

## Dietary Intake of PCDD/Fs from Food in Catalonia, Spain

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### Abstract

Thirty-five food samples from local supermarkets were analyzed for PCDD/Fs. On lipid basis, PCDD/F levels in meat, fish, eggs, and fat and oil were similar or lower than those reported from other countries. However, PCDD/F levels in milk, vegetables, and cereals showed higher levels than those previously reported for other countries. The resulting dietary PCDD/F intake from the population of Catalonia was 210 pg TEQ/day. This value is higher than the dietary intake of PCDD/Fs found in other surveys.

### Introduction

Polychlorinated dibenzo-p-dioxins (PCDD) and polychlorinated dibenzofurans (PCDF) have been detected in almost every matrix: soil, sediments, air, water, animals, and food (primarily meat, dairy products, fish and shellfish).<sup>1, 2)</sup> This widespread presence is due to the numerous sources that produce these chemicals, as well as the fat solubility of the compounds, long range transport, and resistance to biotransformation.

Only a few studies have determined PCDD/Fs levels in Spanish foods, limiting the ability to establish background exposure levels for Spain.<sup>3,4)</sup> The purpose of the present study was to increase the knowledge of the PCDD/F dietary intake by an Spanish population. The food items were selected based on the dietary habits in Catalonia.<sup>5)</sup>

### Experimental Methods

During 1996, 35 food samples were analyzed. The pool of the different subsamples consisted of: beef (*hamburger, steak, liver*), pork (*sausage, hot dogs, steak, liver, hamburger*), chicken (*liver, breast, sausage*), lamb (*steak*), fish (*angler fish, hake, trout*), seafood (*mussel, prawn*), tinned fish (*mussel, tuna, sardine*), blue fish (*sardine, tuna, salmon, mackerel*), milk (*whole, semiskimmed*), dairy products (*cheese, petit-swiss, yogurt, cream*), vegetables (*lettuce, chard, spinach, chickpeas*), pulses (*lentils, beans*), cereals (*spaguettis, rice, bread*), fruits (*orange, banana, apple*), fat and oil (*margarine, sunflower, olive, corn*), and eggs. Samples were obtained from local supermarkets and seafood markets. For all groups, the quantity of each food was quantified according to the dietetic habits of the region.<sup>5)</sup>

Food analyses were performed in series of 5 samples and 1 blank. Lyophilisate

Table 1. PCDD/F concentrations in foods

Congener	Vegetables <sup>*</sup> (n = 2)	Pulse and Cereals <sup>*</sup> (n = 4)	Fruits <sup>*</sup> (n = 2)	Fish (n = 8)	Meat (n = 7)	Eggs (n = 2)	Milk and dairy products (n = 6)	Fat and oil (n = 4)
2378-TCDD	0.1	<0.1	<0.05	1.2	0.1	0.1	<0.1	0.4
12378-PeCDD	0.1	<0.1	<0.05	0.8	<0.1	0.1	<0.1	0.1
123478-HxCDD	0.4	0.3	<0.05	0.4	0.4	0.3	0.1	0.2
123678-HxCDD	0.8	0.8	<0.05	1.5	1.1	1.7	0.6	0.3
123789-HxCDD	0.5	0.3	<0.05	1.2	0.3	0.6	0.1	<0.1
1234678-HpCDD	10.3	5.6	0.2	96.2	16.6	10.2	2.6	1.9
OCDD	111.5	51.7	1.7	126.9	224.6	61.6	26.7	17.8
2378-TCDF	0.6	0.1	1.0	11.1	1.8	2.1	2.1	0.3
12378-PeCDF	0.6	0.2	0.6	3.7	0.9	1.0	1.5	0.4
23478-PeCDF	0.7	0.2	0.5	4.4	0.7	0.5	1.5	0.1
123478-HxCDF	3.3	1.0	0.5	2.2	2.1	2.0	2.0	0.6
123678-HxCDF	0.9	0.5	0.4	0.7	0.7	0.4	0.8	0.4
234678-HxCDF	<0.1	<0.1	0.05	<0.1	<0.1	<0.1	<0.1	<0.1
123789-HxCDF	0.9	0.3	0.06	0.5	0.1	0.1	0.2	<0.1
1234678-HpCDF	23.5	5.6	0.2	2.0	1.9	0.9	1.1	0.8
1234789-HpCDF	2.2	0.6	0.2	0.3	0.5	0.5	0.4	<0.3
OCDF	119.5	42.5	0.8	8.8	8.2	3.7	3.5	1.8
Sum PCDD	156.5	74.5	2.7	188.4	255.2	95.0	33.7	23.9
Sum PCDF	228.5	50.7	7.1	220.4	24.7	21.0	20.6	7.1
Sum PCDD/ Sum PCDF	0.68	1.47	0.38	0.85	10.33	4.52	1.64	3.37
Total I-TEQ	1.85	0.65	0.51	6.61	1.39	1.22	1.49	0.56

Results are given as ng/kg fat weight. Those marked with asterisks are given as ng/kg dry weight  
n = number of samples analyzed.

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samples were spiked with 17  $^{13}\text{C}_{12}$ -labelled PCDD/F- isomers and soxhlet-extracted with toluene for 24 h. The extract was evaporated under vacuum to constant weight. The residue, which represents the fat content, was weighed, redissolved in hexane and cleaned up with standard methods, including several types of modified silicagels and activated charcoal. The purified extract was analyzed by HRGC/HRMS.

## Results and Discussion

The results of the congener-specific and homologue analyses, the sum for PCDD and PCDF, the ratios sum PCDD/sum PCDF, as well as the I-TEQ for each group of food samples are given in Table 1. The highest level of 2,3,7,8-TCDD was found in seafood. Meat, eggs and fat samples showed the highest ratios for dioxins/furans (10.33, 4.52 and 3.37). For the remaining samples, ratios were < 1 or about 1.

Table 2. Concentrations of PCDD/Fs (ng/kg fresh weight) in food categories (average value from duplicates)

Food category	ng TEQ/kg fat	ng TEQ/kg wet weight
Vegetables	---	0.14
Pulses	---	0.19
Cereals	---	0.25
Fruits	---	0.09
White fish	5.39	0.27
Seafood	10.59	0.42
Tinned Fish	2.57	0.24
Blue Fish	7.90	0.76
Pork and pork products	0.90	0.11
Chicken and chicken products	1.15	0.11
Beef and beef products	1.76	0.13
Lamb	1.76	0.13
Eggs	1.22	0.12
Dairy products	1.25	0.04
Whole milk	2.02	0.18
Semiskimmed milk	1.20	0.06
Oil	0.64	---
Margarine	0.49	---

The results from 35 food samples collected in supermarkets from Catalonia are presented according to fat and wet weight basis (Table 2). Based on I-TEQ, PCDD/F levels in seafood (10.59 ng TEQ/kg fat) were higher than those found in blue fish (7.90 ng TEQ/Kg fat) and tinned fish (2.57 ng TEQ/kg fat). Although most of the results here found in this study (Table 2) are in agreement with recent data<sup>1,6,7,8)</sup>, whole milk showed higher levels than those of previous studies. Moreover, vegetables, pulses, and cereals showed also higher levels of PCDD/Fs (0.14, 0.19 and 0.25 ng TEQ/kg wet weight) than those reported in previous studies.<sup>1,8)</sup>

## Dioxin '97, Indianapolis, Indiana, USA

Table 3. Estimated daily intake of PCDD/Fs for an adult general population of Catalonia, Spain

Food group	Consumption rate <sup>a</sup> (g/day)	Daily TEQ intake (pg)	Contribution (%) from each food group
Vegetables	122 (10.7)	17.08	8.13
Pulses	16 (1.4)	3.04	1.45
Cereals	194 (17.0)	48.50	23.09
Fruits	269 (23.6)	24.21	11.53
Fish and seafood	72 (6.3)	30.38	14.46
Meat	173 (15.1)	20.76	9.88
Eggs	29 (2.5)	3.48	1.66
Dairy products	44 (3.9)	1.76	0.84
Whole Milk	178 (15.6)	32.04	15.25
Oils	45 (3.9)	28.80	13.71
<b>TOTAL INTAKE</b>	<b>1142 (100)</b>	<b>210.05</b>	<b>100</b>

<sup>a</sup>In brackets, percentage of the total consumption.

Consumption rate, daily TEQ intake, and contribution (%) from each food group to total PCDD/F intake are summarized in Table 3. Although fish and fat and oils are the most representative products of PCDD/F accumulation, the great consumption of vegetables and cereals included in the mediterranean diet, would explain the comparatively high PCDD/F ingestion through the present diet (8.13% and 23.09%, respectively). These results are not in agreement with those of previous studies where contributions from fruits and vegetables were almost negligible.<sup>1,8)</sup> In relation to the same issue, the average daily intake of PCDD/Fs in Italy<sup>11)</sup> was reported to be higher than the present one (Table 4). It agrees well with the significant exposure to PCDD/Fs from vegetables found in that survey.<sup>11)</sup> However, when only fish, meat, eggs, fat and oils, and milk and dairy products were considered as main components of the present diet (as it has been estimated in previous surveys), the total PCDD/F intake was 117 pg TEQ/day. In contrast, when the remaining components of the diet (vegetables, pulses, cereals and fruits) were included the total daily intake rised up to 210 pg TEQ/day. It means that cereals and vegetables should be taken into account when the total PCDD/F intake through the diet is determined.

As in most EU countries, standard brands are distributed throughout Catalonia. Consequently, in this country food origin is often quite different from the place of consumption. This fact, together with the circumstance that the mediterranean diet is also characteristic of most spanish regions, the results here obtained could be considered as representative of Spain.

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Table 4. Total dietary intake of PCDD/Fs in a number of countries

Country	Estimated dietary intake (pg TEQ/day)	Reference	Year
Spain			
<i>Catalonia</i>	210	This study	1997
<i>Madrid</i>	120	(3)	1995
<i>Basque Country</i>	128	(4)	1990-95
UK	125	(9)	1992
Germany	90.2	(1)	1989
Italy	260-480	(11)	1990
Canada	92	(7)	1989
Japan	63	(10)	1987

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