



## 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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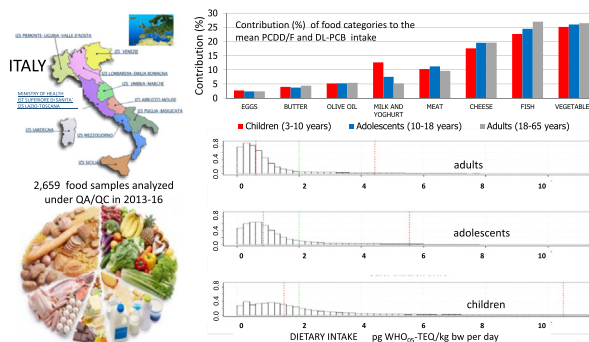
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### HIGHLIGHTS

- 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-TEQ<sub>5</sub>/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.

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## 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 <sup>13</sup>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<sub>05</sub>-TEQ/g for vegetables, 0.01–0.5 pg WHO<sub>05</sub>-TEQ/g for fish, and 0.06–0.4 pg WHO<sub>05</sub>-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

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