



Human exposure to dioxins through the diet in Catalonia, Spain: carcinogenic and non-carcinogenic risk

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Abstract

The main objectives of this study were to estimate the dietary intake of dioxins by the population of Catalonia, Spain, to determine which food groups showed the greatest contribution to this intake, and to assess the health risks potentially associated with the dietary dioxin intake. From June to August 2000, food samples were randomly acquired in seven cities of Catalonia. Dioxin concentrations were determined in 108 samples belonging to the following groups: vegetables, fruits, pulses, cereals, fish and shellfish, meats and meat products, eggs, milk and dairy products, and oils and fats. Estimates of average daily food consumption were obtained from recent studies. Total dietary intake of dioxins for the general population of Catalonia was estimated to be 95.4 pg WHO-TEQ/day (78.4 pg I-TEQ/day), with fish and shellfish (31%), dairy products (25%), cereals (14%) and meat (13%) showing the greatest percentages of contribution to dioxin intake. The contribution of all the rest of food groups to the total dietary intake was under 20%. The non-carcinogenic risk index of dioxin intake through the diet was in the range 0.34–1.36, while the carcinogenic risk level was 1360 excess cancer over a lifetime of 70 years. Our results corroborate the decreasing tendency in dietary intake of dioxins found in recent studies (2000–2001) from various countries.

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1. Introduction

Polychlorinated dibenzo-*p*-dioxins (PCDD) and dibenzofurans (PCDF), commonly known as dioxins, are a lipophilic group of organic compounds, which are ubiquitous environmental pollutants (Hutzinger and Fiedler, 1989). Dioxin origin comes from many sources,

especially from combustion processes in the presence of chlorine and from industrial processes which are based on the chemistry of chlorine (Fiedler, 1996; Baker and Hites, 2000; Vikelsoe and Johansen, 2000; Buekens et al., 2001). Due to the notable toxic properties of PCDD/PCDF (Kogevinas and Janer, 2000; Kogevinas, 2001; Steenland et al., 2001), the potential adverse effects of these contaminants raises a great public concern.

Although human exposure to PCDD/PCDF can occur by various routes; food is the primary source. While a number of studies have reported that 90–95% of the exposure is from food (Fries, 1995; Sweetman et al., 2000), we recently estimated that even for a population

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living in the vicinity of a municipal waste incinerator, dietary exposure accounted for more than 98% of total PCDD/PCDF exposure. While pathways of PCDD/PCDF entry into food chain are atmospheric transport of emissions and their subsequent deposition on plants, soils and water, the major food sources of PCDD/PCDF have been reported to be fat-containing animal products, and fish and shellfish (Fries, 1995).

In relation to the prevention of health risks from PCDD/PCDF exposure, in December 1990 the WHO established a tolerable daily intake (TDI) of 10 pg/kg body weight for the 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD), based on liver toxicity, reproductive effects and immunotoxicity in experimental animals, and making use of kinetic data in humans and animals. However, in May 1998 and taking into account new epidemiological and toxicological data, the WHO's TDI for dioxins was revisited. The health risk assessment was focused on the most sensitive effects that are considered adverse at low doses in animal studies. According to this, a TDI range of 1–4 pg TEQ/kg body weight was established (Van Leeuwen et al., 2000). This range is only applicable to general toxic effects (other than cancer) of PCDD/PCDF. Recently, The European Commission proposed target levels and action levels for the concentration of dioxins in food and feed. A tolerable weekly intake (TWI) of dioxins and dioxin-like PCBs of 14 pg TEQ/kg body weight was also established (European Commission, 2001). On the other hand, using recent epidemiological data on exposed human populations and experimental carcinogenicity bioassays in animals, the IARC evaluated in 1997 the congener TCDD as carcinogenic to humans (IARC group 1 classification) (McGregor et al., 1998).

Data from various countries show that in recent years human dietary exposure to dioxins and dioxin-like PCB congeners is being reduced (Vieth et al., 2000; Kiviranta et al., 2001; Tsutsumi et al., 2001). However, incidents such as the Belgian dioxin and PCB contamination of feeding stuff in 1999 (Van Larebeke et al., 2001), or the incident of the citrus pulp pellets in 1998 (Malisch, 2000), show that PCDD/PCDF continue to be a risk of accidental contamination of the food chain.

The main goal of the present study was to estimate the dietary intake of PCDD/PCDF by the population of Catalonia, Spain and to compare the results with those from the most recent international reports on this issue. Although in the nineties, the dietary dioxin intake was determined in a number of countries, most surveys were mainly focused only on those food groups in which the highest levels of PCDD/PCDF could be expected. The present study is the one of the scarce surveys in which all food groups included in generalized diets have been included.

2. Experimental

2.1. Sample collection

During the months of June–August 2000, food samples were randomly acquired in local markets, big supermarkets, and grocery stores from the cities of Barcelona, Tarragona, Lleida, Girona, L'Hospitalet de Llobregat, Badalona and Terrassa. All these cities belong to Catalonia (NE Spain) and have a large number of inhabitants (150 000–1 800 000).

For collection of food samples, two groups were made up. The first group included: meat of beef (steak, hamburger), pork (loin, sausage), chicken (breast) and lamb (steak); fish (hake, sardine) and shellfish (mussel); vegetables (lettuce, tomato, potato, green beans, cauliflower); fresh fruits (apple, orange, pear), and eggs. The second group included cow milk (whole, semiskimmed) and dairy products (yogurt, cheese); cereals (bread, pasta, rice); pulses (lentils, beans); fats (margarine) and oils (olive, sunflower); tinned fish (tuna, sardine), and meat products (ham, hot dogs, salami).

Because in the first group most products are usually retailed, their origins could be very diverse in the different cities. Therefore, in that group 4 composite samples were analyzed for each food item. Each composite was made up by 10 individual samples. However, since most food items included in the second group corresponded to brands/trademarks that could be acquired in many sampling points, only 2 composite samples were analyzed for each food item. Each composite was made up by 8 individual samples. The total number of food samples collected were 1008, and after the formation of composites, a total of 108 samples were analyzed.

2.2. Analytical procedure

The determination of PCDD/PCDF was performed in accordance to the German VDI 3499 and US EPA 1625 methods. Food analyses were performed in series of 5 samples and 1 blank. Analytical methods, including preparation of samples for analysis and clean-up, were described previously and are not repeated in detail here (Domingo et al., 1999). Measurement and quantification were performed by high-resolution gas chromatography/high-resolution mass spectrometry (HRGC/HRMS), model Fisons 8000 GC coupled with a VG Autospec Ultim system (EI and multiple ion determination mode resolution 10 000). Quantitative determinations were carried out using internal standards. Detection limits were between 0.02 and 0.16 ng/kg dry weight depending on the specific food sample and PCDD/PCDF congener.

2.3. Calculations

Estimates of average daily food consumption were obtained from recent studies (Capdevila et al., 2000; Cucó et al., 2001). Estimates of PCDD/PCDF intake for each food group were performed assuming that non-detected isomer concentrations were equal to half of the respective limit of detection ($ND = 1/2 \text{ LOD}$). Toxic equivalents (WHO-TEQ) were calculated using the toxicity equivalent factors (TEF) established by the WHO in 1998.

3. Results and discussion

Table 1 shows the concentrations of PCDD/PCDF in eight groups of food samples. Results are presented for each of the 17 most toxic congeners. The sum for PCDDs and PCDFs, the ratio sum PCDD/sum PCDF, as well as the WHO-TEQ for each food group are also shown. TCDD, the most toxic congener, was not detected in pulses, cereals and fruits, while fish and shellfish showed the greatest level (0.26 ng TCDD/kg fat) of this congener. OCDD was the predominant congener in all food groups. The highest OCDD values corre-

sponded to fish and shellfish (15.7 ng/kg fat), eggs (15.03 ng/kg fat) and meat (12.39 ng/kg fat). In contrast, 1,2,3,7,8-PeCDD, 1,2,3,7,8,9-HxCDD and 1,2,3,7,8,9-HxCDF were only detected in fish and shellfish, meats, eggs, and milk and dairy products. Fruits, followed by pulses and cereals, were the food groups in which more congeners were under their respective detection limits. The greatest sum PCDD/sum PCDF ratio corresponded to eggs (6.54), fats and oils (4.93), meat (1.95) and vegetables (1.01), while in the remaining groups the ratios were <1.

The maximum WHO-TEQ values corresponded to fish and shellfish (5.57 ng/kg fat), followed at a great distance by milk and dairy products (0.80 ng/kg fat), eggs (0.58 ng/kg fat), meat (0.47 ng/kg fat), and oils and fats (0.18 ng/kg fat). The concentrations of PCDD/PCDF (WHO-TEQ values) in a number of food items are presented in Table 2. Data are given in ng/kg dry weight, ng/kg fat and pg/kg wet weight. When the PCDD/PCDF levels were expressed in pg WHO-TEQ/kg wet weight, blue fish (655), shellfish (302), tinned fish (247), and margarine (236) were the items showing the highest WHO-TEQ values.

Data for the estimation of the total dietary intake of PCDD/PCDF for the general population of Catalonia

Table 1
PCDD/PCDF concentrations in food samples from Catalonia, Spain^a

| Congener | Vegetables and tubercles* (n = 20) | Pulses and cereals* (n = 12) | Fruits* (n = 12) | Fish and shellfish (n = 16) | Meat (n = 30) | Eggs (n = 4) | Milk and dairy products (n = 8) | Fats and oils (n = 6) |
|----------------------|------------------------------------|------------------------------|------------------|-----------------------------|---------------|--------------|---------------------------------|-----------------------|
| 2378-TCDD | 0.01 | <0.02 | <0.02 | 0.26 | 0.03 | 0.17 | 0.06 | 0.04 |
| 12378-PeCDD | <0.04 | <0.04 | <0.04 | 0.44 | 0.06 | 0.12 | 0.18 | <0.1 |
| 123478-HxCDD | 0.02 | <0.04 | <0.04 | 0.20 | 0.09 | 0.15 | 0.15 | <0.1 |
| 123678-HxCDD | 0.03 | <0.04 | <0.04 | 0.81 | 0.22 | 0.35 | 0.28 | 0.07 |
| 123789-HxCDD | <0.04 | <0.04 | <0.04 | 0.31 | 0.06 | 0.12 | 0.12 | <0.1 |
| 1234678-HpCDD | 0.19 | 0.06 | 0.04 | 4.03 | 2.11 | 3.38 | 1.23 | 0.24 |
| OCDD | 0.83 | 0.29 | 0.16 | 15.57 | 12.39 | 15.03 | 3.32 | 1.83 |
| 2378-TCDF | 0.09 | 0.05 | 0.03 | 7.41 | 0.32 | 0.36 | 0.40 | 0.11 |
| 12378-PeCDF | 0.03 | 0.03 | 0.01 | 2.83 | 0.16 | 0.14 | 0.21 | 0.05 |
| 23478-PeCDF | 0.03 | 0.02 | 0.01 | 3.89 | 0.14 | 0.17 | 0.44 | 0.05 |
| 123478-HxCDF | 0.13 | 0.08 | 0.08 | 13.62 | 1.25 | 0.22 | 1.02 | 0.27 |
| 123678-HxCDF | 0.04 | 0.03 | 0.02 | 2.01 | 0.26 | 0.10 | 0.37 | 0.12 |
| 123789-HxCDF | <0.04 | <0.04 | <0.04 | 0.40 | 0.09 | 0.04 | 0.12 | <0.1 |
| 234678-HxCDF | 0.09 | 0.06 | 0.06 | 2.17 | 0.37 | 0.20 | 0.51 | 0.25 |
| 1234678-HpCDF | 0.17 | 0.07 | 0.04 | 4.33 | 0.84 | 0.25 | 1.06 | 0.14 |
| 1234789-HpCDF | 0.03 | 0.02 | <0.04 | 1.13 | 0.20 | 0.07 | 0.27 | <0.1 |
| OCDF | 0.27 | 0.13 | 0.11 | 6.60 | 1.25 | 0.37 | 1.28 | 0.31 |
| Sum PCDD | 1.69 | 0.60 | 0.25 | 37.72 | 15.96 | 22.10 | 7.20 | 2.92 |
| Sum PCDF | 1.67 | 0.75 | 0.43 | 122.99 | 8.20 | 3.38 | 11.59 | 2.01 |
| Sum PCDD/ sumPCDF | 1.01 | 0.79 | 0.57 | 0.31 | 1.95 | 6.54 | 0.62 | 4.93 |
| WHO-TEQ | 0.11 | 0.07 | 0.06 | 5.57 | 0.47 | 0.58 | 0.80 | 0.18 |

^a Results are given in ng/kg fat. Those marked with an asterisk are given in ng/kg dry weight, n = number of samples analyzed.

Table 2

Concentrations of PCDD/PCDF (WHO-TEQ values) in a number of foodstuffs acquired in a number of locations of Catalonia, Spain

| Food group | ng WHO-TEQ/kg dry weight ^a | ng WHO-TEQ/kg fat | pg WHO-TEQ/kg wet weight |
|-------------------------|---------------------------------------|-------------------|--------------------------|
| Vegetables | 0.11 (0.22–0.05) | – | 7.37 |
| Tubercles | 0.07 (0.08–0.05) | – | 12.17 |
| Pulses | 0.06 (0.09–0.04) | – | 13.68 |
| Cereals | 0.08 (0.12–0.041) | – | 66.78 |
| Fruits | 0.06 (0.09–0.04) | – | 9.20 |
| White fish | 0.59 (1.55–0.26) | 6.06 | 109.09 |
| Shellfish | 1.80 (4.04–0.85) | 10.78 | 301.93 |
| Tinned fish | 0.58 (1.73–0.09) | 2.01 | 247.46 |
| Blue fish | 1.99 (2.41–1.36) | 6.98 | 655.81 |
| Pork and pork products | 0.20 (0.50–0.07) | 0.31 | 71.20 |
| Chicken | 0.21 (0.51–0.09) | 1.32 | 54.27 |
| Beef and beef products | 0.22 (0.53–0.12) | 0.45 | 64.01 |
| Lamb | 0.19 (0.26–0.13) | 0.42 | 49.82 |
| Eggs | 0.30 (0.45–0.18) | 0.58 | 69.79 |
| Dairy products | 0.42 (0.87–0.22) | 1.21 | 220.04 |
| Whole milk | 0.11 (0.13–0.08) | 0.32 | 12.24 |
| Semiskimmed milk | 0.07 (0.08–0.07) | 0.45 | 7.13 |
| Vegetable oils and fats | 0.17 (0.19–0.15) | 0.17 | 172.36 |
| Margarine | 0.24 (0.29–0.19) | 0.19 | 235.55 |

^a In parentheses: maximum and minimum values.

Table 3

Estimated daily intake of PCDD/PCDF for the general population of Catalonia, Spain

| Food group | Food consumption ^a (g/day) | Daily intake (pg WHO-TEQ) |
|-------------------------|---------------------------------------|---------------------------|
| Tubercles | 74 (5.1%) | 0.90 |
| Vegetables | 226 (15.7%) | 1.67 |
| Pulses | 24 (1.7%) | 0.33 |
| Cereals | 206 (14.3%) | 13.76 |
| Fruits | 239 (16.6%) | 2.20 |
| Fish and shellfish | 92 (6.4%) | 28.74 |
| Meat | 185 (12.8%) | 12.09 |
| Eggs | 34 (2.4%) | 2.37 |
| Milk | 217 (15%) | 2.10 |
| Dairy products | 106 (7.3%) | 23.32 |
| Vegetable oils and fats | 41 (2.8%) | 7.93 |
| Total Intake | 1444 | 95.4 |
| Caloric basis | 2500 kcal | |

^a In parentheses: percentage of the total consumption.

are summarized in Table 3. Total dietary intake of PCDD/PCDF was 95.4 pg WHO-TEQ/day. The greatest percentages of contribution to total daily intake corresponded to fish and shellfish (31%), and dairy products (25%), followed by cereals (14%) and meat (13%) (Fig. 1). With respect to cereals, although the PCDD/PCDF concentration in this group was comparatively low (66.8 pg WHO-TEQ/kg wet weight), the notable consumption of cereals (bread, pasta, rice) associated with the *Mediterranean diet* could explain the high contribution of this group in this survey.

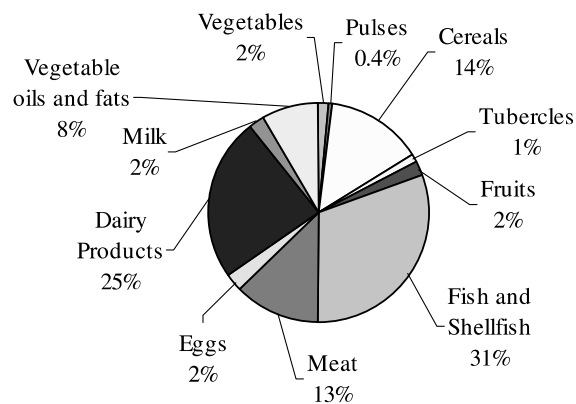


Fig. 1. Percentages of contribution from each food group to the total dietary intake of PCDD/PCDF by the general population of Catalonia, Spain.

Table 4 summarizes the estimated PCDD/PCDF daily intake for the population of Catalonia according to sex and age. Total daily intake ranged between 72 pg WHO-TEQ/kg for females older than 65 years and 99 pg WHO-TEQ/kg for males in the group of 20–34 years. In any case, the total daily intake of PCDD/PCDF was always greater in males than in females, which is very probably due to the lower food ingestion by females.

The variations with age in total PCDD/PCDF dietary intake are depicted in Fig. 2 (pg WHO-TEQ/day) and Fig. 3 (pg WHO-TEQ/kg/day). In males, this intake increased during childhood and adolescence, and decreased from the adult age (Fig. 2). In females, the dif-

Table 4

Estimated daily intake of PCDD/PCDF (pg WHO-TEQ/kg) for males and females from the general population of Catalonia, Spain, according to sex and age

| | Age group (years) | | | | | | | | | | | |
|-------------------------|-------------------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|-------|
| | 4–9 | | 10–19 | | 20–34 | | 35–50 | | 51–65 | | >65 | |
| | M | F | M | F | M | F | M | F | M | F | M | F |
| Tubercles | 0.84 | 0.71 | 1.06 | 0.80 | 0.96 | 0.72 | 0.90 | 0.67 | 0.84 | 0.69 | 1.10 | 0.60 |
| Vegetables | 0.96 | 0.88 | 1.21 | 1.19 | 1.39 | 1.28 | 1.84 | 1.60 | 1.76 | 1.59 | 1.41 | 1.39 |
| Pulses | 0.36 | 0.34 | 0.34 | 0.31 | 0.31 | 0.30 | 0.38 | 0.36 | 0.29 | 0.27 | 0.34 | 0.26 |
| Cereals | 13.36 | 13.42 | 17.56 | 11.95 | 15.83 | 10.82 | 13.82 | 9.02 | 11.69 | 7.88 | 11.89 | 9.02 |
| Fruits | 1.83 | 1.78 | 1.89 | 1.83 | 1.94 | 1.89 | 2.24 | 1.92 | 2.40 | 2.45 | 2.74 | 2.19 |
| Fish and shellfish | 16.87 | 15.31 | 19.99 | 18.74 | 23.43 | 22.80 | 29.67 | 24.05 | 33.11 | 27.49 | 24.36 | 25.61 |
| Meat | 9.15 | 9.15 | 11.83 | 10.00 | 14.70 | 9.02 | 10.85 | 8.30 | 10.65 | 7.19 | 7.91 | 6.99 |
| Eggs | 2.37 | 1.33 | 2.09 | 1.47 | 2.65 | 1.61 | 2.16 | 1.74 | 2.37 | 1.54 | 2.02 | 1.12 |
| Milk | 3.16 | 2.83 | 2.93 | 2.23 | 2.22 | 2.35 | 2.17 | 2.34 | 1.92 | 2.66 | 2.12 | 2.79 |
| Dairy products | 24.86 | 25.30 | 29.92 | 23.98 | 27.28 | 19.58 | 22.66 | 19.14 | 20.02 | 21.56 | 15.18 | 16.50 |
| Vegetable oils and fats | 6.38 | 6.58 | 7.93 | 6.00 | 8.32 | 6.00 | 8.32 | 6.19 | 7.16 | 5.80 | 5.42 | 5.80 |
| Total | 80.13 | 77.62 | 96.76 | 78.50 | 99.04 | 76.36 | 95.02 | 75.33 | 92.21 | 79.13 | 74.49 | 72.26 |

M = males; F = females.

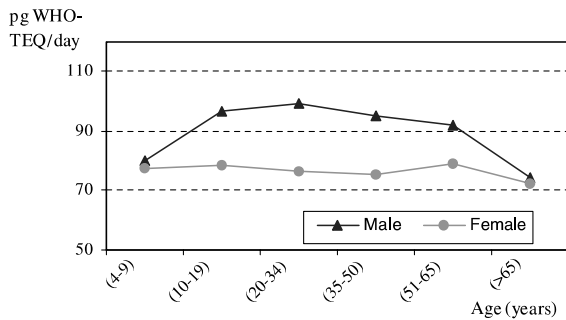


Fig. 2. Dietary intake of PCDD/PCDF (pg WHO-TEQ/day) by the general population of Catalonia, Spain: variation with age.

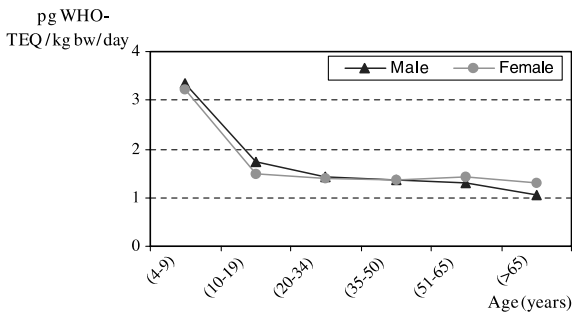


Fig. 3. Total dietary intake of PCDD/PCDF (pg WHO-TEQ/kg body weight/day) by the general population of Catalonia, Spain: variation with age.

ferences in PCDD/PCDF intake with age were practically irrelevant, excepting a reduction in the group >65 years. However, when the data were analyzed according

to the respective average body weight for each age group, a continued decrease of PCDD/PCDF intake with age was noted in both sexes (Fig. 3). According to the results of the current study, a Catalan adult of 70 kg body weight would ingest, on average, 1.36 pg WHO-TEQ/kg/day (1.12 pg I-TEQ/kg/day), value that is clearly within the WHO-TDI range.

The non-carcinogenic risk index of PCDD/PCDF intake through the diet was calculated by dividing the total daily intake by the WHO-TDI. This risk was in the range 0.34–1.36. For carcinogenic effects, the risk is expressed as the probability of contracting cancer over a lifetime. In the present investigation, a value of 1×10^{-3} per pg I-TEQ/kg/day was used as an estimator of upper bound cancer risk (US EPA, 2000). In an adult population of one million, the risk level due to PCDD/PCDF exposure through the diet would be 1360 excess cancer over a lifetime of 70 years. Catalonia has a population of approximately 6 million people, which would mean 8160 cancers in 70 years or 117 cancers per year. Taking into account that in this region the annual number of new cases of cancer detected is about 25 000, the contribution of PCDD/PCDF dietary intake to this amount would be of 0.47%.

Based on previous results on a cohort of workers who were primarily exposed to TCDD, a considerable higher risk could be estimated. Becher et al. (1998) reported that a lifetime exposure (70 years) to 1 pg TCDD/kg/day entailed an incremental lifetime cancer mortality risk between 1.3 and 7.7 per 1000. Based on these estimations and assuming a linear dose-risk model, 1768–10 472 death cancers per million of inhabitants, or 10 608–62 832 cases for the total population of Catalonia could be expected. It would be equivalent to 152–898

cases per year. However, this estimation seems to be less realistic since the occupational exposure to PCDD/PCDF is different to that for a non-exposed population.

A comparison of the results on total dietary PCDD/PCDF intake of recent reports from a number of countries, and those of the present study, is shown in Table 5. It can be seen that PCDD/PCDF dietary intake is currently in a range between 30.4 pg TEQ/day for Korea (Kim et al., 2000) and 87–123 pg TEQ/day for USA (Schechter et al., 2001). The intake for Catalonia would be within this range. However, taking into account the notable differences in the methodologies used in these studies, all these data must be examined and compared very carefully. PCDD/PCDF dietary intake is the result of two factors: the concentrations of PCDD/PCDF in the food items, and the specific consumption of the respective food groups by the populations in each country or region assessed. Moreover, in each study there are also remarkable differences in the number and type of food items collected and analyzed. For example, in the recent Finish survey (Kiviranta et al., 2001) 40 samples were analyzed, while in the German study Vieth et al. (2000) presented their results on the basis of about 3000 dioxin data from food samples, but collected

through 5 years (1995–1999). Between 1995 and 1999, probably there were important decreases in the concentrations of PCDD/PCDF in food, which entails another distortion factor for the estimation of the dietary PCDD/PCDF intake. In addition, cereals and pulses were not included in that study.

In the recent Korean survey (Kim et al., 2000), only 23 different food samples were collected and analyzed. One of the most striking findings of that study was the notable contribution to the total dietary PCDD/PCDF intake (30.4 pg I-TEQ/day) of cereals and grains, 42%. This contribution was higher than that found for fish and shellfish, 39%. In USA, Schechter et al. (2001) analyzed 110 food samples divided into pooled lots by category. Only 12 separate analyses were conducted. In both, males and females, meat and meat products, dairy products, and vegetables were the groups showing the greatest contribution to total PCDD/PCDF dietary intake: 123 and 87 pg TEQ/day for males and females, respectively.

In addition to the current survey, the widest recent study for estimation of PCDD/PCDF dietary intake was conducted in Japan by Tsutsumi et al. (2001). The intakes reported by these authors were 44.7 and 81.9 pg

Table 5
Total dietary intake of PCDD/PCDF and percentages of exposure from the main food groups

| Country | Food groups analyzed | Main food groups contributing to total PCDD/PCDF dietary intake | PCDD/PCDF intake (pg/day) ^a | Reference |
|-------------------|---|--|---|-------------------------|
| Germany | Meat and meat products, fish, vegetables and fruits, eggs, milk and dairy products, oils and fats | Milk and dairy products, 38.7% meat and meat products, 29.9% fish, 11.4% vegetables and fruits, 7.7% | 50.0 (I-TEQ) | Vieth et al. (2000) |
| Korea | Meats and meat products, fish and shellfish, vegetables and grains, fruits, eggs, milk, cereals, pulses, oils and fats | Vegetables and grains, 42.0% fish and shellfish, 39.4% eggs, 4.8% | 30.4 (I-TEQ) | Kim et al. (2000) |
| Finland | Meat and meat products, fish and derivative products, vegetables, flour, eggs, milk | Fish and derivative products, 82.3% milk, 7.8% meat and meat products, 7.2% | 46 (I-TEQ) ^b | Kiviranta et al. (2001) |
| Japan | Meat and meat products, fish and shellfish, milk and dairy products, vegetables, fruits, cereals, pulses, oils and fats | Fish and shellfish, 37.0% meat and meat products plus eggs, 11.6% vegetables, 9.1% milk and dairy products, 6.2% | 81.9 (WHO-TEQ) | Tsutsumi et al. (2001) |
| USA | Meat and meat products, fish, eggs, milk and dairy products, vegetables | Meat and meat products, 32.1–36.1% dairy products, 28.7–30.5% vegetables, 21.3–23.6% | 123 (WHO-TEQ) (adult males) 87 (WHO-TEQ) (adult females) | Schechter et al. (2001) |
| Catalonia (Spain) | Meat and meat products, fish and shellfish, vegetables, fruits, cereals, pulses, eggs, milk and dairy products, oils and fats | Fish and shellfish, 29.1% dairy products, 26.3% cereals, 13.7% meat and products, 12.4% | 78.4 (I-TEQ) 95.40 (WHO-TEQ) | This study |

Comparison of the results reported in the period 2000 and 2001 for various countries with those of the present study.

^a Calculated assuming that non-detected congener concentrations are equal to half of the limit of detection (ND = 1/2 LOD).

^b Calculated assuming that non-detected congener concentrations are equal to 0 (ND = 0).

TEQ/day, using $ND = 0$ or $ND = 1/2$ LOD (limit of detection), respectively. The intake found assuming non-detected congener concentrations to be equal to one-half of the LOD is very similar to that found in the present study, 95.4 pg TEQ/day, which was also calculated assuming $ND = 1/2$ LOD. In both studies, fish and shellfish showed the greatest contribution to PCDD/PCDF dietary intake. However, while in our study dairy products (25%) and cereals (14%) were also important contributors to the total PCDD/PCDF dietary intake, these groups did not have a similar impact in Japan.

Some interesting conclusions can be drawn from the above. In general terms, and in comparison with data from a number of surveys conducted in the nineties, an important reduction in the dietary intake of PCDD/PCDF can be observed. With a few exceptions, the results of the studies published in 2000 and 2001 show a lower PCDD/PCDF dietary intake than those reported in the nineties. For example, in Germany Malisch (1996) found an intake of 65.1 pg TEQ/day vs. 50.9 pg TEQ/day recently reported by Vieth et al. (2000), while in Finland there was a decrease from 95 pg TEQ/kg/day (Hallikainen and Vartiainen, 1997) to 46 pg TEQ/day (Kiviranta et al., 2001). In Catalonia, there was a reduction from 210 pg I-TEQ/day (Domingo et al., 1999) to the current 78.4 pg I-TEQ/day. However in our previous survey, the number of analyzed samples was lower (35 vs. 108) and the population included in the study was focused only on Tarragona Province with approximately 500 000 inhabitants.

In all recent surveys, PCDD/PCDF intake has been found within the TDI for dioxins established by the WHO (1–4 pg TEQ/kg/day), with a prevailing tendency towards the lower value of the range, 1 pg TEQ/kg/day. The current dietary intakes are also below the TWI for dioxins (and dioxin-like PCBs) of 14 pg/kg body weight, as well as the recent tolerable monthly intake of 70 pg/kg body weight recommended by the Scientific Committee on Food of the European Union and the evaluation made by the Joint FAO/WHO Expert Committee on Food Additives (JECFA) in 2001. It is important to remark the significant contribution to PCDD/PCDF intake of fish and shellfish, which has been observed in most studies. However, the impact of other food groups such as vegetables, fruits and cereals should not be minimized especially in those countries or regions in which the consumption of these items is notable. Nowadays, in a global world market, the incidence of local products in the dietary intake of contaminants is obviously being reduced. Many food items are being consumed in countries that are not the producers. It means that increasingly, the differences in PCDD/PCDF dietary intake between countries will be mainly due to the different consumption habits among countries. Therefore, in order to compare results from different countries or regions, it is essential to include all food groups in the

surveys. In the past, only those food groups with high percentages of fat were included in the analyses, while products such as vegetables, pulses, cereals or fruits were minimized or even excluded in spite of the important contribution to total PCDD/PCDF intake of these groups in many places. This notable contribution is probably due to the remarkable consumption of these food groups in some regions and countries rather than to hypothetical high PCDD/PCDF levels in food samples.

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