NATIONAL STUDY ON POLYCHLORINATED BIPHENYL LEVELS IN BLOOD OF FRENCH FRESHWATER FISH EATERS: FIRST EXPOSURE RESULTS

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Introduction

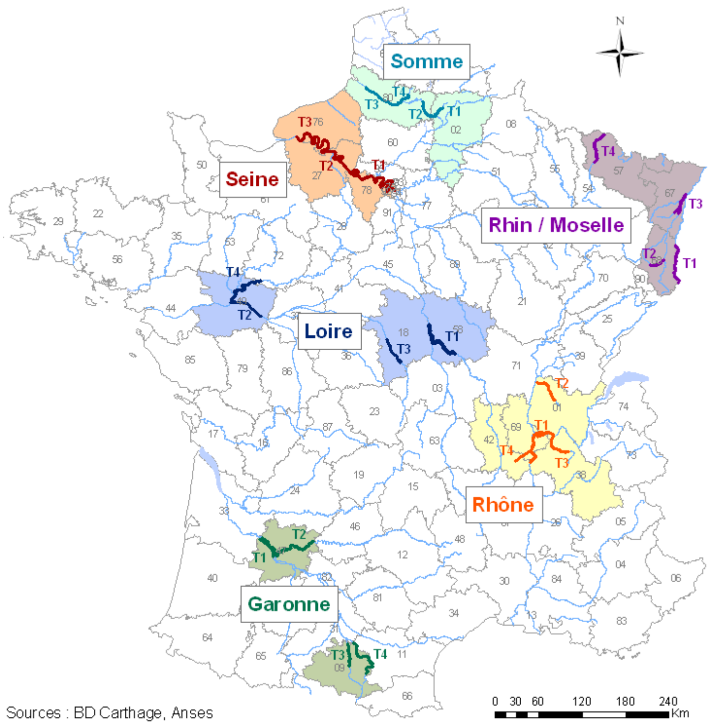


Figure 1 : Rivers and parts of rivers of the study

Polychlorinated biphenyls (PCBs) are persistent and bioaccumulative environmental contaminants that have been banned in France since 1987. These chemicals are associated with a wide range of health effects. Due to past uses, these lipophilic substances are still widely spread in the environment and in foodstuffs. In general population, the diet represents 90% of PCB exposure. Fish consumption especially appears as a major contributor to this total food intake, leading to an exceeded tolerable daily intake for a specific fringe of French consumers[1]. In December 2006, the European Commission set maximum levels for certain contaminants in foodstuffs, in particular for dioxins (PCDD/Fs) and PCBs like dioxins (DL-PCBs) in fishes[2]. Since this regulation, freshwater fishes exceeding maximum regulatory levels have been fished in several rivers in France. In 2008, the French agency for food, environmental and occupational health safety (Anses) and the French Institute for Public Health Surveillance (InVS) set up a national study to describethe freshwater fish consumption and estimate PCB and dioxin blood levels of members of recreational fishermen households. The aim of this article is to describe freshwater fish consumption, contamination and assess the PCB and dioxin exposure from more or less contaminated rivers.

Materials and methods

The study population lived near six rivers or parts of rivers (Figure 1). A total of 900 km are expected to be representative of the different contamination levels of PCBs encountered in the sediments: two rivers with high levels of PCBs (the Seine and the Somme), two rivers with medium levels (the Rhone and the Rhine / the Moselle) and two rivers with low levels (the Loire and the Garonne). In these 6 areas, 3 or 4 sections were examined, a total of 23 parts of rivers.

Fish contamination data used in this study were obtained from the national freshwater fish sampling plan of ONEMA (French National Agency for Water and Aquatic Environments). This plan focused specifically on freshwater fish species. For this plan, the Anses proposed in February 2008 a methodology to select sampling stations to investigate, mainly based on sediment contamination data. A total of 47 stations, located in study areas, were examined. Target compounds were the 17 PCDD/Fs, the 12 DL-PCBs and the 7 marker PCBs (PCB-28, 52, 101, 118, 138, 153, 180), analysis were made by the LABERCA. Only edible parts (i.e. filets) were

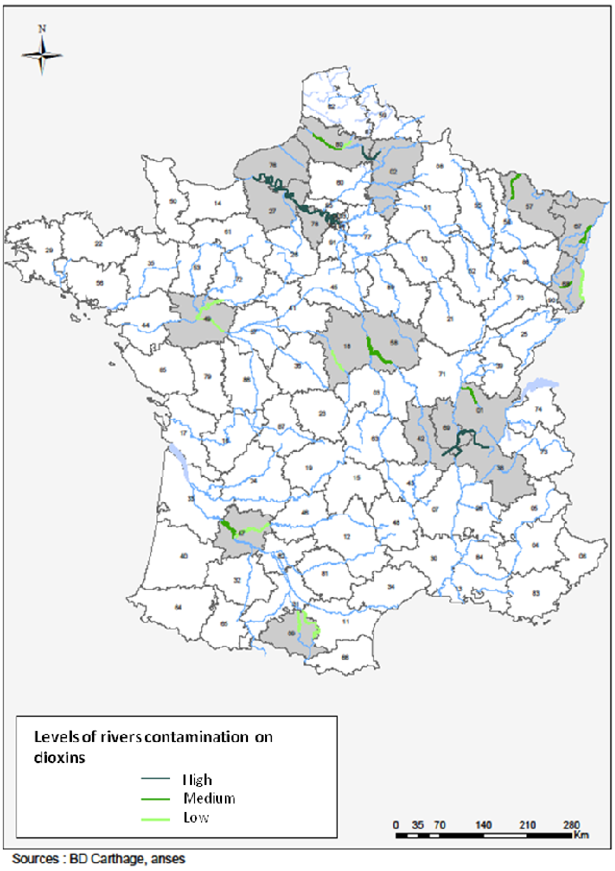


Figure 2 : Rivers clustering according to dioxin contamination levels

analyzed. Globally, the sample preparation method was the one commonly used in the field, details from which can be found elsewhere[3]. Briefly, fish filets were freezedried and powdered. Extraction of fat and target compounds was carried out using Pressurized Liquid Extraction. Purification and fractionation included three chromatographic steps with successively silica, Florisil and carbon columns. Gas chromatography was used for analysis coupled to high-resolution mass spectrometry, with isotopic dilution before quantification. The fish contamination data completed sediment contamination data previously used to select the 6 areas of the study. It appears that fish contamination levels are different between parts of same river. Considering the few enrollments of fishermen household members on river sections (at all 606 persons) and levels of fish contamination, sections were grouped on the basis of fish contamination data to increase robustness of analysis and make easier result interpretation. A hierarchical agglomerative algorithm built clusters using Euclidean distances (as metric) and Ward's method (as linkage criterion)[4], avoiding chaining effect. This new geographic variable was computed separately for fish contamination by mPCBs PCCD/Fs and DL-PCBs. These results were confirmed by using a partitional clustering method, fuzzy c-means clustering[5].,

	mPCBs ng/g of fresh weight (mean)	**Mean confidence interval**	**PCDD/Fs and DL-PCBs pg WHO-TEQ$_{98}$/g of fresh weight (mean)**	**Mean confidence interval**
eels	1103.8***	[934.4 ; 1273.2]	46.8***	[40.1 ; 53.5]
High contaminated area	1708..4	[1455.0 ; 1961.7]	72.1	[61.0 ; 83.1]
Medium contaminated area	604.0	[494.5 ; 713.5]	28.3	[23.3 ; 33.3]
Low contaminated area	242.8	[204.5 ; 281.1]	15.6	[13.2 ; 18.1]
high bioaccumulating fish	221.1***	[184.8 ; 257.4]	11.4***	[9.6 ; 13.1]
High contaminated area	364.1	[304.2 ; 424.0]	18.7	[15.8 ; 21.5]
Medium contaminated area	76.5	[35.5 ; 117.6]	4.1	[2.4 ; 5.9]
Low contaminated area	53.0	[38.5 ; 67.5]	3.1	[2.6 ; 3.7]
low bioaccumulating fish	92.4***	[65.7 ; 119.1]	4.8***	[3.8 ; 5.8]
High contaminated area	171.4	[101.9 ; 241.0]	7.7	[5.5 ; 10.0]
Medium contaminated area	80.5	[60.5 ; 100.5]	4.2	[3.4 ; 5.1]
Low contaminated area	27.2	[22.8 ; 31.5]	1.6	[1.4 ; 1.8]

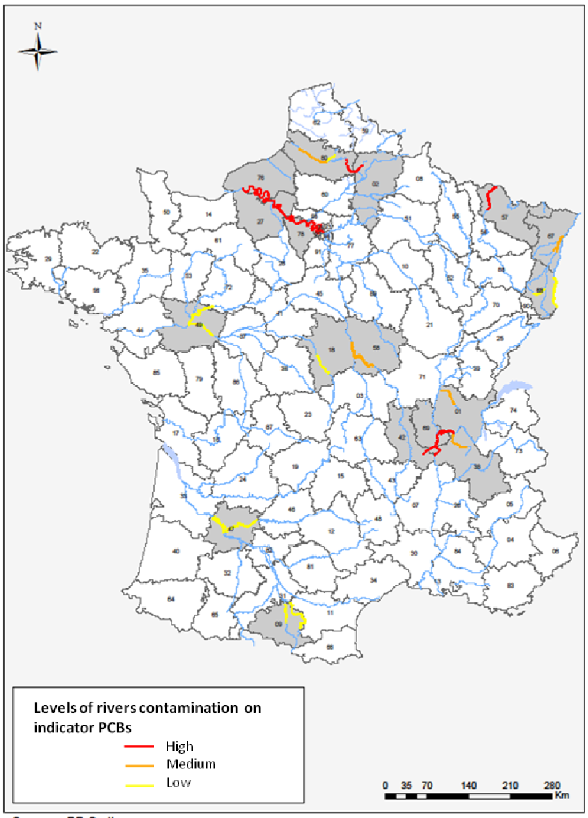
*** significant at the 0.001 level

Table 1 : Fish contamination by PCDD/Fs+DL-PCBs and mPCBs

Consumption data

606 persons, fishermen or members of their family, agreed to participate and were selected with a complex sampling design. They represented 21 180 fishermen household: the whole population. During the survey, they

were asked at home about demographic characteristics (age, gender, BMI), general dietary habits and more exactly the freshwater fish consumption (data collected with Food Frequency Questionnaire). High bioaccumulating fishes grouped eels *Anguilla anguilla*, barbells *Barbus barbus*, breams *Abramis brama*, carps *Cyprinus carpio*, sheatfishes *Silurus glanis* and roaches *Rutilus rutilus*. In low bioaccumulating fishes, there were bleaks *Alburnus alburnus*, gudgeons *Gobio gobio,* pikes *Esox lucius,* black-basses *Micropterus salmoide*s, crucian carps *Carassius carassius,* chubs *Squalius cephalus,* hotus *Chondrostoma nasus,* perches *Perca fluviatilis,* catfishes *Ameiurus nebulosus,* pike perches *Sander lucioperca,* tenches *Tinca tinca*, trouts *Oncorhynchus clarkii clarkii*, daces *Leuciscus leuciscus*, and minnows *Phoxinus phoxinus*. A consumer was someone who had a consumption of high bioaccumulating fish higher than 2 times a year.



Sources : BD Carthage, anses

Figure 3 : Rivers clustering according to mPCB contamination levels

Results and discussion

The clustering methods built 3 classes, corresponding to 3 levels of contamination (high, medium and low), for mPCBs and for PCDD/Fs and DL-PCBs (Figures 2 and 3). Contamination levels for all kind of fishes varied significantly from area to area (p-value < 0.0001, table 1). These results and their consistency with Opinions related to fish regulations, expressed by Anses, increased the relevance of the clustering. Eels were the most contaminated wherever areas. Nevertheless this specie was very little consumed. (Table 2). In the whole population, freshwater fish were little consumed, about 8 times per year for all species (low and high bioaccumulating fishes) and in the consumer population, less than one time per week (33 times per year). The whole population consumption varied from area to area except for low bioaccumulating fishes (p=0.09). In the consumer population, only eel consumption varied

yearly intakes (number of eating occasions)	**high contaminated area**		**medium contaminated area**		**low contaminated area**		**total**		**comparison**
	mean	sem	mean	sem	mean	sem	mean	sem	p-value
fresh water fishes									
whole population	5.5	0.9	7.5	1.1	10	1.2	7.7	1.0	0.01
consumers only	33.9	3.9	28.5	3	34.3	4	32.7	3.6	0.39
high bioacccumulating fishes									
whole population	0.8	0.1	1.1	0.2	2.4	0.7	1.4	0.3	0.03
consumers only	8.0	1.1	6.1	0.7	12.4	3.4	9.7	1.7	0.09
eels									
whole population	0.3	0.1	0.3	0.1	0.6	0.1	0.5	0.1	0.01
consumers only	2.9	0.6	1.7	0.3	2.8	0.3	2.6	0.2	0.01
low bioaccumulating fishes									
whole population	4.4	0.8	6.1	0.9	7.0	0.8	5.8	0.5	0.09
consumers only	23.0	3.2	20.6	2.7	19.1	1.9	20.4	2.6	0.57

sem: standard error mean

Table 2 : Yearly fish intake in whole population (n=606) and consumers only (n=322)

from area to area (p=0.01). For whole and consumer populations, consumption of low bioaccumulating fish was higher than high bioaccumulating ones.

	whole population		**consumer population**	
	PCDD/Fs and DL-PCBs (pg TEQ_{98} /kg of body weight/day)	mPCBs (ng /kg of body weight/day)	PCDD/Fs and DL-PCBs (pg TEQ_{98} /kg of body weight/day)	mPCBs (ng /kg of body weight/day)
Exposure	0.37** [0.3 ; 0.45]	7.79 *** [6.15 ;9.44]	1.85 *** [1.49 ; 2.21]	39.36 *** [30.54 ; 48.19]
high contaminated area	0.57 [0.39 ; 0.76]	12.36 [8.4 ;16.33]	4.93 [3.26 ; 6.59]	108.05 [75.1 ; 141.1]
medium contaminated area	0.29 [0.20 ; 0.37]	7.28 [4.8 ;9.76]	1.63 [1.34 ; 1.92]	28.26 [22.81 ; 33.71]
low contaminated area	0.24 [0.19 ; 0.30]	3.43 [2.65 ;4.2]	0.88 [0.71 ; 1.04]	13.31 [11.07 ; 15.56]

*** significant at the 0.001 level, ** significant at the 0.01 level

Table 3 : PCDD/F+DL-PCB and mPCB exposure in the whole and consumer populations

Exposures caused by freshwater fish consumption varied significantly from area to area for PCDD/Fs and DL-PCBs and mPCBs (Table 3) and were higher in the most contaminated areas. Not surprisingly, consumer population exposures were higher than the whole population ones. Toxicological reference doses were established to ensure that people were not exceeding a certain serum level. Accordingly, JECFA set up a tolerable daily intake[6] in 2001 for PCDD/Fs of 2.33 pg WHO-TEQ kg^{-1} $b.w.day^{-1}$. All of PCDD/Fs and DL-PCBs exposures were under the reference value except for consumers fishing in the high contamination area (mean=4.93 [3.26 ; 6.59]). A guidance value of 10 ng kg^{-1} $b.w.day^{-1}$ was previously proposed for mPCBs [6]. Whole population exposure was in average, under the guidance value, except for high contamination area. For consumer population, iPCB exposure was over the guidance value wherever areas, even for low contamination area. To conclude, just taking account freshwater fish consumption, exposures were higher than the general French population ones, except for the whole population exposure to PCDD/Fs and DL-PCBs [7].

This analysis showed that consumers of freshwater fishes were highly exposed to PCDD/Fs and DL-PCBs and mPCBs because of high contamination levels of fish and even if freshwater fish consumption is quite low. This high exposure could lead to high PCDD/F and DL-PCB and PCB serum levels. The national study on PCBs levels in blood of French freshwater fish eaters will pursue to estimate a possible association between PCB serum levels and consumption of PCB bioaccumulating fish, taking into account all factors that could be associated to PCB exposure. The aim is to propose safe consumption recommendations. Final results are planned for summer 2011.

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