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PCDD/Fs, DL-PCBs and NDL-PCBs in European catfish from a northern Italian lake: the contribution of an alien species to human exposure



S. Squadrone*, M. Prearo, R. Nespoli, T. Scanzio, M.C. Abete

Istituto Zooprofilattico Sperimentale del Piemonte, Liguria e Valle D'Aosta, via Bologna 148, 10154 Torino, Italy

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ABSTRACT

PCDDs/Fs, DL-PCBs and NDL-PCBs are environmentally persistent substances that have been associated with adverse effects on human health. Humans are mainly exposed to these pollutants through ingestion of contaminated fish and fishery products; the consumption of fatty fish such as European catfish can contribute considerably to the intake of dioxins and PCBs.

Samples of fish muscle of the top-of-the-food-chain predator *Silurus glanis* from the northern Italian Lake Varese were analyzed to detect the levels of 17 PCDD/F and 18 PCB congeners. All samples presented detectable levels of PCDD/Fs and PCBs, with concentrations ranging from 0.001 to 1.310 pg g $^{-1}$ wet weight (w.w.) for Σ PCDD/Fs, 0.031 to 21.000 pg g $^{-1}$ w. w. for Σ DL-PCBs, and 0.397 to 117.910 ng g $^{-1}$ w. w. for Σ NDL-PCBs. One sample exceeded the maximum levels of 6.5 pg g $^{-1}$ w. w. for the sum of PCDD/Fs and DL-PCBs set by European regulations for fish (1259/2011 EU), while other samples exceeded the action levels of 0.75 pg g $^{-1}$ w. w. for the sum of PCDD/Fs, and 2 pg g $^{-1}$ w. w. for the sum of DL-PCBs (277/2012 EU).

Consuming contaminated catfish may pose a risk for human health, especially for the subpopulation traditionally accustomed to eating this fish, as well as sensitive individuals, such as children and pregnant women.

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1. Introduction

Polychlorinated dibenzo-p-dioxins and polychlorinated dibenzofurans (PCDDs/Fs) are persistent and lipophilic organic compounds widely distributed in the environment. PCDD/F refers to a family of 210 congeners, 17 of which are considered to be very highly toxic for living organisms. PCDD/Fs can be formed as unintentional by-products in a number of chemical processes as well as in almost every combustion process. Polychlorobiphenyls (PCBs) are persistent organic pollutants of great concern because of their high toxicity and tendency to bio-accumulate through the food chain. PCBs include 209 congeners, which exhibit different degrees and patterns of chlorination (WHO, 1993). During the 1930s and for approximately 50 years, these chemicals were commercially produced in different industrialized countries, mainly for use in dielectric fluids, organic diluents, plasticizers, adhesives, and flame retardants. Although banned in the 1970s and 1980s in the USA and Europe, respectively, PCBs are still present in the environment and can be traced in animal tissues. According to their toxicological properties, PCBs are usually identified by having a

dioxin-like (DL-PCB) or a non-dioxin-like (NDL-PCB) activity. The DL-PCB group comprises 12 congeners characterized by non- or mono-ortho chlorosubstitution. These congeners exert their toxicity primarily by binding the aryl hydrocarbon receptor (AhR), in a similar manner to the PCDDs and PCDFs (van den Berg et al., 2006). The NDL-PCB group includes the remaining congeners, analytically predominant in environmental matrices and animal tissues (EFSA, 2010).

PCDD/Fs and PCBs are considered to pose a substantial risk for human health due to their toxicological properties, associated with damage to the liver and the immune system, in addition to exerting adverse effects on the reproductive system and promoting cancer (Gascon et al., 2013; Leng et al., 2014; Taylor et al., 2013).

Food can be a major source of human dietary exposure to these toxins, especially fish and fish by-products (EFSA, 2012), as these substances are lipophilic and preferentially separate from water and adsorb to sediments at the bottoms of lakes and rivers. Aquatic organisms then ingest and accumulate these persistent organic pollutants, resulting in bio-concentration upwards in the food chain.

The Commission Regulation (EU) No 1259/2011 (in enforcement since January 1st 2012) set the following maximum levels (MLs): 3.5 pg World Health Organization toxic equivalent (WHO

^{*} Corresponding author.

E-mail address: stefania.squadrone@izsto.it (S. Squadrone).

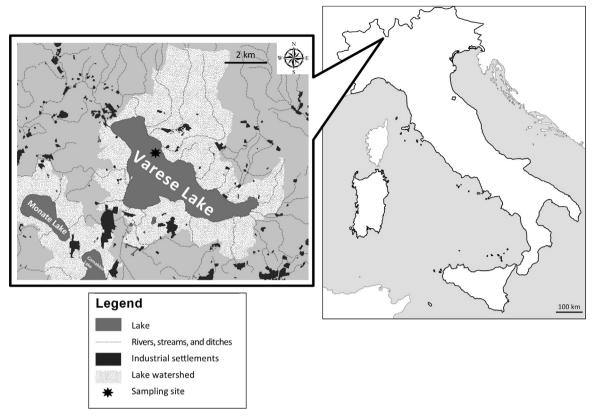


Fig. 1. Study area.

TEQ) g $^{-1}$ (expressed on wet weight) for the sum of PCDD/Fs; 6.5 pg WHO TEQ g $^{-1}$ (expressed on wet weight) for the sum of PCDD/Fs and DL-PCBs, and 125 ng g $^{-1}$ (expressed on wet weight) for the sum of the six NDL-PCBs. Moreover, the Commission Regulation (EU) No 277/2012 set the action level (AL) for PCDD/Fs to 0.75 pg WHO-TEQ g $^{-1}$ w.w. and for DL-PCBs to 2.0 pg WHO-TEQ g $^{-1}$ w.w. in muscle meat of wild-caught freshwater fish utilized as animal feed, and Recommendation (EU) N°663/2014 set the action level (AL) for PCDD/Fs to 1.50 pg WHO-TEQ g $^{-1}$ w.w., and for DL-PCBs to 2.5 pg WHO-TEQ g $^{-1}$ w.w. in muscle meat of farmed fish. These action levels provide a tool for competent authorities and operators to highlight the cases in which it is appropriate to identify a source of contamination and to take measures for its reduction or elimination.

European catfish (Silurus glanis) is a top predator of the foodchain in the freshwater ecosystem, and is popular among European anglers and, for this reason, it has been introduced to many European countries, including France, the Netherlands, Spain, and the UK (Elvira, 2001). In Italy, this species has also received an increasing interest for commercial purposes, as is the case for the Eastern European market, where its flesh is greatly appreciated. In a previous study (Squadrone et al., 2013), we estimated the concentrations of NDL-PCBs in Silurus glanis in four rivers covering the area of the Po river basin (Northern Italy), and we found levels that exceeded the MLs set by the 1259/2011 EU Regulation. Moreover, the presence of the European catfish has been reported in Northern and Central Italian Lakes, and it is causing a progressive depopulation of the lake fish fauna. There have been no previous investigations into the PCDD/F and PCB levels in fish from Lake Varese, a North West Italian Lake. In recent years, the autochthonous fish population of this lake has been impoverished by the expansion of the European catfish that, due to its abundance, has started to be commercialized as a "low-cost" fish in North Italian fish markets. In fact, catfish flesh is highly appreciated by the Austrian, Hungarian, Bulgarian and Romanian populations and in some Northern Italian markets, it has even become necessary to import catfish from abroad due to its high demand.

Fish consumption is one of the primary routes of exposure to PCDD/Fs and PCBs (EFSA, 2012), and wild-caught European catfish are a significant dietary component in several northern European subpopulations that reside in Italy. Moreover, due to its low cost, catfish is even affordable for families with low-income, despite not being culturally accustomed to consuming it.

It is, therefore, essential to characterize the human exposure to PCDD/Fs and PCBs associated with *Silurus glanis* consumption. The objective of this study was to investigate the occurrence of these persistent contaminants in *Silurus glanis*, from Lake Varese, in order to establish the contamination levels in a Northern Italian lake and to assess human dietary exposure through the consumption of this non-native fish species.

2. Materials and methods

2.1. Study species

The European catfish (*S. glanis*), also known as wels catfish, is one of the largest European freshwater fish. This species is native to Eastern Europe and Western Asia and is abundant in the Danube and Volga river basins. The European catfish inhabits the lower reaches of large rivers and muddy lakes, and tends to prey on fish smaller than would be expected for its size and mouth gape (Adámek et al., 1999; Wysujack and Mehner, 2005). *S. glanis* is a bottom-dwelling nocturnal predator, which feeds in the entire water column. Due to its highly developed sense of taste and smell, European catfish has no problems in orientation during complete darkness (Pohlmann et al., 2001). Fry and juveniles are benthic, feeding on a wide variety of invertebrates and fish, while

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