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Polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/Fs) in food and human dietary intake: An update of the scientific literature

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ABSTRACT

It is well established that for non-occupationally exposed populations, dietary intake is, by far, the main way of human exposure to polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzo-*p*-furans (PCDD/Fs), a family of environmental POPs with a well-known potential toxicity –including carcinogenicity-in humans. We here summarize the results of recent studies (2010–2021) (databases: Scopus and PubMed), focused on determining the levels of PCDD/Fs in food samples of different origins, as well as the dietary intake of these pollutants. We have revised studies conducted in various Asian, American and European countries. However, information is rather limited, with no recent data for most countries over the world. Due to the enormous differences in the methodologies of the studies, to conduct a detailed comparison of the results for the different regions and countries has not been possible. Notwithstanding, where data over time are available, important reductions have been observed. These reductions have been linked to the decreases in the environmental emissions of PCDD/Fs noted in recent years. Interestingly, reductions in the levels of PCDD/Fs in biological tissues are also occurring in parallel. In general, the tolerable daily/weekly/monthly dietary intakes of PCDD/Fs are not being currently exceeded where data are available.

1. Introduction

Polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) are a group of persistent organic pollutants (POPs) widely distributed in the environment. Although these contaminants can be released through some natural processes, their environmental presence is mainly due to the atmospheric emissions of a number of industrial processes. These include basically waste (hazardous, municipal and medical) incineration, thermal and combustion-related activities, chlorine bleaching of paper pulp, metal smelting, and cement plants, but also traffic, mainly diesel vehicles (Fuster et al., 2001; Kulkarni et al., 2008; Dopico and Gómez, 2015; Domingo et al., 2020). It is well established that PCDD/Fs are highly toxic compounds, which can cause reproductive and developmental adverse effects, neurodevelopmental impairment, damage to the immune system, and can also interfere with hormones acting as endocrine disruptors. In addition, the possibility that PCDD/Fs may cause cancer is especially worrying.

PCDD/Fs were included in the 1998 UN-EC POP protocol. In recent years, significant reductions in the atmospheric levels of PCDD/Fs have been reported. These decreases have been a direct result of technical improvements in facilities such as waste incinerators, cement kiln plants and smelters, the replacement from coal/diesel in domestic heating to natural gas, as well as modifications of motor vehicles and emission controls (Rahman et al., 2014; van Drooge et al., 2021). Notwithstanding, since PCDD/Fs are environmental persistent substances, human exposure to these compounds, today, is still unavoidable. Exposure may occur through inhalation, dermal contact and ingestion of soils and dust. However, for non-occupationally exposed individuals the diet is quantitatively the main route of exposure (González et al., 2018; 2019). It has been shown that the intake of certain foodstuffs of animal origin such as meat and meat products, dairy products, but mainly fish and shellfish, may surpass 90–95% of the total daily exposure to PCDD/Fs (Domingo and Bocio, 2007; Bocio and Domingo, 2005; Perelló et al., 2015).

Taking into account the potential toxicity of PCDD/Fs and their persistent environmental presence, in recent decades, human exposure/biomonitoring to these substances have been the subjects of a number of studies conducted to assess their human health risks. Thus, the dietary intake of PCDD/Fs has been estimated for different countries and regions, while the concentrations of PCDD/Fs have been determined in

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various human tissues, mainly blood (Marquès and Domingo, 2019), but also breast milk and adipose tissue (Domingo et al., 2017; Schuhmacher et al., 2019).

Based on the above, we have here updated the information on the human dietary exposure to PCDD/Fs. For it, the databases Pubmed (https://pubmed.ncbi.nlm.nih.gov/) and Scopus (https://www.scopus.com/) were used. The terms for the search were "dietary intake", "food consumption", "dioxins", "PCDD/Fs" and "PCDD/PCDFs". The period for the search of the scientific literature covered the papers published since January 2010 to August 2021.

We next summarize data on the available studies. They have been classified by continents and by specific countries in each continent. For every country, results of the studies are presented according to the respective dates of publication, from least to most recent. It does not mean that the studies here reviewed were necessarily conducted in the same year of their publications. On the other hand, we want to note that dioxin-like (DL) PCBs -or other polychlorinated compounds-were not the objective of the current update. However, an important number of the reviewed papers report data on dietary intakes, which correspond to the sum of PCDD/Fs and DL-PCBs. Although data on DL-PCBs corresponding to studies where PCDD/Fs and PCBs were jointly assessed are not specifically discussed, we have here presented total data (PCDD/Fs + DL-PCBs), when in the respective papers the authors did not report the results separately.

2. Studies classified according to continents and countries

2.1. Asia

2.1.1. China

Song et al. (2011) determined the levels of PCDD/Fs (and DL-PCBs) in samples of rice, vegetables, hen eggs, chicken, duck and carps from the cities of Luqiao (LQ) and Yuhang (YH) in the Zhejiang Province. Slight differences were observed for PCDD/Fs (TEQ), with this order: duck > carp > chicken > hen eggs > rice > vegetables in LQ, and duck > hen eggs > carp > chicken > vegetables > rice in YH. The range of ∑PCDD/Fs (pg WHO-TEQ/g fresh weight (fw)) was between non-detected (ND) (rice and vegetables) in YH, and 66.86 (chicken) in LQ. The estimated dietary intakes of PCDD/Fs (plus DL-PCBs) for local residents in LQ and YH were 805.17 and 74.31 (pg WHO-TEQ/day), respectively. This considerable difference was directly related with the uncontrolled e-waste recycling operations in LQ. In turn, Zhang et al. (2013) measured the concentrations of PCDD/Fs (also DL-PCBs) in 96 composite food samples purchased in 12 Chinese Provinces, which included aquatic foods, meat and meat products, egg and egg products, milk and dairy products, cereals, bean products, potatoes, and vegetables. For PCDD/Fs, the highest values (pg TEQ/g fw) were detected in samples of aquatic food (0.44) and meat and meat products (0.17), while the lowest values were found in samples of vegetables and cereals (both 0.001). The dietary intake of PCDD/Fs (plus DL-PCBs) was estimated for 12 age/gender subgroups. It ranged from 15.4 to 38.7 pg TEQ/kg body weight (bw) per month for average individuals, and from 68.5 to 226.1 pg TEQ/kg bw/month for high consumers (97.5th percentile). Zhang et al. (2015) determined the levels of PCDD/Fs (and DL-PCBs) in main Chinese foodstuffs according to the Chinese Total Diet Study (TDS) performed in 2011, being the dietary intake also estimated. The food consumption survey was conducted in 19 provinces of the country. Samples included aquatic foods, meat and meat products, egg and egg products, milk and dairy products, cereals, bean products, potatoes, and vegetables. The highest and lowest concentrations of PCDD/Fs were found in aquatic food (average 0.19 pg TEQ/g fw) and cereals and bean products (0.001 and 0.004 pg TEQ/g fw), respectively. The estimated mean dietary exposure to PCDD/Fs for the 19 Provinces was 0.37 pg TEQ/kg bw/month. Although the authors reported reductions with respect to previous Chinese TDS (2000 and 2007), a notable increase of exposure was also observed in certain regions, which was probably due to potential food contamination.

Shen et al. (2017) measured the concentrations of PCDD/Fs (and PCBs) in 620 samples of foods collected in public markets and near polluted zones (a municipal solid waste incinerator (MSWI) and e-waste disassembling areas) of Zhejiang. The dietary intake of these compounds was subsequently estimated. For PCDD/Fs exclusively, the average TEQ percentage for all 13 analyzed food groups was found to be 42% of the European Union maximum limit (EU ML). Some foods of animal origin were close to the corresponding EU MLs (pork, infant formula and beef). The estimated dietary intake for the general population was 22.0 pg TEQ/kg bw/month, which was below the standard of 70 pg TEQ/kg bw/month set by the Joint FAO/WHO Expert Committee on Food Additives (JECFA). As expected, for the MSWI and e-waste disassembling sites, high levels of PCDD/Fs (and also PCBs) were found in all foods, with estimated dietary intakes for local residents of 244 (near the MSWI) and 240 (near the e-waste disassembling sites) pg TEQ/kg bw/month. These values were approximately 3.5-fold higher than the 70 pg TEQ/kg bw/month set by the JECFA. In turn, Wang et al. (2017) determined the concentrations of PCDD/Fs in 634 food samples (beef and mutton, chicken and duck, pork, fish and seafood, and milk and dairy products) collected from Chinese markets between 2011 and 2014. The mean levels of PCDD/Fs in all groups ranged from 0.291 to 8.468 pg/g whole weight, while the mean TEQ concentrations were between 0.012 pg/g fw for cereals, and 0.367 pg/g fat for marine oil. The estimated mean dietary intake for Chinese rural and urban populations were 0.656 and 0.514 pg TEQ/kg bw/day, respectively, indicating that at the time of the study, the health risks from PCDD/Fs through the diet were low. In order to assess human exposure to PCDD/Fs for the population living in the vicinity of a MSWI, Ben et al. (2017) analyzed the levels of PCDD/Fs in 14 species of local vegetables collected from a garden near a MSWI (Shenzhen, South China), as well as in 21 species of commercial vegetables purchased from a market. It was observed that PCDD/F concentrations in the vegetables grown near the MSWI were higher than those detected in commercial vegetables. The potential dietary exposure to PCDD/Fs for the population living in the vicinity of the MSWI was also estimated. Interestingly, it was noticed that if only the local vegetables were consumed -being other foods acquired commercially-the total dietary intake for a general adult was 0.94 ± 0.41 pg TEQ/kg bw/day (with consumption of local vegetables accounting for 52.3%). In contrast, if all foods consumed -including vegetables-were from commercial sources, the total dietary intake was 0.56 ± 0.30 pg TEQ/kg bw/day (with consumption of commercial vegetables accounting for 20.1%). More recently, Wu et al. (2018) determined the levels and congener profiles of PCDD/Fs (also DL-PCBs) in 226 individual samples of beef, freshwater fish and pork from the Guangdong Province. The dietary intake of PCDD/Fs was also estimated. The median total PCDD/Fs + DL-PCBs levels of these three foodstuffs were 0.174, 0.488, and 0.113 pg TEQ/g fw, respectively. The estimated monthly intakes (pg TEQ/kg bw) of PCDD/Fs (plus DL-PCBs) for children (age 6–18 years) were 19.17 and 23.26, for boys and girls, respectively, being for adults (age 18–70 years) 20.57 (males) and 19.52 (females). All these intakes were lower than the provisional tolerable monthly intake (PTMI) of 70 pg TEQ/kg bw/month set by the Joint FAO/WHO Expert Committee. On the other hand, Fan et al. (2021) recently evaluated the dietary risks of POPs listed in the Stockholm Convention, among which, PCDD/Fs were included. It was observed that the concentrations of PCDD/Fs in foodstuffs from China continued to markedly decrease from 1995, with a coefficient of 0.085 ± 0.02 Ln (ng TEQ/kg)/year ($p < 0.001$). The estimated dietary intake of PCDD/Fs was 17.8 ± 9.5 ng/kg/day, which means a continued decline with time in that country.

Other authors have reported data on human exposure in China to PCDD/Fs, but just through some specific foodstuffs. Thus, Han et al. (2018) assessed the health risks of PCDD/Fs -and PCBs-in Chinese mitten crabs from different lakes in Jiangsu Province. The mean PCDD/F concentration was 42 ± 41 pg/g, while the range for the PCDD/F TEQs was 0.74–9.7 pg/g, with a mean of 2.9 pg/g. Sun et al. (2021) measured the levels of ∑PCDD/Fs (also ∑PCBs) in 354 representative plant origin foodstuffs (cereals, beans, potatoes, leafy vegetables, root and stem vegetables, melon vegetables, legume vegetables, edible fungi, and mixed vegetable oil) obtained from 29 sampling sites in 5 regions of the Chinese mainland. The concentrations of ∑PCDDs and ∑PCDFs ranged from 6.1 to 217.2 pg/kg fw to 32.7–1201.8 pg/kg fw, respectively. Recently, Wang et al. (2021) conducted a controlled feeding experiment aimed at investigating the transfer of dioxin-like compounds from feed to eggs. A probabilistic model was developed to estimate the dietary risk of PCDD/Fs from feed to eggs for the Chinese population. The results showed that PCDD/Fs (and also DL-PCBs) were readily deposited in eggs during exposure. However, the estimated dietary intake, as well as the hazard quotients indicated that dietary risks from feed to eggs were found to be limited for the general population of China.

2.1.2. *Japan*

Nakatani et al. (2011) conducted a TDS aimed at determining the dietary intake of PCDD/Fs (and DL-PCBs). Foods were purchased in Osaka City and belonged to 14 food groups. Although the foodstuffs were collected during 2000–2002, we have included here this survey because of the results were published in 2011. The TEQ dietary intakes (for PCDD/Fs + DL-PCBs) for an adult were estimated to be 104.24 pg TEQ/person/day, 72.73 pg TEQ/person/day, and 87.28 pg TEQ/person/day, in 2000, 2001 and 2002, respectively. These values corresponded to 2.08, 1.45 and 1.74 pg TEQ/kg bw/day for an adult of 50 kg. Fish and shellfish, followed by meat and eggs, showed the highest contribution to the total intake of PCDD/Fs (plus DL-PCBs). Recently, Tsutsumi et al. (2018) estimated the dietary intake of PCDD/Fs (and also DL-PCBs) for the general Japanese population (≥1 year old). The average daily intake of PCDD/Fs (plus DL-PCBs) for a person of 50 kg was 0.54 pg TEQ/kg bw/day, a value that was well below the tolerable daily intake of 4 pg TEQ/kg bw/day set for these compounds in Japan. In accordance with the results of the survey conducted by Nakatani et al. (2011), the main contributor was also the group of fish and shellfish, followed by meat and eggs. On the other hand, Muzembo et al. (2019) determined the blood dioxin concentrations and dietary intake of PCDD/Fs (plus coplanar-PCBs) in Japanese from the 2011–2016 SEDOCCH (Survey on the exposure to dioxins and other chemical compounds in humans) data. Ninety samples were used to calculate the dietary intake. The mean dietary total intake was estimated at 0.49 pg TEQ/kg bw/day, while the median PCDDs/Fs and total TEQ intakes (including co-PCBs) were 0.16 and 0.33 pg/kg bw/day, respectively.

2.1.3. *Other Asian countries*

In the first Hong Kong TDS, Wong et al. (2013) estimated the dietary exposure of various age-gender subgroups of population to PCDD/Fs (and DL-PCBs). For it, 142 composite food samples were previously analyzed. The food group with the highest contribution to total exposure to PCDD/Fs (and DL-PCBs) was fish and seafood and their products, followed by meat, poultry, and game and their products. The mean and 95th percentile exposures to PCDD/Fs (plus DL-PCBs) of the Hong Kong population were 21.9 and 59.7 pg TEQ/kg bw/month. It would mean 31.3% and 85.2% of the PTMI. On the other hand, in 2011, PCDD/Fs were not regulated in the Malaysian Food Act and Regulations. At that time, Leong et al. (2014a) collected 126 food samples (seafood and seafood products, meat and meat products, and milk and dairy products), which were analyzed for PCDD/Fs. The concentration of PCDD/Fs ranged from 0.16 to 0.25 pg WHO-TEQ/g fw. The dietary exposure to PCDD/Fs for the general population of Malaysia was estimated to be in the range 0.777–1.097 pg WHO-TEQ/kg bw/day. Subsequently, the same Malaysian research group (Leong et al., 2014b) conducted another study in 2011 and 2012. Seafood samples including fish, prawn, squid, dried anchovy, as well as other processed seafood products were collected. This study also analyzed DL-PCBs. The levels (pg TEQ/g fw) of PCDD/Fs ranged between 0.13 and 1.03, with a mean value of 0.16. The dietary exposure to PCDD/Fs from seafood for the general population of Malaysia was estimated in 0.042 pg TEQ/kg bw/day.

In Taiwan, Chang et al. (2012) determined the background levels of PCDD/Fs (also DL-PCBs) in 1029 samples of foodstuffs. The highest PCDD/F concentration was found in duck eggs (1.956 pg WHO-TEQ/g fat), followed by beef, and egg products, while the lowest levels corresponded to grape seed oil (0.068 pg WHO-TEQ/g fat). According to age groups of the population, the average dietary intakes of PCDD/Fs (plus DL-PCBs) were in the range between 0.31 and 0.33 pg WHO-TEQ/kg bw/day for adults and 0.62–0.70 pg WHO-TEQ/kg bw/day for children (6–12 years old). The highest percentages of contribution to the total dietary intake were due to the consumption of fish and fishery products, especially by adults (men: 51.6%; women: 47.5%). In South Korea, Shin et al. (2016) analyzed the concentrations of PCDD/Fs (and DL-PCBs) in 480 samples of fish belonging to 37 species consumed by the Korean population. In descending order, gizzard shad, sailfin sandfish and Pacific herring showed the highest TEQ values. The average concentration of PCDD/Fs was 0.13 pg TEQ/g fw. The main dietary and the 95th percentile intakes of PCDD/Fs (plus DL-PCBs) were 0.21 and 0.49 pg WHO-TEQ/kg bw/day, which meant 5.27% and 12.26%, respectively, of the Korean TDI. Recently, Sharma et al. (2021) reported the results of a study from India, in which the dietary intake of various POPs (OCPs, PBDEs, PCDD/Fs and PCBs) was determined. The POP concentrations were measured in samples of food items collected in an urban (Delhi) and a rural area (Dehradun) of the country. Foodstuffs from the urban area were more contaminated than those from the rural zone. The highest estimated daily dietary intakes (EDIs) of the POPs analyzed in that study corresponded to children. We do not summarize here the multiple results of that study, as mixed EDIs were calculated for various groups of population, with significant differences in food consumption (vegetarians, for example). Anyhow, it was noticed that dietary exposure of Indians to the analyzed POPs, was -in general terms-comparable/lower than European data.

2.2. *America*

2.2.1. *Canada and USA*

Surprisingly, in our current search we have not found any study conducted/published in recent years, aimed at estimating the dietary intake of PCDD/Fs in Canada or in the USA. Dabeka and Cao (2013) reported information regarding the Canadian total diet study design (1992–1999), whose results regarding PCDD/Fs for those years, are available in the website of Health Health CanadaBureau of Chemical SafetyGovernment of Canada (2021). In USA, the last data on the dietary intake of PCDD/Fs and related compounds do not belong to the period here reviewed (2010–2021) (Schecter et al., 2001; Lorber et al., 2009), and therefore, they were not included in the current review.

2.2.2. *South America*

Pizarro-Aránguiz et al. (2015) conducted the first study in South America on the levels and congener profiles of PCDD/Fs (also DL-PCBs) in 102 samples of cow's milk and butter collected in Chile between 2011 and 2013. The population exposure to these pollutants through consumption of these foodstuffs was also assessed. The lowest and highest mean values for PCDD/Fs in raw cow milk samples were 0.21 and 0.32 pg WHO-TEQ/g fat (0.02–0.10 pg WHO-TEQ/g fat in butter samples). For Chilean adults, the highest estimated dietary intake of total PCDD/Fs (plus DL-PCBs) was 0.16 pg WHO-TEQ/kg bw/day. The same research group measured the levels of PCDD/Fs and DL-PCBs in samples of meat of bovine, pork, ovine, chicken and turkey collected from 10 Chilean regions (San Martin et al., 2016). The highest mean concentrations of PCDD/Fs were found in samples of bovine and chicken: 0.54 and 0.61 pg WHO-TEQ/g fat, respectively. With respect to the dietary intake of total PCDD/Fs (plus DL-PCBs), it corresponded was 0.09 pg WHO-TEQ/kg/bw/day for adults. In Colombia, Pemberthy et al. (2016) reported the results of a study focused on determining the concentrations of PCDD/Fs (also DL-PCBs) in samples of vegetable oils (soybean and olive), animal oil (fish), and shrimp, commercialized and consumed in that country. The concentrations of the sum of PCDD/Fs ranged from 0.24 pg WHO-TEQ/g fat for soybean oil to 1.71 pg WHO-TEQ/g dry weight for shrimp.

2.3. Europe

2.3.1. Spain

In recent years, most of the studies regarding the dietary intake of PCDD/Fs in European countries have been carried out in Spain, and more specifically in our laboratory placed in Reus (Tarragona Province, Catalonia). Some of our studies have been related with the PCDD/F emissions by a hazardous waste incinerator (HWI), whose regular activities started in 1999. During the construction period of the facility (1996–1998), we estimated the daily intake of PCDD/Fs (baseline survey) through the diet by the population living in the area under potential influence of the emissions from the future HWI (Domingo et al., 1999). A total of 35 food samples (beef, pork, chicken, lamb, fish and seafood, tinned fish, milk and dairy products, vegetables, pulses, cereals, fruits, fats and oils, and eggs) were analyzed. The total dietary PCDD/F intake by the population of the area was estimated to be 210.1 pg I-TEQ/day. Simultaneously, the levels of PCDD/Fs were also determined (background study) in blood of the population living in that area (Schuhmacher et al., 1999). In order to establish the temporal trend in the dietary intake of PCDD/Fs, in recent years we periodically performed dietary surveys (Bocio and Domingo, 2005; Martí-Cid et al., 2008), being two of them (Domingo et al., 2012; González et al., 2018) conducted in the period covered in this review. In 2012, we carried out a sampling in the area under evaluation collecting foodstuffs belonging to the same food groups than those of the background study (Domingo et al., 1999). The dietary intake of PCDD/Fs by the general population of the area was 33.1 pg WHO-TEQ/day, being fish and seafood the groups with the highest contribution to the total TEQ (Domingo et al., 2012). This intake was considerably lower than that found in the baseline study (210.1 pg I-TEQ/day), and lower than that estimated in the 2002 survey (59.6 pg I-TEQ/day) (Bocio and Domingo, 2005). However, it was slightly higher than the intake estimated in the 2006 survey, 27.8 pg WHO-TEQ/day (Martí-Cid et al., 2008). The last study of this series was conducted in 2018 (González et al., 2018). The methodology was quite similar to that employed in the previous surveys. The most striking result was the dramatic reduction in the dietary exposure to PCDD/Fs in the area during the last 20 years (210 pg I-TEQ/day vs. 8.54 pg WHO-TEQ/day). Interestingly, this very important decrease in the dietary exposure to PCDD/Fs occurred in parallel with the notable decline in the concentrations (pg I-TEQ/g lipid) of PCDD/Fs in blood samples of the population living in the area, with mean values of 27.0 (baseline study), 15.7 (2002 study), 9.4 (2007 study), 6.18 (2012) and 6.79 (last study, 2018) (Nadal et al., 2019).

In 2000, we initiated in Catalonia a wide surveillance program focused on measuring the levels of a number of chemical contaminants (including PCDD/Fs) in various groups of foodstuffs. The dietary intake of the pollutants was estimated for various gender/age groups of the Catalan population. Food items belonging to the same food groups assessed in the first (2000) survey were collected and analyzed in 2006 (Llobet et al., 2008) and again in 2008 (Perelló et al., 2012). In the 2008 survey, a total of 65 composite samples, belonging to various food groups, were analyzed for PCDD/Fs. The highest dietary intake of these pollutants corresponded to fish and seafood, followed by dairy products, and oils and fats. The total dietary intake for the adult population was 15.7 pg WHO-TEQ/day, or 0.23 pg/kg bw/day (Perelló et al., 2012). In the previous study, these intakes were 25.7 (pg WHO-TEQ/day (0.37 pg/kg bw/day). In 2012, we collected specific samples of fish and shellfish (16 different species) aimed at determining the temporal trend in the dietary exposure of the Catalan population to PCDD/Fs. In our previous studies, as well as in most surveys conducted over the world, fish and seafood has been the food group showing usually the greatest dietary exposure to PCDD/Fs. In that study (Perelló et al., 2015), sardine, red mullet and mackerel showed the highest concentrations, while canned tuna, cuttlefish and squid were the species with the lowest PCDD/F levels. The estimated dietary intake of PCDD/Fs by adults (70 kg bw) was 3.9 pg WHO-TEQ/day. The continued decline in the dietary exposure to PCDD/Fs by the Catalan population agrees well with the important reductions also observed in the concentrations of these compounds in human blood (Marquès and Domingo, 2019).

On the other hand, Marín et al. (2011) analyzed the concentrations of PCDD/Fs (also DL-PCBs) in 150 individual food (vegetables, cereals, fats and oils, eggs, milk and dairy products, fish products, meat and meat products, and fish oil) samples from the Region of Valencia. The average total exposure to PCDD/Fs through the diet was 1.17 pg WHO-TEQ/kg bw/day. Fish products were the main contributors to the dietary intake of these compounds, followed by dairy products and oils. More recently, the same research group estimated again the dietary exposure of the population of Valencia to PCDD/Fs (and also DL-PCBs) by measuring their levels in a total of 7700 food samples collected in 2010–2011. The highest mean concentration of PCDD/Fs (and DL-PCBs) were also found in fish and seafood, followed by oils and fats, and milk and dairy products. The estimated dietary intake, calculated by a probabilistic approach, showed average intake levels of PCDD/Fs + DL-PCBs (upper-bound scenario) of 1.58 and 2.76 pg TEQ/kg bw/day for adults and young subjects, respectively (Quijano et al., 2018).

2.3.2. Italy

De Filippis et al. (2014) estimated the dietary exposure to PCDD/Fs (also DL-PCBs and PBDEs) of the Italian population by means of a duplicate diet study on prepared meals. Servings from each selected diet were pooled to form a composite and analyzed. The authors found the following mean upper bound intakes for cumulative PCDD/Fs plus DL-PCBs: 0.67, 0.63–0.92, and 0.27-0-63 pg WHO-TEQ/kg bw/day, for toddlers, children and adults, respectively. Diletti et al. (2018) carried out an updated estimate of PCDD/Fs (and DL-PCB) dietary intake in Italy, for which, in the period 2013–2016, a total of 2659 samples of foodstuffs were analyzed. The food items showing a highest contribution to total dietary intake of PCDD/Fs (plus DL-PCBs) were fish and seafood, food of vegetable origin, and cheese. The estimated median cumulative intakes (pg WHO-TEQ/kg bw/day) for PCDD/Fs and PCBs were 1.40–1.52 for children, 0.82–0.85 for adolescents, and 0.64–0.61 for adults, respectively. On the other hand, Barone et al. (2014) determined the levels of PCDD/Fs and PCBs in the main seafood groups (fish, cephalopods and crustaceans) randomly collected from supermarkets of Southern Italy. The greatest mean concentrations of PCDD/Fs were observed in crustaceans (38.4 pg/g lipid fw), being higher than those found in cephalopods (15.1 pg/g lipid fw) and fish (21.5 pg/g lipid fw). The estimated dietary intakes were the following: fish, 0.52 TEQ/kg bw/week; cephalopods, 0.01 TEQ/kg bw/week and crustaceans, 0.02 TEQ/kg bw/week), all below the tolerable weekly intake (TWI) proposed by the European Commission. In a subsequent study conducted by the same researchers (Barone et al., 2018), the concentrations of PCDD/Fs (and PCBs) were measured in Mediterranean Bluefin-tuna, specimens that are considered to be one of the most highly valuable fishery resources. The levels of PCDD/Fs (1.9 pg TEQ/g fw) and their sum with DL-PCBs (2.6 pg TEQ/g fw) remained below the limits for human consumption proposed by the European Union. Recently, this same research group (Barone et al., 2019) also determined the levels of PCDD/Fs (and PCBs) in meats of beef, pork, chicken and turkey purchased from Italian supermarkets. The maximum values of PCDD/Fs were detected in white meats, especially in turkey breast (91.1 pg/g lipid weight), while the lowest levels were found in pork sausage samples (20.1 pg/g lipid weight). Regarding the intake of PCDD/Fs (including also DL-PCBs) through consumption of these meats, this was in the range 0.08–4.16 pg WHO-TEQ/kg bw/week, which is within the limit proposed by the EFSA's expert panel (with the exception of pork sausage, that showed a twofold higher value). Recently, Castellani et al.

(2021) measured the concentrations of PCDD/Fs (and PCBs) in free-range hen eggs from Central Italy. The concentrations of ∑PCDD/Fs (plus DL-PCBs) ranged between 0.463 and 8.028 pg TEQ/g fat. The dietary exposure to PCDD/Fs was subsequently estimated. The weekly dietary intake derived from the locally produced eggs was lower than the TWI currently established by the EFSA.

2.3.3. France

Sirot et al. (2012) assessed the dietary exposure to PCDD/Fs (and DL-PCBs) by the general French population. The human health risks were also evaluated according to the international health-based guidance values. For this, a TDS was conducted and 583 food samples were analyzed. As in most papers here reviewed, the highest PCDD/F levels were found in fish and seafood. The mean dietary exposure (95th percentile) to PCDD/Fs (plus DL-PCBs) was estimated to be 0.57 (1.29) pg TEQ/kg bw/day for adults, and 0.89 (2.02) pg TEQ/kg bw/day in children and teenagers. On the other hand, between 2010 and 2016 the same research group carried out another TDS to determine exposure to PCDD/Fs (and PCBs) by non-breastfed children under 3 years of age (Hulin et al., 2020). PCDD/Fs were analyzed only in those food items known or supposed to contain PCDD/Fs, and for which, data were not available in their previous study (Sirot et al., 2012). The highest mean levels of PCDD/Fs were detected in “infant milk-based desserts” (0.4 pg TEQ/g fw). Mean exposures to PCDD/Fs ranged between 0.22 and 0.44 pg WHO-TEQ/kg bw/day (0.40–0.65 at the 90th percentile), depending on the specific age group.

2.3.4. Other European countries

Windal et al. (2010) assessed the dietary exposure to PCDD/Fs (and DL-PCBs) in the Belgian adult population. The concentrations of PCDD/Fs (and DL-PCBs) were determined in 43 composite samples, belonging most of them to three main groups: meat and meat products, fish and fish products, and dairy and dairy products. The mean dietary intake of PCDD/Fs (plus DL-PCBs) by the Belgian adult population was estimated in 2008 to be 0.72 pg TEQ/kg bw/day. At that time, this value was below the TWI of 14 pg TEQ/kg bw/week set by the Scientific Committee on Food of the European Commission. In Sweden, Törnkvist et al. (2011) analyzed the concentrations of various POPs, including PCDD/Fs and PCBs, in food samples belonging to the groups of fish/fish products, meat/meat products, dairy products, eggs and fats/oils, which are the most consumed by the Swedish population. The highest levels of PCDD/Fs and the rest of analyzed POPs were found in the group of fish/fish products. Dietary intake of the selected POPs was subsequently estimated. For PCDD/Fs (plus DL-PCBs), it was 0.7 pg WHO-TEQ/kg bw/day, or 0.7 pg WHO-TEQ/kg bw/day, when using the TEFs from 1998, or those from 2005, respectively. In Finland, Karjalainen et al. (2012) estimated for the first time in that country the intakes of PCDD/Fs (as well as PCBs and PBDEs) from food in children aged 1–6 years. The analyzed foods were the following: meat (pork, beef and poultry), liver (pig and bovine), milk, eggs (free range and battery farm), oils and fats, fish, and other food items. Food intake and contaminant data corresponded to the period 2002–2005. The long-term upper-bound PCDD/F (plus DL-PCBs) intake ranged between 0.1 and 12.8 pg WHO-TEQ/kg bw/day. The TDI of 4.0 pg WHO-TEQ/kg bw/day was exceeded by 2.5–7.5% of the children. In Greece, Costopoulou et al. (2013) assessed –also for the first time-the dietary intake of PCDD/Fs (and DL-PCBs) in infants of two age groups: 0–6 months, when they are exclusively fed by human milk and/or formula milk, and infants of 6–12 months. Taking into account the main goal of the current review, we here present only the results regarding the second age group. Daily intake estimations (PCDD/Fs + DL-PCBS) were separately done for babies receiving human milk (estimated total daily intake 19.76–24.95 TEQ pg/kg bw) and formula milk (estimated total daily intake 1.60–2.24 TEQ pg/kg bw). The authors highlighted that the risks of this exposure should not be overestimated because nursing is restricted to a very limited period of life. In Austria, Rauscher-Gaberning et al. (2013) estimated -also for the first time-the dietary exposure of the population to PCDD/Fs (and DL-PCBs). A national monitoring program (2005–2011) was conducted including 235 food products (meat, poultry, game and offal, fish and fish products, milk and dairy products, eggs, animal fats and vegetable oils). The mean intakes for PCDD/Fs (plus DL-PCBs) were 0.77, 0.75 and 0.61 pg WHO-TEQ/kg bw/day for children, women and men, respectively. In Ireland, Tlustos et al. (2014) estimated an average upper-bound dietary intake of total WHO-TEQ (PCDD/Fs and DL-PCBs) of 0.3 pg/kg bw/day (95th percentile: 1 pg/kg bw day), while for Germany, Schwarz et al. (2014) reported a dietary intake of PCDD/Fs (plus DL-PCBs) of 2.11/1.53 and 3.56/2.85 pg TEQ/kg bw/day (upper/lower bound) for average and high-end consumers, respectively. Exposure to the pollutants through food was estimated using consumption data from the German food consumption survey, as well as information on the food consumption of 545 individuals. In the United Kingdom, Bramwell et al. (2017) conducted during 2011–2012 a study focused on investigating the dietary exposure to PCDD/Fs (and other 4 groups of POPs) for subjects living in the North East of England. Human health risks derived from the estimated dietary exposures were also assessed. Two different methods were used for estimating dietary exposure to PCDD/Fs and other (POPs): a) the 2012 TDS carried out by the UK Food Standards Agency (FSA), and b) a 24-h duplicate diet (DD) study. Fish and seafood showed the highest levels of PCDD/Fs. The average adult dietary exposure to PCDD/Fs (plus DL-PCBs) in the 2012 TDS was 0.52 pg TEQ/kg bw/day. In turn, in the 2011 DD dietary exposure was 0.27 (lower and upper values) pg TEQ/kg bw/day. Exposure estimates using both TDS and DD data for PCDD/Fs (plus DL-PCBs) were within the recommended TDI. In Poland, Rusin et al. (2019) evaluated the dietary exposure to PCDD/Fs (and PCBs) by the residents in a rural area of the Silesia region in the southern of the country, an area with large and industrialized cities. Two exposure scenarios were considered. The first one assumed that the residents purchase food only from local farms, while the second one assumed that residents purchased foodstuffs only from local stores. Higher levels of PCDD/Fs were detected in the food products analyzed (chicken meat, free-range chicken eggs, and cow milk) from the study area, with respect to those from stores (median contents were 840, 50 and 4 times higher in chicken meat, eggs and milk, respectively). This could be expected taking into account the higher concentrations of PCDD/Fs (and PCBs) found in a previous study in the air of the area. The median level of PCDD/Fs (and DL-PCBs) in chicken meat was 88.2 pg WHO-TEQ/g fat, which was 29.4-fold higher than the normative level. In the worst scenario, the hazard quotient was 71.3 for non-cancer risks and 7.5×10^{-3} for cancer risks.

2.4. Data from other countries: Turkey and Kuwait

In Turkey, Kilic et al. (2011) measured the concentrations of PCDD/Fs (and DL-PCBs) in samples of butter, lamb, beef, eggs and edible oil obtained from two rural and industrial cities (Afyon and Kocaeli) of the country. Although fish samples were not analyzed in that study, data from previous Turkish surveys were used to estimate the dietary intake, which was found to be 0.509 pg WHO-TEQ/kg bw/day. In turn, Kilavuz et al. (2013) assessed dietary intake and health risks of PCDD/Fs for an adult population of Kocaeli (Turkey), based on data of levels of local and non-local grown vegetables and a matrix of environmental exposure factors. The mean PCDD/F intakes via consumption of vegetables were 0.652, 0.672, and 0.661 pg WHO-TEQ/kg bw/day for urban, semi-urban, and rural consumers, respectively.

On the other hand, Husain et al. (2014) determined the levels of PCDD/Fs (and DL-PCBs) in 35 animal feed samples, and in 318 samples of animal origin (lamb, beef, chicken, milk and dairy products, eggs and fish) purchased in 2010 in Kuwait. The dietary intakes of PCDD/Fs (plus DL-PCBs) were estimated for the Kuwaiti population according to four age/gender groups. The highest and lowest intakes, 3.48 and 1.20 pg DR CALUX-BEQ/kg/day, corresponded to the groups of children (girls, 6–9

Table 1
Summary of recent (2010–2021) information on the levels of PCDD/Fs in food and human dietary intake.

Country	Food	∑PCDD/Fs	EDI	Target population	Reference
Luqiao and Yuhang (China)	Rice, vegetable, hen eggs, chicken, duck and carps	ND – 66.86 pg WHO-TEQ/g fw	Luqiao: 805.17 pg WHO-TEQ/day Yuhang: 74.31 pg WHO-TEQ/day	General population	Song et al. (2011)
12 Provinces (China)	Aquatic food, meat and meat products, egg and egg products, milk and dairy products, cereals, bean products, potatoes and vegetables	0.001–0.44 pg TEQ/g fw	15.4–38.7 pg TEQ/kg bw/month*	3–6 years, 7–12 years, 13–17 years, 18–44 years, 45–59 years, >60 years (male and female)	Zhang et al. (2013)
19 Provinces (China)	Aquatic food, meat and meat products, egg and egg products, milk and dairy products, cereals, bean products, potatoes and vegetables	0.001–0.19 pg TEQ/g fw	0.37 pg TEQ/kg bw/month	General population	Zhang et al. (2015)
Zhejiang (China)	Rice, fresh milk, milk powder, infant formula, marine fatty fish, marine shellfish, marine fish oil, herbivorous freshwater fish, omnivorous freshwater fish, chicken egg, pork, pork liver and beef	0.02–3.64 pg WHO-TEQ/g ww	22 pg TEQ/kg bw/month	General population	Shen et al. (2017)
China	Beef and mutton, chicken and duck, pork, fish and seafood, milk and dairy products, pig fat, ruminants and poultry fat, vegetable oils and fats, marine oil, cereals, other food products	0.012–0.367 pg WHO-TEQ/g fw	Rural: 0.656 pg WHO-TEQ/kg bw day Urban: 0.514 pg WHO-TEQ/kg bw day	General population	Wang et al. (2017)
Shenzhen (China)	Meat, fish and shrimp, eggs, cereals, fruits, milk, vegetables and local vegetables grown near a MSWI	0.0003–0.20 pg TEQ/g ww	Including local vegetables: 0.94 pg TEQ/kg bw/day Including commercial vegerables: 0.56 pg TEQ/kg bw/day	General population	Ben et al. (2017)
Guangdong (China)	Beef, freshwater fish and pork	0.113–0.488 pg TEQ/g fw*	19.17–23.26 pg TEQ/kg bw/month*	6–18 years and 18–70 years (male and female)	Wu et al. (2018)
China	Mitten crab	2.9 pg TEQ/g	3.4 pg TEQ/kg bw/day*	General population	Han et al. (2018)
China	Cereals, beans, potatoes, leafy vegetables, root and stem vegetables, melon vegetables, legume, vegetables, edible fungi and mixed vegetable oil	∑PCDDs: 6.1–217.2 pg/kg fw ∑PCDFs: 32.7–1201.8 pg/kg fw	UB: 3.39–4.20 pg WHO-TEQ/kg bw/month LB: 1.57–2.13 pg WHO-TEQ/kg bw/month	General population	Sun et al. (2021)
Japan	Rice and rice products, cereals, seeds, potatoes, sugar and confectionery, fats and oils, pulses, fruits, green vegetables, other vegetables, mushrooms, seaweeds, fish and shellfish, meat, eggs, milk and dairy products, prepared foods, drinking water	–	2000: 2.08 pg TEQ/kg bw/day* 2001: 1.45 pg TEQ/kg bw/day* 2002: 1.74 pg TEQ/kg bw/day*	General population	Nakatani et al. (2011)
Japan	Rice and rice products, cereals, seeds, potatoes, sugar and confectionery, fats and oils, pulses, fruits, green vegetables, other vegetables, mushrooms, seaweed, beverages, fish and shellfish, meat and eggs, milk and dairy products, seasonings	–	0.54 pg TEQ/kg bw/day*	General population	Tsutsumi et al. (2018)
Japan	–	–	0.49 pg TEQ/kg bw/day*	General population	Muzembo et al. (2019)
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, beverages, mixed dishes and other food.	0.011–0.440 pg TEQ/g*	21.9 pg TEQ/kg bw/month*	20–29 years, 30–39 years, 40–49 years, 50–59 years, 60–69 years, 70–84 years and 20–84 years (male and female)	Wong et al. (2013)
Malaysia	Seafood and seafood products, meat and meat products, milk and dairy products	0.16–0.25 pg WHO-TEQ/g fw	0.777–1.097 pg WHO-TEQ/kg bw/day	General population	Leong et al. (2014a)
Taiwan	Pork, beef, mutton, livestock and poultry products, chicken, duck, goose, large marine fish, small-medium marine fish, freshwater fish, other seafood, fishery and seafood products, milk and dairy products, fat and oil, eggs, fruit, vegetables and cereals	0.068–1.956 pg WHO-TEQ/g fat	0.31–0.70 pg WHO-TEQ/kg bw/day*	6–12 years, 13–18 years, 19–64 years, >65 years (male and female)	Chang et al. (2012)
South Korea	Fish and seafood	0.13 pg TEQ/g fw	0.21 pg WHO-TEQ/kg bw/day*	General population	Shin et al. (2016)
Chile	Cow milk and butter	0.21–0.32 pg WHO-TEQ/g fat	0.16 pg WHO-TEQ/kg bw/day*	General population	Pizarro-Aránguiz et al. (2015)
Chile	Bovine, pork, ovine, chicken and turkey	0.1–0.54 pg WHO-TEQ/g fat	<0.001–0.358 pg WHO-TEQ/kg bw/day*	Adults and children	San Martin et al. (2016)
Colombia	Vegetable oils, animal oil and shrimp	0.24–1.71 pg WHO-TEQ/g fat	–	–	Pemberthy et al. (2016)
Tarragona (Spain)	Beef, pork, chicken, lamb, fish and seafood, tinned fish, milk and dairy products, vegetables, pulses, cereals, fruits, fats and oils, eggs	0.003–0.183 pg WHO-TEQ/g ww	0.47 pg WHO-TEQ/kg bw/day	General population	Domingo et al. (2012)
				General population	González et al. (2018)

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Table 1 (*continued*)

Country	Food	∑PCDD/Fs	EDI	Target population	Reference
Tarragona (Spain)	Beef, pork, chicken, lamb, fish and seafood, tinned fish, milk and dairy products, vegetables, pulses, cereals, fruits, fats and oils, eggs	0.003–0.103 pg WHO-TEQ/g ww	0.122 pg WHO-TEQ/kg bw/day		
Catalonia (Spain)	Meat and meat products, fish and shellfish, vegetables, tubers, fruits, eggs, milk and dairy products, cereals, pulses, oils and fats and industrial bakery	0.002–0.120 pg WHO-TEQ/g fw	0.23 pg WHO-TEQ/kg bw/day	General population	Perelló et al. (2012)
Catalonia (Spain)	Fish and shellfish	0.016–0.249 pg WHO-TEQ/g fw	0.056 pg WHO-TEQ/kg bw/day	General population	Perelló et al. (2015)
Valencia (Spain)	Vegetables, cereals, fats and oils, eggs, milk and dairy products, fish products, meat and meat products, and fish oil	0.04–1.22 pg WHO-TEQ/g	1.17 pg WHO-TEQ/kg bw/day	General population	Marín et al. (2011)
Italy	Duplicate diet study	0.0002–0.04 pg WHO-TEQ/g ww*	0.27–0.67 pg WHO-TEQ/kg bw/day*	9–12 months, 3–9 years, 18–64 years	De Filippis et al. (2014)
Italy	Eggs, fish and seafood, vegetables, meat, milk, olive oil, sheep liver	0.006–0.481 pg WHO-TEQ/g	0.28–0.62 pg WHO-TEQ/kg bw/day	3–10 years, 10–18 years, 18–65 years	Diletti et al. (2018)
Italy	Fish, cephalopods, crustaceans	15.1–38.4 pg/g lipid fw	0.01–0.52 pg TEQ/kg bw/week	General population	Barone et al. (2014)
Italy	Bluefin-tuna	1.9 pg TEQ/g fw	–	–	Barone et al. (2018)
Italy	Beef, pork, chicken, turkey	20.1–91.1 pg/g lipid	0.08–4.16 pg WHO-TEQ/kg bw/week	General population	Barone et al. (2019)
Italy	Free-range hen eggs	0.463–8.028 pg TEQ/g fat*	–	–	Castellani et al. (2021)
France	Milk and dairy products, cheese, eggs and egg products, butter, oil, margarines, mean, poultry and game, offal, meat products, fish, crustaceans and molluscs, vegetables, pizza and savoury pastries, sandwiches and hamburgers, mixed dishes, cream desserts, condiments and sauces	0.003–0.187 pg TEQ/g fw	0.192–0.312 pg WHO-TEQ/kg bw/day	3–17 years, >18 years	Sirot et al. (2012)
France	Milk-based beverage, cereals-based food, milk-based dessert, growing-up milk, soup, purée, fruit purée, vegetable-based ready-to-eat meal, meat/fish-based ready-to-eat meal, infant formula, follow-on formula	0.062–0.441 pg TEQ/g lipid	0.218–0.444 pg TEQ/kg bw/day	1–4 months, 5–6 months, 7–12 months, 13–36 months	Hulin et al. (2020)
Belgium	Dairy products, meat and meat products, eggs, fish and fishery products, other food	ND-1.28 pg TEQ/g fat	0.61 pg TEQ/kg bw/day*	General population	Windal et al. (2010)
Sweden	Fish and fish products, meat and meat products, dairy products, eggs, fats and oils	0.007–0.391 pg WHO-TEQ/g	0.6 pg TEQ/kg bw/day*	General population	Törnkvist et al. (2011)
Finland	Meat, liver, milk, eggs, oils and fats, fish and other food	–	0.1–12.8 pg TEQ/kg bw/day*	1–6 years (male and female)	Karjalainen et al. (2012)
Greece	Beef, chicken, olive oil, lamb, eggs, formula milk, human milk	0.33–7.83 pg TEQ/g fat	Human milk: 19.76–24.95 pg TEQ/kg bw/day* Formula milk: 1.60–2.24 pg TEQ/kg bw/day*	6–12 months	Costopoulo et al. (2013)
Austria	Meat, poultry, game and offal, fish and fish products, milk and dairy products, eggs, animal fats and vegetable oils	ND-2.13 pg WHO-TEQ/g fat	0.10–0.36 pg WHO-TEQ/kg bw/day	6–15 years, 19–65 years (male and female)	Rauscher-Gaberning et al. (2013)
Ireland	Dairy products, eggs, fats and oils, fish and fish products, meat and meat products, oil-based supplements	0.01–13.07 pg WHO-TEA/g fat*	0.3 pg WHO-TEQ/kg bw/day*	General population	Tlustos et al. (2014)
Germany	Fish, fruits and nuts, vegetables, beverages, cereals, eggs, oily seeds and fruits, dairy products, non-allocated foods	–	Average consumers: 1.53–2.11 pg WHO-TEQ/kg bw/day* High-end consumers: 2.85–3.56 pg WHO-TEQ/kg bw/day*	General population	Schwarz 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	1.0–191 pg WHO-TEQ/kg ww	TDS: 0.52 pg WHO-TEQ/kg bw/day DD: 0.27 pg WHO-TEQ/kg bw/day	General pooulation	Bramwell et al. (2017)
Poland	Meat, eggs, milk	0.09–92.35 pg WHO-TEQ/g fat*	0.03–49.9 pg WHO-TEQ/kg bw/day*	General population	Rusin et al. (2019)
Turkey	Butter, lamb, beef, eggs and edible oil	0.12–6.12 pg WHO-TEQ/g fat*	0.509–0.588 pg WHO-TEQ/kg bw/day*	General population	Kilic et al. (2011)
Turkey	Rooted vegetables, leafy vegetables, leafless vegetables, fruit, cereals	0.028–0.079 pg/g fw	Urban: 0.652 pg WHO-TEQ/kg bw/day Semi-urban: 0.672 pg WHO-TEQ/kg bw/day Rural: 0.661 pg WHO-TEQ/kg bw/day	General population	Kilavuz et al. (2013)
Kuwait		–			Husain et al. (2014)

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Table 1 (*continued*)

Country	Food	∑PCDD/Fs	EDI	Target population	Reference
	Animal feed samples, lamb, beef, chicken, milk and dairy products, eggs and fish		1.20–3.48 pg DR CALUX-BEQ/kg bw/day*	6–9 years, 10–19 years, 20–49 years, >50 years (male and female)	

∑PCDD/Fs: sum of polychlorinated dibenzo-p-dioxins and dibenzofurans; EDI: estimated dietary intake; UB: upperbound; LB: lowerbound; ND: not detected; TDS: total diet study; DD: duplicate diet.
*DL-PCBs also included in the value.

years old) and adult (females, >50 years old), respectively.

3. Discussion and conclusions

The diet is the main source of human exposure to PCDD/Fs. For non-occupationally exposed individuals, or for very specific subjects, who for some unusual reason could be extraordinarily and unusually exposed to PCDD/Fs, the diet is, by far, the main source of human exposure to PCDD/Fs (EFSA, 2015). In recent decades, tolerable dietary intakes of these compounds have been established, but they have being periodically modified based on new information/data. In 1990, the WHO established a TDI of 10 pg/kg bw/day for 2,3,7,8-tetrachlorodibenzo-p--dioxin (TCDD), the most toxic dioxin, based on liver toxicity, reproductive effects and immunotoxicity in experimental animals, and making use of kinetic data in humans and experimental animals (WHO, 1991). That TDI was modified in 1998 to the range of 1–4 pg TEQ/kg bw/day (WHO, 1998). On the other hand, in 2001 the Committee on Toxicity of Chemical in Food, Consumer Products and the Environment of the UK, proposed a TDI of 2 pg WHO-TEQ/kg bw/day, while the Scientific Committee on Food (SCF) of the European Commission set a TWI of 14 pg WHO-TEQ/kg bw/week. In June 2011, the Joint Expert Committee on Food Additives (JECFA) established a PTMI of 70 pg/kg bw/month. This value is currently used as reference (EFSA, 2015).

In recent years, the environmental levels of PCDD/Fs have followed a continuous reduction in most industrialized countries. Logically, because of these important decreases, the dietary intake of PCDD/Fs has also diminished, as it can be noted for those countries where information is available. Table 1 summarizes the most relevant data of the current update. Unfortunately, due to the enormous differences in the methodologies of the respective studies, to conduct a detailed comparison of the results for the different regions and countries is not possible. However, where data over time are available (for example, our area of residence), a tremendous reduction from 210 pg I-TEQ/day in 1998, to 8.54 pg WHO-TEQ/day in 2018, has been observed (González et al., 2018). Interestingly, it has run in parallel with the considerable reductions in the concentrations of PCDD/Fs in blood of the population of the area (Nadal et al., 2019).

In spite of the importance of the potential adverse health risks (including cancer) derived from human exposure to PCDD/Fs, which is mainly due to dietary intake, the current available scientific information on this topic is limited to a few countries. No recent studies on the dietary intake of PCDD/Fs have been found in the scientific literature (PubMed, Scopus) for countries as diverse as USA, Canada, Australia, Brazil, Russia, or African countries, for example.

Based on the continued reductions in the environmental emissions of PCDD/Fs, it is logical to expect less human environmental exposure to PCDD/Fs in the coming years. In relation to this, an important decrease of the environmental concentrations of PCDD/Fs should have a direct repercussion on the levels of these substances in food, and therefore in dietary exposure. In turn, this would point to a very probable further decline in the levels of PCDD/Fs in biological tissues and consequently on potential health adverse effects. In this sense, Arisawa (2018) assessed the relationship between the decreasing trends of exposure to PCDDs/Fs (plus dioxin-like PCBs) in general populations with diabetes, metabolic syndrome, and gout/hyperuricemia, suggesting that a reduction in the blood levels of these pollutants, which is mainly due to a reduction in their dietary intake, should reduce the association with these diseases.

However, PCDD/Fs are also potential carcinogens, and therefore, the carcinogenic risks of the environmental presence of these substances must be very especially taken into account to decisively bet on the elimination of anthropogenic sources of PCDD/Fs. As we recently stated (Marquès and Domingo, 2019), it is essential to fill the existing gaps related with the lack information on human exposure to PCDD/Fs, both environmental and through the diet. In addition, national and international regulatory organisms should promote actions aimed at eliminating any source of anthropogenic emissions of PCDD/Fs, which will end up being incorporated into food, and therefore, accumulating in the population.

CRediT authorship contribution statement

Neus González: Data curation, Writing – original draft, Table preparation. **José L. Domingo:** Conceptualization, Methodology, Supervision, Writing- Reviewing and Editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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