NATIONAL STUDY ON POLYCHLORINATED BIPHENYL LEVELS IN BLOOD OF FRENCH FRESHWATER FISH EATERS : COMPARISON OF THE FIRST RESULTS WITH OTHER COUNTRIES

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Introduction

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 fish⁽²⁾. Since this regulation, freshwater fish exceeding maximum regulatory levels have been found in several rivers in France. Could consumption of these fishes increase PCB serum levels? Indeed, American studies in the Great Lakes, particularly concerned by PCB pollution, highlight higher PCB serum levels of freshwater fish eaters⁽³⁾.

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 describe freshwater fish consumption and compare PCB and PCDD/F serum levels of fish consumers with non consumers. The aim of this abstract is to present the first results concerning participation and PCB and PCDD/F serum levels of participants.

Materials and methods

The population of recreational fishermen and their family were chosen to include freshwater fish consumers and non consumers. They came from six rivers or parts of rivers, which represent a total of 900 km and expected to be representative of the different PCB contamination levels in the sediments of French rivers: 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).



The study was divided into three parts. Firstly, all the fishermen were surveyed by phone and asked to describe their consumption of high PCB bioaccumulating freshwater fishes (eel *Anguilla anguilla*, barbell *Barbus barbus*, bream *Abramis brama*, carp *Cyprinus carpio*, sheatfish *Silurus glanis* and roach *Rutilus rutilus*)⁽⁴⁾. Then, they described the consumption of these fishes for all the family, adults aged 18 to 75 years old. Subsequently, a random sample of consumers and non consumers were surveyed at home about demographics (age, gender, BMI, smoking, pregnancy history for women) and general dietary habits (all freshwater fish but also sea fish, meat, fatty food, dairy products, fruit and vegetables). At that point, this subset of subjects was invited to give a blood sample for chemical analysis. Informed consent forms were previously signed and the study was approved by the ethical Committee of Protection of People of Ile-de-France IX Henri Mondor.

PCB and PCDD/F determination

For analysis in serum samples, a preliminary addition of formic acid was applied, followed by the extraction procedure performed on C^{18} silica column with two hexane extractions. The total lipid content of the analyzed samples was determined using an enzymatic dosage dedicated to four classes of lipids from 50μ L aliquots. Further clean-up and separation processes were carried out using the classic liquid-solid adsorption chromatography with silica, Florisil and CarbopackC/Celite. The solvents used for the elution were hexane and toluene. The external standard was added for the recovery calculation. According to the isotopic dilution method used for quantification, the 18¹³C-labelled PCB standards (12 dioxin-like PCBs and 6 marker PCBs) and the 17¹³C-labelled PCDD/F standards were added to each sample before extraction. GC-HRMS measurement was performed on an Agilent 7890A gas chromatograph equipped with a DB-5MS capillary column (30 m, Ø 0.25 mm, 0.25 µm) and coupled to a Jeol JMS 800D high-resolution mass spectrometer, at a resolution of 10,000 in the selected ion-monitoring (SIM) mode using Electronic Impact as ionisation technique.

Results and discussion

Participants were 606 fishermen or members of their family composed of 400 men and 206 women. They were between 18 and 75 years old (average age=44.8) and were distributed among the different sites of the study (min: 59; max: 182). Participation rates (table 1) were between 31.9% and 49.6% with a maximum for the Rhône site where the environmental PCB pollution is a matter of great concern to fishermen and local authorities. There were 322 consumers of high PCB bioaccumulating freshwater fish (consumption of these fish more than twice a year) and 284 people were considered as non consumers. Consumers participated more than non consumers (51.6% *vs* 37.8%) probably because they felt more concerned by the PCB fish contamination.

	Seine	Somme	Rhône	Rhine	Loire	Garonne	total
number	59	80	114	92	182	79	606
participation rate (%)	31.9	42.3	49.6	48.2	45.0	44.6	44.0

Table 1: Number of participants and participation rates by site

Three PCB congeners (PCB 138, 153 and 180) were quantified for all participants. Total PCB serum levels were estimated by multiplying the sum of these PCB congeners by a 1.7 factor. This factor was calculated for the present population (quantification of all PCB congeners for 83 randomly sampled participants).

For the whole population, the geometric mean of total PCB serum levels was 491.1 ng/g lipid (95% CI [462.2; 523.5]) and the 95th percentile was 1461.8 ng/g lipid (table 2). PCB 153 congener contributed to 24.2% of total serum level. Exposition of childbearing age women (between 18 and 44 years old) to PCBs is of great concern. Indeed, at the lowest levels, health effects from PCB exposure are neurodevelopmental effects on children exposed during pregnancy and breast-feeding. For this sub-population, the geometric mean of total PCB serum levels was 230.0 ng/g lipid (95% CI [196.0; 271.0]) and the 95th percentile was 647.8 ng/g lipid (table 2). PCB 153 congener contributed to 24.9% of total serum level. Women of childbearing age serum levels were lower than the whole population's, probably because of years of exposure and very slow elimination kinetics of PCBs. We may also suppose a birth cohort effect. Indeed, since banning of PCBs in France in 1987, environmental pollution has progressively declined and human exposure too.

2

	total	population (n=606)	women of childbearing age (n=79)				
congeners	geometric 95% confidence p05		geometric	95% confidence	n95		
congeners	mean	interval	py5	mean	interval P ⁵		
PCB 138	53.7	[50.5; 57.1]	177.9	29.9	[25.8; 34.5]	71.0	
PCB 153	118.6	[111.4; 126.3]	352.5	57.4	[49.1; 67.1]	144.0	
PCB 180	113.0	[105.8; 120.7]	335.6	46.8	[38.9; 56.3]	144.2	
total PCBs	491.9	[462.2; 523.5]	1461.8	230.0	[196.0; 271.0]	647.8	

Table 2: Serum levels of PCB congeners (ng/g lipid) of total population and childbearing age women

The PCB 153 congener was used as a marker to compare serum levels across studies (table 3). Serum levels of the present population were about five or six times higher than those of the North American population (126.1 ng/g lipid vs 19.5 and 24.2 ng/g lipid)^(5,6). These differences could be explained by higher environmental contamination in France and different dietary habits (in North American population, less consumption of fish which is a major contributor to PCB exposure). Conversely, serum levels were lower than those in Czech Republic probably highly contaminated by PCBs (438 ng/g lipid)⁽⁷⁾. Finally, they were within the range of the current French general population (130 ng/g lipid)⁽⁸⁾ and have declined since the eighties (360 ng/g lipid in the general population)⁽⁹⁾. Same trends were observed for women of childbearing age^(10,7).

 Table 3: Median PCB 153 serum levels in ng/g lipid of general population and childbearing age women from selected countries

population	region	country	year of collect	n	age	PCB 153	reference
	North America	United States	2003-2004	1300	> 20	24.2	(5)
	Norui America	Canada	2007-2009	1666	20-79	19.5	(6)
general population		Czech Republic	2006	202	33	438.0	(7)
	Europe	France	2006-2007	386	18-74	130.0	(8)
		France	2009-2010	606	44.8	126.1	this study
	North America	United States	2001-2002	496	16-39	14.0	(10)
childbearing age women	F	Czech Republic	2006	78	33	332.0	(7)
	Europe	France	2009-2010	79	34	57.4	this study

PCDD/Fs (17 congeners) and DL-PCBs (12 congeners) were also selected for analysis in this study. Rate of censored data was low (8%) with a very low contribution to the total TEQ (<0.5%).

For the whole population, the geometric mean of PCDD/F+DL-PCB serum levels was 18.6 pg TEQ_{2005}/g lipid (95% CI [17.8; 19.4]) and the 95th percentile was 43.3 pg TEQ_{2005}/g lipid (table 4). DL-PCB congeners contributed to 44.4% of this sum. For childbearing age women, the geometric mean of PCDD/F+DL-PCB serum levels was 13.0 pg TEQ_{2005}/g lipid (95% CI [11.9; 14.2]) and the 95th percentile was 24.0 pg TEQ_{2005}/g lipid (table 4). DL-PCB congeners contributed to 33.8%. As for PCBs, serum levels of this sub-population were lower.

 Table 4: Serum levels of PCDD/F and DL-PCB congeners (pg TEQ₂₀₀₅/g lipid) of total population and childbearing age women

	tota	l population (n=606)	women of childbearing age (n=79)			
congeners	geometric mean	95% confidence interval	p95	geometric mean	95% confidence interval	p95
PCDD/Fs	10.7	[10.3; 11.1]	22.7	8.3	[7.7; 9.1]	15.4
DL-PCBs	7.4	[7.0; 7.9]	24.2	4.4	[3.9; 5.0]	10.4
PCDD/Fs+DL-PCBs	18.6	[17.8; 19.4]	43.3	13.0	[11.9; 14.2]	24.0

PCDD/F+DL-PCB serum levels were within the range of current French general population (18.5 pg $\text{TEQ}_{2005}/\text{g}$ lipid) and quite within those of North American population (between 6.2 and 37.5 pg $\text{TEQ}_{2005}/\text{g}$ lipid)^(11,12) (table 5). Nevertheless and consistently with previous results, DL-PCB contribution was higher in French population than in North American one.

region	country	year of	n age		PCDD/Fs		DL-PCBs		PCDD/Fs+ DL-PCBs	reference
		conect			mean	%	mean	%	mean	
North America	United States	2001-2002	51*	> 20	-	75 to 90	-	10 to 25	6.2 - 37.5	(11)
Furana	France	2005	1030	52	11.8	62.5	6.4	37.5	18.5	(12)
Europe	France	2009-2010	606	44.8	10.7	55.6	7.4	44.4	18.6	this study

Fable 5 : Geometric mean PCDD/F and DL-PCB serum levels in pg TEQ ₂₀₀₅ /g lipid of general population from
selected countries

*51 pools (total of 1734 people)

Considering total PCBs, 20 people were above the 97th percentile. Their mean age was 63.2 years old, quite higher than total population (average age=44.8). There were 15 consumers of high PCB bioaccumulating freshwater fish (consumption of these fishes more than twice a year). They were advised to reduce their consumption particularly in PCB contaminated rivers.

First results indicate that PCB and PCDD/F+DL-PCB serum levels of the present population is within the range of current French general population and quite higher than North American one. They have declined since the eighties. People with the highest serum levels are mostly consumers of PCB bioaccumulating freshwater fish and are older than total population. Consequently, the study will pursue to estimate a possible association between PCB serum levels and consumption of PCB bioaccumulating fish, using a multivariate analysis and taking into account all factors that could be associated to PCB exposure. The aim is to propose healthy consumption recommendations. Final results are planned for summer 2011.

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4