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Dietary intake of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) by a population living in the vicinity of a hazardous waste incinerator. Assessment of the temporal trend

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ABSTRACT

The concentrations of polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/Fs) were determined in a number of foodstuffs purchased in various locations near a hazardous waste incinerator (HWI) in Tarragona County (Catalonia, Spain). The dietary intake of PCDD/Fs by the population of the area under potential influence of the HWI was subsequently estimated. The results were compared with previous surveys performed in the same area in 1998 (baseline), 2002 and 2006. In the present study, the highest WHO-TEQ corresponded to industrial bakery (0.183 ng/kg wet weight, ww), followed by fish (0.156 ng/kg ww), oils and fats (0.112 ng/kg fat weight), and seafood (0.065 ng/kg ww). In contrast, the lowest values were observed in pulses and tubers (0.003 ng/kg ww), and cereals and fruits (0.004 ng/kg ww). The dietary intake of PCDD/Fs by the general population was 33.1 pg WHO-TEQ/day, having fish and seafood (11.6 pg WHO-TEQ), oils and fats (4.61 pg WHO-TEQ), dairy products (3.79 pg WHO-TEQ), and industrial bakery (3.49 pg WHO-TEQ) as the groups showing the highest contribution to the total TEQ. The lowest daily contributions corresponded to pulses (0.08 pg WHO-TEQ) and tubers (0.25 pg WHO-TEQ). This intake was considerably lower than that found in the baseline study, 210.1 pg I-TEQ/day, and also notably lower than that found in the 2002 survey (59.6 pg I-TEQ/day), but slightly higher than the intake estimated in the 2006 survey, 27.8 pg WHO-TEQ/day. The results of this study show that any increase potentially found in the biological monitoring of the general population living in the area under evaluation should not be attributed to dietary exposure to PCDD/Fs.

1. Introduction

Waste management facilities in general, and incinerators in particular, have been traditionally affected by the NIMBY (Not In My Back Yard) syndrome (Domingo, 2002a; Kikuchi and Gerardo, 2009). Although in comparison with other treatments for processing municipal solid waste (MSW) and hazardous waste (HW), incineration has multiple advantages (i.e., volume reduction, energy recovery, elimination of pathogen agents), public opposition to the siting and permitting of MSW and HW incinerators has been important. The main concern is usually related to the potential negative consequences of the emission of pollutants for both the environment and public health. The adverse health effects associated with stack emissions from waste incinerators have focused basically on metals and semivolatile and volatile organic compounds, mainly polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/Fs) (Domingo, 2002b; Giné-Bordonaba et al., 2011; Schuhmacher and Domingo, 2006; Zheng et al., 2008).

Nowadays, the management of HW generated in EU countries partly relies on incineration processes. In 1996–1998, a new HW incinerator (HWI) was constructed in Constantí (Tarragona County, Catalonia, Spain). Regular operations in the facility started in 1999. In the same area, there is also an important industrial activity with a notable number of potential emission sources of environmental pollutants, including two oil refineries, an important complex of petrochemical industries, and a municipal solid waste incinerator, among others. Because the new facility was the first, and up to date, the only HWI in Spain, the concern about its potential environmental impact and health risks was considerable in the public opinion. In response to that concern, a wide pre- and post-operational program was designed in order to assess the impact of toxic emissions from the new HWI on the neighborhood, as well as to establish the health risks on the population living near the facility. Regarding human health risks, a biological monitoring program concerning metals and PCDD/Fs was designed. Samples of human blood, milk, autopsy tissues, and hair were obtained during the construction period of the HWI from subjects living in the neighborhood of the facility and analyzed for baseline levels of metals and PCDD/Fs (Granero et al., 1998; Llobet et al., 1998; Schuhmacher et al., 1999a,b,c). However, it is well established that for the general

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population, exposure to metals and PCDD/Fs through dietary intake is higher than inhalation and/or dermal contact. Consequently, food samples were also collected to estimate the intake of metals and PCDD/Fs through foods purchased in the area under evaluation (Domingo et al., 1999).

With respect to PCDD/Fs, it has been reported that the contribution of products of animal origin such as meat and meat products, dairy products, and fish and other seafood may surpass 90–95% of the total daily exposure to these environmental pollutants (Bergkvist et al., 2008; Charnley and Doull, 2005; Domingo and Bocio, 2007; Llobet et al., 2003). Taking this into account, in order to establish clearly whether any hypothetical increase in the PCDD/F levels in biological tissues of subjects living near the facility could be due to the emissions of PCDD/Fs by the HWI, or it could be derived from potential increases of PCDD/F concentrations in food, the periodic update of the dietary intake of PCDD/Fs was and continues being essential. During the construction of the facility (1996–1998), a baseline food survey was performed (Domingo et al., 1999), while second and third surveys were carried out in 2002 and 2006, respectively (Bocio and Domingo, 2005; Martí-Cid et al., 2008). We here present the results of a fourth campaign in which the concentrations of PCDD/Fs were again determined in foodstuffs. The dietary intake of these pollutants by the population living in the vicinity the HWI was subsequently estimated. The results were compared with those corresponding to our previous studies (baseline, 2002 and 2006) (Bocio and Domingo, 2005; Domingo et al., 1999; Martí-Cid et al., 2008).

2. Materials and methods

2.1. Sample collection

In January–February 2012, food samples were randomly purchased in local markets, large supermarkets, and grocery stores from different locations of Tarragona County (Catalonia, Spain) within a radius of 15 km from the HWI. It is important to emphasize that, as in the baseline survey (Domingo et al., 1999), most acquired foods were not local products. It is noteworthy that in the area under potential environmental influence of the HWI, there are neither pasture grounds, nor crops of vegetables, grains, or fruits, that could be consumed in significant amounts by the population living in the area. Thus, for the purpose of the study, foodstuffs might be potentially of any origin. Consequently, they were randomly purchased, having PCDD/F exposure from ingestion of local products considered as irrelevant.

A total of 45 samples composed by subsamples of 67 food items were analyzed. The pool of the different subsamples consisted of 22 food subgroups, organized in 12 food groups. The complete list of foodstuffs here analyzed is summarized in Table 1. For the preparation of all composite subsamples, the quantity of each food in each subsample was included according to the dietary habits of the population of the area under evaluation. Ten individual samples of each food were collected in different markets and stores of that area. Each sample was part of a composite of its respective kind of food, in a representative percentage of the consumption by the population of the zone.

2.2. Analytical procedure

Food samples were homogenized and lyophilized. The determination of PCDD/Fs was performed according to German VDI 3499 and US EPA 1625 methods. Five to 10 g of the fresh or freeze-dried solid sample was mixed with a small amount of Na_2SO_4, spiked with a mixture of $^{13}C_{12}$-labeled PCDD/F standards (at least one dioxin and furan for each chlorination degree), and Soxhlet-extracted over 24 h with toluene. The content of lipids in each sample was isolated, and part of them was used for the clean-up procedure. The clean-up and fractionation were carried out by adsorption chromatography as a multistep clean-up, using multilayer silica columns (from top to bottom: sodium sulfate, silica, silica/sulfuric acid, silica, silica/potassium hydroxide, silica) and alumina columns. For oil and margarine, 2 g of sample was dissolved in hexane and immediately used for the clean-up steps as above indicated. The final step was the reduction of the PCDD/F-containing fraction to the analytically needed volume. Prior to analysis, ^{13}C-labeled PCDD/F standards were added for the calculation of

Table 1
Summary of foods here analyzed: groups, subgroups and individual items.

Food items	Food subgroups	Food groups
Hamburger	Beef	Meat
Steak		
Sausage	Pork	
Hot dogs		
Steak		
Salami		
Hamburger		
Boiled jam		
Breast	Chicken	
Sausage		
Thigh		
Rib	Lamb	
Thigh		
Hake	White fish	Fish and seafood
Whiting blue		
Bass		
Angler fish		
Sardine	Blue fish	
Tuna		
Salmon		
Cuttlefish	Cephalopods	
Squid		
Red mullet	Other fish	
Sole		
Mussels	Tinned fish	
Tuna		
Sardine		
Mussels	Seafood	
Prawn		
Whole milk	Whole milk	Milk
Semiskimmed milk	Semiskimmed milk	
Soft cheese	Dairy products	Dairy products
Semi-cured cheese		
Cured cheese		
Yogurt		
Petit-Swiss		
Cream caramel		
Crème brûlée		
Custard		
Lettuce	Vegetables	Vegetables
Tomato		
Spinach		
Green beans		
Cauliflower		
Potato	Tubers	Tubers
Carrot		
Lentils	Pulses	Pulses
Beans		
Chick-peas		
Rice	Cereals	Cereals
Spaghettis		
White bread		
Pan loaf		
Apple	Fruits	Fruits
Orange		
Pear		
Banana		
Eggs	Eggs	Eggs
Croissant	Industrial bakery	Industrial bakery
Cookies		
Muffin		
Olive oil	Oils and fats	Oils and fats
Sunflower oil		
Corn oil		
Margarine		
Butter		
Extra-virgin olive oil	Other oils	

recovery ratios. Measurements and quantifications were performed by high-resolution gas chromatography/high-resolution mass spectrometry (HRGC/HRMS), Model Fisons CE 8000 GC coupled with a VG Autospec Ultima system (EI and multiple ion determination mode resolution > 10,000), using DB5-type nonpolar columns. Detection limits were between 0.001 and 0.020 ng/kg wet weight (ww), depending on the specific food sample and the different PCDD/F congeners.

2.3. Calculations

Toxic equivalents (TEQ) were calculated using the toxicity equivalent factors established by the WHO (WHO-TEQ). For the estimation of the average daily intake of PCDD/Fs, it was assumed that non-detected congener concentrations were equal to one-half of the respective limit of detection (ND = 1/2 LOD). Estimates of average daily food consumption were obtained from a study carried out in the same area (Martí-Cid et al., 2008). Kohonen's Self-Organizing Map (SOM), a special kind of artificial neural network (ANN), was built with the database on PCDD/Fs in foodstuffs. SOM is a "data mining" multivariate technique, whose use has been continuously increasing for data analysis in environmental and agricultural sciences, because of its considerable capabilities to manage large amounts of data (Janaszek and Trajer, 2011; Mari et al., 2010; Ocampo-Duque et al., 2012; Vänninen et al., 2011). Furthermore, this classification technique allows a very friendly visualization and clustering of data (Roig et al., 2011).

3. Results and discussion

The current concentrations of 17 PCDD/F congeners in food samples collected in various locations from Tarragona County (Catalonia, Spain), classified according to 13 food groups are shown in Table 2. The WHO-TEQ values for each group of food samples are also given. Moreover, data obtained in our previous (2006) survey (Martí-Cid et al., 2008) are also included. The highest WHO-TEQ value corresponded to the industrial bakery (0.183 ng/kg ww), followed by fish (0.156 ng/kg ww), oils and fats (0.112 ng/kg fat weight), and seafood (0.065 ng/kg ww). In contrast, the lowest values were detected in pulses and tubers (0.003 ng/kg ww), and cereals and fruits (0.004 ng/kg ww). In the previous (2006) survey, the highest WHO-TEQ values were found in oils and fats (0.147 ng/kg fat weight), eggs (0.134 ng/kg ww), fish (0.086 ng/kg ww) and seafood (0.069 ng/kg ww), while the lowest levels corresponded to fruits and milk (0.003 ng/kg ww) and

Table 2
Concentrations of PCDD/Fs in food samples collected in different locations from Tarragona County (Catalonia, Spain) near to the HWI.[a]

Congener	Vegetables		Tubers		Pulses		Cereals		Fruits		Fish		Seafood	
	(n = 2)		(n = 2)		(n = 2)		(n = 2)		(n = 2)		(n = 6)	(n = 8)	(n = 2)	(n = 4)
	2006	2012	2006	2012	2006	2012	2006	2012	2006	2012	2006	2012	2006	2012
2,3,7,8-TCDD	<0.001	<0.002	–	<0.001	0.003	<0.001	<0.003	<0.001	<0.001	<0.001	0.014	0.025	0.005	0.009
1,2,3,7,8-PeCDD	0.001	<0.001	–	<0.001	0.006	<0.001	<0.003	<0.001	<0.001	0.001	0.020	0.041	0.014	0.015
1,2,3,4,7,8-HxCDD	0.001	<0.001	–	<0.001	<0.004	<0.001	<0.003	<0.001	0.001	0.001	0.008	0.007	0.014	0.006
1,2,3,6,7,8-HxCDD	0.001	<0.001	–	<0.001	<0.004	0.002	<0.003	0.003	<0.001	0.002	0.026	0.032	0.020	0.018
1,2,3,7,8,9-HxCDD	<0.001	<0.001	–	<0.001	0.003	0.002	0.003	0.003	<0.001	0.004	0.014	0.010	0.017	0.018
1,2,3,4,6,7,8-HpCDD	0.006	0.009	–	0.005	<0.019	0.004	0.013	0.015	<0.006	0.019	0.077	0.056	0.100	0.080
OCDD	0.023	0.039	–	0.022	<0.074	0.006	<0.066	0.245	<0.022	0.128	0.218	0.179	0.645	0.332
2,3,7,8-TCDF	0.003	0.018	–	0.002	<0.008	0.002	<0.007	0.004	0.002	0.002	0.221	0.277	0.120	0.185
1,2,3,7,8-PeCDF	0.001	0.002	–	<0.001	<0.004	<0.001	0.003	<0.001	0.002	0.001	0.070	0.059	0.033	0.021
2,3,4,7,8-PeCDF	0.002	0.004	–	0.002	0.004	0.002	<0.003	0.002	0.001	0.003	0.061	0.152	0.036	0.045
1,2,3,4,7,8-HxCDF	0.003	0.004	–	<0.000	0.009	0.001	0.006	0.002	0.003	0.002	0.054	0.057	0.022	0.016
1,2,3,6,7,8-HxCDF	0.002	0.002	–	<0.000	0.003	<0.000	<0.003	0.001	0.001	0.001	0.026	0.023	0.018	0.008
2,3,4,6,7,8-HxCDF	<0.001	<0.001	–	<0.000	<0.004	<0.001	<0.003	<0.000	0.001	0.001	0.004	<0.003	0.006	<0.003
1,2,3,7,8,9-HxCDF	<0.001	0.001	–	<0.000	<0.004	<0.000	<0.003	<0.001	<0.001	0.002	0.029	0.018	0.010	0.008
1,2,3,4,6,7,8-HpCDF	0.014	0.005	–	0.001	0.043	0.001	0.036	0.003	0.015	0.010	0.081	0.035	0.090	0.017
1,2,3,4,7,8,9-HpCDF	<0.005	<0.001	–	<0.001	<0.019	<0.001	<0.017	0.001	<0.006	0.001	0.019	0.010	<0.029	<0.003
OCDF	0.104	0.007	–	0.002	0.325	<0.001	0.270	0.003	0.104	0.017	0.315	0.051	0.500	0.018
WHO-TEQ	0.004	0.007	–	0.003	0.015	0.003	0.007	0.004	0.003	0.004	0.086	0.156	0.069	0.065

Congener	Meat		Eggs		Milk		Dairy products		Industrial bakery		Oils and fats	
	(n = 7)	(n = 8)	(n = 2)		(n = 4)		(n = 2)	(n = 4)		(n = 2)	(n = 4)	(n = 3)
	2006	2012	2006	2012	2006	2012	2006	2012	2006	2012	2006	2012
2,3,7,8-TCDD	0.002	0.003	0.022	0.008	0.000	<0.001	<0.002	0.005	–	0.004	<0.039	<0.015
1,2,3,7,8-PeCDD	0.002	0.003	0.076	0.005	0.001	0.002	0.008	0.011	–	0.011	0.046	0.013
1,2,3,4,7,8-HxCDD	0.002	0.002	0.016	0.003	0.000	0.001	0.005	0.006	–	0.026	0.049	<0.010
1,2,3,6,7,8-HxCDD	0.007	0.007	0.017	0.008	0.001	0.004	0.010	0.016	–	0.063	0.026	0.028
1,2,3,7,8,9-HxCDD	0.003	0.004	0.016	0.006	0.001	0.002	0.006	0.009	–	0.043	<0.039	0.021
1,2,3,4,6,7,8-HpCDD	0.038	0.048	0.025	0.068	0.002	0.010	0.031	0.056	–	1.186	<0.193	0.304
OCDD	0.119	0.168	0.140	0.192	<0.010	0.011	0.066	0.109	–	5.005	1.465	1.019
2,3,7,8-TCDF	0.009	0.011	0.020	0.026	0.001	<0.001	0.007	0.009	–	0.042	0.046	0.113
1,2,3,7,8-PeCDF	0.004	0.003	0.023	0.010	0.001	<0.001	0.004	0.005	–	0.018	0.056	0.019
2,3,4,7,8-PeCDF	0.004	0.008	0.027	0.013	0.001	0.006	0.023	0.030	–	0.041	0.047	0.055
1,2,3,4,7,8-HxCDF	0.012	0.022	0.031	0.042	0.003	0.003	0.021	0.028	–	0.506	0.185	0.186
1,2,3,6,7,8-HxCDF	0.007	0.009	0.071	0.016	0.002	0.002	0.016	0.016	–	0.181	0.073	0.076
2,3,4,6,7,8-HxCDF	<0.002	0.003	0.024	<0.002	<0.000	<0.001	0.002	<0.002	–	0.155	<0.039	0.046
1,2,3,7,8,9-HxCDF	0.002	0.007	0.018	0.016	0.001	0.002	0.006	0.013	–	0.012	0.027	0.070
1,2,3,4,6,7,8-HpCDF	0.050	0.078	0.054	0.269	0.015	0.002	0.052	0.065	–	3.134	0.725	0.698
1,2,3,4,7,8,9-HpCDF	0.007	0.015	0.018	0.029	0.003	<0.001	<0.012	0.013	–	0.624	0.121	0.121
OCDF	0.294	0.143	0.325	0.215	0.100	<0.001	0.320	0.164	–	5.511	5.125	0.831
WHO-TEQ	0.012	0.017	0.134	0.032	0.003	0.007	0.029	0.036	–	0.183	0.147	0.112

[a] Results are given in ng/kg wet weight excepting oils and fats, which are given in ng/kg fat; n: number of analyzed samples.

vegetables (0.004 ng/kg ww) (Martí-Cid et al., 2008). The most notable difference between both studies corresponded to eggs, with a substantial reduction in the current survey (0.134 vs. 0.032 ng/kg ww). By contrast, the WHO-TEQ corresponding to fish was almost twice that of the previous study (0.086 vs. 0.156 ng/kg ww). The remaining differences were, in general terms, more limited. It is important to note that in our baseline, 2002 and 2006 studies, the group of industrial bakery had not been included. In our 2002 survey (Bocio and Domingo, 2005), the highest WHO-TEQ values corresponded to fish (0.270 ng/kg ww), oils and fats (0.238 g/g fat weight), seafood (0.123 ng/kg ww), and dairy products (0.083 ng/kg ww), while the lowest WHO-TEQ values were found in vegetables and fruits (0.006 and 0.008 ng/kg ww, respectively).

The current TEQ values according to 22 subgroups of foodstuffs, as well as those found in our baseline, 2002 and 2006 surveys are shown in Table 3. In comparison with the baseline results, all the current TEQ values are clearly lower, with the only exception of that found in dairy products, which showed similar values in both campaigns (0.040 vs. 0.036 ng/kg ww). In the present survey, the highest TEQ values corresponded to fish and seafood; specifically to the subgroup of other fish species (red mullet and sole), which had not been analyzed as such subgroup in our previous studies. It was followed by blue fish, seafood, tinned fish and white fish. Blue fish was the subgroup of fish and seafood showing the highest TEQ values in our three previous surveys (Bocio and Domingo, 2005; Domingo et al., 1999; Martí-Cid et al., 2008). In addition to fish and seafood, high concentrations of PCDD/Fs were also observed in industrial bakery and other oils (0.183 ng/kg ww and 0.151 ng/kg fat weight, respectively).

To detect any common profile in the PCDD/F levels in food samples, as well as to determine the relationship among PCDD/F congeners, a SOM algorithm was applied. A rectangular grid of 96 units (12 × 8) was built. The learning and tuning phases consisted of 10,000 steps each. The resulting Kohonen's map, together with the associated component planes (c-planes), is shown in Fig. 1. Five clusters were detected.

Table 3
Concentrations of PCDD/Fs (ng WHO-TEQ/kg wet weight) in foodstuffs purchased in Tarragona County (Catalonia, Spain) in 1998, 2002, 2006 and 2012.

Food subgroup	1998 (Domingo et al., 1999)[b]	2002 (Bocio and Domingo, 2005)	2006 (Martí-Cid et al., 2008)	2012 (present study)	Number of samples in the present study
Vegetables	0.14	0.01	0.004	0.007	2
Tubers	–	–	–	0.003	2
Pulses	0.19	0.01	0.015	0.003	2
Cereals	0.25	0.04	0.007	0.004	2
Fruits	0.09	0.01	0.003	0.004	2
White fish	0.27	0.07	0.055	0.076	2
Blue fish	0.76	0.61	0.133	0.121	2
Seafood	0.42	0.12	0.063	0.092	2
Other fish	–	–	–	0.340	2
Tinned fish	0.24	0.13	0.093	0.089	2
Cephalopods	–	–	–	0.037	2
Pork	0.11	0.02	0.013	0.013	2
Chicken	0.11	0.03	0.011	0.008	2
Beef	0.13	0.03	0.012	0.030	2
Lamb	0.13	0.04	0.014	0.019	2
Eggs	0.12	0.04	0.134	0.032	2
Whole milk	0.18	0.02	0.003	0.010	2
Semiskimmed milk	0.06	0.01	0.003	0.004	2
Dairy products	0.04	0.08	0.029	0.036	4
Oils and fats[a]	0.64[c]	0.20	0.097	0.073[d]	2
Industrial bakery	–	–	–	0.183	2
Other oils[a]	–	–	–	0.151[e]	1

[a] ng WHO-TEQ/kg fat.
[b] ng I-TEQ/kg wet weight.
[c] ng I-TEQ/kg fat.
[d] Olive oil, sunflower oil, corn oil, margarine and butter.
[e] Extra virgin olive oil.

The first cluster was associated with the 2 samples of other fish species (sole and red mullet), both presenting very high concentrations of low-chlorinated PCDD/F congeners. The second cluster was associated to most of the remaining samples of fish and seafood – with a few exceptions – all of them characterized by showing a particular increase of 2,3,7,8-TCDF levels. On the other hand, two more clusters included 3 samples (two of industrial bakery and one of other oils), being all of them associated to high-chlorinated congeners of PCDD/Fs. Finally, all the remaining 33 samples were grouped together into a fifth cluster, characterized by lower concentrations of PCDD/Fs. These results confirm that fish and seafood are the groups presenting the highest accumulation of these chemicals, which is in agreement with the scientific literature. In this same line, when applying SOM to a database of PCDD/Fs in human milk and food products from diverse countries, Nadal et al. (2004) already found evidences that those countries with a greater fish consumption showed also higher PCDD/F concentrations in human milk, confirming a direct link between fish and seafood intake and PCDD/F body burdens. Recently, we have also highlighted the predominant role of fish and seafood in the dietary intake of PCDD/Fs, as well as other chlorinated compounds (Perelló et al., 2012).

Table 4 shows the estimated current dietary intake of PCDD/Fs for the population living in the area under potential influence of the stack emissions of the HWI. Data corresponding to the baseline, 2002 and 2006 surveys are also shown. In addition, information on the consumption rate (g/day) of the 12 food groups (fish and seafood are shown together) is also summarized. The current daily intake of PCDD/Fs by the general population through the diet was 33.1 pg WHO-TEQ, being fish and seafood (11.6 pg WHO-TEQ), oils and fats (4.61 pg WHO-TEQ), dairy products (3.79 pg WHO-TEQ), and industrial bakery (3.49 pg WHO-TEQ) with the groups showing the highest contribution to the total TEQ. The lowest daily contributions corresponded to pulses (0.08 pg WHO-TEQ) and tubers (0.25 pg WHO-TEQ). The current total intake was considerably lower than that found in the baseline study, 210.1 pg I-TEQ/day (Domingo et al., 1999), with a reduction of approximately 84%. According to the specific food groups, the highest decreases corresponded to cereals (98%), pulses (97%), fruits (96%), and milk (95%). The only group showing an increase between the baseline and the current study was the group of dairy products (1.80 vs. 3.79 pg TEQ/day). The dietary intake of PCDD/Fs estimated in the present study was also notably lower than that found in our 2002 study (59.6 pg I-TEQ/day) (Bocio and Domingo, 2005), although slightly higher than the intake estimated in the 2006 survey: 27.8 pg WHO-TEQ/day (Martí-Cid et al., 2008). To explain some of these last differences, it is important to note that, in contrast to our previous studies, the present survey included two new food groups: tubers and industrial bakery, which contributed with values of 0.25 and 3.49 pg WHO-TEQ, respectively (in percentages, 0.8% and 10.5%, respectively), to the current total daily intake of PCDD/Fs. Excluding these two groups, the total intake would have been 29.4 pg WHO-TEQ/day, a very similar value to that estimated in 2006 (27.8 pg WHO-TEQ/day).

Fig. 2 shows the percentages of contribution from each food group to the total dietary intake (pg WHO-TEQ/day) of PCDD/Fs by the population living in the area under evaluation. The highest percentage of contribution corresponded to fish and seafood (34.9%), followed by oils and fats (13.9%) and dairy products (11.4%). By contrast, the lowest contributors were pulses (0.2%), tubers (0.8%) and cereals (2.6%). In the baseline survey, the percentages of contribution were notably different to the current ones, having cereals (23.1%) as the highest contributor, followed by milk (15.3%), and fish and seafood (14.5%). The lowest contributors were dairy products (0.84%), pulses (1.45%) and eggs (1.66%) (Domingo et al., 1999). However, in the 2006 study, fish and seafood (28%), and oils and fats (22%) showed also the highest percentages of contribution to the total intake of PCDD/Fs, having pulses (1%) as the group showing the lowest contribution.

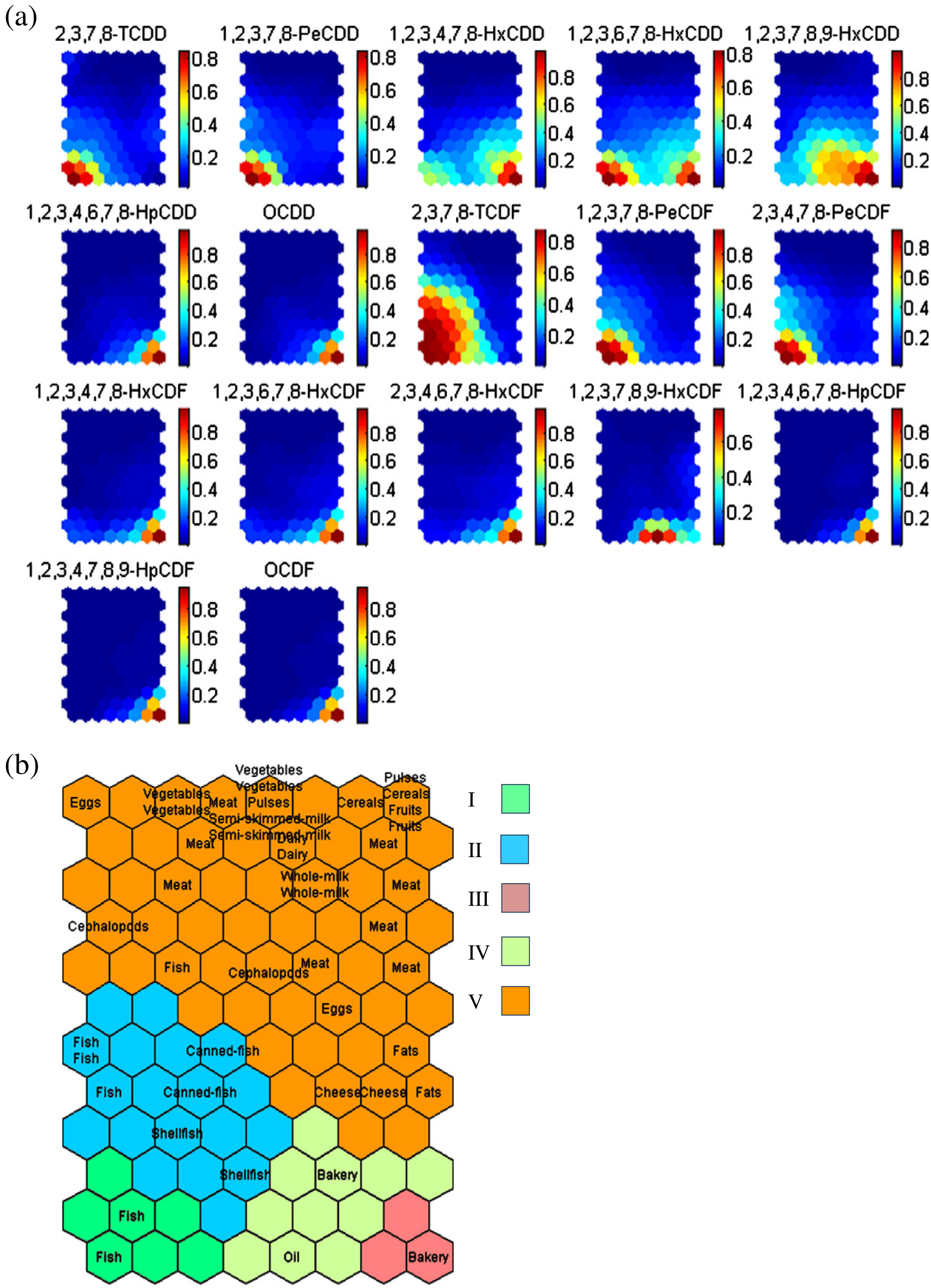

Fig. 1. SOM results for PCDD/F concentrations in food samples: (a) component planes; (b) distribution of samples and clusters. In the 17 component planes, each hexagon represents one map unit. Colors indicate the value of the component in that unit (the higher the value is, the lighter the color is). Hexagons at the same place on different component planes correspond to the same map unit, showing the levels of the components in the weight vector of that unit. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

The estimated dietary intake of PCDD/Fs (pg WHO-TEQ/kg/day) by the general population of the assessed area according to age and gender is depicted in Fig. 3. Children (boys and girls) aged 4–9 years were the group showing the highest daily intake of PCDD/Fs. The remaining five age groups showed rather similar values. In any of the six age groups, gender differences were appreciable. When the calculations

Table 4
Estimated daily intake of PCDD/Fs by the general population living near a HWI in Tarragona County (Catalonia, Spain).

Food group	Consumption rate[a] (g/day)			pg I-TEQ/day	pg WHO-TEQ/day		
	1998[b]	2002[c] and 2006[d]	2012	1998	2002	2006	2012 (present study)
Vegetables	122 (10.7)	226 (16.5)	226 (15.4)	17.1	1.3	0.86	1.67
Tubers	–	–	74 (5.1)	–	–	–	0.25
Pulses	16 (1.4)	24 (1.8)	24 (1.6)	3.0	0.2	0.36	0.08
Cereals	194 (17.0)	206 (15.0)	206 (14.1)	48.5	8.3	1.42	0.87
Fruits	269 (23.6)	239 (17.4)	239 (16.3)	24.2	1.8	0.75	1.01
Fish and seafood	72 (6.3)	92 (6.7)	92 (6.3)	30.4	20.2	7.91	11.6
Meat and meat products	173 (15.1)	185 (13.5)	185 (12.6)	20.8	5.4	2.26	3.18
Eggs	29 (2.5)	34 (2.5)	34 (2.3)	3.5	1.3	4.59	1.11
Milk	18 (15.6)	217 (15.8)	217 (14.8)	32.0	3.9	0.62	1.50
Dairy products	44 (3.9)	106 (7.7)	106 (7.2)	1.8	8.3	3.02	3.79
Oils and fats	45 (3.9)	41 (3)	41 (2.8)	28.8	8.8	6.01	4.61
Industrial bakery	–	–	19 (1.3)	–	–	–	3.49
Total intake	1142 (100)	1370 (100)	1463 (100)	210.1	59.6	27.8	33.1

[a] In parentheses: percentage on the total consumption.
[b] Domingo et al. (1999).
[c] Bocio and Domingo (2005).
[d] Martí-Cid et al. (2008).

were performed taking into account the average body weight estimated for each age/gender group (Table 5), the differences among groups were rather low, ranging between 25.9 pg WHO-TEQ/day for women aged >65 years, and 33.5 WHO-TEQ/day for male adults aged 35–50 years, respectively.

Although human exposure to PCDD/Fs may occur through a number of routes such as inhalation, dermal contact, and ingestion of soils and dust, it is well known that for non-occupationally exposed individuals the diet is quantitatively the main way of exposure (Bilau et al., 2009; Nadal et al., 2004; Passuello et al., 2010; Schuhmacher and Domingo, 2006). Previously, the environmental exposure to PCDD/Fs, derived from the concentrations of dioxins and furans in soil and air samples from the vicinity of the HWI here evaluated, was calculated and compared with the dietary intake. Food exposure accounted for 99% of the total exposure (Vilavert et al., 2010). Furthermore, a global analysis of environmental and biological samples showed that the weight of the PCDD/F stack emissions of the HWI on the environmental burden and on the exposure of the individuals living in the surroundings was not significant (Mari et al., 2010). In recent years, various international organizations have established the tolerable daily intake (TDI) to PCDD/Fs including that of dioxin-like PCBs. Thus, the WHO established a TDI for PCDD/Fs (plus dioxin-like PCBs) in the range of 1–4 pg WHO-TEQ/kg body weight per day for the non-carcinogenic effects of these organic pollutants (van Leeuwen et al., 2000). This TDI was replaced to the previous one, which had been estimated in 10 pg I-TEQ/kg/day. On the other hand, the Committee on Toxicity of Chemical in Food, Consumer Products and the Environment of the United Kingdom proposed a TDI of 2 pg WHO-TEQ/kg body weight per day (COT, 2001), while the Scientific Committee on Food (SCF) of the European Commission (EC) recommended a temporary tolerable intake on a weekly basis (t-TWI) of 7 pg WHO-TEQ/kg body weight. However, based on new findings, the t-TWI was corrected to 14 pg WHO-TEQ/body weight per week (SCF, 2001). In turn, the FAO/WHO joint expert committee on food additives and contaminants (JEFCA) also evaluated the health implications of PCDD/Fs, suggesting a provisional maximum tolerable monthly intake of 70 pg TEQ/kg body weight per month (JEFCA, 2002). In fact, the tolerable daily intake recommended for the sum of PCDD/Fs and dioxin-like PCBs by different international scientific institutions is ~2 pg WHO-TEQ/kg. Based on practical reasons, the weekly and monthly maximum intakes are being handled as if it was a TDI of 2 pg WHO-TEQ/kg. A detailed observation of the results of the present study shows that they are clearly lower than these recommendations, which are not exceeded for any age/gender group of the population living in the vicinity of the HWI here assessed.

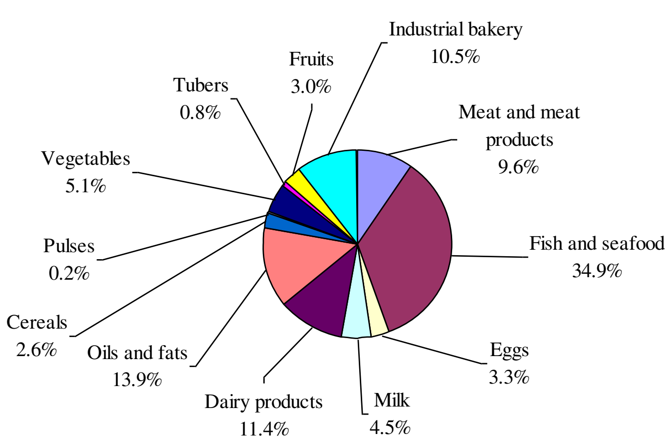

Fig. 2. Percentages of contribution from each food group to the total daily intake (pg WHO-TEQ/day) of PCDD/Fs in 2012 by the population of Tarragona County, (Catalonia, Spain) living near a HWI.

Table 6 summarizes the results of a number of recent studies (2010–2012), in which the concentrations of PCDD/Fs in foods were measured, and where the dietary intake of these compounds was also estimated. Additional information on studies reported before 2010 can be found in Domingo et al. (1999), Bocio and Domingo (2005) and Martí-Cid et al. (2008). It can be seen that the current intakes were lower than those reported for Japan by Nakatani et al. (2011) corresponding to the data from 2000 (104.24 pg TEQ/person/day), 2001 (72.73 pg TEQ/person/day) and 2002 (82.78 pg TEQ/person/day). In the region of Zhejiang, China, Song et al. (2011), determined the levels of PCDD/Fs and dioxin-like PCBs in six food groups, and estimated the dietary intake for the local residents in an area where e-waste had been recycled, and in another area where the agricultural activities were dominating. The estimated daily intakes were considerably different, being 805.2 and 74.3 pg WHO-TEQ in the presumably polluted area and in the agricultural area, respectively. In Sweden, Törnkvist et al. (2011) estimated an intake of 21.8–54.4 pg WHO-TEQ/day in men (equivalent to 0.31–0.78 pg WHO-TEQ/kg/day for a 70-kg male), with values of the same order than those of the present survey. On the other hand, in a total diet study conducted in France, Sirot et al. (2012) reported mean values of 0.57 and 0.89 pg WHO-TEQ/kg body weight/day for the intake of PCDD/Fs through the diet by adults, on one hand, and by children and

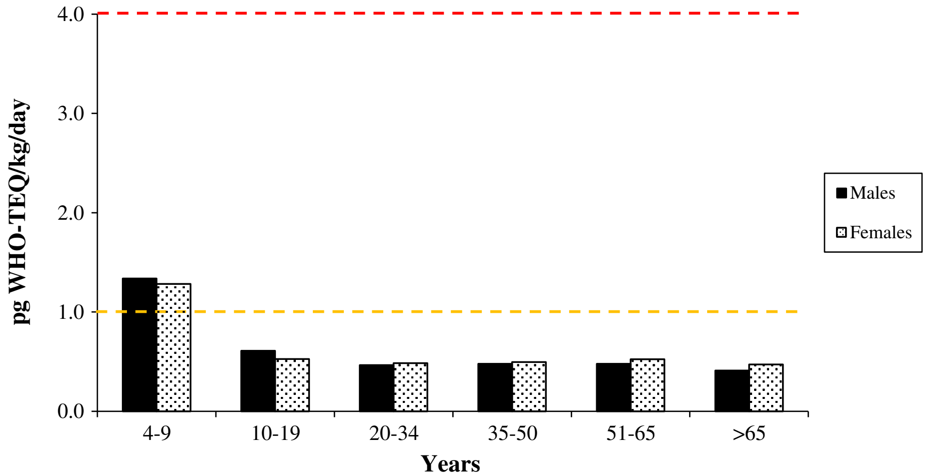

Fig. 3. Estimated daily intake (pg WHO-TEQ/kg body weight/day) of PCDD/Fs by the general population of Tarragona County (Catalonia, Spain) living near a HWI, according to sex and age. Broken lines indicate the lower- and upper-threshold of the TDI range according to the WHO. Average body weight: 4–9 years: 24 kg; 10–19 years: 56 kg (M) and 53 kg (F); 20–34, 35–50, 51–65 and >65 years: 70 kg (M) and 55 kg (F).

teenagers, on the other. For a subject of 70-kg, it would mean a daily intake of 39.9 pg WHO-TEQ, which is also very similar to the intake found in the current survey. In Belgium, Windal et al. (2010) reported a mean dietary intake of PCDD/Fs and dioxin-like PCBs by an adult population of 0.72 pg TEQ/kg/day (using the 1998 TEFs, or 0.61 pg TEQ/kg/day, using the 2005 TEFs), which is also higher than that found for the population living near the HWI here assessed. Finally, in another study also carried out in a Spanish Autonomous Community, Valencia, Marin et al. (2011) established that the dietary intake of PCDD/Fs plus dioxin-like PCBs was 2.86 and 4.58 pg WHO-TEQ/kg/day for adults and children, respectively, intakes that are also clearly higher than those corresponding to the current study. However, it is important to note that the dietary intakes reported in all the above studies refer to PCDD/Fs plus dioxin-like PCBs, while those of the present survey concern only PCDD/Fs, as emissions of PCBs, as those of other organic pollutants, are not included in the surveillance program of the HWI. Among the emitted pollutants by the HWIs, heavy metals and PCDD/Fs are those raising the major concern with respect to human health risks. According to a recent study performed for the entire population of Catalonia, in which we also determined the dietary intake of dioxin-like PCBs, we found that the contribution of dioxin-like PCBs to the total intake of both group of pollutants was approximately twice that of PCDD/Fs: 52.4 and 25.7 pg WHO-TEQ/day, respectively (Llobet et al., 2008).

The results of the present survey show a continued decrease in the dietary exposure to PCDD/Fs of the population living in the neighborhood of the HWI. This reduction agrees well with the results of other recent studies also performed in Catalonia. Thus, Perelló et al. (2012) found that, for a standard adult man (body weight 70 kg), the mean intake of PCDD/Fs through the consumption of 11 food groups was 15.72 pg WHO-TEQ/day, an intake lower than the 25.7 pg WHO-TEQ/day found in a previous study carried out with the same general characteristics (Llobet et al., 2008), and also lower than the intake found in this study. In conclusion, the dietary intake of PCDD/Fs by the population living near the HWI here evaluated should not mean any additional increase in the concentrations of these organohalogenated contaminants in those biological tissues (plasma, milk, autopsy tissues) periodically analyzed in the surveillance program. Consequently, any potential increase hypothetically observed in those periodical measurements should not be attributed to the dietary exposure to PCDD/Fs, being very probably due to environmental exposure to these pollutants.

Acknowledgments

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Table 5
Estimated daily intake of PCDD/Fs (pg WHO-TEQ/day) by the population of Tarragona County (Catalonia, Spain) living near a HWI, according to sex and age.

Age (years)	4–9		10–19		20–34		35–50		51–65		>65	
Sex (kg; average body weight)	M (24)	F (24)	M (56)	F (53)	M (70)	F (55)	M (70)	F (55)	M (70)	F (55)	M (70)	F (55)
Vegetables	0.96	0.89	1.22	1.19	1.40	1.29	1.85	1.61	1.77	1.60	1.42	1.39
Tubers	0.24	0.20	0.30	0.22	0.27	0.20	0.25	0.19	0.24	0.19	0.31	0.17
Pulses	0.09	0.08	0.08	0.08	0.08	0.07	0.09	0.09	0.07	0.07	0.08	0.06
Cereals	0.84	0.85	1.11	0.76	1.00	0.68	0.87	0.57	0.74	0.50	0.75	0.57
Fruits	0.84	0.82	0.87	0.84	0.89	0.87	1.03	0.88	1.10	1.13	1.26	1.01
Fish and seafood	6.79	6.16	8.05	7.55	9.43	9.18	11.95	9.69	13.3	11.1	9.81	10.3
Meat and meat products	2.41	2.41	3.11	2.63	3.87	2.37	2.86	2.18	2.80	1.89	2.08	1.84
Eggs	1.10	0.61	0.97	0.68	1.22	0.74	1.00	0.81	1.10	0.71	0.93	0.52
Milk	2.26	2.02	2.10	1.59	1.59	1.68	1.55	1.68	1.37	1.91	1.52	2.00
Dairy products	4.04	4.11	4.86	3.90	4.43	3.18	3.68	3.11	3.25	3.50	2.47	2.68
Oils and fats	3.71	3.82	4.61	3.48	4.83	3.48	4.83	3.60	4.16	3.37	3.15	3.37
Industrial bakery	8.83	8.83	6.84	5.02	3.49	2.91	3.49	2.91	3.49	2.91	5.02	2.02
pg WHO-TEQ/day	32.1	30.8	34.1	27.9	32.5	26.7	33.5	27.3	33.4	28.8	28.8	25.9

M: Males; F: Females.

Table 6
Dietary intake of PCDD/Fs and foodstuffs assessed: a summary of recent reports (2010–2012) of various countries, as well as results of our previous surveys in the area.

Country	Foods analyzed	pg WHO-TEQ/day	pg WHO-TEQ/kg/day	Reference
Tarragona, Spain[c]	Meat, fish and seafood, milk, dairy products, vegetables, tubers, pulses, cereals, fruits, eggs, industrial bakery, oils and fats	33.1	0.47[a]	This study
France[d]	Milk, other dairy products (including yogurt), cheese, eggs and egg products, butter, oil, margarine, meat, poultry and game meat, offal, meat products, fish, crustaceans and molluscs, vegetables, pizza, pretzels, burgers and sandwiches, main courses, desserts and cream, condiments and sauces		0.57 (adults) 0.89 (children and teenagers)	Sirot et al. (2012)
Sweden[c]	Fish/fish products, meat/meat products, dairy products, eggs, oils and fats	21.8–54.4	0.31–0.78	Törnkvist et al. (2011)
Japan[d]	Rice, seeds, potatoes, sugar and confectionery, fats and oils, vegetables, fruits, green vegetables, vegetables, mushrooms and seaweed, condiments and beverages, seafood, meat, eggs, milk and dairy products, other food drinking water	72.73–104.24	1.45–2.08	Nakatani et al. (2011)
Valencia, Spain[d]	Vegetables, cereals, fats and oils, eggs, milk and dairy products, fish products, meat and meat products and fish oil		2.86 (adults) 4.58 (children)	Marin et al. (2011)
China[d]	Hornbeam red, duck, chicken eggs, chicken, rice and vegetables	74.31–805.17	1.24–13.42	Song et al. (2011)
Belgium[d]	Milk and dairy products, meat and fish		0.72	Windal et al. (2010)
Tarragona, Spain[c]	Meat and meat products, fish and seafood, eggs, milk, dairy products, oils and fats, bread and cereals, pulses, vegetables, fruits	27.8	0.40[a]	Martí-Cid et al. (2008)
Tarragona, Spain[c]	Meat and meat products, fish and seafood, eggs, milk, dairy products, oils and fats, bread and cereals, pulses, vegetables, fruits	63.8	0.91[a]	Bocio and Domingo (2005)
Tarragona, Spain[c]	Meat and meat products, fish and seafood, eggs, milk, dairy products, oils and fats, bread and cereals, pulses, vegetables, fruits	210[b]	3.0[a,b]	Domingo et al. (1999)

[a] Estimated weight: 70 kg.
[b] In pg I-TEQ/day.
[c] PCDD/Fs only.
[d] PCDD/Fs + dioxin-like PCBs.

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Glossary

ARC: Agència de Residus de Catalunya
ANN: Artificial Neural Network
COT: Committee on Toxicity of Chemical in Food
c-planes: component Planes
EC: European Commission
JEFCA: FAO/WHO Joint Expert Committee on Food Additives and Contaminants
FAO: Food and Agriculture Organization
HW: hazardous waste
HWI: hazardous waste incinerator
LOD: limit of detection
MSW: municipal solid waste
ND: no detected
NIMBY: Not In My Back Yard
PCBs: polychlorinated biphenyls
PCDD/Fs: polychlorinated dibenzo-*p*-dioxins and dibenzofurans
SCF: Scientific Committee on Food
SOM: Kohonen's Self-Organizing Map
t-TWI: Temporary Tolerable Intake on a Weekly Basis
TDI: Tolerable Daily Intake
TEF: Toxic Equivalency Factor
TEQ: Toxic Equivalents
US EPA: United States Environmental Protection Agency
ww: wet weight
WHO-TEQ: World Health Organization-Toxic Equivalents