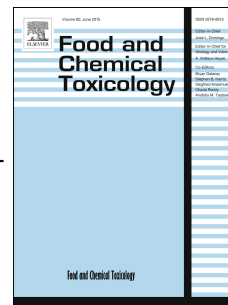


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Levels of PCDD/Fs in foodstuffs in Tarragona County (Catalonia, Spain): Spectacular decrease in the dietary intake of PCDD/Fs in the last 20 years

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1 **Levels of PCDD/Fs in foodstuffs in Tarragona County**
2 **(Catalonia, Spain): Spectacular decrease in the dietary**
3 **intake of PCDD/Fs in the last 20 years**

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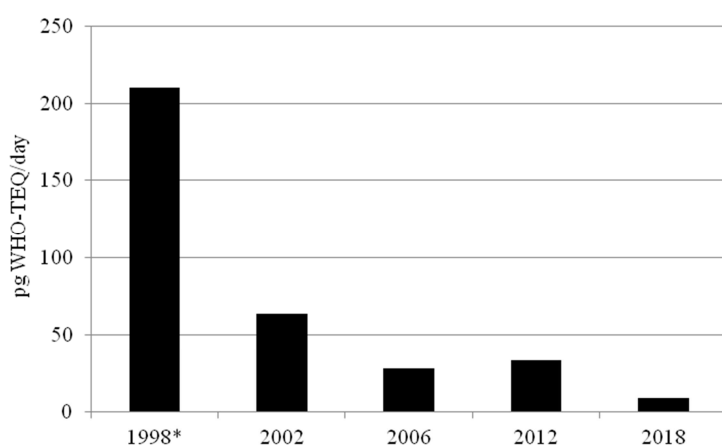
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29

30 Highlights

- 31 ▪ Dietary exposure to PCDD/Fs was assessed for the Tarragona County
- 32 population
- 33 ▪ Major food group contributor to the exposure was fish and seafood
- 34 ▪ Spectacular decrease in the exposure to PCDD/Fs was observed in the last 20
- 35 years

36 Graphical abstract



37

38 * In pg I-TEQ/day

39 **ABSTRACT**

40

41 In this study, we determined the levels polychlorinated dibenzo-*p*-dioxins (PCDDs) and
42 and dibenzofurans (PCDFs) in 45 “composite” samples of food items belonging to 12
43 food groups. Foodstuffs were purchased in various locations of Tarragona County,
44 Catalonia, Spain. The daily dietary intake of PCDD/Fs was subsequently estimated for
45 the population living in that County. The highest toxic equivalent (WHO-TEQ) value
46 corresponded to fish (0.103 ng/kg ww), followed by seafood (0.071 ng/kg ww) and eggs
47 (0.070 ng/kg ww). In contrast, the lowest PCDD/F concentrations corresponded to
48 vegetables, tubers, pulses, and fruits (each, 0.003 ng/kg ww). The current dietary intake
49 of PCDD/Fs by the adult population was estimated in 8.54 pg WHO-TEQ/day, being
50 fish and seafood the food groups with the greatest contribution to the dietary exposure
51 of PCDD/Fs, followed by eggs, as well as by meat, oils and fats. The current study
52 shows a continued decrease in the exposure to PCDD/Fs by the general population of
53 Tarragona County since the first survey in the area conducted in 1998. The reduction
54 observed in the last 20 years (from 210 pg I-TEQ/day to 8.54 pg WHO-TEQ/day) is

55 certainly spectacular. The current daily intake (0.122 pg WHO-TEQ/kg bw) is also
56 lower than most intakes recently reported in a number of regions and countries.

57

58

59 *Keywords:* PCDD/Fs, foodstuffs, human exposure, dietary intake, Tarragona County (Catalonia,
60 Spain).

61

ACCEPTED MANUSCRIPT

62 Abbreviations

63 AECOSAN, spanish agency of consumption, food safety and nutrition; EC, European
64 commission; ENALIA, spanish national food survey in children and adolescents;
65 ENALIA2, spanish national food survey in adults, elderly and pregnant women; FAO,
66 food and agriculture organisation; HWI, hazardous waste incinerator; HRGC/HRMS,
67 high resolution gas chromatography/high resolution mass spectrometry; IARC,
68 international agency for research on cancer; JECFA, joint expert committee on food
69 additives; MSWI municipal solid waste incinerator; PCDDs, polychlorinated dibenzo-
70 *p*-dioxin; PCDFs, polychlorinated dibenzofurans; POPs, persistent organic pollutants;
71 PTMI, provisional tolerable monthly intake; SCF, scientific committee on food; TCDD,
72 2,3,7,8- tetrachlorodibenzo-*p*-dioxin; TDI, tolerable daily intake; TEF, toxic
73 equivalence factors; TEQ, toxic equivalent; TWI, tolerable weekly intake.

74 **1. Introduction**

75

76

77 Polychlorinated dibenzo-*p*-dioxins (PCDDs) and dibenzofurans (PCDFs) are well
78 known environmental pollutants, which belong to the called “*dirty dozen*”, a group of
79 dangerous persistent organic pollutants (POPs). PCDD/Fs are of notable concern given
80 their important toxic potential, which can be cause of reproductive and developmental
81 problems, damage to the immune system, interferences with hormones, and can also
82 cause cancer (WHO, 2016). Regarding this, 2,3,7,8- tetrachlorodibenzo-*p*-dioxin
83 (TCDD) was evaluated by the International Agency for Research on Cancer (IARC) in
84 1997 and 2012, being classified as a “known human carcinogen” (WHO, 2016).

85 PCDD/Fs can be released in a number of natural processes, such as volcanic
86 eruptions and forest fires. However, they are always unwanted by-products, whose
87 environmental presence is mainly due to emission of industrial processes such as
88 incineration of hazardous, municipal and medical wastes, cement plants, chlorine
89 bleaching of paper pulp or smelting, as well as traffic of motor vehicles (Fiedler, 2016;
90 Nadal et al., 2009; Rovira et al., 2014; Schuhmacher et al., 2004).

91 The only hazardous waste incinerator (HWI) in Spain is located in Tarragona
92 County (Catalonia). It was built during the period 1996-1998, starting regular operations
93 in 1999. A municipal solid waste incinerator (MSWI) is also situated in the same area,
94 together with most important chemical/petrochemical complex in southern Europe. In

95 addition, a heavy traffic flows through this area. This combination of potential emission
96 sources could mean, in principle, an increase of the PCDD/F concentrations in the zone.
97 Taking into account that more than 90% of human exposure to PCDD/Fs is due to food
98 consumption, in 1998 we conducted a first food survey, whose main goal was to
99 estimate the dietary exposure to PCDD/Fs by the population living in Tarragona County
100 (Domingo et al., 1999). In order to establish the temporal trend in human dietary
101 exposure to PCDD/Fs in Tarragona County, we conducted a second, third and fourth
102 surveys in 2002, 2006 and 2012, respectively (Bocio and Domingo, 2005; Domingo et
103 al., 2012; Martí-Cid et al., 2008). The results of our fifth campaign, carried out in 2018,
104 in which the concentrations of PCDD/Fs were again determined in foodstuffs, are here
105 presented. As in our previous surveys, we also estimated the dietary intake of PCDD/Fs
106 by the population of the zone. The current results have been compared with those
107 obtained in our previous studies (1998, 2002, 2006 and 2012) (Bocio and Domingo,
108 2005; Domingo et al., 1999, 2012; Martí-Cid et al., 2008). Data from recent (2012-
109 2018) studies carried out in other regions and countries around the world have been also
110 reviewed.

111

112 **2. Materials and methods**

113 *2.1. Sampling*

114 In January-February 2018, food samples were randomly purchased at various
115 hypermarkets, supermarkets, local markets and grocery stores from 8 cities and villages
116 (Reus, La Canonja, Tarragona, Constantí, Bonavista-Torreforta, La Pobla de Mafumet,
117 Sant Salvador, and El Morell) in Tarragona County. It is important to emphasize that, as
118 in all the previous surveys, most food items were not local products, as their
119 consumption is relatively minor. In the area, there are neither pasture grounds, nor crops
120 of vegetables, grains, or fruits, products that are consumed in significant amounts by the
121 individuals living in the area. Therefore, for the objective of the study, we decided that
122 foodstuffs would be of any origin. Thus, they were randomly purchased, being PCDD/F
123 exposure from ingestion of local products considered as irrelevant. The only exception
124 was the olive oil of a local cooperative (Constantí, Tarragona County).

125 A total of 67 types of foods were purchased, being classified in the following
126 groups: 1) vegetables: lettuce, tomato, green bean, spinach and cauliflower;

127 2) tubers: potato and carrot; 3) pulses: lentils, white beans and chickpeas;
128 4) bread and cereals: white bread, tin loaf, rice, and pasta; 5) fish and seafood: white
129 fish (hake, whiting, sea bass and anglerfish), blue fish (salmon, sardine and tuna),
130 seafood (mussels and prawns), others (red mullet and sole), canned fish (tuna, sardine
131 and mussels), cephalopods (cuttlefish and squid); 6) meat: pork (fillet, sausage,
132 hamburger, sweet ham, *frankfurt* sausage and *chorizo*), chicken (breast, thigh and
133 sausage), beef (fillet and hamburger), lamb (thigh and rib); 7) eggs: hens eggs; 8) milk
134 and dairy products: whole milk, semi-skimmed milk, dairy products (yoghurts, *petit-*
135 *Swiss*, custards), cheeses (tender, semi-cured, cured); 9) fruits: apple, orange, pear and
136 banana; 10) oils and fats: olive oil, sunflower oil, margarine and butter; 11) industrial
137 bakery: croissant, cookies and muffins; 12) specific extra virgin olive oil of Constantí.
138 Each final sample or "composite" of the respective food item was formed by individual
139 sub-groups. In total, the samples were purchased in 14 different commercial
140 establishments, all of which were considered to be sufficiently representative for the
141 zone.

142 A total of 938 individual food samples (67 different food items x 14 establishments)
143 were processed for PCDD/F analysis. A "composite" of each food consisted of 50 g of
144 each of the 14 individual samples, which were acquired in the 8 locations.
145 Subsequently,, each "composite" for a specific food group, consisted of the same
146 proportion of the different subunits for that group to a final weight of 500 g. In
147 summary, 45 composite samples were analyzed (22 duplicate samples plus one
148 composite of extra virgin olive oil (Cosntantí), belonging to 67 different food items.

149

150 2.2. Chemical analysis of PCDD/Fs

151

152 The analysis of the levels of PCDD/Fs in food samples was based on the US EPA
153 1625 method. Previously to analyses, samples were homogenized and kept in a freezer.
154 From 5 to 10 g of dry and frozen samples were mixed with a small amount of Na₂SO₄,
155 marked with ¹³C₁₂-labeled congeners of PCDD/Fs and extracted with an organic solvent
156 for 24 hours. The lipid fraction of the sample was isolated, being a part used for the
157 purification process. The purification and fractionation of the aliquot of the samples was
158 carried out by means of adsorption chromatography with multiple steps, using
159 multilayer columns of silica (top to bottom: sodium sulfate, silica, silica/sulfuric acid,
160 silica, silica/potassium hydroxide, silica) and alumina. The final step was the reduction

161 of the fractions containing PCDD/Fs to the necessary volume. Before analysis, a
162 PCDD/F standard marked with $^{13}\text{C}_{12}$ was added to calculate the recovery percentages.
163 The purified extract was analyzed by high resolution gas chromatography/high
164 resolution mass spectrometry (HRGC/HRMS), Model Fisons CE 8000 GC coupled to a
165 VG Autospec Ultima (Electronic Impact Mode and Multiple Detection of Ions,
166 resolution > 10,000). The quantification of the PCDD/Fs was carried out by means of
167 internal standards. The detection limits ranged from 0.0007 pg/g (congener 1,2,3,6,7,8-
168 HxCDF in vegetables and semi-skimmed milk) to 0.082 pg/g (congener OCDD in oils
169 and fats).

170

171 2.3. Calculations

172

173 Toxic equivalents (TEQ) were calculated using the toxic equivalence factors
174 established by the WHO (WHO-TEF) (van den Berg et al., 2006). For the quantification
175 of PCDD/F concentrations in different food groups, and in order to be able of
176 comparing the current results with the limits proposed by the European Commission, we
177 assumed that non-detected congener concentrations were equal to one-half of the
178 respective limit of detection ($\text{ND} = \frac{1}{2} \text{LOD}$). The estimation of the average
179 consumption of food was obtained from the Spanish National Food Survey in Children
180 and Adolescents (ENALIA) and the Spanish National Food Survey in Adults, Elderly
181 and Pregnant Women (ENALIA2), both of them conducted by the Spanish Agency of
182 Consumption, Food Safety and Nutrition (AECOSAN) (ENALIA, ENALIA 2, 2016).

183

184

185 3. Results and discussion

186

187 The concentrations of the 17 most toxic PCDD/F congeners in the 13 food groups
188 are summarized in Table 1. The WHO-TEQ values for each group are also shown. For
189 comparative purposes, we have also included data of the previous study conducted in
190 2012 (Domingo et al., 2012). The highest WHO-TEQ value corresponded to fish (0.103
191 ng/kg wet weight), followed by seafood (0.071 ng/kg ww) and eggs (0.070 ng/kg ww).
192 In contrast, the lowest PCDD/F concentrations corresponded to vegetables, tubers,
193 pulses, and fruits (each, 0.003 ng/kg ww). In the 2012 survey, industrial bakery, fish

194 and oils and fats showed the greatest levels of PCDD/Fs (0.183 ng/kg ww, 0.156 ng/kg
195 ww and 0.112 ng/kg fat weight, respectively). The comparison of both set of data shows
196 that decrease in the levels of PCDD/Fs can be observed in almost all food groups, being
197 the industrial bakery the group in which the greatest decreased was mostly noted (from
198 0.183 to 0.006 ng/kg ww). By contrast, eggs and seafood showed slight increases in the
199 concentration of PCDD/Fs. In a previous study performed in our laboratory in 2006
200 (Martí-Cid et al., 2008), the highest PCDD/F levels corresponded to oils and fats (0.147
201 ng/kg fat weight), followed by eggs (0.134 ng/kg ww). These levels were considerably
202 higher than those found in the present study

203 The dietary intakes of PCDD/Fs by the general population living in Tarragona
204 County in 1998, 2002, 2006, 2012 and 2018 are summarized in Table 2. The current
205 intake was estimated in 8.54 pg WHO-TEQ/day, being fish and seafood the food the
206 group with the greatest contribution to the dietary exposure of PCDD/Fs (2.82 pg
207 WHO-TEQ/day), followed by eggs (1.21 pg WHO-TEQ/day), and by the groups of
208 meat, and oils and fats (both with 0.89 pg WHO-TEQ/day). In turn, the lowest
209 contributions of the dietary intake of PCDD/Fs corresponded to pulses (0.02 pg WHO-
210 TEQ/day), industrial bakery (0.07 pg WHO-TEQ/day), and tubers (0.12 pg WHO-
211 TEQ/day).

212 The comparison of these results with those from our previous surveys shows that the
213 current total dietary intake (8.54 pg WHO-TEQ/kg/day) is dramatically lower than that
214 found in the baseline study (210 pg I-TEQ/day) (Domingo et al., 1999), meaning a 96%
215 decrease. The dietary intake of PCDD/Fs is also much lower than that estimated in the
216 immediately previous survey, conducted in 2012 (33.1 vs 8.54 pg WHO-TEQ/day)
217 (Domingo et al., 2012) (reduction of 74%). A considerable general decrease can be
218 noticed in all food groups, ranging from 38%, for cereals, to 98%, for industrial bakery.
219 The only food group that shows an increase regarding 2012 survey are the eggs, which
220 represents an increase of 9%. With respect to the 1998 study, the decreases are still
221 much more relevant, being all of them higher than 90% (97-99% for vegetables, pulses,
222 cereals, milk, fruits, and oil and fats). The most modest reduction corresponded to dairy
223 products (64%).

224 The tremendous differences in the total dietary exposure to PCDD/Fs can be
225 explained not only by the reduction of the concentrations of these pollutants in

226 foodstuffs, but also taking into account that the consumption data used for the 2012 and
227 2018 have notable differences. Thus, total consumption in 2012 was accounted for 1463
228 g/day, while in 2018, the consumption of the food items included in the study was
229 considerably lower 666 g/day, with a reduction of more than the 50%. Dairy products,
230 pulses, and fish and seafood, have been the food groups with the greatest reduction
231 (67%). However, it is important to remark that even if the same consumption rates are
232 used for estimating the dietary intake of PCDD/Fs, the current total daily intake for
233 these environmental contaminants would be still lower than that found in 2012 (33.1 vs
234 21.1 pg WHO-TEQ/day). Consequently, the important decrease in the human dietary
235 exposure to PCDD/Fs is due to both variables, the notable reduction in the consumption
236 of the foodstuffs here analysed and the decrease in the PCDD/Fs concentrations in the
237 analysed foods.

238 The percentages of the contribution of each food group to the total dietary intake of
239 PCDD/Fs are depicted in Fig. 1. It can be seen that by far, fish and seafood (33%) is the
240 main contributor group to the total exposure to PCDD/Fs, followed by eggs (14.1%),
241 and meat, and oils and fats (both with 10.4%). The lowest contributions corresponded to
242 pulses (0.3%) and industrial bakery (0.8%). The current contributions differ notably
243 from those found in the 2012 survey, when industrial bakery and eggs contributed
244 10.5% and 3.3% to the total exposure, respectively, being the contribution of the
245 remaining food groups rather similar to those estimated in the 2012 study (Domingo et
246 al., 2012).

247 The estimated daily intake for PCDD/Fs by the general population of Tarragona
248 County, classified according to various age groups, is depicted in Fig. 2. The group
249 aged 12-36 months showed the highest daily intake (0.626 pg WHO-TEQ/day). It was
250 followed by the groups aged 6-11 months and 3-9 years, respectively. In general terms,
251 children had higher PCDD/F dietary intakes than adult groups, because of the lower
252 body weight which the former own. To correct the differences in body weight with the
253 age, we have also estimated exposure to PCDD/Fs by the same age groups without, but
254 considering also the average body weight for each group. With these calculations, the
255 highest dietary intake of PCDD/Fs corresponded to adolescents aging 10-17 years (11.2
256 pg WHO-TEQ/day), while the lowest corresponded to the group of babies aged 6-11
257 months (3.67 pg WHO-TEQ/day) (Table 3). As for pregnant women, which are a risk

258 group, exposure to PCDD/Fs is estimated to be 7.89 pg WHO-TEQ/day, lower than that
259 estimated for the general population.

260 It is well known that the primary source of human exposure to PCDD/Fs is the diet
261 (EFSA, 2015). To avoid the wide range of potential adverse health effects caused by
262 exposure to these chemicals, in 2001 the Committee on Toxicity of Chemicals in Food,
263 Consumer Products and the Environment of the United Kingdom proposed a tolerable
264 daily intake (TDI) of 2 pg WHO-TEQ/kg of body weight per day. At the same time, the
265 Scientific Committee on Food (SCF) of the European Commission (EC) set a tolerable
266 weekly intake (TWI) of 14 pg WHO-TEQ/kg of body weight. Moreover, in June 2011,
267 the Joint Expert Committee on Food Additives (JECFA) of the WHO and the UN Food
268 and Agriculture Organisation (FAO) recommended a provisional tolerable monthly
269 intake (PTMI) for PCDD/Fs of 70 pg/kg of body weight. These are the references that
270 are still being used (EFSA, 2015). In the present survey, any of the age groups exceeded
271 these recommendations.

272 While we have observed a considerable decrease between the studies conducted
273 2012 and 2018 in Tarragona County, what happened in other places? In Table 4, we
274 have summarized the results of a number of studies conducted between 2012 and 2018,
275 the years in which we assessed human exposure to PCDD/Fs in Tarragona County. Our
276 current estimated dietary intake is rather similar to that found in France by Sirot et al.
277 (2012), which was 0.192 pg WHO-TEQ/kg/day. However, in other studies also
278 conducted in European countries, data on estimated daily intake was higher than that
279 calculated in the present survey. The highest value was reported in Germany (0.71
280 WHO-TEQ/kg/day), followed by the United Kingdom (0.52 WHO-TEQ/kg/day),
281 Ireland (0.3 WHO-TEQ/kg/day) and Italy (0.28 WHO-TEQ/kg/day) (Bramwell et al.,
282 2017; Diletti et al., 2018; Schwarz et al., 2014; Tlustos et al., 2014). Anyhow, all these
283 intakes are of the same order of magnitude than that estimated for Tarragona County.
284 On the other hand, studies conducted in Asian countries also reported higher –although
285 of the same order- levels than the current one. In Hong Kong, Wong et al. (2013) found
286 a monthly intake of 21.9 WHO-TEQ/kg/month, while in China, Shen et al. (2017) also
287 reported a monthly intake of 22 WHO-TEQ/kg/month, which are equivalent to 0.73
288 WHO-TEQ/kg/day (Shen et al., 2017; Wong et al., 2013). In contrast, a recent study
289 performed in Valencia (Spain) reported a dietary intake of PCDD/Fs of 1.31 WHO-
290 TEQ/kg/day (Quijano et al., 2018), which is almost 10-times higher than our current

291 estimated daily intake of PCDD/Fs. With respect to the differences between the studies,
292 it is important to remark that in all the surveys summarized in Table 4 the
293 methodologies are different, including number of foodstuffs, number of samples, and
294 food groups, among others. It means the comparison between the results of these studies
295 should be done carefully and individually.

296 4. Conclusions

297 In summary, the results of our current study show a continued decrease in the
298 exposure to PCDD/Fs by the general population of Tarragona County since our first
299 survey conducted in 1998 (Domingo et al., 1999), being particularly dramatic the
300 reduction after 20 years (210 pg I-TEQ/day vs 8.54 pg WHO-TEQ/day). Considering
301 the dietary PCDD/F intakes according to various age groups, it can be noticed that all of
302 them are notably lower than the reference/safety values established by international
303 organizations. It means that the dietary exposure to the population of Tarragona County
304 to PCDD/Fs should not be a concern from a human health perspective. In spite of this, it
305 is always important to conduct periodical surveys to ensure population safety to these
306 environmental pollutants.

307
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Table 1. Concentrations of PCDD/Fs in food samples collected in different locations from Tarragona County (Catalonia, Spain) (in ng/kg wet weight)

Congener	Vegetables (n=2)		Tubers (n=2)		Pulses (n=2)		Cereals (n=2)		Fruits (n=2)		Fish (n=8)		Seafood (n=4)	
	2012	2018	2012	2018	2012	2018	2012	2018	2012	2018	2012	2018	2012	2018
2,3,7,8-TCDD	< 0.002	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001	0.025	0.014	0.009	0.010
1,2,3,7,8-PeCDD	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001	0.002	0.001	< 0.001	0.041	0.028	0.015	0.017
1,2,3,4,7,8-HxCDD	< 0.001	0.003	< 0.001	0.003	< 0.001	0.002	< 0.001	0.004	0.001	0.003	0.007	0.010	0.006	0.011
1,2,3,6,7,8-HxCDD	< 0.001	< 0.001	< 0.001	< 0.001	0.002	< 0.001	0.003	0.002	0.002	< 0.001	0.032	0.023	0.018	0.019
1,2,3,7,8,9-HxCDD	< 0.001	< 0.001	< 0.001	< 0.001	0.002	< 0.001	0.003	0.002	0.004	< 0.001	0.010	0.007	0.018	0.014
1,2,3,4,6,7,8-HpCDD	0.009	0.008	0.005	0.003	0.004	0.003	0.015	0.007	0.019	0.003	0.056	0.027	0.080	0.068
OCDD	0.039	0.041	0.022	0.014	0.006	0.014	0.245	0.023	0.128	0.013	0.179	0.076	0.332	0.203
2,3,7,8-TCDF	0.018	0.004	0.002	0.001	0.002	0.001	0.004	0.006	0.002	0.004	0.277	0.177	0.185	0.200
1,2,3,7,8-PeCDF	0.002	0.001	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001	0.002	0.001	< 0.001	0.059	0.044	0.021	0.025
2,3,4,7,8-PeCDF	0.004	< 0.001	0.002	< 0.001	0.002	< 0.001	0.002	0.002	0.003	0.001	0.152	0.110	0.045	0.047
1,2,3,4,7,8-HxCDF	0.004	0.002	< 0.000	0.001	0.001	0.001	0.002	0.003	0.002	0.002	0.057	0.024	0.016	0.021
1,2,3,6,7,8-HxCDF	0.002	0.001	< 0.000	< 0.001	< 0.000	0.001	0.001	0.001	0.001	0.001	0.023	0.013	0.008	0.010
2,3,4,6,7,8-HxCDF	< 0.001	0.001	< 0.000	< 0.001	< 0.001	< 0.001	< 0.000	< 0.001	0.001	0.001	< 0.003	0.014	< 0.003	0.012
1,2,3,7,8,9-HxCDF	0.001	< 0.001	< 0.000	< 0.001	< 0.000	< 0.001	< 0.001	< 0.001	0.002	< 0.001	0.018	< 0.002	0.008	< 0.002
1,2,3,4,6,7,8-HpCDF	0.005	0.002	0.001	0.001	0.001	0.002	0.003	0.003	0.010	0.001	0.035	0.017	0.017	0.021
1,2,3,4,7,8,9-HpCDF	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001	< 0.001	0.001	< 0.001	0.001	< 0.001	0.010	< 0.004	< 0.003	< 0.002
OCDF	0.007	0.003	0.002	< 0.003	< 0.001	0.005	0.003	< 0.004	0.017	< 0.003	0.051	0.017	0.018	0.015
WHO-TEQ	0.007	0.003	0.003	0.003	0.003	0.003	0.004	0.005	0.004	0.003	0.156	0.103	0.065	0.071

Table 1 (continued). Concentrations of PCDD/Fs in food samples collected in different locations from Tarragona County (Catalonia, Spain) (in ng/kg wet weight)

Congener	Meat (n=8)		Eggs (n=2)		Milk (n=4)		Dairy products (n=4)		Industrial bakery (n=2)		Oils and fats ^a (n=3)	
	2012	2018	2012	2018	2012	2018	2012	2018	2012	2018	2012	2018
2,3,7,8-TCDD	0.003	0.002	0.008	0.047	< 0.001	< 0.001	0.005	0.002	0.004	< 0.001	< 0.015	< 0.006
1,2,3,7,8-PeCDD	0.003	0.003	0.005	0.015	0.002	0.002	0.011	0.006	0.011	< 0.001	0.013	0.012
1,2,3,4,7,8-HxCDD	0.002	0.004	0.003	0.007	0.001	0.003	0.006	0.007	0.026	0.004	< 0.010	0.029
1,2,3,6,7,8-HxCDD	0.007	0.009	0.008	0.005	0.004	0.003	0.016	0.010	0.063	< 0.001	0.028	0.020
1,2,3,7,8,9-HxCDD	0.004	0.003	0.006	0.003	0.002	0.002	0.009	0.005	0.043	0.003	0.021	0.014
1,2,3,4,6,7,8-HpCDD	0.048	0.047	0.068	0.016	0.010	0.008	0.056	0.024	1.186	0.024	0.304	0.056
OCDD	0.168	0.213	0.192	0.158	0.011	0.015	0.109	0.057	5.005	0.815	1.019	0.269
2,3,7,8-TCDF	0.011	0.009	0.026	0.022	< 0.001	0.001	0.009	0.006	0.042	0.008	0.113	0.069
1,2,3,7,8-PeCDF	0.003	0.002	0.010	0.009	< 0.001	< 0.001	0.005	0.002	0.018	0.003	0.019	< 0.007
2,3,4,7,8-PeCDF	0.008	0.007	0.013	0.007	0.006	0.004	0.030	0.015	0.041	0.003	0.055	0.028
1,2,3,4,7,8-HxCDF	0.022	0.008	0.042	0.007	0.003	0.002	0.028	0.009	0.506	0.005	0.186	0.022
1,2,3,6,7,8-HxCDF	0.009	0.004	0.016	0.004	0.002	0.002	0.016	0.007	0.181	0.002	0.076	0.014
2,3,4,6,7,8-HxCDF	0.003	0.003	< 0.002	0.004	< 0.001	0.002	< 0.002	0.006	0.155	0.002	0.046	0.013
1,2,3,7,8,9-HxCDF	0.007	< 0.001	0.016	< 0.002	0.002	< 0.001	0.013	< 0.001	0.012	< 0.001	0.070	< 0.005
1,2,3,4,6,7,8-HpCDF	0.078	0.011	0.269	0.005	0.002	0.002	0.065	0.006	3.134	0.006	0.698	0.020
1,2,3,4,7,8,9-HpCDF	0.015	< 0.001	0.029	< 0.002	< 0.001	< 0.001	0.013	< 0.002	0.624	< 0.001	0.121	< 0.006
OCDF	0.143	0.015	0.215	< 0.003	< 0.001	< 0.001	0.164	< 0.002	5.511	0.015	0.831	< 0.024
WHO-TEQ	0.017	0.012	0.032	0.070	0.007	0.005	0.036	0.018	0.183	0.006	0.112	0.046
^a In				ng/kg				fat				weight

Table 2. Estimated daily intake of PCDD/Fs by the general population of Tarragona County

<i>Food group</i>	<i>Consumption rate (g/day)</i>				<i>Dietary intake</i>					<i>Variation (%)</i>	
	<i>1998^a</i>	<i>2002^b, 2006^c</i>	<i>2012^d</i>	<i>2018</i>	<i>pg I-TEQ/day</i>	<i>pg WHO-TEQ/day</i>				<i>1998-2018</i>	<i>2012-2018</i>
					<i>1998</i>	<i>2002</i>	<i>2006</i>	<i>2012</i>	<i>2018</i>		
Vegetables	122	226	226	79	17.1	1.40	0.86	1.67	0.25	-98	-85
Tubers	-	-	74	48	-	-	-	0.25	0.12	-	-52
Pulses	16	24	24	7.9	3.04	0.30	0.36	0.08	0.02	-99	-75
Cereals	194	206	206	99	48.5	9.20	1.42	0.87	0.54	-99	-38
Fish and seafood	72	92	92	30	30.4	21.5	7.91	11.6	2.82	-91	-76
Meat	173	185	185	74	20.8	5.60	2.26	3.18	0.89	-96	-72
Eggs	29	34	34	17	3.48	1.30	4.59	1.11	1.21	-65	9
Milk	178	217	217	141	32.0	4.10	0.62	1.50	0.78	-98	-48
Dairy products	44	106	106	35	1.76	8.80	3.02	3.79	0.63	-64	-83
Fruits	269	239	239	103	24.2	2.00	0.75	1.01	0.32	-99	-68
Oils and fats	45	41	41	19	28.8	9.70	6.01	4.61	0.89	-97	-81
Industrial bakery	-	-	19	12	-	-	-	3.49	0.07	-	-98
Total	1142	1370	1463	666	210	63.8	27.8	33.1	8.54	-96	-74

^aDomingo et al. (1999)^bBocio and Domingo (2005)^cMartí-Cid et al. (2008)^dDomingo et al. (2012)

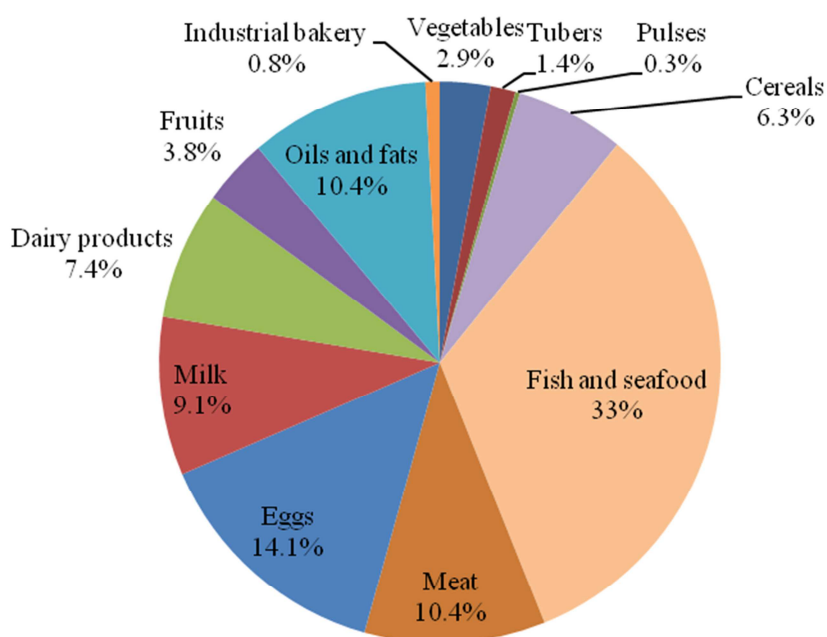


Fig. 1. Percentages of contribution from each food group to the total daily intake (pg WHO-TEQ/day) of PCDD/Fs in 2018 by the population of Tarragona County, (Catalonia, Spain)

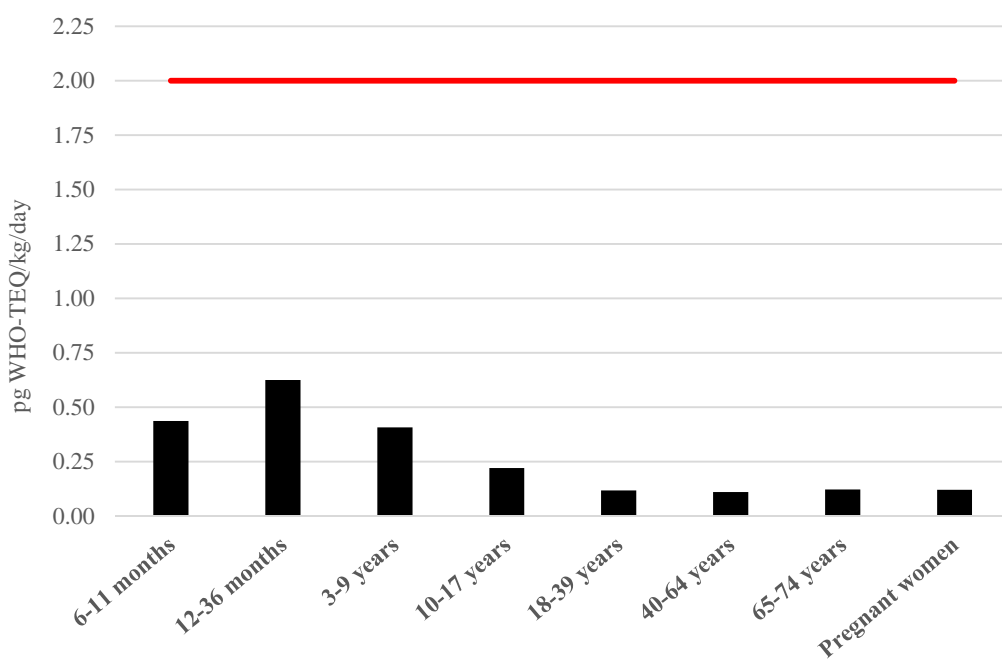


Fig. 2. Estimated daily intake (pg WHO-TEQ/kg body weight/day) of PCDD/Fs by the general population of Tarragona County (Catalonia, Spain) according to different age groups. The red line indicates the TDI.

Table 3. Estimated daily intake of PCDD/Fs by the population of Tarragona County (Catalonia, Spain) according to different age groups.

	<i>6-11 months</i>	<i>12-36 months</i>	<i>3-9 years</i>	<i>10-17 years</i>	<i>18-39 years</i>	<i>40-64 years</i>	<i>65-74 years</i>	<i>Pregnant women</i>
Vegetables	0.05	0.05	0.11	0.18	0.20	0.29	0.25	0.25
Tubers	0.25	0.16	0.15	0.18	0.12	0.12	0.13	0.12
Pulses	0.00	0.02	0.03	0.04	0.02	0.02	0.03	0.02
Cereals	0.02	0.21	0.53	0.77	0.55	0.53	0.48	0.49
Fish and seafood	0.99	2.75	3.08	3.45	2.73	2.78	3.24	2.45
Meat	0.44	0.64	0.98	1.23	1.00	0.86	0.66	0.76
Eggs	0.10	0.89	1.31	1.70	1.24	1.18	1.22	1.03
Milk	0.04	0.85	1.39	1.36	0.81	0.77	0.68	0.85
Dairy products	0.98	1.25	1.09	1.09	0.66	0.63	0.57	0.69
Fruits	0.40	0.32	0.30	0.31	0.27	0.34	0.46	0.23
Oils and fats	0.37	0.47	0.70	0.82	0.82	0.93	0.90	0.92
Industrial bakery	0.03	0.09	0.11	0.11	0.08	0.07	0.05	0.07
pg WHO- TEQ/day	3.67	7.70	9.79	11.2	8.50	8.52	8.66	7.89

Table 4. Dietary intake of PCDD/Fs and foodstuffs assessed: A summary of recent (2012-2018) reports of various countries

Country	Analyzed foods	Major contributor	pg WHO-TEQ/kg/day	Reference
Tarragona, Catalonia	Meat, fish and seafood, eggs, milk and dairy products, oils and fats, cereals, pulses, vegetables, tubers, fruits, industrial bakery	Fish and seafood	0.122 ^a	This study
France	Milk, dairy products, cheese, eggs and egg products, butter, oil, margarines, meat, poultry and game, offal, meat products, fish, crustaceans and molluscs, vegetables, pizzas and savoury pastries, sandwiches and hamburgers, mixed dishes, cream desserts, condiments and sauces	Fish	0.192	Sirot et al. (2012)
Hong Kong	Cereals and their products, meat, poultry, game and their products, eggs and their products, fish and seafood and their products, dairy products, fats and oils, non-alcoholic beverages, mixed dishes, others	Fish and seafood and their products	21.9 ^b	Wong et al. (2013)
Ireland	Dairy products, eggs, fats and oils, fish and fish products, meat and meat products, and oil-based supplements	Fish and fish products	0.3	Tlustos et al. (2014)
Germany	Fish, fruits and nuts, vegetables, beverages, cereals, eggs, oily seeds and fruits, dairy products, and non-allocated foods	Dairy products and fish	0.71	Schwarz et al. (2014)
Malaysia	Seafood and seafood products, meat and meat products, milk and dairy products	Seafood and seafood products	0.983	Leong et al. (2014)
United Kingdom	Bread, cereals, meat, poultry, carcass meat, offal, meat products, fish and seafood, fats and oils, eggs, sugar and preserves, green vegetables, potatoes, other vegetables, canned vegetables, fresh fruit, fruit products, milk and dairy products, nuts	Fish and seafood	0.52	Bramwell et al. (2017)
China	Rice, fresh milk, milk powder, infant formula, marine fatty fish, marine shell fish, marine fish oil, herbivorous freshwater fish, omnivorous freshwater fish, chicken egg, pork liver, pork and beef meat	Fish	22.0 ^b	Shen et al. (2017)
Italy	Eggs, fish and seafood, vegetables, meat, milk, olive oil, sheep liver	Fish and vegetables	0.28	Diletti et al. (2018)
Valencia, Spain	Fish and seafood, milk and dairy products, cereals, pulses, tubers, nuts, oils and fats, non-alcoholic drinks, prepared dishes, fruits, vegetables, sweeteners and condiments, meat and meat products, eggs and alcoholic drinks	Fish and seafood	1.13	Quijano et al. (2018)

^aEstimated weight: 70 kg. ^bIn pg WHO-TEQ/kg/month