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Assessment of seasonality in exposure to dioxins, furans and dioxin-like PCBs by using long-term food consumption data

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Key words: dioxins, dioxin-like PCBs, dietary exposure, seasonality, long-term dietary survey

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Abstract

According to the European Food Safety Authority (EFSA) guidance related to uncertainties in dietary exposure assessment [The EFSA Journal (2006) 438:1-54], exposure assessment based on short-term food consumption surveys, such as 24-hour recalls or 2 days records, tend to overestimate the long term exposure because of the assumption that the dietary pattern will be similar day-after-day over a lifetime. The aim of this study was to make an assessment of dietary exposure to polychlorinated dibenzodioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) also called 'dioxins' and 'dioxin-like PCBs' using, long-term household purchase and consumption survey data collected by TNS-Secodip. Weekly purchases of the major dioxins and dl-PCB vector products of these contaminants were collected for 328 single person households, who participated at TNS-Secodip consumption surveys from 2003 to 2005 and who were single person households in order to better estimate their consumption. These data were combined with average contamination levels of food products. Weekly gross average exposure was estimated at 10.2 pg Toxic Equivalent (WHOTEQ)/kg body wt/week (95% confidence interval [9.6, 10.9]). According to the typical shape of the distribution of individual weekly exposures, it is sensible to fit an exponential law to these data. The mean value was therefore 12.1 pg WHOTEQ /kg bw/week. This value is higher than the arithmetic mean because it better takes into account the inter-individual variability. It was estimated that about 20% persons of this sample were exceeding the current Health Based Guidance Value mainly due to high consumption of seafood and/or dairy products. Thanks to long survey duration (3 years) and the weekly recording of food consumption, it was possible to demonstrate the actual seasonality of dietary exposure to dioxins and dl-PCBs with a maximum between March and September; similar seasonality is observable for fish consumption. Autoregressive integrated moving average or ARIMA models were adjusted to the time series and it could be demonstrated that the number of times the upper limit of confidence intervals exceeds Provisional tolerable weekly intake (PTWI) is about 15 weeks per year in average. Finally, compared to the results obtained from data collected in the short term surveys (one week), this study does not suggest that short term consumption surveys tend to overestimate the long-term exposure.

53 Introduction

Introduction

The term “dioxins” encompasses a group of 75 polychlorinated dibenzo-*p*-dioxin (PCDD) (among which 2,3,7,8-tetrachlorodibenzo-*p*-dioxin or TCDD plays a key-role), and 135 polychlorinated dibenzofuran (PCDF) congeners. Polychlorinated biphenyls (PCBs) are chlorinated aromatic hydrocarbons which have been classified according to their biochemical and toxicological properties. Non-*ortho* and mono-*ortho* substituted PCBs show toxicological properties that are similar to dioxins. They are therefore often termed “dioxin-like PCBs” (dl-PCBs).

Dioxins and dl-PCBs are lipophilic compounds. They are extremely resistant towards chemical and biological degradation processes and therefore persist in the environment and accumulate in the food chain. It is considered that 95% of the human exposure to dioxins and related compounds is from the diet (De Mul, et al. 2008; Parzefall W, 2002). When ingested the compounds accumulate in the body and any adverse effects are likely to appear after years and to be related to the total body burden. This property underlines the importance of long-term dietary exposure surveys.

Many toxic effects are related to dioxin exposure like reproductive effects (Mocarelli, et al. 2008), neurodevelopmental effects and endocrine effects via thyroid toxicity (Giacomini, et al. 2006) even if various studies show controversial results (Wilhelm, et al. 2008). Recently a metabolic syndrome including increase blood pressure, high triglyceride levels as well as glucose intolerance was described and related to exposure to dioxins (Uemura, et al. 2009). Finally, 2,3,7,8-TCDD is considered to be carcinogen for human (Schwarz and Appel, 2005; (IARC, 1997) but for relatively high exposure compared with the one of the general population (Kociba, et al. 1978). The toxicity of all dioxins and dioxin-like PCBs is calculated relatively to 2,3,7,8-TCDD, one of the most potent of the chemicals on which most toxicological and epidemiological information is available. The analytical results are converted into toxic equivalents (TEQ). This conversion is based on the assumption that all 2,3,7,8-substituted PCDDs and PCDFs, as well as the dioxin-like PCBs, bind to the same Ah receptor, and show comparable qualitative effects, but with different potencies (Schwarz, 2005). The differences in toxicity are expressed in the toxic equivalency factors (TEFs).

Within all the congeners, 17 dioxins and 12 dioxin-like PCBs are of toxicological concern. These 29 substances are considered all together for risk assessment.

For the current study, the TEFs of dioxins and dioxin-like PCBs established in 1998 and revised in 2006 by the World Health Organization (WHO) were considered and noted WHOTEQ (Van den Berg, et al. 1998; Van den Berg, et al. 2006). WHO also established a Provisional Tolerable Monthly Intake (PTMI) of 70 pg WHOTEQ/kg body weight (BW) for dioxins and dl-PCBs (WHO, 2002). Similarly the EU established a Provisional Tolerable Weekly Intake (PTWI) of 14 pg WHOTEQ/kg body weight (BW) for dioxins and dl-PCBs (SCF, 2001).

The results of the most recent dietary exposure assessments show that this health based guidance value is often exceeded by a significant part of the population (SCF, 2001; WHO, 2001; AFSSA, 2005). However these assessments rely on food consumption data collected over short periods of time between 1 and 7 days which are expected to overestimate the exposure due to the under-estimation of the intra-individual variability i.e. food consumption observed on short period of time are likely to vary on longer term and therefore the resulting exposure is likely to be lower (WHO, 2005).

The objective of the present study was to estimate the long-term dietary exposure to dioxins and related compounds based on French household food consumption data collected over multiple years of consumption recording. This approach should allow highlighting the inter-individual variability as well as the specific contribution to dietary exposure of rarely consumed food like fish.

Material and methods

Food consumption survey

Various types of surveys can be used to assess dietary exposure (Kroes et al., 2002) but most of them are based on survey durations between 1 and 7 days. The current study used the household budget survey performed by TNS-Secodip (Boizot, 2005) and hereafter called TNS-Secodip in the text. In household consumption surveys, the amounts of foods and drinks purchased into the household are recorded. The TNS-Secodip panel corresponds to French households included by random sampling combined with quotas method in order to obtain a correct representativeness together with a relatively high response rate (EFSA, 2006;

Feinberg, et al. 2006). Household surveys do collect data every week during multiple years. Unfortunately, they do not provide information on actual consumption by household's individual members. Therefore, following rules were applied for selecting the sample of households adapted to this study:

- Select households made of only one individual, i.e. single person households;
- Select same single person households who regularly reported over the three years between 2003 and 2005;
- Discard households with less than 40 weeks/year;
- Discard households with partial recording of total consumption controlled from total energy intake;
- Discard households with missing data, such as absent body weight.

In the rest of the text we used the term “household” for “single person household”. In addition to the food consumption, the TNS-Secodip survey provides information on subjects like their socio-professional categories, their age, sex, body weight and place of residence.

Within the food items recorded by the survey participants, only those which can significantly contribute to the exposure to dioxins i.e. product of animal origin containing fat (De Mul, et al. 2008; Parzefall W, 2002, WHO, 2002), were further analyzed. In practice, all food containing less than 30% of ingredients potentially contaminated by dioxins and related compounds were excluded. For example, a pizza containing 10% of ham was not taken into account. Finally 31 food categories were considered and listed in Table 1.

(Table 1)

Within these food categories, 19 are foods containing fat from animal origin and which are therefore the main vectors for dioxins and related compounds. The other 12 food groups are composite foodstuffs containing at least 30 % of the main vectors as ingredient. For these composite foods, an adjustment factor of 30 % was systematically applied on the level of occurrence in the absence of accurate recipes.

The consumption was calculated per week, per person and per food category over 3 years.

139 **Contamination data**

140 Analytical results were collected by the French Ministry of Agriculture and Fisheries between
 141 2002 and 2006 on foods on the French market. Seven congeners were analyzed for PCDDs,
 142 10 congeners for PCDFs and 12 congeners for dl-PCBs (Commission Regulation, 2006).
 143 Results are expressed in pg WHOTEQ/g of fresh weight.

144 The weekly exposure was assessed for each household in a deterministic way by multiplying
 145 the individual weekly consumption of the food categories by the corresponding mean level of
 146 contamination. It should be mentioned that results below the Limit of Reporting (left-censored
 147 data) were replaced by the corresponding Limit of Reporting (Limit of Detection or Limit of
 148 Quantification). Due to the relatively low number of non-quantified results, the impact of
 149 substitution by the limit of reporting was limited when compared with the substitution by 0.
 150 This approach has also the advantage of not underestimating the exposure (WHO, 1995).

151 For dairy products, dioxin concentrations were extrapolated from the value analyzed in fat
 152 milk. For animal fat, dioxin concentrations were extrapolated from the values analyzed in
 153 meat and standardized accordingly to fat content. The percentage of fat in each food group
 154 was extracted from the French table of composition (REGAL) (Feinberg, et al. 1991).

155 **Dietary exposure assessment**

156 In order to be able to use household consumption survey data in the purpose of assessing
 157 dioxin dietary exposure several assumptions must be made:

- 158 — Food quantities that were purchased and recorded correspond to food quantities that
 159 were consumed and edible part was assumed to be 100% for all foods; this hypothesis
 160 may generate an overestimate of exposure;
- 161 — Only foods consumed at home are taken into account and selected foods when
 162 consumed out of home (restoration) are neglected; this hypothesis may generate an
 163 underestimate of exposure.

164 Individual household weekly exposure to PCDD/F and dl-PCB is calculated using a
 165 deterministic approach, given by equation (Eq. 1):

$$E_{i(k)} = \frac{\sum_{j=1}^J C_{ij(k)} \times T_j}{P_i} \quad \text{Eq.1}$$

Where:

- $E_{i(k)}$ household exposure i ($1 < i < I$) for week k ($1 < k < K$), expressed in pg WHOTEQ/kg BW/week;
- $C_{ij(k)}$ food quantity consumed for food category j ($1 < j < J$), for week k , for individual i , expressed in g/week;
- T_j estimated contaminant concentration for food category j , in pg kg⁻¹ fresh weight (FW);
- P_i body weight (BW) of individual i , in kg.

Statistical data processing was performed using JMP software release 7.0.1 (SAS Institute, Cary, NC, USA).

Results

Sample description

Three consecutive household consumption surveys were carried out by TNS-Secodip during 3 years, from 2003 up to 2005, i.e. over 157 weeks. A preliminary step consisted in harmonizing the different coding systems used over this period of time in order to merge the three surveys. Several single person households participated over all survey period and were selected. A preliminary sub-sample of 1161 single person households was then sorted out. But after applying the selection criteria described above, a working sample of 328 single person households was selected and used for the study.

Major descriptive characteristics of the working sample are given in Table 2. When referring to classical Body Mass Index (BMI) criterion, 50% of individuals are normal weight, 35% are overweight ($25 \text{ kg/m}^2 < \text{BMI} < 30 \text{ kg/m}^2$), 12% are obese ($\text{BMI} > 30 \text{ kg/m}^2$), and 3% are underweight ($\text{BMI} < 18.5 \text{ kg/m}^2$). Most represented geographical regions are Paris and its surroundings (25%), East-central region (10%), and South-east region (8%). Moreover, 14 French departments were not represented in the working sample, and TNS-Secodip purposely discards several departments, such as overseas departments, from the household purchase surveys because local consumption behavior is known to be different from the rest of the country. Main occupational categories of selected single person households were: 50%

occupied government and private positions and 44% were retired. Only 6% of the panel members were unemployed (not including retired persons), and no student participated.

(Table 2)

When considering these raw figures, it is obvious that the working sample was not representative of whole French population, as it is usually defined when using quota sampling for sampling plan. This bias was considered as negligible because the purpose of this study was to assess a possible seasonal effect on dioxin food exposure and the number of weeks with consistent food records was considered as the most important objective.

Household consumption estimate

According to notations used in equation 1, the working sample consisted in $I = 328$ households, $J = 31$ food categories, and $K = 157$ weeks. If all households should have consumed, at least, one time each food category each week, the theoretical number of records $C_{ij(k)}$ should be equal to $328 \times 31 \times 157 \approx 566\,000$ food consumption, expressed in g/kg BW/week. However, despite the selection criteria used to sample households, many individuals never consumed certain foods over this long period of time (3 years). The importance of non consumers is indicated in the column of Table 1 where percents of consumers are reported for each food category. Altogether, about 425 000 weekly food consumption were calculated for the 328 households, each value being, by itself, the weekly summation of several possible daily consumption.

Food categories consumed in larger quantities, i.e. purchased, by at least 95% of households over the survey period were fishes, milk and dairy products, beef meat, pork meat and poultry. On the contrary, foods consumed in smaller quantities were animal fats excluding butter and creams (less than 10%), butter and composite dishes containing cephalopods and mutton offal.

When looking at the gross average food consumption, results are consistent with those observed in other French food consumption surveys based on individual surveys. By range of magnitude, most frequent food categories were milk and yoghurts (14 g/kg BW/week), pork meat (5.33 g/kg BW/week), beef meat (3g/kg BW/week), and cheese (2.37 g/kg BW/week). When grouping food categories by principal types – dairy products, animal foods, seafood, eggs, dishes excluding seafood, and fats – it appears in the bottom of Table 1, that dairy products and animal foods embody the major part of the total consumption of dioxin food

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sources with almost 91% of total diet, complemented by seafood and composite dishes based on seafood (8%) and eggs (3%); fats and others composite dishes are negligible.

Because several foods, such as vegetable and fruits or beverages, were removed from the selection, it must be reminded that these data are not representative of some “average diet”. Only food categories that were recognized as potential vectors for dioxins were selected and used in the calculation of the global food consumption. Considering the aim of the study, it was a realistic choice, as a poorly consumed highly contaminated foods category may represent an important source for exposure.

Average dioxin concentration in foods

The average concentrations in the 31 food categories were estimated from analytical results, when available. The major source of data consisted in analytical results collected during different official control surveys performed. These values are put together in the last columns of Table 1.

For several foods, the average sum of pollutants (PCDD/F + dl-PCB) is not equal to the sum of average concentrations of individual pollutants (PCDD/F and dl-PCB). This is due to the fact that the number of analytical data used to calculate each mean is not the same for both pollutants. In column “Number”, first number indicates the total number of measurements, the second the number of measurements below LOD/LOQ. When data were left-censored, i.e. below LOD/LOQ, censored data were substituted by LOD or LOQ reported by the laboratories. For several food categories i.e. dairy products, animal fats and composite foods, no analytical measurements were available and “average” values were imputed as described in section Material and methods; they are marked by letter “E”.

According to these data, fishes are most contaminated foods with an average content close to 2.5 pg WHOTEQ/g fresh weight (FW). Other seafood are also significantly contaminated, but weakly consumed. All average PCDD/F + dl-PCB concentrations of animal foods are below 1 pg WHOTEQ/g FW. Because of their high fat contents, offal is always more polluted than meat, except for poultry. By decreasing order, the concentrations in animal foods are mutton, beef, pork and poultry. Average concentration in eggs is 0.14 pg WHOTEQ/g FW and is comparable to mutton meat while milk, with 0.06 pg WHOTEQ/g FW, is slightly polluted.

Relative proportions of PCDD/F compared to dl-PCB were estimated from available data (but are not given here) and are highly variable depending on the food category. This can be

explained by the fact that the origins of both these types of chemicals are different as they are differently accumulated in the various environmental compartments. As a rule of thumb, dl-PCB represents about 80% total contaminants burden for fishes, and falls to 50 to 60% for fats, animal foods, and eggs.

Estimation of dietary exposure

Average population exposure

A rough point estimate of dietary exposure can be directly calculated from data in Table 1 by multiplying food consumption by food concentration. The average exposures to PCDD/F + dl-PCB for each food categories are reported in the last column of the table. Thus, total average exposure is about 10.42 pg WHOTEQ /kg BW/week; this value can be compared to the Provisional tolerable weekly intake (PTWI) 14 pg WHOTEQ/kg BW/week, and the global situation seems to be satisfactory. The mean exposure from dairy products, fish and other sea foods represents 88% of the total (9.35 vs. 10.42 pg WHOTEQ/kg body weight/week). The first category is a major contributor because of its high and widely distributed consumption across the French population. The second is a major contributor because of a high mean level of contamination and despite of lower levels of consumption as well as percentage of consumers. In the bottom of Table 1, the contributions of the six principal food types were estimated in the same way. The major role of fishes and seafood can then be exhibited whereas they only represent 8% of total food consumption but 62% of dietary exposure. Next, dairy products contribute to 27% of dietary exposure but represent 65% of total food consumption.

Individual weekly exposures

However, for purpose of identifying any group at risk, average population exposure may be misleading as it does not give any indication on individual exposure distribution nor seasonal influence. Therefore, individual weekly exposure, denoted $E_{i(k)}$, were calculated from consumption data and average concentrations of PCDD/F + dl-PCB reported in Table 1 by applying equation 1. Because the number of weeks was 157 and the number of households 328 the total number of values should be $328 \times 157 = 51\,496$. But several records were missing due to holiday periods. The total number of individual weekly exposures was 41 789, i.e. 81% of expected number. This percent is large enough to be considered as representative of the seasonal/individual variations of dietary exposure to dioxin. Figure 1 illustrates the

histogram of $E_{i(k)}$ whatever the week or the household. Thus, it can be shown that there is a very large dispersion of data as they are ranging from 0.11 pg WHOTEQ/kg BW/week, while about 26% of estimated weekly dietary exposures are above PTWI and 0.5% above 100 pg WHOTEQ/kg BW/week.

(Figure 1)

The right-skew of data distribution is typical of many risk assessment studies on food contaminants when individual exposures are estimated instead of average population exposure (Tressou, et al. 2004; Crépet, et al. 2005). Obviously, data are not distributed following normal distribution. Because, the mean of normal distribution is estimated by the arithmetic mean, this means that this statistic is not well adapted to estimating population exposure. According to the typical shape of the histogram, it is sensible to fit an exponential law to these data. The probability density function of the exponential distribution is described by the following function (when $x \geq 0$) and fully defined by a single parameter λ :

$$f(x) = \lambda \exp(-\lambda x)$$

Then, the mean of an exponential law is equal to $1/\lambda$. Exponential probability density function was adjusted to individual weekly exposures and confirmed by a Kolmogorow-Smirnov adjustment test. The mean was estimated thanks to this new parameterization and observed value was 12.1 pg WHOTEQ /kg BW/week. This value is higher than the former arithmetic mean because it better takes into account the inter-individual variability.

It is also possible to use this statistical modeling to estimate the percent of weekly exposure data which are above the PTWI; about 27.1% weekly exposures exceed this Health Based Guidance Value. Separate exponential distribution laws were also fitted for each year and the percent of exceeding exposure values were estimated; they are ranging from 21 to 22%; this is still very high but also demonstrates the great stability of this percent over time.

At this stage it can be concluded from these data that there is a real risk of sporadic overexposure to dioxin for a non negligible portion of French population: about 1/5 weekly dietary exposures may be too high. It means that in average about 10 weeks per year, consumers are exposed to too high dioxin intake.

Contribution of fish to the overall dietary exposure

More in detail, the analysis of contributions in the case of dietary exposure exceeding the Health Based Guidance Value shows that it is mainly related to an increase in the consumption of fish and other seafood.

A complementary approach consists in selecting within all the individual weekly exposures, the ones in which a consumption of fish and other seafood was reported. This happens for about half of the cases, i.e. 19,070 persons $\times$ weeks over 41,789. It is therefore possible to estimate the contribution from fish and seafood noted $E_{i(k)}^*$, relative to the overall exposure noted $E_{i(k)}$ by calculating the ratio $B_{i(k)}$ for a considered individual during a considered week:

$$B_{i(k)} = \frac{E_{i(k)}^*}{E_{i(k)}} \quad \text{Eq. 2}$$

Figure 2 illustrates the frequency distribution of this ratio. It shows that in about 3% of cases fish and other seafood are the only contributors to the dietary exposure. In 25% of cases they represent 88% and in half of cases they represent 75% of the total dietary exposure.

These results globally emphasize the predominant role of fish consumption regarding dietary exposure to dioxins and related compounds. However, they may hide many specific situations where fish consumption poorly contributes to overall exposure as illustrated by the minimum value of the distribution, at 0.6%.

(Figure 2)

Average individual weekly exposures

When considering the distributions of $E_{i(k)}$ for each individual 157 weeks, the same typical exponential distribution can be observed. Average individual weekly exposures were then calculated for each of the 328 households, all years and weeks confounded, and reported in Figure 3. A log-normal distribution can be fitted to the data of this new histogram and confirmed by a Kolmogorow-Smirnov adjustment test. The mean of a log-normal distribution can be estimated by the geometric mean. The estimated value of the geometric mean is 10.2 pg WHOTEQ/kg BW/week and the 95% confidence interval is [9.6, 10.9]. Most interesting percentiles values are: 99.5 % is 77.8 pg; 97.5 % is 38.9 pg, and 50.0 % (median) is 10.5 pg.

(Figure 3)

Modeling weekly exposure as a function of time

Also starting from $E_{i(k)}$ values, it is possible to estimate the weekly average by summing over all households. The 157 obtained data can be represented as a function of the week when they were calculated. They form a time series that can be used to show evidence of seasonal variation. In order to ascertain this dependence with time, time series statistical techniques were used as described in (Pandit and Wu, 1983). It can be assumed that there is no trend because it was demonstrated that global average exposure was stable over year as illustrated in Table 3. Thus, it is not necessary to use detrending techniques and a simple exponential smoothing is sufficient to show the seasonality as illustrated by Figure 4. Observed averages and averages predicted by using the smoothing model with their 95% confidence intervals are simultaneously presented on the graphics.

(Table 3)

Average exposure to dioxin is actually constant over the three-year period of the study (around 12 WHOTEQ/kg BW/week), but increase up to a maximum of 13 pg WHOTEQ/kg BW/week between March and September and decrease down to a minimum of 11 pg WHOTEQ/kg BW/week between December and February. The magnitude of these average variations is small if confidence intervals are not taken into consideration. But if they are considered, the risk to exceed PTWI is rather frequent. For instance, the upper limits of the confidence intervals are exceeding PTWI about 35 weeks per year.

(Figure 4)

Complete and partial autocorrelation functions were also calculated in order to extract parameters for what is called the seasonal Autoregressive integrated moving average (ARIMA) model in time series theory. This kind of model can be used for predicting no observed situations, and the results observed during 2003 to 2005 were used to build the model and predict exposures for 2006. The seasonal coefficient used for the ARIMA model was 13 and the order coefficient 7; according to classical nomenclature, this model is defined as ARIMA (7,0,7)₁₃ and illustrated on Figure 5. Confidence intervals are narrower because this model is more explicative than simple smoothing as it takes into account the role of seasons. Therefore, the number of times the upper limit of confidence intervals are exceeding PTWI is smaller and only 15 weeks per year in average. However, the variations from one week to another may still be very large and can be explained by the uncertainty of exposure estimates.

(Figure 5)

Obviously, the observed stability over time is emphasized by the fact point estimate and unique dioxin concentrations were used for exposure assessment. The application of probabilistic risk assessment technique and several concentration levels should increase the variability and slightly modify these conclusions, although no actual decrease of dioxin concentration in fish was observed during the considered years.

Discussion and conclusion

Long-term single person household purchase survey was used to estimate long-term food consumption for 328 single person households. These data were combined to average concentrations of PCDD/F + dl-PCB in 31 selected foods categories. It was then possible to derive different dietary exposure values. It was mainly possible to demonstrate that dietary exposure to dioxin may vary with the season and that, if the global point estimates may lead to conclude that they are below PTWI, individual variations and seasonal effect are important and may induce frequent exposure above the Health Based Guidance Value. Several important methodological conclusions may also be drawn from this study.

- 1) Household purchase surveys are interesting to show evidence of seasonality for long-term dietary exposure and consequently food risk assessment. Because they are performed over a period of time much longer than few days records or 24-hour recalls, it is possible assess more accurately the consumption habits of a large panel. For this study, it appears that fish consumption is not constant over months and influences the exposure to dioxin. Recent report from the French fish marketing board (Office FranceAgrimer) which is in charge of the regulation of fish markets in France, partially confirms the seasonality of fish consumption in relation to the seasonal variations of stocks and fishing campaigns (FranceAgrimer, 2008).
- 2) Because seasonality of dioxin exposure covers one year collecting data over a short period of time, likewise few days record or 24-hour recalls may conceal the season effect and neglect the modifications of consumption behavior as a function of time On the contrary, it may be sufficient to collect data over one year or to correct the sampling for seasonal variations over a period of 1 year. A recent study realized by Tard *et al.* 2007 has been used to define the French exposure to total dioxins expressed in toxic equivalents (TEQs). The study combines concentrations data from French

monitoring programs (2001-2004) with the first French Individual Consumption Survey (INCA 1999). This survey is based on seven 24-hour diet records corrected for seasonal variations by a sampling of subjects distributed over 1 year. The daily exposure to total dioxins was estimated to be on average 1.8 pg WHOTEQ/kg BW/day, i.e. 12.6 pg WHOTEQ/kg BW/week, and very similar to our own average estimates. Therefore, it seems that there is a good convergence of results for average point estimates whatever the food survey methodology. But only long-term surveys may help identifying a seasonal effect.

3) This study also confirms the results of other risk assessment studies and demonstrates that the possibility for an individual to exceed PTWI is a rather frequent event as it is observable in about 20% of weeks; it is not a rare event.

From a general point of view, the estimation of the uncertainty of dietary exposure simultaneously depends on uncertainty of consumption data and uncertainty of contamination data. Obviously, this study may present some major sources of uncertainty that may explain the large variability of observed data.

Firstly, when considering consumption data:

□ Consumption outside home, auto-consumption and non-consumption were not accounted and total food consumption is certainly systematically biased. Food categories were selected for their assumed usual high concentration in dioxin. But, other food categories may also contribute to global exposure. For instance, in its study of 2005, AFSSA stated that cereals and fruits and vegetables may represent 1% and 6% respectively of total PCDD-PCDF + dl-PCB dietary exposure for adults (Moccarelli, et al. 2008). As consumption of vegetal foods was neglected and consumption of butter seemed very low in our study, some underestimation of total dioxin dietary exposure is likely. Although vegetal foods are little contaminated, they are highly consumed in France and they may represent a non negligible contribution to total exposure.

□ The fact that we were able to use three year-survey must be put into balance. Such a long-term survey cannot be easily achieved by other recording methods. The working sample was sampled by only selecting single person household. Consequently, it is a small sample which presents its own characteristics and extrapolation to the rest of

French population must be cautiously made, as it is not evident that single person household consumption behavior is comparable to other households.

- However, no food survey methodology can be claimed as unbiased and each may underestimate or overestimate some aspects of food consumption. Therefore, we were always very cautious in contrasting our results with those obtained by other means. This was only possible for global average exposure estimates and presented results can be considered as very consistent. But the great advantage of used data was their ability to demonstrate the seasonality of dioxin exposure.

Secondly, when considering contamination data:

- Only “average” values were used and exposure was calculated in a deterministic way while it is more interesting to apply a probabilistic approach (Feinberg, et al. 2006). But this requires more contamination data than we were actually able to gather (mainly for these food categories where average values were imputed) and a more important computational effort.
- Because left-censored data were substituted by detection limit, exposure was not underestimated. But, for some foods, the small number of analytical data for calculating the average, or the absence of any analytical data, greatly contributes to total uncertainty.

Despite these precautions, if one extrapolates the results to entire French population, it can be concluded that, if the estimate is restricted to global average, the average weekly exposure is 12 pg WHO TEQ/kg BW/week and below the health based guidance value of 14 pg WHO TEQ/kg BW/week. This value is comparable to the estimates published by the Scientific Committee on Food (SCF) of the European Commission in 2001 (SCF, 2001). These estimates were derived from various short-term studies, and it seems unnecessary to use long-term studies to assess average exposures and reduce the uncertainty. However, our study shows that in average, people who consume foods contaminated by dioxin are above PTWI 10 weeks/year, because about 20% of individual estimated weekly exposures are above PTWI each year.

Finally, because dioxins and related compounds are accumulating in the body, this study could be completed by Kinetic Dietary Exposure Model (Verger et al. 2007; Bertail et al. 2010) which take into account the period of time between two food consumption and the half-life of PCDD/F and dl-PCB in the body, in order to evaluate the total body burden of

consumers and estimate more realistically the probability to exceed the Health Based Guidance Value. Dynamic models were developed for methyl mercury from fish but they require more sophisticated modeling to calculate the dietary exposure from various inter-dependent food groups.

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Tables

Table 1: Food categories selected for the study: average weekly consumption expressed as g/kg Body Weight/week, % of consumers and average dioxin contents. Remarkable values are underlined

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Food categories	Food consumption		Average contamination pg WHOTEQ/g FW ⁽¹⁾				Exposure pg/kg BW/week
	Average g/kg BW/week	% consumers	Number (²)	PCDD/F	dl-PCB	PCDD/F + dl- PCB	
Fishes	2.8	100%	578 – 583	0.340	<u>2.130</u>	<u>2.470</u>	<u>5.9360</u>
Mollusks	0.27	46%	42 – 43	0.290	0.570	0.860	0.2295
Shellfishes	0.53	79%	70 – 71	0.240	0.220	0.460	0.2385
Cephalopods	0.02	15%	55	0.180	0.370	0.550	0.0170
Milk	<u>14.1</u>	98%	35	0.020	0.040	0.060	0.8442
Butter	0.001	3%	24	0.200	0.350	0.550	0.0006
Cheese and creams	2.4	98%	E ⁽³⁾	0.150	0.330	0.480	<u>1.1376</u>
White cheese	0.49	58%	E	0.080	0.170	0.250	0.1225
Yoghurts, dairy desserts and ice-creams	<u>15.0</u>	100%	E	0.020	0.040	0.060	0.7485
Pork meat and delicatessen	5.3	99%	13	0.020	0.020	0.040	0.2665
Pork offal	0.16	52%	7	0.060	0.030	0.090	0.0144
Mutton offal	0.03	18%	2	0.420	0.460	0.880	0.0264
Mutton meat	0.58	75%	10	0.040	0.070	0.110	0.0638
Poultry offal	0.15	70%	6	0.020	0.130	0.150	0.0225
Poultry meat	1.97	99%	46	0.020	0.020	0.040	0.0788
Beef meat	3.0	97%	23	0.030	0.060	0.090	0.3000
Beef offal	0.27	64%	8	0.040	0.070	0.110	0.0297
Animal fats excluding butter and creams	0.01	8%	E	0.450	0.540	0.990	0.0099
Eggs	1.7	94%	155	0.070	0.070	0.140	0.2366
Dishes with >30% Fishes	0.11	51%	E	0.100	0.640	0.740	0.0704
Dishes with >30% Mollusks	0.004	9%	E	0.090	0.170	0.260	0.0010
Dishes with >30% Shellfishes	0.01	15%	E	0.070	0.070	0.140	0.0014
Dishes with >30% Cheese	0.09	35%	E	0.040	0.100	0.140	0.0135
Dishes with >30% Pork meat	0.09	47%	E	0.010	0.010	0.020	0.0009
Dishes with >30% Pork offal	0.01	16%	E	0.020	0.010	0.030	0.0003
Dishes with >30% Mutton offal	0.02	13%	E	0.120	0.140	0.260	0.0052
Dishes with >30% Poultry offal	0.0001	1%	E	0.005	0.040	0.045	0.0000
Dishes with >30% Poultry meat	0.14	54%	E	0.005	0.010	0.015	0.0014
Dishes with >30% Beef meat	0.17	54%	E	0.010	0.020	0.030	0.0051
Dishes with >30% Beef offal	0.002	4%	E	0.010	0.020	0.030	0.0001
Dishes with >30% Eggs	0.01	16%	E	0.020	0.020	0.040	0.0004
Grouping							
Dairy products	31.9	65%					<u>2.85</u>
Animal foods	11.5	23%					0.80
Seafood	3.7	8%					<u>6.49</u>
Eggs	1.7	3%					0.24
Dishes (excluding seafood)	0.5	1%					0.03
Fats	0.01	0%					0.01
Total	49.4						10.42

Footnotes

- (1) Data from official control plans of Direction Générale de l'Alimentation (DGAL) a division of French Ministry of Agriculture. FW stands for fresh weight.
- (2) Number: first number indicates the number of quantified data and the the second number includes left-censored data below determination limits (LOD or LOQ).
- (3) E extrapolated concentration

Table 2: Description of the survey sample

	Women	Men	Total
Number	225	103	328
% total	69%	31%	
Average age (years)	62.1 (± 14.3)	54.3 (± 14.4)	59.7 (± 14.8)
- More than 50 years	80%	58%	
Average BMI	24.90 (± 4.52)	25.40 (± 3.77)	25.06 (± 4.30)

Footnotes

BMI : Body mass Index expressed as kg/m² (SD between parentheses)

Table 3: Average individual weekly exposure per year to PCDD/PCDF/dl-PCB (expressed in pg WHOTEQ /kg BW/week

	Average	Standard deviation	Minimum	Maximum	(1)	%	(2)	%
2003	10.7	7.8	1.4	54.5	71	22%		
2004	10.5	7.2	0.8	49.2	64	20%		
2005	10.3	7.3	1.0	56.6	67	20%	99	30%
2003 et 2004	10.6	7.3	1.4	48.7	65	20%		
2004 et 2005	10.4	7.0	0.9	52. 6	56	17%	59	18%
2003 à 2005	10.5	7.1	1.6	50.2	61	19%	44	13%

Footnotes

- (1) Number of persons exceeding PTWI
- (2) Number of persons with at least one yearly average exposure that exceeds PTWI

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