ASSESSMENT OF DIETARY INTAKE OF DIOXINS, FURANS AND DIOXIN-LIKE PCBS FOR THE FRENCH POPULATION

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Introduction

The only estimation of dietary exposure to dioxins in France was done in 2000[1]. But this estimate didn't take into account the dioxin-like PCBs. It was thus necessary to update this first study in order to begin a monitoring of dietary exposure to dioxins and PCBs in France.

Materials and methods

Sampling of foods

Data on occurrence of PCDD/Fs and DL-PCBs in food products (fishes, molluscs and crustaceans, meat, offals, milk, butter, eggs, fruit and vegetables, cereal products) consumed in France were collected from national monitoring programs carried out by food administrations during 4 years (2001 to 2004). The foods were sampled randomly in order to represent the French consumption. For the fishes, the sample was stratified according to the harbour of origin in order to be sure that the different fishing areas (Mediterranean Sea, North Sea, English Channel, Atlantic Ocean, other fishing areas) were correctly represented.

Chemical analysis

The specifications defined by the EU directive 2002/69 were applied. For all the analyses, ^{13}C labelled congeners were added before extraction for quantification according to the isotopic dilution method. The lipidic fraction was extracted by accelerated solvent extraction (ASE). Solvent were evaporated in order to determine the amount of fat. After fat extraction and purification steps, a quantification standard was added in order to evaluate the recovery yields. The fractions were analysed by gas phase chromatography coupled to high-resolution mass spectrometry (HRGC/HRMS). The analyses were done by LABERCA and CARSO laboratories. The analyses were provided to Afssa by the French department of Agriculture (animal products), the French department of Consumption (vegetal products), and the National Centre of the Dairy Industry (CNIEL) for the dairy products.

Exposure assessment method

The total concentration has been calculated with two methods, firstly assuming non detects equal zero (lower bound estimate) secondly assuming non detects equal LOD (upper bound estimate). Using a deterministic exposure modelling, those contamination data were combined with individual food consumption. Intake data was obtained from a diary record on 7 consecutive days from the last national dietary survey (INCA survey, 1999) including 2492 individuals (from 3 years old). The mean contamination of each food was combined with its consumption for each of the 23 food categories studied. Real individual body weight recorded in the survey were used in the calculation of the exposure. The following formula was used :

$$E_i = (1/p_i) x \Sigma_j C_{ij} T_j$$

Where :
E_i is the exposure of individual i
p_i is the body weight of individual i
C_{ij} is the intake of the food j by the individual i
T_j is the mean contamination of the food j

Two exposure studies were done. In order to be as comparable as possible to the 2000 study on PCDD/F exposure, a first estimate was done without any use of recipes to convert foods to primary agricultural products. The second exposure assessment for PCDD/F and DL-PCBs used a new exposure assessment method with recipes and conversion of foods to primary agricultural products.

Results and discussion

Food contamination

Analytical results were available for 797 individual food samples : 219 wild fishes of sea and freshwater, 148 farmed fishes, 98 crustaceans, molluscs and cephalopods, 2 fish oils, 17 meats except chicken, 38 chickens, 39 offals (livers), 91 eggs, 102 milks and milk products, 22 fruit and vegetables, 21 cereal products.

Upper bound concentrations of PCDD/Fs + DL-PCBs for animal products (meat, milk, butter, eggs) are between 0.8 and 1.1 pg WHO-TEQ/g fat; livers exceed 2.5 pg WHO-TEQ/g fat. Fish products are around 2.8 pg WHO-TEQ/g fresh weight excepted farmed trout (0.8 pg WHO-TEQ/g fresh weight); same concentrations are found in other fishery products (crustacean, shellfish). Vegetables concentrations do not exceed 0.01 pg WHO-TEQ/g fresh weight. Contaminations of foods in general and of farmed, wild fishes and molluscs are closed to other recent European data ([2,3,4]).

Table 1 : Mean contamination (SD) of 24 food groups for PCDD/Fs, DL-PCBs (in pg WHO-TEQ/ g product except for * in pg WHO-TEQ/ g fat), upperbound estimate (ND=LOD)

Food group	PCDD/Fs(pg WHO-TEQ/g product or * fat)	DL-PCBs(pg WHO-TEQ/g product or * fat)
Wild salt-water fishes	0.42 (0.79)	2.29 (6.51)
Farmed salmons and salt water fishes	0.56 (0.33)	2.33 (2.40)
Farmed trout	0.17 (0.12)	0.58 (0.33)
Other fresh-water fishes	0.38 (0.51)	2.35 (3.05)
Molluscs (oysters, mussels)	0.40 (0.41)	0.94 (1.49)
Cephalopods	0.18 (0.19)	0.55 (1.00)
Crustaceans	0.57 (0.85)	0.70 (1.11)
Meat *	*0.36 (0.25)*	*0.74 (0.61)*
- Beef *	*0.42 (0.21)*	*0.77 (0.23)*
- Mutton *	*0.50 (0.33)*	*1.25 (0.66)*
- Pork *	*0.21 (0.18)*	*0.37 (0.70)*
Liver *	*1.26 (1.21)*	*1.29 (2.35)*
Chicken *	*0.39 (0.28)*	*0.56 (0.73)*
Eggs *	*0.51 (0.62)*	*0.58 (1.98)*
Butter *	*0.29 (0.03)*	*0.52 (0.13)*
Milk *	*0.38 (0.13)*	*0.74 (0.38)*
Fruit	0.01 (0.01)	0.01 (0.01)
Vegetables	0.01 (0.01)	0.01 (0.00)
Bread	0.01 (0.01)	0.00 (0.00)
Rice	0.00 (0.00)	0.01 (0.01)
Pastas	0.00 (0.00)	0.00 (0.00)
Other cereal products	0.01 (0.00)	0.00 (0.00)
Oils *	*0.17 (0.03)*	*0.18 (0.04)*
Fish oil *	0.74 (0.11)	0.35 (0.46)

Exposure of the French population to PCDD/Fs in 2000 and 2005
Considering PCDD/Fs only, the calculated intake (whole population : children and adults) is 0.53 pg WHO-TEQ/kg bw/d that means about 60% less than the earlier estimation made in 2000. The results between 2000 and 2005 are not strictly comparables because the use of NATO TEQ in the first study instead of WHO TEQ. However, NATO TEQ are usually lower than WHO TEQ. So the decrease of the exposure is probably underestimated. This decrease of the exposure results from a diminution of the contribution for all food groups. However, we observed highest decreases for the contributions of milk products, meat products and eggs. We used lowerbound contamination figures for this comparison but for the year 2005 where both upperbound and lowerbound figures are available there was no difference of exposure between the two methods.

Table 2 : Mean exposure to PCDD/Fs of the French population in 1999 and 2005 and contributions of food groups to the exposure

	2000			2005		
Food groups	Mean PCDD/F exposure of the French population aged 3 y old and over (NATO TEQ)		Contribution	Mean PCDD/F exposure of the French population aged 3 y old and over (WHO TEQ)		Contribution
	(pg/pers/ day)	(pg/kg b.w./day)	%	(pg/pers/ day)	(pg/kg b.w./day)	%
Meat and meat products	9,83	0,20	15,3	3,24	0,06	10,5
Fish and seafood	17,22	0,32	24,4	13,97	0,24	44,7
Fats excluding butter	0,52	0,01	0,8	0,36	0,01	1,1
Dairy products including butter	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
Cereal 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

Exposure of the French population to PCDD/Fs and DL-PCBs in 2005
The mean calculated intake of PCDD/Fs + DL-PCBs is 1.8 pg WHO-TEQ/kg bw/d (median = 1.5) for adults and 2.8 pg WHO-TEQ/kg bw/d (median = 2.4) for children. The 95th percentile of intake is 3.9 pg WHO-TEQ/kg bw/d for adults aged 15+ and 6.0 pg WHO-TEQ/kg bw/d for children aged 3-14.

These exposure figures are closed to other recent publications in European countries ([5, 6]).

The contribution of different food groups to the total intake of PCDD/Fs + DL-PCBs show the great importance of fish and milk products, respectively 48% and 31% for adults and 34% and 43% for children. The total of animal products (including meat) represents more than 85% of total intake.

On the total intake, the proportion of PCDD/Fs-DL-PCBs is on average 30%-70% for both children and adults. Considering the life-long average intake, 20 to 28% of the population has an intake above the tolerable monthly intake for dioxins and dioxin-like PCBs of 70 pg WHO-TEQ/kg bw/month settled in 2001 by the JECFA.

Taking into account those new figures, the exposure to dioxins has significantly decrease since the last 5 years, being now lower than the reference dose of 1 pg WHO-TEQ/kg bw/d for almost the whole adult population. Regarding the dioxin-like PCBs, strain still have to be achieved given that estimation of both dioxins and dioxin-like PCBs is above the tolerable monthly intake of 70 pg WHO-TEQ/kg bw/month. Major reduction in

exposition is due to PCDD/F; diminution of DL-PCB exposure will have to be the upcoming objective with the setting up of the EU maximum levels and of targets of food contamination.

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