Dioxins, furans and dioxin-like PCBs: occurrence in food and dietary intake in France

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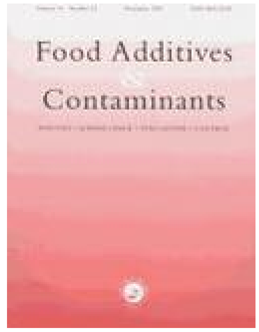

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Abstract

PCDD/Fs and DL-PCBs contamination data in food products consumed in France collected from national monitoring programs (2001 to 2004) and representing analytical results for almost 800 individual food samples were combined with food consumption data from the French national dietary survey to estimate PCDD/Fs and DL-PCBs dietary intakes, expressed as toxic equivalents (WHO-TEQs). The mean PCDD/Fs and DL-PCBs intake was estimated at 1.8 and 2.8 pg WHO-TEQ/kg bw/d respectively for adults (15 years and over) and children (3-14 years). Main contributors to total intake were fish and milk products for both children and adults (respectively 48% and 31% for adults and 34 and 43% for children). DL-PCBs constituted the largest contributor to contamination in most of foodstuffs. Life-long intake estimate showed that a non-negligible part of the French population (between 20 to 28%) had an intake above the tolerable monthly intake for dioxins and dioxin-like PCBs of 70 pg WHO-TEQ/kg bw/month (JECFA, 2001).

Introduction

Dioxins (polychlorinated dibenzo-p-dioxins, PCDDs) and furans (polychlorinated dibenzofuran, PCDFs) are chlorinated polycyclic aromatic hydrocarbons. There are 75 and 135 congeners for PCDDs and PCDFs respectively depending on the number and position of chlorine atoms in the aromatic rings. Those congeners are very similar with common characteristics. They have a strong thermal stability, are insoluble in water but very soluble in lipids and are poorly biodegradable. As a result, they accumulate in adipose tissues, especially of animal origin, where they can remain for a long time. This leads to their classification as persistent organic pollutants. This category of molecules is generally called “dioxins”.

Polychlorinated biphenyls (PCBs) are also chlorinated aromatic compounds. 209 congeners form this group of closely related chemicals with different toxicity levels. As with dioxins, PCBs are poorly biodegradable and have strong chemical and physical stability, which explains why they accumulate in tissues. But unlike dioxins, which are by-products (occurring during thermal process), PCBs were used in industry and agriculture until the 1970s. Some PCBs, called dioxin-like PCBs (DL-PCBs), have the same type of toxicity as dioxins. This enabled the overall intake of dioxins and DL-PCBs to be estimated using the Toxic Equivalent Factor (TEF) based on an additivity model (World Health Organization [WHO], 1998). 17 out of the (75+135) PCDD/F congeners and 12 DL-PCBs are recognised as biologically active and have a TEF value.

The toxic equivalency concept uses the relative effect potency determined for each of the 17 PCDD/F and 12 DL-PCB congeners in relation to the 2,3,7,8-TCDD, the most toxic element, identified as the reference compound. The toxicity of a mixture equals the sum of the products of the concentration of each congener multiplied by its TEF value. TEF values can often be re-estimated due to new toxicological insights (1988, NATO I-TEQ; 1998, WHO-TEQ; 2005, WHO Re-evaluation of TEFs, July 2006). For example, between 1988 and 1998, the same PCDD/PCDF congeners are included, but the main difference is for 1,2,3,7,8 – Pentachlorodibenzodioxin whose TEF has been multiplied by 2 (from 0.5 to 1); also 2 others TEF congeners (Octachlorodibenzodioxin and 1,2,3,4, 7,8,9 – Heptachlorodibenzofuran) fell from a factor 10 (0.001 to 0.0001). Considering DL-PCBs, they did not have TEF values before 1998. This study uses the 1998 WHO-TEQ since 2005 TEFs were not published when this assessment was performed.

Dioxins and DL-PCBs are ubiquitous components in the environment and their characteristics result in bioaccumulation in adipose tissues. Dietary intake is therefore the major source of exposure (90%) (Liem et al., 2000), with lipid-rich food elements (food of animal origin) making up a considerable contribution.

Dietary intake in France was first estimated in 2000 for dioxins only (CSHPF-AFSSA, 2000). This estimation was based on contamination data from the late 1990s (between 1996 and 1998) and on 1994 consumption survey, ASPCC (Rigaud et al., 1997). With regard to DL-

PCBs in the estimation of total dioxin exposure, re-evaluation of TEF by WHO in 1998, better knowledge of the toxicological mechanisms of those molecules and changing dioxin emissions into the environment have led to an update of the estimated dietary intake of PCDD/Fs and DL-PCBs in France.

This study presents PCDD/F and DL-PCB levels in most of the foods making up the French diet. For the survey of dietary intake in France (INCA, 1999), we estimated dietary intake among the general population, for children and adults, and the contribution of each food group. The results are compared with previous French data and international intake assessments.

Materials and methods

Food sampling

Contamination data used to estimate dioxin exposure in France comes mainly from food control and food monitoring programmes. The European food monitoring programme called "Market basket" gathers data on almost all food products (mostly of animal origin): meat (including poultry) and offal (liver), milk and butter, eggs, shellfish (mussels and oysters), farmed fish and also some fruit, vegetables and vegetable oils. About 350 analytical results from 2001 to 2004 come from this programme. From another national food monitoring programme, we added wild fish, crustaceans and cephalopods taken from a landing site including some freshwater fish. This represents 260 analyses from 2002 and 2003.

To complete the set of data, dairy analysis from the joint-trade organization were added; data from an egg control plan including organic and free-range eggs sampled on the farms were included; cereals (bread, rice, pasta, etc.) were also sampled. In total, almost 800 (797) samples were analysed over 11 food groups. Table 1 presents the number of samples broken down into food groups. Food products not expected to contain dioxins or PCB-DLs due to their source and extremely low fat content (soft drinks, alcoholic beverages, sweets, etc.) were not considered for this exposure assessment.

[insert Table 1 about here]

Chemical analysis

The methods used to take samples and analyse dioxins and DL-PCBs in food were based on EU Directive 2002/69/EC. Extraction isolates the fat fraction containing dioxins and PCBs. Extracts are then purified and quantification is performed using a mass spectrometer equipped with gas chromatography. Analytical methods have already been described elsewhere and are not presented in detail here (AFSSA, 2005).

The toxic equivalent factors (TEF) (WHO, 1998) of the 17 PCDD/F congeners and 12 DL-PCB congeners were multiplied by the analytical concentration in order to express the result in toxic equivalent (TEQ). The concentration of each sample was then calculated by adding up the concentrations of each of the 29 congeners (PCDD/F + PCB-DL).

Non-detects are considered as equal to zero (lower bound estimates) or to LOD (upper bound estimates). As there is no significant difference between lower bounds and upper bounds, the results reported in this paper are lower bound estimates. TEQs for animal products and oils were calculated on a lipid weight basis (pg TEQ/g fat), while vegetable and fish products were calculated on a fresh weight basis (pg TEQ/g FW).

Dietary intake

Food consumption data is based on the INCA survey (individual and national survey of dietary intake in France) (Volatier et al., 2000). This national survey was conducted across France from August 1998 to June 1999, so as to incorporate seasonal effects. Food consumption was reported in a diary record over 7 consecutive days. The identification of the type of food and portion size consumed were estimated with a validated photographic book. The food nomenclature was based on the nomenclature used in the food composition tables and contains about 1000 codes and 44 food groups.

The total sample (3003 people) was composed of 1985 adults aged 15 years and over and 1018 children and young teenagers (3-14 years). There is no data available for children under the age of 3. The representativeness of the sample is assured by a geographical stratification (based on region of residence, size of urban area) and by the quota sampling method (with the factors age, gender, profession and size of household).

In this survey, we recorded the body weight of almost all the participants so that intake per kg of body weight could be calculated using the real weight of each person and not a fixed weight.

Recipes.

In order to take account of the ingredients in composite foods, recipes (Calamassi, 2004) were used to assign their ingredients to the appropriate reported food. For example, in a pastry, the cereal contamination value will be assigned to flour and the butter value to butter. The use of recipes gave the total consumption by type of ingredients (vegetables, meat, cereals, etc.) instead of food group.

Comments on food groups

- Dairy products: As the analysed samples did not include all dairy products, contamination of cheese and yoghurts for example was estimated by multiplying the milk contamination (in pg TEQ/g fat) by their lipids percentage as recorded in the French composition tables (Favier J.C. et al, 1995). We assume that contamination in milk (raw food) ends up in the processed food as a proportion of the amount of fat in the product.
- Fish products: Many species of fish were analysed and contamination means were allocated to each. When this was not possible, a mean fish contamination (taking account of its consumption level) was assigned. Breaded fish was assigned the mean contamination of cod and pollock, since these are the main types of fish used. With regard to contamination differences between various types of wild fish according to their fishing area, a stratification contingent based on the volume of fish landed has been applied on the basis of data from the fishery joint trade organisation.

- Vegetable products: as fruits and vegetables are generally far less contaminated, only a few analyses were conducted on these products. For example, as we did not have occurrence data on potatoes, despite their consumption level, we assume that they have the same contamination data as vegetables. Cereals were categorised into 4 contamination groups: bread, pasta, rice and other.

Exposure assessment

Dietary intake was calculated using the standard deterministic method for chronic exposure (WHO/GEMS-Food, 1997). On the basis of food consumption specific to each individual and the contamination vector of various foods, the exposure of each person for each food is obtained. In order to calculate this exposure per kilogram body weight (kg bw), this value is divided by the weight of each consumer. All the descriptive statistics presented are produced from real individual data. No use is made of means or conventional data derived from other sources. The use of real individual data makes it possible to assess the reality of exposure variability, incorporating real correlations between the various components (body weight, consumption of different foods).

Here, we make the assumption that there is an absence of correlation between body weight and dietary habits on the one hand and food contamination on the other. Irrespective of whether the consumer of a type of food – for example, UHT semi-skimmed milk – is small, medium-sized or large, it is assumed that the average contamination of the UHT semi-skimmed milk bought is, over a long period of time, equal to the average contamination for UHT semi-skimmed milks. This assumption is justified by the fact that this study does not take account of populations who, due to their proximity to a source of dioxin emissions, may systematically consume foods that are more contaminated than average. Another methodology must be used to assess the exposure of these populations (InVS, AFSSA, 2003).

This standard deterministic method to estimate exposure can be described by the following formula for the individual i:

$$E_i = \frac{\sum_j C_{ij} T_j}{w_i}$$

where:

wi is the individual body weight of the individual i

Cij is the consumption of food j by the individual i

Tj is the average PCDD/F or DL-PCB contamination of food j

Exposure is calculated for both PCDD/Fs and DL-PCBs. Total exposure was calculated by adding the two together.

The results presented here concern the overall population. Since PCDD/Fs and DL-PCBs are found in a wide range of foods, all the individuals included in the dietary survey were revealed to have been exposed to PCDD/Fs and DL-PCBs.

Results

Contamination data

Table 2 shows the analysis results per food group. Results are expressed in pg WHO-TEQ/g fresh weight for fish products and vegetables and in pg WHO-TEQ/g fat for land-based animals (meat, including poultry and offal, dairy products, eggs).

- Meat (of which poultry and offal), dairy products and eggs: the total concentration (PCDD/F+DL-PCB) remains around 1 pg TEQ/g fat, except for offal (liver) whose concentration reaches 2.5 pg TEQ/g fat. Lamb has the highest level (1.5 pg/g fat) and pork the lowest (0.4 pg/g fat). For meat products, DL-PCBs represent 85% of total contamination. In dairy products and eggs, the proportion PCDD/Fs – DL-PCBs is better-balanced: 30%-70% and 45%-55% respectively.
- Fish and fish products: these are the most contaminated food products. Sea (wild and farmed) and freshwater fish have a total concentration of between 2.7 and 2.9 pg TEQ/g fresh weight. Farmed trout is far less contaminated (0.75), as are shellfish (range between 0.73 and 1.34). DL-PCBs account for 80 to 85% of total contamination for sea fish and freshwater fish and 55 to 75% for other fish products.

- Vegetable products (fruits, vegetables, cereals): PCDD/F+DL-PCB concentrations do not exceed 0.01 pg WHO-TEQ/g fresh weight. The dioxin part (in %) is higher on average than for other products.

Of the 800 analyses, the highest PCDD/F and DL-PCB level is found in fish, irrespective of whether it is wild or farmed, from the sea or freshwater. Only farmed trout is less contaminated, perhaps because of its special feeding and breeding conditions. Even molluscs and crustaceans are more highly contaminated than meat (especially lamb, which is more fatty) or milk. We found that offal (liver) is also highly contaminated (2.52 pg TEQ/g LW), probably because of its high lipid content and also because of the role liver plays in the organism (metabolite accumulation). The other food groups (land-based animals) are far less contaminated: around 1 pg WHO-TEQ/g LW for animal products (only lamb is slightly more contaminated) and 0.01 pg WHO-TEQ/g FW for vegetable products. In spite of a slight decrease in fish contamination levels since 1999, we found the same order of magnitude for PCDD/Fs between 1999 and 2005. On the contrary, the decline is greater in meat and dairy products: from 2 to 3 times less in offal and pork; we found the same decline for eggs, milk and butter.

[insert Table 2 about here]

Dietary intake of dioxins and DL-PCBs

Intakes are calculated for PCDD/F on the one hand and for DL-PCBs on the other, and for the sum of the two. Results presented here represent the whole population. As dioxins and dioxin-like-PCBs are widely spread in the environment, all consumers in the survey are exposed to these contaminants.

Dietary intake of dioxins.

In order to compare our results with the previous assessment (2000), dietary intake of PCDD/Fs only was assessed first (no available data on DL-PCBs in 2000). For this estimation, we used the lower bound estimate (ND=0) and recipes were not used (non-existent in 2000). For the same reason, the whole population (aged 3 years and over) was considered for dioxin intake estimation. The same methodology was used in the consumer survey used in 2000 and the INCA survey: a diary record over 7 consecutive days.

Table 3 and figure 1 present the comparison results between PCDD/F exposure in 2000 and in 2005. Food groups are broken down into 7 types of product.

[Insert Table 3 and figure 1 about here]

In 2000, total exposure to dioxins and furans was estimated at 1.31 pg I-TEQ/kg bw/d (I-TEQ which correspond to NATO TEF (NATO/CCMS, 1988), were used for the 2000 French dioxins assessment). In 2005, it was 0.53 pg WHO-TEQ/kg bw/d. This indicates a 60% reduction in 5 years. Moreover, increases in the toxicity factors (I-TEF vs WHO-TEF) since 1998 mean that the previous exposure has been underestimated, and a comparison (AFSSA, 2002) between the 2 consumption surveys used in 2005 and 2000 (INCA and ASPCC respectively) with the same concentration data indicates that use of the INCA survey leads to a 15% decrease in the intake because of the decline in consumption of almost all food groups, but especially in dairy products (butter) and fish products (shellfish).

Analysis of detailed exposure indicates that the contribution of each food group has fallen in 5 years. The greatest decreasing trends concern dairy products, meat products and eggs.

Figure 2 presents the contribution of food groups for dioxin intake between 1999 and 2005.

In 2000, the highest contributing food groups were dairy products (40.5% of the total exposure) and seafood products (24.4%). In 2005, these groups reversed. Animal products (including fish) are still major contributors with 85% of total exposure, but fish products are now the leading contributor (almost 45% of total ingestion). We can see that, in general, land-based products such as milk, meat, eggs and vegetables have registered a greater downward trend than seafood products. The fall in dioxin emissions in the early 2000s (CITEPA, 2005) had a greater consequence for those products than for fish, for which exposure sources are not the same.

[Insert Figure 2 about here]

Dietary intake of dioxins and dioxin-like PCBs.

Table 4 shows the results of total exposure for children (3-14 years) and adults (15 years +). For adults, the mean daily intake was estimated at 1.8 pg WHO-TEQ/kg bw/d; the median is 1.5 pg WHO-TEQ/kg bw/d for a 95th percentile of 3.9 pg WHO-TEQ/kg bw/d. For children, the mean daily intake is 2.8 pg WHO-TEQ/kg bw/d; the median equals 2.4 pg WHO-TEQ/kg bw/d and the 95th percentile 6.0 pg WHO-TEQ/kg bw/d. Intake is therefore higher in children than adults. This is partly due to the difference in food intake and partly to their relative body weight.

[Insert Table 4 about here]

Life-long intakes.

Toxicological reference doses are established to ensure that people are not exceeding a certain body burden. Accordingly, JECFA set up a tolerable daily intake in 2001 for PCDD/Fs and DL-PCBs of 70 pg WHO-TEQ/kg bw/month (2.33 pg WHO-TEQ/kg bw/d). A life-long daily intake (from 3 years and over) has been estimated with the same food consumption survey, INCA, by building up individual cohorts at different life ages (AFSSA, 2002) In this approach, the main conservative hypothesis is that each individual has the same position in the statistical distribution of exposure during his whole life. The combination of children and adult consumers enabled the construction of life-long individuals, resulting in an estimation of 2.0 pg WHO-TEQ/kg bw/d (median=1.9) over a person's lifetime. The most exposed population (95th percentile) has an intake of 3.0 pg WHO-TEQ/kg bw/d. This would be the case throughout a lifetime if we assume that food intake and dioxin and DL-PCB concentrations remain the same throughout a person's lifetime.

[Insert table 5 about here]

If we convert this into the number of people exceeding the TDI, around 20 to 28% of people exceed the TDI for their entire lifetime. It depends on whether consumption, but not contamination, remains stable through a person's life (someone who eats a lot of a particular food category as a child continues to eat this amount as an adult), leading to 20% of people exceeding the TDI. If there is a correlation between contamination levels during a lifetime (someone who eats contaminated food as a child consumes as much contaminated food as an adult), then 28% of people exceed the TDI.

Contribution of different food groups.

Figure 3 shows the detailed contributions to total intake of PCDD/Fs + DL-PCBs of the 7 food groups for children and adults. As seen earlier with dioxin intake, the major contributors are dairy products and fish products, consistent with other international studies (Fattore et al, 2006). The greatest contribution comes from fish products for adults (48%), while for children it comes from dairy products (43%). This is due to their different consumption patterns. For others products, contributions are the same for both: meat products represent 8% of total intake, vegetables account for 5-6%, fats for 4-5% and eggs for 2-3%.

[Insert Figure 3 about here]

Considering dioxins and DL-PCBs separately, contributions to total intake are slightly different, as seen in Table 4. Table 6 shows in greater detail that DL-PCB levels are much higher than PCDD/F levels (3/4-1/4) in most products, particularly animal products i.e. fish, meat, dairy products and fats. For eggs, as for fruits and vegetables, dioxin and DL-PCB contributions are about the same. Cereal products demonstrate the opposite: PCDD/F levels are higher (almost 75%). The same distribution is observed for adults and children.

[Insert Table 6 about here]

Discussion

Overall, very few analyses, as reported in this paper, are not in accordance with the Commission Regulation (No 199/2006). This result is probably due to the decline in atmospheric emissions since the late 1990s (CITEPA, 2005), notably with the lockout of old domestic waste incinerators in 2003, and consistent with what is carried forward in different publications (EU-SCOOP, 2000; Fernandez et al, 2002). The decline noted in the environment following the protective measures taken over recent years has paid off a few years later with the decreasing trend in land-based products, but is still not reflected in the aquatic milieu which remains much more contaminated. Dioxin sources are not the same and seem to evolve slowly here (sediments).

An important difference is the ratio between PCDD/Ds and DL-PCBs in food products. In fish products, DL-PCB levels are high (between 85 and 77% of total TEQ for fish and up to 56% for crustaceans), whereas for milk, butter, meat and poultry, DL-PCBs account for 2/3 of the total and for liver, eggs and vegetable products, for half of the total. This converges with different sources of exposure between aquatic and land-based products.

Between countries, contamination data is wide-ranging. For Sweden and Finland (Darnerud, et al., 2006; Kiviranta et al., 2001), fish contamination data is only marginally less than that in France, whereas for land-based animals, French data is several times higher. On the contrary, Spanish and Dutch data is higher than French data for meat or dairy products, but about the same for fish products (Fernandez et al., 2004; Baars et al., 2004).

This resulted in the whole population being exposed to two and a half times less PCDD/F between 2000 and 2005: from 1.31 pg TEQ/kg b.w./d to 0.53 pg TEQ/kg b.w./d. Moreover, the use in 2000 of I-TEQ makes the decrease marginally greater. Indeed, studies have shown that exposure calculated with I-TEQ is 10% less than when calculated with WHO-TEQ (WHO, 1998; InVS, 2000).

Even if the intake assessment methodology is identical (individual consumption survey with diary record, same methodology) for the 2 studies (AFSSA, 2000; this study), the consumption survey is not the same between the 2 exposure assessments, and a study (AFSSA, 2002) demonstrated that this led to a decrease of around 15% in general dioxin exposure with the INCA survey which was used for the intake calculations reported in this paper. This reflects the downward trend of fatty foods observed in several countries.

As seen in figure 1, there has been a decline in the contribution of all food groups, but the greatest decrease concerns dairy products, followed by meat, fruits and vegetables. There has also been a downward trend in fish and fish products, but the relative contribution of this food group is increasing, as seen on figure 2. We can note a contrast between land-based products whose contribution is decreasing and fish products whose relative contribution is on the rise.

In terms of total PCDD/F and DL-PCB exposure, adults and children in France respectively reach an average of 1.8 pg WHO-TEQ/kg bw/d and 2.8 pg WHO-TEQ/kg bw/d (table 4). No such previous estimation has been done in France. It is difficult to conduct a comparison with international studies because methodologies used are not exactly the same, but intake among the French population seems to be within the same range as other European countries. In the UK (FSA, 2003), the mean adult 19-64 years intake is 0.9 pg WHO-TEQ/kg bw/d; the Netherlands (Baars et al., 2004) has estimated the median intake at 1.1 pg WHO-TEQ/kg bw/d for 40 year olds; in Finland (Kiviranta et al., 2001), the mean is 1.3 pg WHO-TEQ/kg bw/d for adults 25-64 years, while in Spain (Fernandez et al., 2004), it reaches 2.8 pg WHO-TEQ/kg bw/d for a person weighing 70 kg and in Italy (Fattore et al, 2006), the mean is 2.3 pg WHO-TEQ/kg bw/d for 13 to 94 years old. All these studies include DL-PCBs. The Dutch study seems to resemble ours the most. In a comparison of occurrence data, fish contamination levels are about the same between these 2 studies, which is a logical outcome if we consider that the fish probably comes from the same fishing areas. However, a lot of other contamination data is higher in the Netherlands, particularly butter, eggs and poultry. This means that, in France, there is a higher proportion of fish and fish product in total intake (48% for adults versus 16% in the Netherlands). The UK intake level has been based on a methodology (Total Diet Study) assumed to give lower results because foods are sampled and analysed “as consumed”. These differ considerably from produced or purchased foods. For example, the British study does not take account of the inedible part, and cooking foods reduces dioxin and DL-PCB contamination by 14 to 44% (Hori et al., 2005). The Spanish study used household food purchasing data instead of individual consumption data. The consumption levels may be overestimated since non-consumed foods may be included in this study.

Other international studies outside Europe, assessed total dioxin and dioxin-like compound intake. In the USA (Schecter, 2001), total intake is comparable to adult intake in France, estimated at 2.2 and 2.4 pg/kg bw/day respectively for men and women aged between 20 and 79 years old. The small difference may be due to different dietary habits and the time period of the analysis: for the US population, less consumption of fish products which have approximately the same contamination values, but higher levels of dioxins and dioxin-like PCBs in meat which are major contributors to total intake, especially for men. Total intake among children is also markedly higher than for adults (from 6.3 pg WHO-TEq/kg bw/d for boys aged between 1 and 11 years old to 2.7 for 12 to 19 year-old girls). Their relatively high consumption level in proportion to their body weight explains this difference. More recent study (FDA, 2004) based on TDS data, shows dietary PCDD/F exposure estimate between 9.6 pg WHO-TEQ/kg bw/month (0.32 pg WHO-TEQ/kg bw/d) and 26.8 pg WHO-TEQ/kg bw/month (0.89 pg WHO-TEQ/kg bw/d) for the whole population. This is quite a large range depending on whether non-detects equals zero, half the LOD or LOD, however, those results are in accordance with ours for dioxins only,

Considering the methodology used for this study, these results provide reference data for the background dioxin and DL-PCB exposure in France from 2002 to 2004. The consumption survey dates from 1999 but the individual and national survey of food intake in France (INCA)is representative for early 2000s food consumption. Diary recording, a validated method, was used; foods as ingredients were included; with regard to the sub-samples of adults and children, their characteristics were compared with national data from the population census (organised by INSEE: National Institute of Statistics and Economic Studies) and the differences were very small. Contamination data covers almost 90% of total diet (excluding beverages), several samples for one item. The fact that it dates from 2001 to 2004 makes it representative of a slightly earlier exposure. Note that the results presented here are calculated using data from the early 2000s (2001 to 2004), the 2000 estimation was also made with previous data. As the decline is probably continuous, today's exposure is presumably lower.

French dietary exposure has to be compared with the Provisional Tolerable Monthly Intake of 70 pg WHO-TEQ/kg bw/m i.e. 2.33 pg WHO-TEQ/kg bw/d (JECFA, 2002). On average, adult exposure is lower than this reference dose; children exceed the tolerable limit but the PTMI has to be understood as a life-long intake. This means that an excessive intake during childhood does not raise an additional health risk. An estimation of the life-long intake (from age 3 and over) provides an excess for 20 to 28% of the population (average life-long intake estimate of 2.0 pg WHO-TEQ/kg bw/d). As in the Italian study (Fattore et al, 2006), a non negligible part of the population is above the PTMI.

For dioxins only, intake for adults and children does not exceed the limit. For DL-PCBs, intake is above the limits. At present, no maximum value is established for DL-PCBs only, but if we consider that the new bound of 2.33 pg WHO-TEQ/kg bw/d is for both PCDD/Fs and DL-PCBs, it remains 1.33 pg WHO-TEQ/kg bw/d for DL-PCBs. This mean that on average, adults are below the threshold (1.2 pg WHO-TEQ/kg bw/d) while high consumers are above (p90=2.2 pg WHO-TEQ/kg bw/d). For children, the problem is evidently greater and related to their higher consumption level in proportion to their body weight: the mean is already over the limit (1.9 pg WHO-TEQ/kg bw/d for DL-PCBs only). Comparison of average intakes with PTMI seems inappropriate and results have to be interpret carefully: indeed, when intakes are above limits, it does not mean that there is a risk for the population because they are point estimates whereas limits are constructed for a life-long intake. Average intake allow comprehension about general exposure and trend analysis.

As for dioxin and dioxin-like PCB contamination levels in food products, for both populations (adults and children), the proportion of DL-PCBs is predominant in total exposure: 70% of the total.

Conclusion

This study must be viewed as a current reference point for the French population. Indeed, the methodology is recognised and validated, and data is representative: firstly the food consumption survey, INCA 1999, gives the most complete and up-to-date information about food consumption in France; secondly, occurrence data covers 90% of total diet; comparison is made with new regulations for occurrence data and total exposure. However, this study only reflects the primary exposure of the French population through their diet, since people who might be exposed to other key sources are not considered here.

It shows that, since 1999, exposure has decreased sharply (around 60% for dioxins only). As the data used for this estimation dates from 2001-2004, we can assume that the actual estimation for 2006 is probably lower in view of the rapidly decreasing trend. Moreover, the dioxin exposure level of 1 pg WHO-TEQ/kg bw/d has now been reached. Future efforts will have to focus on secondary emission sources like road traffic or wood burning fires.

However progress still has to be made in terms of total exposure, particularly for DL-PCBs. Indeed, the recent inclusion of DL-PCBs in the PTMI requires data on dioxin-like compounds to be collected in order to estimate overall exposure. The result is that they are major contributors (around 70%). The first step to reduce these compounds was the establishment of maximum levels in foods at a European level [amended Regulation (EC) N°466/2001]. There is still a need to monitor regularly exposure to dioxins and DL-PCBs in France.

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Figure 1. Comparison of levels of exposure to PCDD/Fs. estimated in 1999 and 2005 for the general population for the 7 food categories

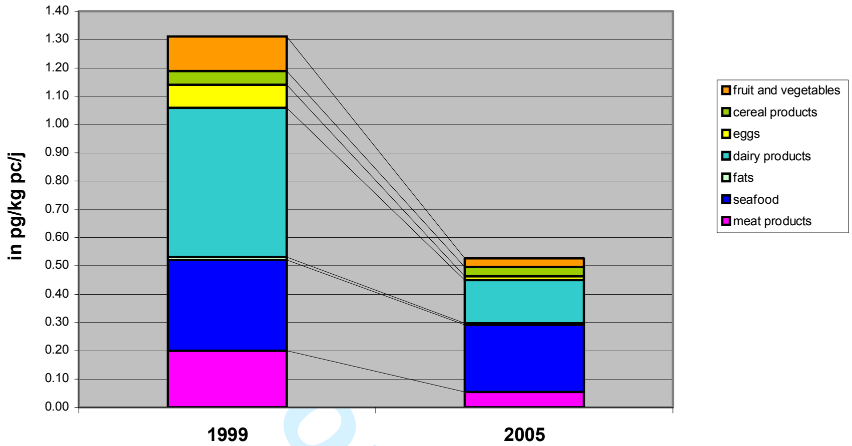
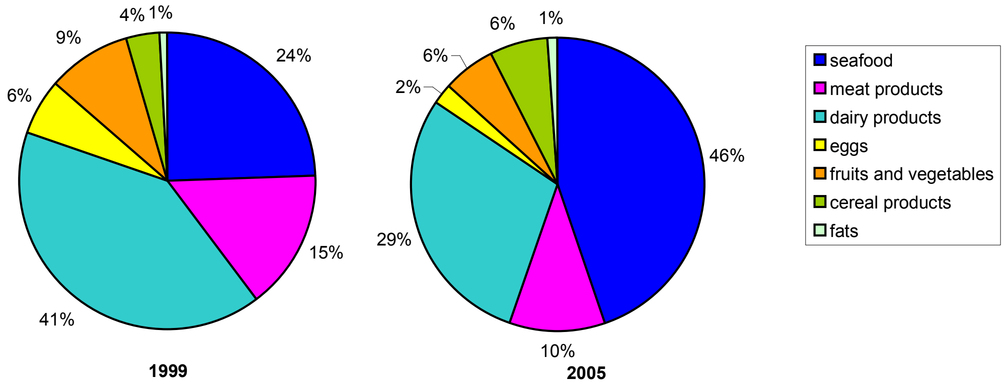
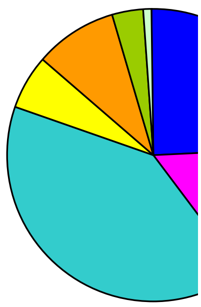


Figure 2. Contribution of the different food groups to PCDD/F exposure:



1999



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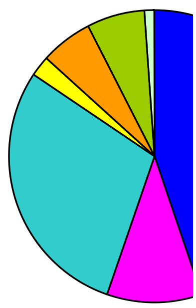

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Figure 3. Relative contribution of 7 food categories to total exposure (PCDD/F + DL-PCB) for adults (15 years and older) and children (3-14 years).

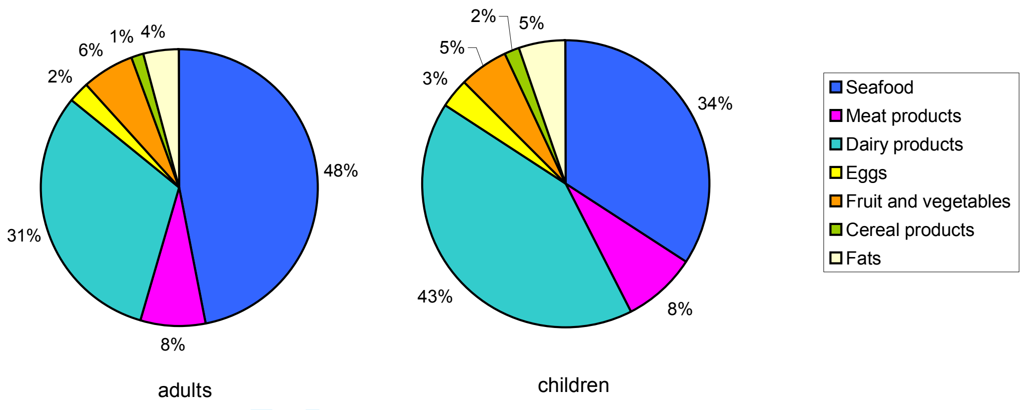

Table 1. Number of analysed samples per food group:

Products	**Number of samples**
Wild sea fish and wild freshwater fish	219
Farmed sea fish and farmed freshwater fish	148
Other sea food products (crustaceans. molluscs and cephalopods)	98
Fish oil	2
Meat (except poultry)	17
Poultry	38
Offal (liver)	39
Eggs	91
Milk and dairy products	102
Vegetables products (including fruits)	22
Cereal products	21
Total	**797**

Table 2. Mean and standard deviation of PCDD/F. DL-PCB or both PCDD/F+PCB-DL. expressed in pg TEQ/g fresh weight (FW) or lipid weight (LW) (indicated with * and in italic)

	PCDD/F (in pg TEQ/g LW or FW) (SD)	PCDD/F (in pg TEQ/g LW or FW) (SD)	DL-PCB (in pg TEQ/g LW or FW) (SD)	DL-PCB (in pg TEQ/g LW or FW) (SD)	total PCDD/F+PCB-DL (in pg TEQ/g LW or FW)	total PCDD/F+PCB-DL (in pg TEQ/g LW or FW)
Food groups	**Lowerbound (ND=0)**	**Upperbound (ND=LOD)**	**Lowerbound (ND=0)**	**Upperbound (ND=LOD)**	**Lowerbound (ND=0)**	**Upperbound (ND=LOD)**
Wild sea fish	0.42 (0.79)	0.42 (0.79)	2.29 (6.51)	2.29 (6.51)	2.72	2.72
Farmed sea fish	0.56 (0.33)	0.56 (0.33)	2.33 (2.40)	2.33 (2.40)	2.89	2.89
Farmed trout	0.17 (0.12)	0.17 (0.12)	0.58 (0.33)	0.58 (0.33)	0.75	0.75
Freshwater fish	0.38 (0.51)	0.38 (0.51)	2.35 (3.05)	2.35 (3.05)	2.73	2.73
Molluscs (mussels. oysters)	0.39 (0.41)	0.40 (6.97)	0.94 (1.49)	0.94 (1.49)	1.34	1.34
Cephalopods	0.18 (0.19)	0.18 (0.19)	0.55 (1.00)	0.55 (1.00)	0.73	0.73
Crustaceans	0.57 (0.85)	0.57 (0.85)	0.70 (1.11)	0.70 (1.11)	1.28	1.28
Meat*	0.24 (0.19)	0.36 (0.25)	0.74 (0.61)	0.74 (0.61)	0.99	1.11
- beef*	0.41 (0.14)	0.42 (0.21)	0.77 (0.23)	0.77 (0.23)	1.18	1.18
- lamb*	0.25 (0.08)	0.50 (0.33)	1.25 (0.66)	1.25 (0.66)	1.5	1.75
- pork*	0.05 (0.02)	0.21 (0.18)	0.37 (0.71)	0.37 (0.70)	0.42	0.59
Offal*	1.23 (1.21)	1.26 (1.21)	1.29 (2.35)	1.29 (2.35)	2.52	2.55
Poultry*	0.31 (0.25)	0.39 (0.28)	0.56 (0.73)	0.56 (0.73)	0.87	0.95
Eggs*	0.48 (0.61)	0.51 (0.62)	0.58 (1.98)	0.58 (1.98)	1.05	1.08
Butter*	0.29 (0.03)	0.29 (0.03)	0.52 (0.13)	0.52 (0.13)	0.82	0.82
Milk*	0.36 (0.13)	0.38 (0.13)	0.74 (0.28)	0.74 (0.28)	1.1	1.12
Fruit	0.01 (0.00)	0.01 (0.01)	0.01 (0.01)	0.01 (0.01)	0.01	0.02
Vegetables	0.01 (0.00)	0.01 (0.01)	0.01 (0.00)	0.01 (0.00)	0.01	0.02
Bread	0.01 (0.01)	0.01 (0.01)	0.00 (0.00)	0.00 (0.00)	0.01	0.01
Rice	0.00 (0.00)	0.00 (0.00)	0.01 (0.01)	0.01 (0.01)	0.01	0.01
Pasta	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0.00 (0.00)	0	0
Cereals	0.01 (0.00)	0.01 (0.00)	0.00 (0.00)	0.00 (0.00)	0.01	0.01
Vegetable oil*	0.07 (0.01)	0.17 (0.03)	0.18 (0.04)	0.18 (0.04)	0.25	0.36
Fish oil*	0.57 (0.16)	0.74 (0.11)	0.35 (0.46)	0.35 (0.46)	0.92	1.09

Values are based on 1998 WHO-TEFs.

Table 3. Dietary intake of PCDD/F: Comparison of exposure between CSHPF/Afssa study in 2000 and Afssa study in 2005 in the general population.

	ASPCC food survey CSHPF/Afssa 2000			INCA food survey Afssa 2005		
Food groups	**Mean dioxin exposure**		**% contribution**	**Mean dioxin exposure**		**% contribution**
	(pg TEQ_{NATO}/ pers/d)	**(pg TEQ_{NATO}/ kg b.w./jour)**		**(pg WHO-TEQ/ pers/d)**	**(pg WHO-TEQ/ kg b.w./d)**	
Meat products	9.83	0.20	15.3	3.24	0.06	10.5
Fish and fish products	17.22	0.32	24.4	13.97	0.24	44.7
Fats	0.52	0.01	0.8	0.36	0.01	1.1
Dairy Products	25.81	0.53	40.5	8.87	0.15	29.1
Eggs	4.04	0.08	6.1	0.77	0.01	2.5
Cereals products	2.21	0.05	3.8	1.94	0.03	6.4
Fruit and vegetables	6.12	0.12	9.2	1.78	0.03	5.7
Total	**65.73**	**1.31**	**100.0**	**30.93**	**0.53**	**100.0**

Table 4. Estimation of daily exposure to PCDD/Fs+DL-PCBs. PCDD/Fs and DL-PCBs for adults (15 years +) and children (3-14 years)

Population	**Total exposure in pg TEQ_{WHO}/kg bw/d**					% Contribution
	Mean	Median	Standard deviation	p90	p95	
Adults	**1.8**	**1.5**	**1.1**	**3.1**	**3.9**	
PCDD/F	*0.5*	*0.5*	*0.3*	*0.9*	*1.1*	*30.6*
DL-PCB	*1.2*	*1.0*	*0.8*	*2.2*	*2.8*	*69.4*
Children	**2.8**	**2.4**	**1.6**	**4.8**	**6.0**	
PCDD/F	*0.9*	*0.8*	*0.4*	*1.5*	*1.8*	*31.8*
DL-PCB	*1.9*	*1.6*	*1.1*	*3.3*	*4.2*	*68.2*

Table 5. Estimation of life-long intakes to PCDD/Fs+DL-PCBs. PCDD/Fs and DL-PCBs

Total exposure in pg TEQ_{WHO}/kg bw/d				
Mean	Median	Standard deviation	p90	p95
2,0	**1,9**	**0,5**	**2,6**	**3,0**

Table 6. Contribution of 7 food categories to total exposure. expressed in pg TEQ/kg bw/d and relative share of PCDD/Fs and DL-PCBs. expressed as a percentage. in adults (15 years and +): N=1474

Food category	Exposure in pg/kg bw/d					Percentage of consumers (in %)	DL-PCBs (in %)	PCDD/Fs (in %)
	Mean	Median	Standard deviation	p90	p95			
Seafood	**0.8**	**0.5**	**1.0**	**2.0**	**2.8**	**92.4**	**75.5**	**24.5**
Dairy products	**0.6**	**0.5**	**0.2**	**0.8**	**1.0**	**100.0**	**65.6**	**34.4**
Meat products	0.1	0.1	0.1	0.2	0.3	99.9	70.3	29.7
Fruit & vegetables	0.1	0.1	0.1	0.2	0.2	100.0	54.1	45.9
Fats	0.1	0.1	0.0	0.1	0.1	100.0	72.9	27.1
Eggs	0.0	0.0	0.0	0.1	0.1	98.8	54.5	45.5
Cereal products	0.0	0.0	0.0	0.0	0.0	100.0	26.8	73.2
Total exposure	**1.8**	**1.5**	**1.1**	**3.1**	**3.9**	**100.0**	**69.4**	**30.6**

Table 7: Summary of reference values:

	PCDD/F	DL-PCB
WHO, 1990	10 pg I-TEQ/ kg bw/d	-
WHO, 1998	1-4 pg WHO-TEQ/kg bw/d	
SCF, 2001	14 pg WHO-TEQ/kg bw/week (2 pg WHO-TEQ/kg bw/d)	
JECFA, 2002	70 pg WHO-TEQ/kg bw/month (2.33 pg WHO-TEQ/kg bw/d)	