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Polychlorobiphenyls in freshwater fish: a new strategy to set maximum contamination limits

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Keywords

PCB, areas of health concern, consumption recommendations

Abstract

Polychlorinated biphenyls (PCBs) are persistent organic pollutants accumulating along the food chain and particularly in fish. Consequently, the European Commission has set regulatory limits for PCBs in both sea and freshwater fish. Focusing on freshwater fish, the French Agency for Food Environmental and Occupational Health & Safety has developed a method to determine the areas in France where the consumption of locally caught freshwater fish is not recommended due to PCB contamination. To determine these areas of potential health concern, an existing statistical model of the relationship between the consumption of local fish by freshwater anglers and their PCB body burden is linked to a newly determined critical PCB body burden threshold for the population. The main conclusions of this study are that the consumption of 2 freshwater fish per week from rivers in the areas of France where the median contamination level in fish is greater than 250 ng/g could lead to some exceedance of the critical body burden threshold. In these areas, eels, known to have a very high PCB accumulation potential, should only be consumed on rare occasions.

1. Introduction

Polychlorinated biphenyls (PCBs) are chlorinated aromatic compounds constituting a family of 209 compounds or congeners. In the past, different congeners were formed during the production process, in highly variable concentrations, to the composition of commercial mixtures used for their insulating properties (electrical transformers) and their chemical and physical stability (cutting oils, inks, and paints). These uses were restricted to closed systems (transformers and capacitors) during the 1970s, and the production and use of PCBs was then banned in France in 1987.

Chronic PCB toxicity is primarily correlated with cumulative tissue burden (PCB concentrations in the body, usually expressed in ng PCBs/g of plasma lipids) and not directly with the amount an individual is exposed to at a given time. The toxicity of PCBs varies considerably depending on the

considered species and the different congeners within the same species. The number of chlorine atoms on the phenyl rings determines the toxicity. The persistence of PCBs in the environment and in the body increases with the number of chlorine atoms. The higher the number of chlorine atoms of a congener, the slower its elimination and the longer its persistence in the body (EFSA 2005).

PCBs are chemically stable and weakly biodegradable; consequently, they are classified as persistent organic pollutants (POPs). They are lipophilic substances that become concentrated in food chains (Borgå et al., 2001) and are mainly found in animal fats (Bernard et al., 2002). In humans, exposure occurs mainly through food consumption (EFSA, 2005; Bemrah et al., 2008; Afssa 2007) and mainly through fish consumption, as demonstrated by the results of the most recent French total diet study (Sirot et al., 2012).

Food is a vector of essential nutrients for our metabolism. Fish are known to be valuable sources of n-3 Long Chain-Poly Unsaturated Fatty Acids (LC-PUFAs, particularly eicosapentaenoic acid -EPA and docosahexaenoic acid-DHA). However, fish can also be vectors of chemical contaminants such as PCBs. To protect consumer health, a balance must therefore be maintained between the benefits and risks of consuming some food groups. The strategy is first to prevent nutritional imbalances and second to limit exposure to certain chemical contaminants.

In 2010, the French Agency for Food, Environmental and Occupational Health and Safety (ANSES) released an opinion analyzing the benefits and risks of fish consumption based on these two aspects (Afssa 2010). Briefly, it was recommended for the entire population and as part of a balanced diet, to consume 2 servings of fish per week (one serving being on average 100 g for adults and around 75 g for children, Dubuisson et al., 2010; Lioret et al., 2010), including one with high EPA and DHA levels, and varying the species and source (wild, farmed, fishing location, etc.). This consumption optimally meets nutritional requirements while limiting the risk of over-exposure to chemical contaminants.

Due to the local accumulation of PCBs in sediments of rivers, freshwater fish may accumulate PCBs. Due to the lipophilicity of these compounds, fatty fish are the most likely to accumulate them. Fish species in contact with the sediments such as eels, carps or breams are also among the most contaminated. Starting in 2008, the French Ministry of Health set up an action plan aimed at determining the different areas where lean fish should not be consumed. ANSES was asked to recommend appropriate management decisions based on the PCB concentration in fish. These recommendations were based on a statistical approach predicting the maximum weight or length of the fish having a PCB concentration lower than the maximum limits (Afssa 2009) set by Commission Regulation (EC) No 1881/2006 (EC, 2006). For non-dioxin like PCBs (ndl-PCBs), these limits are established on the basis of the sum of PCB-28, -52, -101, -138, -153 and -180, and are as of today, 125 ng ndl-PCB/g fresh weight (fw) for all freshwater fish, except for eels for which the limit is 300 ng ndl-PCB/g fw. Considering PCB contamination data in fish, a hierarchical grouping (Ward, 1963) was performed that clustered 3 groups of fish based on their PCB concentrations. The first group included fish with high PCB concentrations. The following fish were part of this group: barbel (*Barbus barbus*), bream (*Abramis brama*), carp (*Cyprinus carpio*), wels catfish (*Silurus glanis*) and roach (*Rutilus rutilus*). The second group was for fish with lower PCB concentrations, including the following: bleak (*Alburnus alburnus*), gudgeon (*Gobio gobio*), pike (*Esox lucius*), largemouth black bass (*Micropterus salmoides*), crucian carp (*Carassius carassius*), chub (*Leuciscus cephalus*), common nase (*Chondrostoma nasus*), European perch (*Perca fluviatilis*), black bullhead (*Ameiurus melas*), pike-perch (*Sander lucioperca*), tench (*Tinca tinca*), rainbow trout (*Oncorhynchus mykiss*), dace (*Leuciscus leuciscus*) and minnow (*Phoxinus phoxinus*). Eel (*Anguilla anguilla*) was considered separately due its specific maximum limit. The statistical approach was performed for each group independently. This analysis was performed on all water bodies in France, and enabled restriction of consumption of fish from areas in which the predicted fish contaminations were higher than the regulatory thresholds.

In 2015, Desvignes et al. established a relationship between serum PCB concentrations and anglers' consumption of freshwater fish with high PCB bioaccumulation potential (Desvignes et al., 2015). This relationship was the basis to update recommendations with regard to safe consumption of freshwater fish. The established regression model explained 66.5% of the variation in total serum PCB concentrations. The two factors with the highest significant and positive association with total serum PCB concentrations were age (explaining 60% of the model) and to a much lower extent, the consumption of freshwater fish with high PCB accumulation potential (explaining 2% of the model). In addition, the study showed that total serum PCB levels in the study population (anglers and their family) and in women of childbearing age were in the same range as those observed in the general population in France. This last result is explained by the low prevalence of regular consumption of fish with high PCB accumulation potential.

The impact of PCBs on human health has been investigated in various studies conducted in the United States, Canada and Europe. In 2007, the National Public Health Institute of Quebec conducted a critical review of epidemiological studies published since 1997 (INSPQ, 2007) to analyze the causal relationships between exposure to PCBs (assessed in terms of levels in the body, which are estimated in most of these studies by measuring the PCBs in plasma lipids and in maternal milk) and the incidence of health problems in exposed subjects. Dose-response relationships have been established by correlating markers of exposure with markers of effects. Considering the entire set of data, ANSES concluded that critical body burdens could be proposed (ANSES 2010). Regarding pregnant women or women of childbearing age (i.e., women aged from 18 to 44), as well as breastfeeding women, available literature converge to propose a critical concentration level between 700 and 1000 ng total PCB/g of maternal plasma lipids. This value corresponds to the level of prenatal PCB body burden (plasma concentration in pregnant women) above which the occurrence of significant effects on the mental and motor development of children exposed in utero cannot be ruled out. ANSES consequently proposed to set the critical concentration threshold for pregnant women, women of childbearing age, breastfeeding women, and children less than three years of age at 700 ng total PCB/g of plasma lipids. Due to the persistence of PCBs in the body and thus the gradual increase in concentrations with age, this threshold value also applies to young and adolescent girls.

For boys over three years of age, adult men, and women past childbearing age, data are fragmentary and sometimes contradictory, making clinical interpretation difficult. However, based on the study by Schantz et al (1999) showing that below 1890 ng total PCB/g of plasma lipids, the effect of PCBs on neurological function (manual stability and/or mnemonic functions) can be excluded and on the PCB-RISK study, showing that variations of thyroid hormones occur in individuals with PCB body burden higher than 2300 ng/g plasma lipids, the value of 1800 ng total PCB/g of plasma lipids was considered as the critical concentration level for boys over three years of age, adult men, and women over 45 years (considered not of childbearing potential). However, due to the

difficulties in determining a threshold for the general population, this value was considered as indicative.

Taking advantage of the amount of available PCB contamination data in freshwater fish and of the statistical model previously developed (Desvignes et al., 2015), an

analysis was performed aiming to determine the criteria to use to identify areas that should be considered as of health concern, i.e. areas where the consumption of fish could lead to exceeding the critical body burden, and ultimately, to propose health-based management measures rather than measures based on regulations.

2. Material and Methods

2.1. Data

All fish contamination data collected since 2008 in different rivers from the 6 water bodies in metropolitan areas (Artois-Picardie, Rhin-Meuse, Rhône-Méditerranée-Corse, Seine-Normandie, Loire-Bretagne and Adour-Garonne) were used to generate a database containing more than 5,000 fish PCB contamination values after database cleaning. The contamination values of fish from the parts of rivers studied to set the regression model were aggregated with those from the different surveillance plans and specific PCB plans performed from 2005 to 2013. Altogether, 824 contamination values were available over the 23 parts of rivers previously studied; 228 values from fish with high PCB accumulation potential were available and 596 from fish with low PCB accumulation potential (Desvignes et al., 2015). For these 23 parts of rivers, biomonitoring and freshwater fish consumption data were available for 606 anglers or members of their families.

The same areas as those studied in a previously published article (Desvignes et al., 2015) were considered in this study. A hierarchical agglomerative algorithm defined clusters using Euclidean distances (as the metric) and Ward's method (as the linkage criterion) (Ward, 1963), avoiding any chaining effect. The analysis resulted in three clusters of contamination levels (high, moderate, and low).

2.2. Regression model

The model used to predict the body burden correlated with the frequency of consumption of freshwater fish with high PCB accumulation potential coming from areas with different levels of contamination was fully described previously (Desvignes et al., 2015). The covariates age, current consumption of fresh

water fish with high PCB bioaccumulation potential, gender and area were positively associated with serum PCB level.

The strategy here was to determine the areas where the consumption of two servings of fish with high PCB accumulation potential per week could lead to body burden exceeding the critical values. Fish with high PCB accumulation potential were: barbel (*Barbus barbus*), bream (*Abramis brama*), carp (*Cyprinus carpio*), wels catfish (*Silurus glanis*) and roach (*Rutilus rutilus*). Body burden values were predicted using the regression model in varying age, gender, current consumption of fresh water fish with high PCB bioaccumulation potential and area and in setting the other covariate at the mean of the studied population. Four different populations were considered: 44-year-old women and men, and 60-year-old women and men. The 44-year-old female population was considered as the most at-risk population considering that this is the oldest population (and therefore, the one that accumulated the most PCBs over the years) for which the critical body burden value was set at 700 ng/g lipids. Three different consumption frequencies were tested: 1 serving every 2 months (this frequency was the one ANSES recommended for women of childbearing potential based on the previously reported study, Desvignes et al., 2015), 2 servings per week (this frequency corresponded to the recommendation for the general population based on a benefit-risk approach, ANSES, 2010), and 1 serving per week (this last frequency was arbitrarily selected between the first two). These scenarios were carried out in taking account of the contamination level cluster described previously. Statistical analyses were performed using SAS 9.2 software (SAS Institute Inc., Cary, NC, USA); R 2.12 software (R Core Team, 2010) and Stata v10 software (StataCorp, 2007).

2.3. Part 3

3. Results and Discussion

3.1. Simulated body burden

Based on the previously published regression model of the individual determinants of blood PCB levels (Desvignes et al., 2015), the body burden can be simulated in different conditions (different areas with different levels of fish PCB contamination, different populations and different frequencies of fish consumption). Body burden predictions were performed for four different populations (44-year-old men and women, and 60-year old men and women). The populations of interest were 44-year-old women and 60-year-old men. The 44-year-old and below female population is the population of women of childbearing potential and consequently the associated critical

body burden recommended by ANSES is 700 ng/g plasma lipids. Moreover, this population is the oldest for which the 700 ng/g plasma lipids critical level applies, meaning that accumulation of PCBs over the years is more pronounced. On the other hand, the population of 60-year-old males is amongst the oldest within the general population and for which the critical body burden recommended by ANSES is 1800 ng/g plasma lipids, and for which age-related PCB accumulation is limited compared to an older population. The results of these simulations are presented in Table 1.

It appeared that for both males and females, the body burden predicted at the age of 60 is twice that at the age of 44, for the

same consumption of freshwater fish with high PCB bioaccumulation potential. As described previously (Desvignes et al., 2015), consumption of fish with high PCB concentrations is responsible for approximately 2% of overall body burden, whereas 60% of PCB body burden results from age. The covariate age represented birth cohort effect, past exposure (i.e. over the entire lifespan, body accumulation of PCBs) and dietary exposure over the 16 years between 44 and 60. Unfortunately it was not possible to estimate separately the contribution of each factor.

The simulations showed that, in highly contaminated areas, the consumption of 2 servings of fish with high PCB accumulation potential per week can lead to an exceedance of the critical body burden in both considered populations. However, in the low contamination areas, the same frequency of consumption of fish with high PCB accumulation potential does not lead to any exceedance of the critical values. In a moderately contaminated area, the consumption of 2 servings of fish with high PCB accumulation potential per week led to body burden below or very close to the critical values (mean values being 1,540 versus 1,800, and 720 versus 700 ng/g plasma lipids for the 44-year-old female population and the 60-year-old male population, respectively). The upper bound of the 95% CI for the 44-year-old female population exceeded the critical body burden (1,040 ng/g plasma lipid). However, this should not be considered as a public health concern since it was shown that the average consumption of freshwater fish with high PCB accumulation potential in anglers' families is on average 14.8 (95% CI: 11.3, 18.4) servings per year (Desvignes et al., 2015), far from the frequency we considered to perform the predictions that were used to draw conclusions. Moreover, the study showed that this average low consumption of fish with high PCB accumulation potential was associated with body burdens very close to those measured within the general population (492 ng/g in the present study versus 480 in the general French population; InVS, 2010).

In conclusion, in moderately contaminated areas, the consumption of two servings of fish with high PCB accumulation potential per week is not anticipated to lead to any significant exceedance of the critical body burden in women of childbearing age. Since the simulations were performed considering the worst case scenario, i.e. the most exposed (freshwater fish consumers) and the most at-risk population (44-year-old female population), this conclusion also applies to the general population.

3.2. Fish contamination database

Concentration data from the 3 categories of fish were available (fish with low PCB

accumulation potential, fish with high PCB accumulation potential, and eels). Focusing on the population of anglers and their family and associated areas (in total, 23 zones representing 900 km across France) studied in the context of the previously published study (Desvignes et al., 2015), three clusters were created depending on fish contamination according to the Ward approach. The contamination data of fish with low PCB accumulation potential were used to perform the hierarchy since this category is the most represented amongst the three groups of fish. Within each of the clusters, the descriptive statistics of PCB concentrations of fish with high PCB accumulation potential are shown in Table 2. Eel was not included in this analysis due to its highly variable PCB contamination within the same body of water (Blanchet-Letrouvé et al., 2014). In some zones, the amount of available data was not sufficient to calculate descriptive statistics; consequently Table 2 presents descriptive statistics for 14 zones.

The most contaminated zones were zones 1 and 4 of the Rhone-Méditerranée-Corse water body as well as zone 1 of the Seine-Normandie water body with median PCB concentrations ranging from 278 to more than 700 ng/g. In these areas, as shown above, the weekly consumption of two fish servings with high PCB accumulation potential can lead to an exceedance of the critical body burden concentration in the most at-risk population (women of childbearing potential). With the objective of proposing a threshold to characterize highly contaminated zones, the lowest PCB concentration median value within the highly contaminated zone was selected. The median value was selected instead of the mean to circumvent any non-normal distribution of the contamination data. This value was 278 ng ndl-PCB/g fresh weight (zone 1 of the Rhône-Méditerranée-Corse water body). On the basis of these data, 250 ng ndl-PCB/g in fish was considered as the threshold to discriminate the fishing zones. Below this value, respecting the consumption of two fish servings per week, the body burden is not expected to exceed the critical value set for the most sensitive population, i.e. women of childbearing age. However, areas with median PCB concentrations in fish above 250 ng ndl-PCB/g are considered at risk for public health in case of consumption. Considering the approach used in the present article (values of PCB concentrations based on anglers' consumption and body burdens), this threshold should apply to fish caught by anglers and not to fish available on the market. Altogether, these data allowed us to determine areas where consuming fish would be of health concern, since the consumption of two servings of fish with high accumulation potential per week could lead to exceedance of the critical body burden set for women of childbearing potential, bearing in mind that ANSES's recommendation to meet optimal nutritional requirements is two servings of fish per week

4. Conclusion

In Europe, certain maximum limits have been established for PCBs in fish and are currently 125 ng ndl-PCB/g for all fish, except eels for which the maximum limit is 300 ng ndl-PCB/g. These values, set according to the ALARA approach (as low as reasonably achievable), were also those applied as a default value to fish harvested by anglers. The present study describes a new approach to set PCB thresholds in fish above which the recommended consumption could lead to

exceedance of a critical body burden. On the one hand, this was made possible thanks to the amount of data collected over almost 10 years in different water bodies in France. On the other, the availability of body burden values associated with dietary data was also needed to reach the conclusions. It appears that due to the low consumption of freshwater fish in France and particularly of fish with high PCB accumulation potential, the PCB contamination limit could be 250 ng ndl-PCB/g. Due to the approach used, this limit would apply to fish

caught by anglers and would not exceed the regulatory limits set by the European Commission that apply to fish supplied on the market. Moreover, considering that dl-PCBs and PCDD/Fs fish concentrations have been shown to be highly correlated with those of ndl-PCBs (Anses, 2015), in absence of industrial contamination, the limit set at 250 ng PCB/g appears valid.

The approach presented here changes the paradigm to set health-based regulatory limits, most of the time set according to the ALARA approach or to dietary toxicological credit

calculation. In this study, the limit for fish contamination was established using a body burden determinants regression model and simulations relying on this model. The model allowed us to predict body burden values based on the frequency of fish consumption, sex, age and the contamination of the area. In France, various recommendations not to consume fish based on the regulatory limits currently apply; this limit could possibly be doubled without significant risk to consumers, should the approach described above be selected.

Table 1. Simulated body burdens for different areas, frequencies of consumption of highly contaminated fish and populations

Areas	Recommendation	Simulated mean body burdens (95% CI) (ng PCB g ⁻¹ plasma lipids)			
		44-year-old women	60-year-old women	44-year-old men	60-year-old men
High contamination	1 serving of fish every 2 months	546 (488–611)	1330 (1120–1600)	774 (659–917)	1450 (1230–1690)
	1 serving of fish per week	834 (665–1050)	1630 (1290–2100)	950 (759–1200)	1780 (1420–2230)
	2 servings of fish per week	1090 (798–1490)	2130 (1550–2990)	1240 (915–1680)	2320 (1660–3110)
	1 serving of fish every 2 months	486 (435–540)	1080 (884–1320)	627 (522–760)	1170 (1000–1400)
	1 serving of fish per week	619 (476–815)	1210 (933–1560)	705 (545–928)	1320 (1000–1670)
	2 servings of fish per week	720 (487–1040)	1410 (928–2090)	821 (549–1190)	1540 (1060–2240)
Moderate contamination	1 serving of fish every 2 months	463 (420–510)	991 (865–1140)	577 (505–653)	1080 (956–1230)
	1 serving of fish per week	551 (478–636)	1080 (921–1290)	628 (539–738)	1180 (1020–1370)
	2 servings of fish per week	615 (497–745)	1200 (960–1470)	701 (575–860)	1310 (1080–1600)
	1 serving of fish every 2 months	463 (420–510)	991 (865–1140)	577 (505–653)	1080 (956–1230)
Low contamination	1 serving of fish per week	551 (478–636)	1080 (921–1290)	628 (539–738)	1180 (1020–1370)
	2 servings of fish per week	615 (497–745)	1200 (960–1470)	701 (575–860)	1310 (1080–1600)

Notes: Critical body burden is 1800 ng g⁻¹ plasma lipids except for the 44-year-old women for whom it is 700 ng g⁻¹ plasma lipids. Values are rounded to three significant figures. Values in bold are those exceeding the respective critical body burden.

Table 2. Descriptive statistics of fish concentrations (ng g⁻¹) in the different studied areas

	Zone 1	Zone 2	Zone 3	Zone 4
<i>Adour-Garonne</i>				
Minimum	4.56	10.2	25.3	
P25	8.89	15.9	42.4	
Median	13.2	23.1	72.8	
Mean	13.3	24.6	88.7	
P75	17.7	31.7	103	
Maximum	22.1	42.0	197	
<i>Loire-Bretagne</i>				
Minimum	37.6	13.4	11.7	16.1
P25	44.0	18.3	12.8	21.8
Median	64.4	33.2	14.4	33.4
Mean	90.7	37.2	14.3	64.4
P75	111	55.6	15.8	109
Maximum	197	66.6	16.7	198
<i>Rhin-Meuse</i>				
Minimum	35.4	23.1		
P25	48.9	51.2		
Median	62.3	79.3		
Mean	62.3	63.1		
P75	75.8	83.1		
Maximum	89.2	86.9		
<i>Rhône-Méditerranée-Corse</i>				
Minimum	13.7	14.9	123	401
P25	171	29.8	181	607
Median	278	41.1	239	710
Mean	360	155	219	1030
P75	489	97.9	267	1070
Maximum	1660	784	294	2450
<i>Seine-Normandie</i>				
Minimum	413			
P25	495			
Median	665			
Mean	705			
P75	876			
Maximum	1080			

Notes: In some zones, the amount of available data was not sufficient to calculate descriptive statistics. Values are rounded to three significant figures.

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