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Intake estimates of dioxins and dioxin-like polychlorobiphenyls in the Italian general population from the 2013-2016 results of official monitoring plans in food

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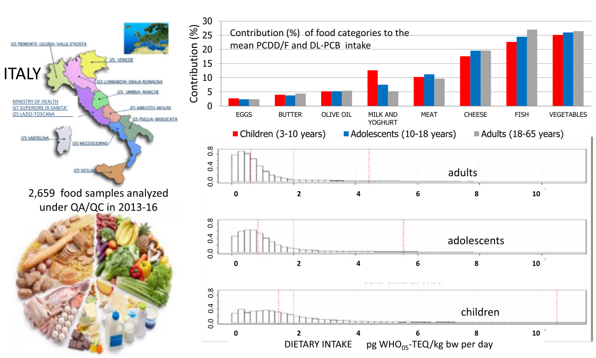
HIGHLIGHTS

- PCDD/Fs + DL-PCBs analyzed in 2659 official food samples in 2013–16 in Italy.
- National food consumption database allowed probabilistic intake estimates.
- A mean intake of 1.98–0.90 pgWHO-TE_{05}/kg bw per day, computed in children/adults.
- Food of vegetable origin contribution to the intake up to 18–25%.
- Decreasing time trends with respect to previous 2006 and 2012 estimates.

GRAPHICAL ABSTRACT



Abbreviations: AAQ, Average Daily Food Consumption; AL, Action Levels; bw, Body weight; CL, Contamination Levels; CPE, Contribution as Percentage to the alimentary Exposure; DL-PCBs, Dioxin-Like PolyChlorinated Biphenyls; EC, European Commission; EFSA, European Food Safety Authority; EU, European Union; HBGV, Health-Based Guidance Value; lb, Lipid basis; LB, Lower Bound; LoQ, Limit of Quantification; MB, Medium Bound; ML, Maximum Levels; MoS, Margin of Safety; NRSP, National Residues Surveillance Plan; PCDD/Fs, PolyChlorinated Dibenzo-Dioxins and -Furans; TDI, Tolerable Daily Intake; TEF, Toxic Equivalency Factors.; TEQ, Toxic EQuivalents; TL, Target Levels; WHO, World Health Organization; UB, Upper Bound.

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ABSTRACT

The implementation of the European Union strategy for polychlorodibenzo-dioxins and -furans (PCDD/Fs), and dioxin-like polychlorobiphenyls (DL-PCBs) is determining a general reduction of their presence in the environment and in the food chain. The most important route for human exposure to these substances is food consumption and, as a consequence, a progressive decrease of their dietary intake has been observed in the last decades. In this context, it seemed worth updating the PCDD/F and DL-PCB intake estimation for the Italian population. A total of 2659 samples of food of animal and vegetable origin analyzed for PCDD/Fs and DL-PCBs in the period 2013–2016 by accredited official laboratories and the national food consumption database were considered for the dietary intake assessment in different age groups of the Italian general population The median cumulative intake estimates expressed as pg WHO-TEQ/kg body weight per day and computed with a deterministic and a probabilistic approach were 1.40–1.52 for children, 0.82–0.85 for adolescents, and 0.64–0.61 for adults, respectively. Such results confirm the decreasing trend of PCDD/F and DL-PCB dietary intake even though the Tolerable Daily Intake (TDI) value of 2 WHO-TEQ/kg body weight per day is exceeded at the 95th percentile for all age groups, with children as sensitive group. Most contributing food categories to the intake resulted fish, food of vegetable origin, and cheese. A sensitivity analysis was also performed to calculate the target contamination levels able to keep the dietary exposure below the TDI. Computed target levels fall between P50 and P97 of the occurrence distribution of the main food groups, meaning that most of the Italian food production can be considered safe.

1. Introduction

Since the Belgium dioxin crisis in 1999, the European Union (EU) adopted a risk management strategy for monitoring the presence of polychlorinated dibenzo-dioxins and -furans (PCDD/Fs) and polychlorinated biphenyls (PCBs) in feed and food, in line with the farm-to-fork approach (Covaci et al., 2008). This strategy is aimed at reducing the levels of PCDD/Fs and PCBs in the environment and in the food chain and at decreasing the exposure of the European population to levels of these chemicals that are below a health-based guidance value (HBGV) related to food intake (EC, 2001). For this purpose, the European Commission has established an HBGV of 2 pg WHO-TEQ/kg body weight per day. This was derived from the results of toxicity studies on laboratory animals in a margin of exposure approach related to the toxic effects on the reproductive and immune systems at doses 10–100 folds lower than those provoking genotoxicity (EC, 2000; Van den Berg et al., 1998, 2006). Owing to this, legislative limits have been set within EU with maximum levels (MLs; EC Regulation no. 1881/2006) and action levels (ALs; EC Recommendation 2014/663) for feed and food. MLs have been established according to the principle "strict but feasible" on the basis of background levels, while ALs represent an early warning of higher than desirable levels of PCDD/Fs and dioxin-like PCBs (DL-PCBs) that require the health authorities to identify the contamination source and to take actions for its reduction or elimination.

In the general context of the significantly decreasing trend of human dietary exposure to PCDD/Fs and DL-PCBs at the EU level, it seemed appropriate to update the intake estimation of the Italian population with respect to previous studies (EFSA, 2012; Fattore et al., 2006).

A set of 2659 results from 2013 to 2016 related to food samples of animal and vegetable origin that were analyzed by a network of Italian official laboratories and national food consumption data were used for this purpose.

Therefore, the aim of this paper is to provide an updated estimate of PCDD/F and DL-PCB dietary intake in Italy in order to be compared with the results obtained from similar studies recently performed in other European countries (Schwarz et al., 2014; Sirot et al., 2012). The information derived from this study can be used for the development of risk-oriented sampling programs that improve cost efficiency.

2. Materials and methods

2.1. Food sampling

Data on contamination levels in foodstuffs were collected from both analytical official controls and research activities performed by the Italian network of official laboratories (Istituti Zooprofilattici Sperimentali) between 2013 and 2016. From this dataset, samples coming from those areas known to be highly contaminated with PCDD/Fs and PCBs (i.e. hot spots) and samples collected under targeted sampling were excluded, resulting in 2659 remaining samples.

In detail, a total of 2116 food samples of animal origin were collected from 2013 to 2015 using the framework provided by the National Residues Surveillance Plan (NRSP). Data from monitoring plans at the regional and national level on fruit, vegetables, and edible marine species that were not investigated by the NRSP were also included: 224 fruit and vegetable samples from the Campania region (2014–2015), 80 olive oil samples from different areas of Italy, and 160 samples of wild fish from a monitoring plan based on environmental contaminants in fishery products from the Mediterranean Sea (2013). Furthermore, 79 samples of mussels from specific surveillance plans performed by local health authorities (2013–2016) were considered. The details of the geographical origin of the samples are reported in the Supplementary Materials (Table 3S and Fig. 2S).

2.2. Analysis

Samples were tested by validated and accredited methods (EN ISO/IEC 17025) routinely used for PCDD/F and PCB analysis in food; these methods have successfully been tested in a number of inter-laboratory studies.

The analytical methodology used followed international standards and was in accordance with the Commission Regulation (EU) N. 589/2014 which replaced the Commission Regulation (EU) N. 252/2012. All analyses were carried out using ^{13}C–labelled internal standards and measurement was made using Gas Chromatography - High Resolution Mass Spectrometry (HRGC-HRMS). Generally, the limit of quantifications (LoQs) for examined food groups were less than one fifth of the corresponding action levels. In particular, LoQ values for PCDD/Fs + DL-PCBs were in the range 0.005–0.04 pg WHO_{05}-TEQ/g for vegetables, 0.01–0.5 pg WHO_{05}-TEQ/g for fish, and 0.06–0.4 pg WHO_{05}-TEQ/g fat for other food. The concentration of contaminants in food items was mostly referred to as unprocessed food. According to the European legislation, analytical results were reported as "upper bound" levels (not detects posed equal to the LoQ) using the toxic equivalency factors proposed by the World Health Organization in 2005 (Van den Berg et al., 2006).

2.3. Consumption dataset

Consumption data were taken from the European Food Safety Authority Comprehensive European Food Consumption (EFSA, 2015) and was based on information of the dietary habits in the total population and consumers in the 2005–2006 Italian national food consumption survey INRAN-*SCAI* (Leclercq et al., 2009).

The population groups were classified according to the age category as follows: a) children 3–10 years, average body weight 26.1 ± 8.3 kg as S.D.; b) adolescents 10–18 years (average body weight 52.6 ± 12.5 kg); c) adults 18–65 years (average body weight 69.7 ± 13.5 kg).

With regards to foods of animal origin, literature data on fishery products reported a wide range of contamination levels (EFSA, 2012) depending on the species and the origin of fish (i.e. wild or farmed fish). Therefore, a more detailed consumption dataset that included different fish species was used. In cases where contamination data were not available, literature data on species of the same trophic level in the Mediterranean Sea were used (Karl et al., 2016; Miniero et al., 2014; Perelló et al., 2015).

Detailed consumption data were also considered for other food items. Cheeses, for example, are generally characterized by lipid contents above 20% and may significantly contribute to the daily intake of lipophilic contaminants despite eating small amounts.

For eggs, literature data reported that eggs coming from battery rearing were significantly less contaminated by PCDD/Fs and DL-PCBs than those from other farming systems (free range, organic production, and outdoor growing production). In Italy, non-battery farming accounts for a small scale economy and cannot provide a realistic picture of human exposure through diet. As a consequence, only eggs from battery hens were included in the dataset.

Regarding the food of vegetable origin category, samples were divided into five groups: a) fruit and fruit products (apricot, orange, cherry, strawberry, lemon, tangerine, apple, pear, peach, plum, and grapes); b) grains and grain based products (maize and wheat); c) legumes, nuts, and oilseed (bean, peas, hazelnut, walnut, and olive); d) starchy roots and tubers (potato); and e) vegetables and vegetable products (salad, aubergines, peppers, tomato, parsnip, radish, and zucchini).

In conclusion, food items were divided into 7 food categories and 42 food groups. The considered food groups encompassed >90% consumption of each related food category.

2.4. Methodology to calculate exposure through daily diet

Consumption data were referred to as processed or unprocessed foods and the values were expressed on a fresh weight basis, while occurrence data for PCDD/Fs and PCBs in foodstuffs were referred to unprocessed foods and the values reported on the basis of fat weight, except for food of vegetable origin and fish products (expressed on fresh basis).

To calculate human exposure through diet, contamination levels in food were transformed from the lipid to product basis and from not processed to processed food (e.g. from milk to cheese and from pork meat to preserved meat). With respect to unprocessed food (milk, meat, and eggs), the lipid percentage of each sample was used to calculate contamination levels at the product level. For processed food (cheese and preserved meat), the different lipid contents of each food item were taken into account to calculate contamination data, making the assumption that a quantitative transfer of contaminants from unprocessed to processed food occurred. Data on food composition were taken from the National Institute for Food and Nutrition (dataset CREA http://nut.entecra.it/646/tabelle_di_composizione_degli_alimenti.html) or from product specifications.

2.4.1. Deterministic approach

In order to calculate daily intake, the following parameters were taken into consideration: mean value, median, 75th and 95th percentiles (P75 and P95) of the contamination level; average consumption per food category and age group, and mean weights of the different age groups (children, adolescents, and adults).

2.4.2. Probabilistic approach

A probabilistic model, taking into account the uncertainty related to different sample sizes in consumption and contamination surveys, was developed to estimate the daily PCDD/F and DL-PCB intake for each age group.

Consumption (C) for each food group (j) and age group (i) was extracted from truncated Normal distributions (with mean and standard deviation derived from consumption surveys). Contamination levels (CL) in each food group were assumed to follow a log-normal distribution (for a few fish species the distributions were not available and the mean values, derived from literature, were considered constant in the model).

The probability (p_{ij}) of an age group consuming a food group was estimated through a Beta distribution (s + 1, n-s + 1), where n equals the number of interviewed people in the age group, s = the number of people in the age group consuming the food category. The Bernoulli (p_{ij}) distribution was used to establish if a food group is consumed or not by an age group.

The model was built on the assumption that there is independent consumption among different food categories, and the daily intake for each age group was calculated as the sum of the product of consumption and the contamination level of each food group.

To calculate the daily intake per kg of body weight, the total intake was divided by the weight of each age group, extracted from a truncated Normal distribution. The model was run in an R environment (R Core Team, 2017; Trautmann et al., 2014), using 50,000 iterations. Fig. 1S (supplementary material) shows the model structure for each food group *j* and age group *i*.

2.4.3. Sensitivity analysis and identification of target levels

The sensitivity analysis was performed to estimate the tolerable contamination (target level) able to maintain a dietary exposure below the tolerable daily intake (TDI) of 2 pg WHO-TEQ/kg bw per day guidance value set out in the framework of the European strategy for reduction of human exposure to PCDD/Fs and PCBs (EC, 2001). Target levels were calculated by applying the following formula: TDI x CPE/AAQ, where: TDI equals 2 pg WHO-TEQ/kg bw per day, CPE equals the percentage contribution of the food item to the dietary exposure, AAQ equals the mean daily consumption of food (g/kg bw per day) in the considered population group (children, adolescents, or adults), referred to as the total population and consumers.

The following main food categories for which a regulatory limit (or action level in the case of vegetables) has been set, have been considered: food of vegetable origin, fish, milk, cheese, and eggs. For those food categories, for which maximum limits were set on the basis of fat, a standard lipid percentage was considered (i.e. 3.8% for milk, 30% for cheese, and 10% for eggs) for the appropriate conversion of values on fresh basis.

Taking the group population "children" and the food category "eggs" as an example, a daily consumption of 0.76 g/kg bw per day, the average daily food consumption (AAQ) was deducted from the Italian national food consumption survey INRAN-*SCAI* and a contribution to dietary intake of 2.7% (CPE) was calculated. The corresponding target level is equal to $2 \times 0.027/0.76 = 0.071$ pg WHO-TEQ/g on a product basis and 0.71 pg WHO-TEQ/g on a lipid basis (with a lipid percentage in eggs equal to 10%).

The calculated target values were interpolated with the contamination distributions provided by the probabilistic approach, in order to gain information regarding which food categories are more relevant to human exposure, taking into consideration the dietary habits of the total population and not the high consumers (Frey and Patil, 2002).

3. Results and discussion

3.1. WHO-TEQ contamination levels

Table 1 reports the statistical descriptors of PCDD/F and DL-PCB occurrence as pg WHO_{05}-TEQ/g in the different food groups. In accordance with the EU legislation on food, contamination was reported on a lipid basis for all of the food commodities assessed, with the exception of fish, liver, and foods of vegetable origin, where results are expressed on a fresh basis. All the values have been expressed as upper-bound concentrations. To allow a direct comparison with inventoried data from other countries, those samples of animal origin with a fat percentage < 2% ($n = 188$) have been excluded from the inventory. For such low-fat content samples, EU legislation requests that results are reported on a fresh basis, thus, hampering a sound scientific literature comparison within the same food category for samples containing a fat percentage above such thresholds.

With respect to the opinion of EFSA (EFSA, 2012), the descriptors shown in Table 1 are generally lower. Among the congeners, PCDD/F data show the most marked decrease, while DL-PCB values remain almost aligned with those reported in the EFSA assessment. The descriptors confirm the general decreasing trend of PCDD/F and DL-PCB contamination in food commodities since 2000, as already reported in other papers (Malisch and Kotz, 2014; Perelló et al. 2015; Sirot et al., 2012).

Among the different food categories, the lower mean contamination observed in Italian eggs than was reported in the EFSA inventory (0.75 vs. 1.62 pg WHO_{05}-TEQ/g fat (EFSA, 2012) may be reasonably ascribed to the focus of the Italian sampling scheme being on conventional poultry farms rather than on rural ones, the latter mostly based on free-range flock rearing systems. The lower contamination associated with conventional eggs is well-consolidated in scientific literature (Lambiase et al., 2017; Holt et al., 2011; Hoogenboom et al., 2016; Van Overmeire et al., 2009) and explained by PCDD/F and DL-PCB intake from the soil and worms in rural flocks in addition to the contribution from feedstuffs that are a unique source of exposure in intensively reared hens. This point is reprised while considering uncertainties in the estimates.

A similar mismatch in contamination levels was found for wild vs. farmed fish species, where the mean values in farmed species, such as trout, sea bream, and sea bass are generally lower than those found in other wild fish species (EFSA, 2012; Miniero et al., 2014; Costopoulou et al., 2016). This relies again on improving the quality of aquaculture feeds with the use of less-contaminated fish meals and fish oils and the partial replacement of these animal feed materials with plant proteins and vegetable oils that are progressively reducing the PCDD/F and DL-PCB burden in farmed species. Among wild fish of commercial interest, swordfish, pilchards, and anchovies show the highest mean values (1.844, 1.767, and 1.186 pg WHO_{05}-TEQ/g, respectively). In contrast, clams are less contaminated on average (0.216 pg WHO_{05}-TEQ/g) in relation to their position in the trophic chain.

Among food from terrestrial animals, the bovine and ovine meat had higher contamination than pork meat (0.790, 0.734, and 0.325 pg WHO_{05}-TEQ/g fat, respectively). The short economic life of fattening pigs (6 months on average) and their fat mass in fattening pigs may mitigate bioaccumulation and dilute the contamination of the lipophilic contaminants, as already observed by Malisch et al. (1999).

Contamination in poultry meat from chicken and turkey averages at 0.5 pg WHO_{05}-TEQ/g fat, while this was higher for game meat, represented almost exclusively by wild boars, at 0.852 pg WHO_{05}-TEQ/g fat. However, the latter value may be affected by the limited sample size. ($n = 9$).

In dairy products, the cumulative contamination expressed on a lipid basis is generally higher than that recorded in meat products; mean contamination ranges from the 1.532 pg WHO_{05}-TEQ/g found in goat milk to the 0.538 pg WHO_{05}-TEQ/g fat in buffalo milk.

In fruit and vegetables, contamination values of 0.021 pg WHO_{05}-TEQ/g expressed on a fresh basis are close to the best performance limits of the analytical techniques currently used, accounting for the expression of the results in the upper-bound mode. In this context, the

Table 1
Statistical descriptors of PCDD/Fs, DL-PCBs, and cumulative occurrence in the different samples analyzed ranked according to food category and group: the pg WHO_{05}-TEQ values are expressed as upperbound on g lipid basis, with the exception of those referred to fish and seafood, liver, and products of vegetable origin, expressed on g fresh weight.

Food category	Food group	N samples	PCDD/Fs					DL-PCBs					PCDD/Fs + DL-PCBs				
			Mean	P25	P50	P75	P95	Mean	P25	P50	P75	P95	Mean	P25	P50	P75	P95
Eggs	Eggs	389	0.282	0.124	0.190	0.260	0.876	0.465	0.119	0.150	0.312	1.142	0.747	0.265	0.360	0.570	2.072
Fish	Anchovy	49	0.127	0.030	0.074	0.136	0.438	1.059	0.173	0.740	1.460	3.078	1.186	0.254	0.779	1.570	3.206
	Sea Bass	32	0.039	0.020	0.036	0.048	0.118	0.446	0.141	0.218	0.464	1.396	0.485	0.162	0.250	0.496	1.451
	Sea Bream	24	0.062	0.022	0.038	0.056	0.227	0.285	0.125	0.263	0.318	0.650	0.347	0.151	0.306	0.394	0.823
	Clam	37	0.046	0.027	0.036	0.049	0.066	0.171	0.036	0.103	0.115	0.221	0.216	0.071	0.135	0.153	0.436
	Cod	39	0.065	0.011	0.026	0.054	0.340	0.495	0.061	0.314	0.720	1.144	0.561	0.109	0.540	0.857	1.254
	Mussel	42	0.125	0.077	0.102	0.148	0.257	0.287	0.166	0.210	0.363	0.591	0.411	0.269	0.323	0.499	0.823
	Other seafood	22	0.106	0.034	0.087	0.146	0.255	0.913	0.257	0.598	1.380	1.944	1.019	0.332	0.658	1.528	2.189
	Sardine and pilchard	37	0.254	0.150	0.266	0.360	0.410	1.513	0.900	1.560	2.150	2.750	1.767	1.220	1.800	2.439	3.017
	Swordfish	13	0.278	0.034	0.160	0.255	0.962	1.565	0.460	1.190	2.112	3.790	1.844	0.495	1.450	2.340	4.700
	Trout	86	0.070	0.018	0.036	0.060	0.350	0.149	0.067	0.135	0.199	0.354	0.219	0.105	0.165	0.277	0.553
All fish		381	0.104	0.027	0.050	0.117	0.370	0.577	0.106	0.217	0.660	2.180	0.682	0.141	0.307	0.773	2.440
Vegetables	Fruit and fruit products	64	0.008	0.003	0.005	0.008	0.025	0.011	0.003	0.006	0.011	0.034	0.020	0.008	0.011	0.019	0.052
	Grains and grain based products	14	0.014	0.004	0.006	0.011	0.047	0.017	0.005	0.007	0.018	0.061	0.032	0.009	0.013	0.050	0.084
	Legumes. nuts and oilseed	33	0.017	0.007	0.010	0.022	0.052	0.021	0.004	0.007	0.025	0.074	0.038	0.013	0.025	0.050	0.110
	Starchy roots and tubers	22	0.006	0.004	0.005	0.007	0.014	0.008	0.003	0.006	0.011	0.020	0.014	0.007	0.011	0.016	0.032
	Vegetables and vegetable products	91	0.007	0.004	0.005	0.007	0.021	0.008	0.002	0.004	0.008	0.029	0.015	0.007	0.010	0.015	0.058
All vegetables		224	0.009	0.004	0.005	0.008	0.037	0.012	0.003	0.005	0.011	0.054	0.021	0.007	0.011	0.022	0.080
Meat	Beef and veal meat	262	0.197	0.090	0.180	0.240	0.410	0.593	0.330	0.460	0.660	1.522	0.790	0.470	0.640	0.928	1.848
	Boar meat	9	0.313	0.200	0.210	0.240	0.762	0.539	0.180	0.270	0.330	1.878	0.852	0.380	0.470	0.660	2.420
	Pig meat	203	0.172	0.170	0.170	0.180	0.259	0.152	0.140	0.140	0.150	0.250	0.325	0.310	0.320	0.330	0.537
	Poultry meat	198	0.215	0.092	0.170	0.210	0.536	0.300	0.140	0.178	0.301	0.715	0.514	0.290	0.371	0.529	1.129
	Sheep meat	34	0.217	0.091	0.155	0.273	0.567	0.517	0.192	0.371	0.648	1.551	0.734	0.301	0.534	0.948	2.060
	Turkey meat	94	0.211	0.154	0.180	0.230	0.497	0.322	0.150	0.280	0.428	0.674	0.533	0.330	0.500	0.664	0.911
All Meat		800	0.199	0.108	0.170	0.210	0.470	0.373	0.140	0.239	0.450	1.051	0.572	0.310	0.410	0.660	1.343
Milk	Buffalo milk	111	0.194	0.100	0.140	0.250	0.420	0.344	0.180	0.260	0.390	0.665	0.538	0.305	0.450	0.640	1.106
	Cow milk	303	0.271	0.180	0.225	0.293	0.630	0.771	0.370	0.530	0.828	2.113	1.042	0.578	0.770	1.125	2.657
	Goat milk	23	0.258	0.130	0.180	0.290	0.586	1.274	0.350	0.430	0.800	3.353	1.532	0.510	0.640	1.055	3.897
	Sheep milk	142	0.245	0.146	0.190	0.250	0.620	0.681	0.160	0.231	0.487	2.020	0.926	0.331	0.455	0.739	2.563
All Milk		579	0.249	0.149	0.210	0.280	0.618	0.687	0.230	0.420	0.680	1.976	0.936	0.410	0.640	0.950	2.433
Olive oil	Olive oil	80	0.047	0.030	0.040	0.050	0.111	0.053	0.024	0.040	0.063	0.130	0.100	0.060	0.081	0.120	0.247
Sheep liver	Sheep liver	18	0.481	0.145	0.465	0.640	1.167	0.402	0.135	0.335	0.528	1.040	0.883	0.275	0.845	1.240	2.184

Table 2
Cumulative and detailed PCDD/F, DL-PCB intake estimates based on the deterministic approach in the different groups of the Italian general population. Mean from the national food consumption database and mean, P50, P75, and P95 contamination. Values expressed as pg WHO_{05}-TEQ/kg bw per day.

Population groups	Mean	P50	P75	P95
PCDD/Fs + DL-PCBs				
Children	1.98	1.40	2.33	4.98
Adolescents	1.16	0.82	1.36	2.90
Adults	0.90	0.64	1.05	2.24
PCDD/Fs				
Children	0.62	0.44	0.62	1.66
Adolescents	0.37	0.26	0.37	0.98
Adults	0.28	0.20	0.28	0.75
DL-PCBs				
Children	1.36	0.96	1.71	3.32
Adolescents	0.79	0.56	0.99	1.92
Adults	0.62	0.44	0.77	1.49

improvement of analytical performance in the past years does not allow a direct comparison with the data inventoried by EFSA (0.05 pg WHO_{05}-TEQ/g), possibly affected by higher LoQs.

Among congeners, DL-PCB contribution is generally greater than that from PCDD/Fs in the food of animal origin, where mean values for the DL-PCB:PCDD/F ratio spans from 1.6 in eggs to 5.5 in fish, while in the food of vegetable origin the ratio is close to 1.

In contrast, in sheep liver, the DL-PCB:PCDD/F ratio is 0.8. This finding is consistent with previous studies and could be explained by the preferential binding of PCDD/F congeners to this tissue (Bruns-Weller et al., 2010; Fernandes et al., 2010; Fernandes et al., 2011; Mortimer, 2012).

There is consolidating evidence that PCBs in Europe may represent the main risk to be managed in food (EFSA, 2012; Lorenzi et al., 2016; Schwarz et al., 2014; Sirot et al., 2012; Windal et al., 2010). In contrast, one study carried out in Spain (Marin et al., 2011) reports the prevalence of the PCDD/F contribution to the cumulative WHO-TEQ in all food categories, excluding fish. PCB mixtures such as Arochlor were produced in Europe until the 1980s and PCB-containing products, such as paints, insulators, sealing materials, and old transformers are still present in rural farms (Bogdal et al., 2017). The input from such PCB-containing products to agricultural soils through the use of topsoil improvers recovered from the sludges of civil wastewater treatment plants should also not be overlooked (Brambilla et al., 2016; Zennegg et al., 2013) as a possible source of contamination in the food chain for grazing- and forage-fed animals.

3.2. Alimentary intake assessment

The deterministic estimates of alimentary intake are reported in Table 2. They have been computed based on mean consumption data from the Italian Food Consumption Database and mean, P50, P75, and P95 occurrence levels of food items, detailed by congener groups and the considered age classes. Fig. 1 shows the mean contribution to the total WHO_{05}-TEQ intake for the main food categories, while Table 3 lists the average contribution of each of the 42 food items considered in the present study. The overall probabilistic intake estimates for the three selected age groups is shown in Fig. 2, highlighting the mean, P50, P75, and P95 values. The sensitivity analysis was carried out on the most vulnerable group —children— and the identified target levels were compared with the EU regulatory limits (Table 4). Target levels equated to the maximum tolerable contamination of the food commodity tailored to the related consumption levels, bringing the exposure equal to the toxicological guidance value of 2 pg WHO-TEQ/kg bw per day.

3.3. Uncertainties

Before the discussion of the results, the following sources of uncertainties that can affect the intake assessment should be acknowledged.

3.3.1. Sampling

Official residue monitoring plans are mostly focused on foods of animal origin due to their economic relevance. Sampling schemes are not targeted on low contaminated but highly consumed items, such as fruit, legumes, cereals, and vegetables, even if they could contribute to the overall WHO_{05}-TEQ intake as much as foods that are more contaminated but consumed less. The lack of a regular monitoring plan for PCDD/Fs and DL-PCBs in food of vegetable origin has been overcome

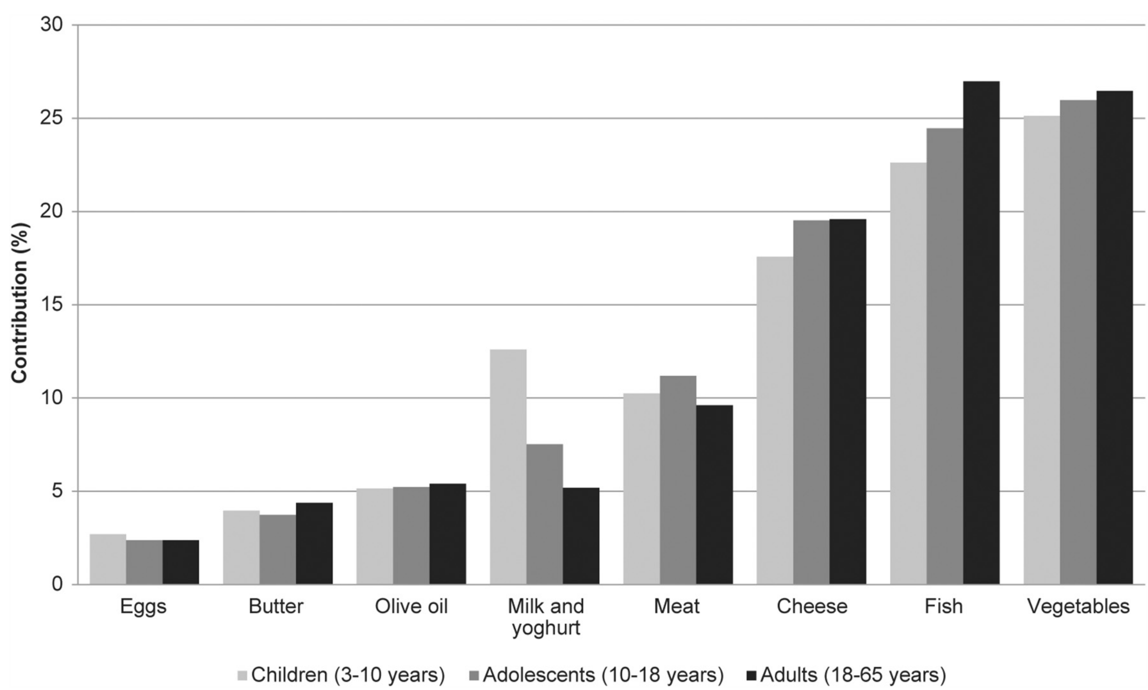

Fig. 1. Percentage contribution of the food categories to daily intake using the mean concentrations of PCDD/Fs and DL-PCBs stratified for three age groups.

Table 3
Relative mean contribution (%) to the cumulative intake from the contamination recorded in the different food groups.

Food category	Food group	Children (3–10 y)	Adolescents (10–18 y)	Adults (18–65 y)
Butter	Butter	4.0	3.7	4.4
Meat	Beef and veal not preserved	4.8	5.3	4.4
	Chicken meat (*Gallus domesticus*)	1.1	0.8	0.7
	Ham. salami. sausages	3.0	3.7	3.1
	Pork not preserved	1.2	1.2	1.1
	Turkey meat (*Meleagris gallopavo*)	0.2	0.2	0.2
Cheese	Cheese	1.7	1.8	2.0
	Cheese. Emmental	0.4	0.6	0.8
	Cheese. Grana Padano	0.6	0.3	0.5
	Cheese. Mozzarella	9.3	10.4	9.9
	Cheese. Parmigiano Reggiano	3.4	3.4	3.2
	Cheese. Pecorino Romano	0.5	0.6	0.9
	Cheese. processed spreadable	1.5	1.8	1.5
	Cheese. Ricotta	0.3	0.7	0.7
Milk and yoghurt	Cow milk. <1% fat (skimmed milk)	0.0	0.0	0.0
	Cow milk. 1–2.9% fat (semi-skimmed milk)	1.7	1.5	1.1
	Cow milk. 3–4% fat (whole milk)	10.0	5.3	3.3
	Yoghurt. cow milk. >3% fat	0.1	0.2	0.2
	Yoghurt. cow milk. with fruit. >3% fat	0.7	0.5	0.5
Olive oil	Olive oil	5.2	5.2	5.4
Fish	Anchovy (*Engraulis*)	1.6	1.2	4.1
	Sea Bass (*Sparus*)	1.8	2.3	1.8
	Sea Bream (*Dicentrarchus*)	1.5	1.6	1.7
	Clam (*Mya arenaria*)	0.3	0.5	0.4
	Cod and whiting (*Gadus spp.*)	7.8	6.5	6.4
	Cuttlefish (*Sepia officinalis*)	0.2	0.2	0.3
	Fish fingers	0.6	0.2	0.1
	Fish meat	2.3	4.7	5.3
	Mussel (*Mytilus edulis*)	0.9	1.8	0.6
	Octopus (*Octopus vulgaris*)	0.3	0.2	0.2
	Salmon and trout (*Salmo spp.*)	0.3	0.4	0.8
	Sardine and pilchard (*Sardina*)	1.7	0.9	1.9
	Shrimp (*Crangon crangon*)	0.7	1.1	1.0
	Sole (*Limanda*; *Solea*)	1.4	1.0	1.2
	Squid (*Loligo vulgaris*)	0.9	1.4	0.9
	Tuna (*Thunnus*)	0.3	0.4	0.4
Eggs	Eggs and egg products	2.7	2.4	2.4
Vegetables	Fruit and fruit products	5.0	4.6	6.1
	Grains and grain-based products	14.0	14.5	12.9
	Legumes. nuts and oilseeds	0.8	0.7	0.8
	Starchy roots and tubers	1.2	1.3	1.1
	Vegetables and vegetable products	4.1	4.7	5.6

regional level investigations in the Campania region, one of the most relevant agricultural districts in Italy for vegetables. In this context, no relevant uncertainties can be ascribed to such food items representativeness in the occurrence database.

Official sampling is mainly targeted at the farm level, therefore processed products are not primarily considered. Therefore, the assessment of contamination for processed products has been derived from that of raw food by applying the appropriate transfer factors. For lipophilic contaminants, such factors can be reasonably shaped based on differences in the lipid content between the raw and the processed food commodity. In the cheese making process, however, it could be possible to have PCDD/F and DL-PCB enrichment in cheese resulting from the hydrophobic clotting of caseins of milk during ripening (De Filippis et al., 2013). Such enrichment usually does not surpass 10% of the original contamination present in the lipids of unprocessed milk and can be reasonably included as a systematic error in the analytical procedure.

In fish, the criteria for the analysis of PCDD/Fs and DL-PCBs indicate that the test sample is the edible part composed of the meat and the subcutaneous fat removed from the skin. Subcutaneous fat is not consumed in all types of fish, therefore it is reasonable to consider uncertainties directed towards a slight overestimation of the real WHO_{05}-TEQ intake.

As already considered from the occurrence dataset, eggs from rural and free-range rearing systems have a significantly higher burden of contamination (EFSA, 2012). The lack of consumption figures referring to rural eggs does not allow for a more detailed intake assessment. The related uncertainties address higher intakes only in those groups of the population that regularly purchase eggs at a local level.

3.3.2. Analysis

The expression of the results in the upper-bound mode determines an overestimation in the WHO_{05}-TEQ values in those low-contaminated food items, where some of the congeners are analytically reported below the LoQ. This is the case with food of vegetable origin, where the difference between the results expressed as upper-bound and lower-bound values may be as high as 65%. Consequently, there is uncertainty that intake from food of vegetable origin is overestimated; the dietary contribution under the lower-bound approach would drop to the 18% from the reported 25% in upper-bound (Fig. 1). This means a reduction of 8–10% in the overall mean estimates across the age groups considered. More detailed information is given in the Supplementary Materials (Table 1S).

Negligible uncertainties can be ascribed to the lack of data on PCDD/F and DL-PCB contamination in drinking water, soft drinks, alcoholic beverages, sweets, and sugar, whose contribution to the overall intake spans between 1 and 2% (Kiviranta et al., 2004; Liem et al., 2000).

3.3.3. Computing

The Italian Food Consumption Database reports rather limited data referring to child food habits in consumers only ($n = 193$). Such data are further reduced when broken down from the food category to the single food item (for instance, from fish consumption to trout consumption). At this point, the overall variability related to the probabilistic estimates can be greater when moving towards higher percentiles that are not supported by an adequate number of observations (Kroes et al., 2002). Therefore, the computed higher percentiles for intake by children may be biased due to an overestimation.

Other uncertainties are ascribed to food preparation. Cooking may represent a source of TEQ mitigation or enrichment, as is the case for lipid losses during food heating or for additional contamination from open-flame grilling, roasting, or baking. In this sense, ongoing Total Diet Studies at the national level might provide an on average insight into the extent and direction of such uncertainty towards under- or overestimates (Cubadda, 2016 personal communication).

3.4. PCDD/F and DL-PCB intake assessment

Previous estimates of food exposure to PCDD/Fs and DL-PCBs in the Italian population were performed by Fattore et al. (2006) and by EFSA (2012). Both the evaluations were based on the EU food occurrence dataset, and in 2006, the old WHO_{98}-TEF (Van den Berg et al., 1998) scale was used to compute the intake. The adoption of the old scale instead of the WHO_{05}-TEF (Van den Berg et al., 2006) may have led to a 15% overestimation in values, as assessed by EFSA (EFSA, 2010). In the EFSA 2012 assessment, the contribution from food of vegetable origin was not included, thus possibly underestimating around 20% the total WHO-TEQ intake. Despite this, a clear reduction in intake can still be noted when the mean estimates from this and from the other two available studies are compared. In children, from the 3.37 pg WHO_{98}-TEQ/kg bw per day in 2006 and the 2.15 pg WHO_{05}-TEQ/kg bw per day in 2012, the value has dropped to 1.98 pg WHO_{05}-TEQ/kg bw per day in 2016 A similar decreasing trend was also observed in adults (2.28, 1.21, and 0.90 pg WHO-TEQ/kg bw per day for 2006, 2012, and 2016 respectively).

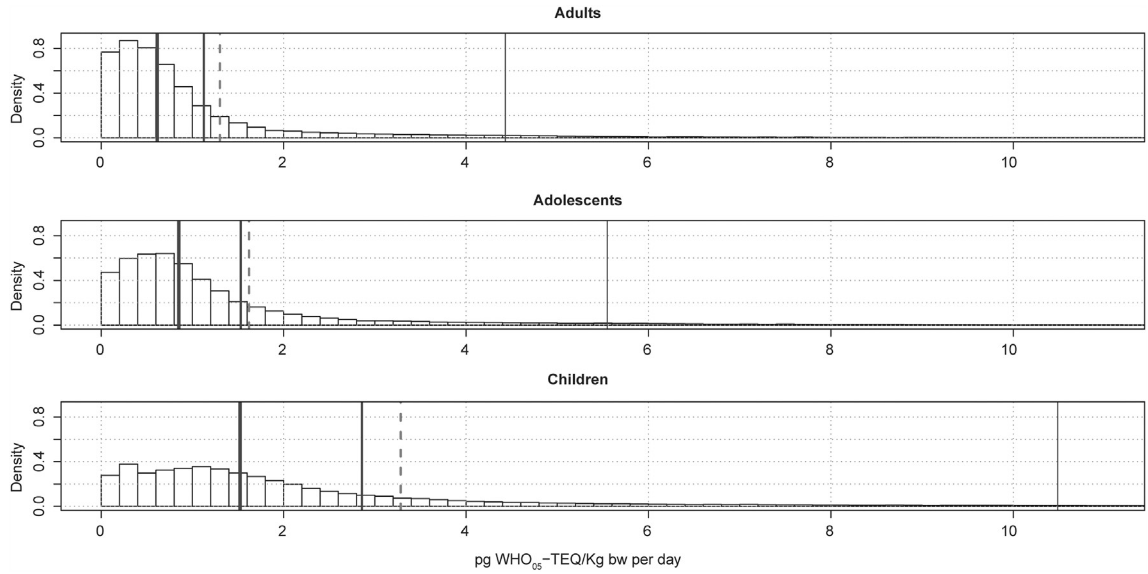

Fig. 2. Probabilistic estimates of alimentary intakes for three age groups (from bottom to top children, adolescents, and adults) for PCDD/Fs and DL-PCBs. From left to right of each distribution curve, the vertical lines correspond to P50, P75, mean (dotted) and P95 values.

In 2014, a surrogate of a duplicate diet study (De Filippis et al., 2014) based on lunch meals served weekly from Monday to Friday in five different school canteens in Italy indicated an intake range of 0.63–0.92 pg WHO_{05}-TEQ/kg bw per day. However, the contribution from breakfast, dinner, and weekend meals was not accounted for. In the same paper, from the assessment of one-day duplicate diets in eight adults, intakes fell within 0.33–0.63 pg WHO_{05}-TEQ/kg bw per day, with the lower value referring to a vegetarian diet.

Such comparisons highlight the possibility that the data derived from regular monitoring plans can be used to calculate intake estimates more representative of long-term periods and are not restricted to a limited timeframe as found for duplicate diets, and to a lesser extent, total diet studies, where sampling activities cover a limited timeframe.

Table 4
Cumulative target levels (TLs) in selected food groups/items able to bring the alimentary exposure below the Health-Based Guidance Value of 2 pg WHO-TEQ/kg bw per day in the most vulnerable group considered – Children 3–10 y. Comparison of TLs with the Regulatory Levels (RLs) in place in the European Union and interpolation with the related percentile from the occurrence distribution curves. Values computed as pgWHO_{05}-TEQ/g lipid weight basis (*lb*), with the exception of fish and seafood and food of vegetable origin expressed on fresh weight basis (*fb*).

Food group/item	TLs			RLs	TL Percentile
	Children		Lipid (%)[a]		
	fb	*lb*			
Food of vegetable origin	0.02		–	0.4[b]	P79
Fish and seafood	0.30		–	6.5	P49
Eggs	–	0.71	10	5.0	P83
Cheese	–	1.03	23[c]	5.5	P77
Whole bovine milk	–	1.06	3.6	5.5	P72
Meat and meat products	–	0.64	8.7[d]		P73
Beef and veal not preserved	–	1.25	5.2	4.0	P89
Chicken meat (*Gallus domesticus*)	–	0.84	3.4	3.0	P92
Turkey meat (*Meleagris gallopavo*)	–	1.45	2.4	3.0	P97
Pork not preserved	–	0.66	8.0	1.25	P97
Ham, salami and sausages	–	0.33	20	1.25	P77
Olive oil	0.10	–	100	1.25	P66
Butter	–	1.06	83.4	5.5	P72

[a] Lipid content derived from the national food composition database http://nut.entecra.it/646/tabelle_di_composizione_degli_alimenti.html.
[b] Action Level.
[c] Lipid percentage weighted on different cheese consumption.
[d] Lipid percentage weighted on different meat and meat products consumption.

At the European level, the Italian PCDD/F and DL-PCB intake assessment presented in this paper can be compared with the most recent studies carried out in France (Sirot et al., 2012) and in Germany (Schwarz et al., 2014). The French study was based on 212 diets formed by 1319 food samples representative of the most consumed items. Each food sample was formed by pooling up to 15 elementary samples. The mean and P95 intake estimates spanned from 0.57 to 0.89 pg WHO_{98}-TEQ/kg bw per day in adults and from 1.29 to 2.02 pg WHO_{98}-TEQ/kg bw per day in children and teenagers. <4% of the population were potentially overexposed with respect to a TDI of 2 pg WHO-TEQ/kg bw per day. The French estimates are lower than those presented in this study and could also be due to the different WHO-TEF scales used. The differences may be explained by the pool approach that can dilute the occurrence of PCDD/F and DL-PCB present in a single highly contaminated sample. This can be of particular relevance when defining the worst cases. The contribution to contamination intake from products of vegetable origin was <5% in the estimates for France, compared to the 25% in the present study (Fig. 1). This can be explained by the PCDD/F and DL-PCB occurrence computed with the medium-bound approach and with the differences in the consumption figures of such items between the French and the Italian food consumption databases. In contrast, in the German study, the contribution from food of vegetable origin was reported to be 20% of the average WHO_{98}-TEQ total intake in the general population across all age groups, equating to 2.11 pg WHO_{98}-TEQ/kg bw per day, with 5% of individuals potentially overexposed.

A comparison of the alimentary intake estimates described in the scientific literature from 2010 with those from this study is reported in Table 4S of the Supplementary Materials.

The intake assessment based on the probabilistic approach, when supported by an adequate number of observations (Kroes et al., 2002) could allow quantification of the risk related to the tails of distribution in the occurrence and consumption datasets, thus, helping health managers to orient monitoring activities in a risk-based way to reduce the percentage of potentially overexposed groups. Accounting for the uncertainties related to the abovementioned Italian Food Consumption Database in children, the probabilistic P95 intake estimate in this study indicated a value of 10.5 pg WHO_{05}-TEQ/kg bw per day, which is higher than the 5.7 pg WHO_{05}-TEQ/kg bw per day computed by EFSA in 2012. Similarly, overexposure is considered as 38.7% in the current study and 26.4% in the EFSA (2012) study, not accounting for the contribution from products of vegetable origin. Such discrepancies are increasingly reduced considering the other age groups. The P95 in adolescents: 5.55 vs 4.30 pg WHO_{05}-TEQ/kg bw per day with a 17.9% vs 15.4% of overexposed. The P95 in adults:

4.43 vs 3.70 pg WHO_{05}-TEQ/kg bw per day; 13.4% vs 17.3% of overexposed. Therefore, the risk characterization, expressed as Margin of Safety (MoS = the ratio between the related HBGV of 2 pg WHO_{05}-TEQ/kg bw per day and the estimated intake of this work) at the P95 would result in 0.19 in children, 0.36 in adolescents, and 0.45 in adults, respectively. More details are reported in Table **2S** of the Supplementary Materials.

It is worthy of note that in this paper the consumption data on important food categories, such as dairy, fish, and seafood products, were reduced to those referred to as single food items (Table 3). By doing this, in some cases, the related consumption figures were restricted to a few consumers. This explains that the progressively reduced differences between the calculated values and the EFSA P95 estimates, starting with children and ending with adult intakes, involves the increased representativeness of consumption data across the considered age groups.

Fig. 2 clearly highlights how the intake distribution estimates achieved under the probabilistic approach are skewed in all the three age groups, with mean intakes falling to the right of the computed P75 percentiles. Considering this perspective, the median values can be viewed as more robust, with less influence from outlier results.

The median intake values for the sum of PCDD/Fs and DL-PCBs were comparable between the deterministic and probabilistic model (see Table 2S in supplementary material and Table 2), with values of 1.40–1.52 for children, 0.82–0.85 for adolescents, and 0.64–0.61 for adults (expressed as pg WHO_{05}-TEQ/kg bw per day).

From the assessment of the contamination contribution to the WHO_{05}-TEQ intake shown in Fig. 1, vegetal items should be not overlooked due to high levels of daily consumption. The associated contamination of food of vegetable origin is more recalcitrant to be reduced than that present in food of animal origin. Reduction of the amount of contamination in the food of animal origin, with the exclusion of products from wild and free-range animals, has been progressively achieved through improved feedstuff quality administered to conventionally farmed feedlots and flocks. Moreover, the relevance of cheese to exposure, owing to elevated lipid content, is a consequence of the breakdown of dairy product consumption into single food items (Table 3). To the best of our knowledge, this point has been not raised in previous papers dealing with dioxin-like compound intake estimates.

The sensitivity analysis carried out for the most vulnerable group—children (Table 4)—underlines that the target levels that are compatible with tolerable intakes fall within the P50 and P97 of the occurrence distribution of the main food categories. This indicates that most Italian alimentary production can be considered safe. The target levels, susceptible to updates according to changes in alimentary habits, will provide useful guidance for reducing contamination and or the intake, through responsible choice and consumption of the food items. Such empowerment could be channeled through the release of health-based advisories targeting the most sensitive/vulnerable groups.

4. Conclusions

In this work, a general decreasing trend from 2006 to 2016 in PCDD/F and DL-PCB intake in the Italian population was observed. In the assessment, a more complete food consumption database targeting children is envisaged in order to reduce the related uncertainties. The presence of a valuable dataset for food samples analyzed under quality assurance and control schemes that are supported by descriptions regarding origin and provenience could be used to support the evolution of personalized food habits. This could be used to maximize nutritional benefits in potentially overexposed groups through an evidence-based reduction of the health risks associated with the intake of such classes of persistent organic pollutants.

Conflict of interest

The authors declare the absence of any conflicts of interest.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2018.01.181.

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