HUMAN DIETARY EXPOSURE TO PCDD/PCDFs AND PCBs IN CATALONIA, SPAIN

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Introduction

Humans are primarily exposed to most persistent organic pollutants (POPs) by eating food contaminated by these chemicals[1]. Polychlorinated dibenzo-p-dioxins (PCDD) and dibenzofurans (PCDFs), and polychlorinated biphenyls (PCBs) are among the most known and investigated POPs. During 2000-2002, we measured the levels of a number of chemical contaminants in several groups of foodstuffs purchased from Catalonia (Spain). Dietary intake of the pollutants, including PCDD/PCDFs and PCBs, was subsequently estimated for various age and gender groups of the population of that country[2]. The purpose of the present study was to establish the temporal trend in the total dietary intake of the same environmental pollutants by the population of Catalonia, some years after our first market basket study. For it, foodstuffs belonging to the food groups assessed in the previous survey were analyzed and the dietary intake of PCDD/PCDFs and PCBs was again determined.

Materials and methods

Sampling

In March-June 2006, food samples were randomly acquired in local markets, big supermarkets, and grocery stores from 12 representative cities of Catalonia. For collection of samples, 2 groups were made up. The first group included meat of beef (steak, hamburger), pork (loin, sausage), chicken (breast), and lamb (steak); vegetables and tubers (lettuce, tomato, cauliflower, string bean, potato); fruits (apple, orange, pear, banana), and eggs. Because in the first group most products are usually retailed, their origins could be very diverse in the different cities. The second group included cow milk (whole, semi-skimmed) and dairy products (yoghurt, cheese); cereals (French and sandwich bread, pasta, rice); pulses (lentil, haricot bean); oils (olive, sunflower) and fats (margarine, butter); meat products (boiled ham, hot dogs, salami), and bakery products (croissant, cookie, fairy cake).

Analytical techniques

Analytical procedure was based on the US EPA method 8290 for PCDD/PCDFs, and on the US EPA Method 1668 and JIS K 0311 for PCBs. Toxic equivalents (TEQ) of the analyzed PCDD/PCDFs and dioxin-like PCBs (DL-PCBs) were calculated using the WHO-TEF values[3].

Dietary exposure estimates

Consumption data by the general population of Catalonia of the analyzed foodstuffs were obtained from recent studies[4, 5]. The population was divided into four age groups: children (4–9 years), adolescents (10–19 years), adults (20–65 years), and seniors (> 65 years). In turn, each group was subdivided according to gender. To estimate dietary PCDD/PCDF and PCB intake, only the food items analyzed in the present survey were considered. For each food group, intake was calculated by summing the results of multiplying the PCDD/PCDF and PCB concentrations in the specific food item by the amount (proportionally estimated) consumed of that item. Finally, estimated total dietary intake of PCDD/PCDFs and PCBs was obtained by summing the respective intakes from each food group. When a concentration was under the limit of detection (LOD), daily intakes were calculated assuming the respective values would be equal to one-half of that LOD (ND = 1/2 LOD).

Results and discussion

Levels of PCDD/PCDFs

The highest level of 2,3,7,8-TCDD, the most toxic congener, corresponded to oils and fats (0.057 ng/kg) followed by fish and seafood (0.024 ng/kg), while the lowest levels of this congener were found in vegetables, tubers, and fruits (0.001 ng/kg). For each food group, the highest TEQ values corresponded also to oils and fats (0.223 ng WHO-TEQ/kg), followed by fish and seafood (0.131 ng WHO-TEQ/kg) and dairy products (0.057 ng WHO-TEQ/kg), while the lowest levels were found in fruits (0.003 ng WHO-TEQ/kg), as well as in vegetables and milk (0.009 ng WHO-TEQ/kg). In our previous (2000) survey[6], the maximum WHO-TEQ values corresponded also to the fish and seafood group (fresh plus tinned products), followed by oils and fats, and dairy products, whereas the lowest concentrations were found in semi-skimmed milk, vegetables, and fruits.

Levels of PCBs

The highest sum (ng/kg of fresh weight) of the 18 PCB levels, as well the highest WHO-TEQ values (ng WHO-TEQ/kg of fresh weight) corresponded to the groups of fish and seafood (16,265 and 0.76) followed by oils and fats (674 and 0.17) and dairy products (321 and 0.04). In contrast, the lowest sum (ng/kg) and WHO-TEQ values (ng/kg of fresh weight) were observed in fruits (11 and 0.004), and vegetables (24 and 0.005) and tubers (17 and 0.006). Taken individually, the highest levels corresponded generally to PCB 153 (meat and meat products, fish and seafood, milk and dairy products, cereals, pulses, and oils and fats) and PCB 28 (vegetables and tubers, fruits, eggs, and bakery products). In those food groups showing the highest TEQ values of DL-PCBs, the greatest contribution to the TEQ corresponded to PCB 118 and PCB 156. Among the PCB congeners analyzed in our previous survey[7], generally, PCB 153 showed also the highest concentrations. As in the present study, the lowest levels corresponded mainly to PCB 126 and PCB 169.

Dietary exposure estimates

The dietary intake (pg WHO-TEQ/kg body weight/day) of PCDD/PCDFs, DL-PCBs, and PCDD/PCDFs plus DL-PCBs by the population of Catalonia according to gender and age is depicted in Fig. 1. The highest intake corresponded to children (4-9 years old), with boys showing slightly higher intakes than girls. By contrast, the lowest intake was observed in individuals aged > 65 years, with females showing lower values than males. The intake through the diet of PCDD/PCDFs and DL-PCBs by a standard adult male living in Catalonia is summarized in Table 1. The current dietary intakes of PCDD/PCDFs, DL-PCBs, and PCDD/PCDFs plus DL-PCBs were estimated to be 25.7, 52.4, and 78.1 pg WHO-TEQ/day vs. 95.4, 150.1, and 245.5 pg WHO-TEQ/day found in our previous survey[6, 7]. It means reductions of 73%, 65%, and 68%, for PCDD/PCDFs, DL-PCBs, and PCDD/PCDFs plus DL-PCBs, respectively. Given in pg/kg body weight/day, for an adult male of 70 kg body weight, the intake of PCDD/PCDFs plus DL-PCBs was reduced from 3.51 in 2000 to the current 1.12.

The results of the present study show a very notable reduction in the dietary exposure to PCDD/PCDFs and DL-PCBs between 2000 and 2006 (68% for a standard adult male). However, it is very important to note that the sources for consumption data in Catalonia were different in both surveys. In the 2000 survey, data were taken from Capdevila et al.[8], while in the current study calculations of estimated PCDD/PCDF and DL-PCB intake were based on more recent consumption data[4]. Although decreases in the intake of these pollutants were observed in all food groups, the important quantitative decline was mainly due to the reductions in the contribution to total intake of fish and seafood, and dairy products, followed by cereals, meat and meat products, and oils and fats. Some reduction was probably due to the decrease in the total consumption of the different foodstuffs between the 2000 and 2006 surveys. In recent years, other studies concerning the concentrations of PCDD/PCDFs in foodstuffs and human dietary intake have been performed in a specific area of Catalonia, Tarragona County. In a Total Diet Study in which food samples were collected in 2002, the mean PCDD/PCDF intake was 63.8 pg WHO-TEQ/day[9], a value notably lower than that found in a previous study carried out by our research group at the same geographical zone, 210.1 pg I-TEQ/day[10]. Recently, total dietary intake in that area was estimated to be 27.8 pg WHO-TEQ/day[11]. Although the studies performed in Tarragona County did not include PCBs, the estimated intakes of PCDD/PCDFs only, are quite analogous to those reported by Martí-Cid et al.[11] and the intake of the current study. In the survey performed by Martí-Cid et al.[11], fish and seafood (28%),

oils and fats (22%), eggs (17%), and dairy products (11%) were the most important contributors to PCDD/PCDF intake, while pulses (1%), milk (2%), vegetables (3%) and fruits (3%) showed the lowest contribution-TEQ.

With respect to the current health risks derived from dietary exposure to PCDD/PCDFs and DL-PCBs, the non-carcinogenic risk index was calculated by dividing the total daily intake by the tolerable intake established by the WHO in the range 1-4 pg WHO-TEQ/kg of body weight/day[12]. For a standard adult male of Catalonia weighing 70 kg, this risk was in the range 0.28-1.12 vs. a range of 0.88-3.51 found in our 2000 survey[8, 9]. For carcinogenic effects, the risk is expressed as the probability of contracting cancer over a lifetime. In the present study, a value of 1×10^{-3} per pg TEQ/kg/day was used as an estimator of upper bound cancer risk[13]. In an adult population of one million, the risk level due to PCDD/PCDF plus DL-PCB exposure through the diet would be 1,120 excess cancer over a lifetime of 70 years vs. 3,550 cases in our previous survey. Catalonia has a population of approximately 7.25 million people, which would mean 8,120 cancers in 70 years, or 116 cancers per year.

The results of this study show a notable decline in human dietary exposure to PCDD/PCDFs and DL-PCBs in Catalonia. The current estimated intake for an adult male, 1.12 pg WHO-TEQ/kg body weight per day, is lower than most intakes recently reported in a number of countries over the world: Italy[14], Sweden[15], France[16], and Japan[17]. China was the only country where a lower intake than the current one was found[18]. The above differences among the results can be explained by multiple reasons: dietary habits of each specific region/country, number of foodstuffs included in the surveys, PCB congeners analyzed, etc. Another reason could be the time elapsed between sample collection and results publication, which in our case has been comparatively short. Anyhow, it seems quite obvious that according to the current results, but also those of other recently published studies, the decline in human dietary exposure to PCDD/PCDFs and DL-PCBs is genuine.

Acknowledgements
This study was supported by the Catalan Food Safety Agency, Department of Health, Generalitat de Catalunya, Barcelona, Catalonia, Spain.

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Table 1: Estimated dietary intake of PCDD/PCDFs, DL-PCBs and PCDD/PCDFs plus DL-PCBs by the adult population of Catalonia, Spain[a]

food group	daily consumption[b] (g)		PCDD/PCDF intake (pg WHO-TEQ/day)		PCB intake (pg WHO-TEQ/day)		PCDD/PCDF + DL-PCB intake (pg WHO-TEQ/day)	
	2000	2006	2000	2006	2000	2006	2000	2006
meat and meat products	185 (12.8)	172 (14.0)	12.09	2.62	8.85	2.24	20.94	4.86
fish and seafood	92 (6.4)	68 (5.5)	28.74	6.53	82.87	38.77	111.61	45.30
vegetables	226 (15.7)	160 (13.0)	1.67	1.28	1.07	0.70	2.74	1.98
tubers	74 (5.1)	73 (5.9)	0.90	0.77	0.83	0.45	1.73	1.22
fruits	239 (16.6)	194 (15.8)	2.20	0.65	2.10	0.79	4.30	1.44
eggs	34 (2.4)	31 (2.5)	2.37	0.43	0.84	0.55	3.22	0.99
milk	217 (15.0)	128 (10.5)	2.10	1.47	1.78	1.72	3.88	3.19
dairy products	106 (7.3)	76 (6.2)	23.32	1.81	29.38	1.24	52.70	3.05
cereals	206 (14.3)	224 (18.3)	13.76	6.33	11.36	2.27	25.12	8.60
pulses	24 (1.7)	30 (2.5)	0.33	0.43	0.37	0.22	0.70	0.65
oils and fats	41 (2.8)	27 (2.2)	7.93	2.50	10.67	2.48	18.60	4.98
bakery products		45 (3.7)		0.85		0.98		1.82
total intake (TI)	1444 (100)	1228 (100)	95.41	25.67	150.12	52.40	245.53	78.07
TI (pg/kg body weight/day)			1.36	0.37	2.14	0.75	3.51	1.12

[a]Results are given for a male adult of 70 kg body weight. [b]Food consumption data were taken from Capdevila et al.[8] and from Serra-Majem et al.[4], for the 2000 and 2006 surveys, respectively. In parentheses, percentages of total consumption.

Figure 1: Estimated dietary intake of PCDD/PCDFs, DL-PCBs and PCDD/PCDFs plus DL-PCBs by the general population of Catalonia, Spain, in relation to age and gender. Calculations were done assuming ND = 1/2 LOD