations were detected in the muscle and in the liver; results for bluefin tuna male gonads were also reported (this tissue is used to prepare the “lattume” in Sicily). Its use is moderated and rare and it cannot be considered as a diet component. We compare results with a commercial fish species, the Antarctic toothfish from open waters remote from industrial pollutants. This species is managed by the Commission for the Conservation of Marine Living Resources of Antarctica (CCAMLR); its catching is strictly regulated by the Catch Documentation Scheme (CDS) for toothfish, 4 November 1999 (CCAMLR Conservation Measure 170/XVIII).

2. Materials and methods

2.1. Collection of samples

Details of sampling and biometric data are reported in Table 1 and Fig. 1. The bluefin tuna (*Thunnus thynnus*, Linnaeus 1758) were collected off the Ionian coast of Sicily, Italy; samples of muscle ($n=5$), liver ($n=7$) and male gonads ($n=6$) were excised from specimens caught in early November 1999. Tuna can reach more than 600 kg of weight and can be 15 years old; it feeds primarily on small schooling fishes (Atlantic herring, Atlantic mackerel, sand lance, squid [11]). The specimens sampled weighed 86 ± 80 kg. Bluefin tuna is pelagic (depth range: 0–100 m) and prefers subtropical climates (70°N–40°S); it

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