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## OCCURENCE OF PCDD/FS AND PCBS IN FOOD OF ANIMAL ORIGIN FROM ITALY: YEARS 2013-2015

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### Introduction

Polychlorinated dibenzo-p-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs) and polychlorinated biphenyls (PCBs) are ubiquitous toxic contaminants, that persist in the environment and bioaccumulate in the food chain. General population is exposed to these contaminants through the diet (more than 90%). Within this context, foodstuffs monitoring is a useful tool to measure the time trend of the occurrence and levels of PCDD/Fs and PCBs and, to update predictions of human exposure to these contaminants through the diet.

In Italy, the analysis of PCDD/Fs and PCBs on feed and food has been introduced by the Italian Ministry of Health in the National Residues Surveillance Plan (NRSP) since 2000, according to the Directive 96/23/EC.

In this paper we report results from Italian NRSP on the presence of PCDD/Fs and PCBs in food of animal origin. Data have been collected from 2013 to 2015 and represent a valuable information about current levels of dioxins in domestically produced food, in particular eggs, meat, milk and fish.

### Material and methods

Between 2013 and 2015, a total of 2447 samples were collected by the regional veterinary services, covering the national territory. Samples were taken at slaughterhouses, farms, production and transformation facilities. In order to prevent contamination, samples were placed in clean, inert glass or polyethylene containers. The samples were frozen and shipped in sealed boxes to the network of Italian official laboratories (Istituti Zooprofilattici Sperimentali) that performed the analyses.

Samples were tested by validated and accredited methods (EN ISO/IEC 17025) routinely used for PCDD/Fs, DL-PCBs and NDL-PCBs analysis in food and, successfully tested in a number of inter-laboratory studies. The procedure and requirements for collection, storage and analyses of dioxins and PCBs in food were in accordance with Commission Regulation (EU) No 252/2012 replaced in 2014 by Commission Regulation (EU) No 589/2014.

Analysis of PCDD/Fs and DL-PCBs was performed by HRGC/HRMS while for six NDL-PCB indicators both HRGC/HRMS and GC/MS-MS were used. PCDD/Fs and DL-PCBs toxic equivalent (TEQ) values were calculated using the World Health Organization Toxic Equivalency Factors (WHO-TEFs2005), while the sum of six indicator congeners was calculated for NDL-PCBs. According to the European legislation, WHO-TEQs and the sum of six NDL-PCBs were expressed as upper bound (UB) concentrations, assuming that all the values of the different congeners below the limit of quantification (LOQ) are equal to the respective LOQ.

Statistical evaluation for the three classes of contaminants was performed at TEQ level for PCDD/Fs and DL-PCBs, and the sum of six congeners for NDL-PCBs, considering only upper bound values. To compare results, mean, median and 95<sup>th</sup> percentile values, were calculated.

### Results and discussion

From 2447 samples, 187 were not included in the statistical analysis (meat of terrestrial animals with a percentage of fat below 2%). The final dataset was of 2260 samples: 184 fish samples (119 trout, 33 sea bass, 29 sea bream, 3 sturgeon), 439 eggs, 988 meat samples (322 beef, 265 chickens, 226 pigs, 118 turkeys, 47 sheep, 10 wild boars), 633 milk samples (330 bovine, 164 sheep, 116 buffaloes, 23 goats), and 16 sheep livers.

In 1950 samples PCDD/Fs, DL-PCBs and NDL-PCBs were analyzed simultaneously, while the remaining 310 samples were analyzed only for NDL-PCBs.

Table 1 summarizes the distribution of PCDD/F and DL-PCB contamination for each food category. The analytical results are reported as pg WHO-TEQ/g fat for food samples from terrestrial animals (except liver) and pg WHO-TEQ/g product for fish and sheep liver, in conformity with the European legislation. Regarding NDL-PCBs, because of the high proportion of not quantified congeners (more than 60% of samples reported LOQ values for all congeners) and great heterogeneity in LOQs (about one order of magnitude), the corresponding results of statistical analysis were not summarized in table 1. In fact, estimates of mean and median were strongly influenced by the high percentage of not detected congeners. Overall, relatively low contamination levels were recorded in each group. Median contamination levels of PCDD/Fs and DL-PCBs in food of terrestrial animals ranged from 0.14 to 0.22 and from 0.14 to 0.53 pg WHO-TEQ/g fat respectively while, in aquatic animals ranged from 0.036 to 0.36 and from 0.14 to 0.26 pg WHO-TEQ/g respectively. Comparing data from Italian monitoring 2013-2015 and EFSA 2012 inventory<sup>1</sup> (see table 2 for details), contamination levels, expressed in total TEQ, found in Italy were lower than those reported by EFSA for most of food categories, in particular eggs, bovine meat and trout samples. Generally, the lower values of total TEQ are ascribed to low levels of PCDD/Fs while contamination levels of DL-PCBs are in agreement with those reported by EFSA<sup>2</sup>.

With reference to non-compliance, a total of eight samples (5 eggs, 2 goat milk and 1 beef meat) exceeded the corresponding maximum level set by European legislation<sup>3</sup>. In all the cases, non-compliance was related to total TEQ and sum of six NDL-PCBs, except for one egg sample where high levels of PCDD/Fs were also recorded (3.5 pg WHO-TEQ/g fat).

It is important to underline that most of non-compliant samples were collected under targeted sampling, when an environmental contamination of dioxin-like compounds was suspected. In non-compliant eggs, total TEQ ranged from 10.4 to 12.7 pg WHO-TEQ/g fat and NDL-PCBs from 35.8 to 208 ng/g fat; in milk samples total TEQ ranged from 14.3 to 29.3 pg WHO-TEQ/g fat and NDL-PCBs from 77.2 to 170 ng/g fat while in beef meat total TEQ was 5.9 pg WHO-TEQ/g fat and NDL-PCBs were 20.4 ng/g fat.

Among food from terrestrial animals, the DL-PCBs were the main contributor to the total TEQ, accounting for 58% - 85% of the total TEQ, except for eggs, pig meat, chicken meat and sheep liver, where the contribution of DL-PCBs was between 45 % and 52 %. The same trend was observed in the aquatic animals, except for sturgeon, where PCDD/Fs accounted for 72 % of the total TEQ. In non-compliant samples, the contribution of the DL-PCBs to the total TEQ increased from 66% to 96%.

There was considerable variation in contamination levels within food groups, except for pig meat, sheep liver and aquatic animals. In some cases, this difference was mainly due to the source of data that included results from both random and targeted monitoring.

### Acknowledgements

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### References

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2. European Food Safety Authority; Results of the monitoring of dioxin levels in food and feed. EFSA Journal 2010; 8(3):1385 [36 pp.]. doi:10.2903/j.efsa.2010.1385.
3. Commission Regulation (EC) No 1881/2006 of 19 December 2006.

**Table 1. Number of samples analyzed, mean, median and 95<sup>th</sup> percentile values for TEQ PCDD/F and DL-PCB levels across food groups (pg WHO-TEQ/g fat, except liver and fish pg WHO-TEQ/g product)**

Matrix	Species	N. samples	PCDD/Fs			Dioxin like PCBs		
			Mean	Median	P 95	Mean	Median	P 95
Egg	Hen	389	0.28	0.19	0.88	0.47	0.15	1.1
Meat	Bovine	270	0.20	0.18	0.41	0.59	0.46	1.5
	Poultry	210	0.23	0.17	0.60	0.30	0.17	0.73
	Pig	208	0.17	0.17	0.26	0.15	0.14	0.25
	Turkey	95	0.21	0.18	0.50	0.32	0.28	0.67
	Sheep	37	0.23	0.17	0.58	0.53	0.37	1.5
	Wild Boar	10	0.30	0.21	0.72	0.50	0.26	1.8
Milk	Cow	304	0.27	0.22	0.63	0.77	0.53	2.1
	Sheep	142	0.25	0.19	0.62	0.68	0.23	2.0
	Buffalo	111	0.19	0.14	0.42	0.34	0.26	0.67
	Goat	23	0.26	0.18	0.59	1.3	0.43	3.4
Liver	Sheep	16	0.50	0.47	1.21	0.38	0.34	0.83
Fish	Trout	85	0.070	0.036	0.35	0.15	0.14	0.36
	Bass	32	0.039	0.036	0.083	0.45	0.22	1.4
	Sea Bream	24	0.062	0.038	0.23	0.29	0.26	0.65
	Sturgeon	3	0.38	0.36	0.41	0.15	0.15	0.16

**Table 2. Mean, median and 95<sup>th</sup> percentile values for total-TEQ levels from EFSA 2012 inventory and from Italian 2013-2015 results, for the selected food items (pg WHO-TEQ/g fat, except liver and fish pg WHO-TEQ/g product)**

PCDD/Fs + Dioxin like PCBs							
Matrix	Species	Italy 2013-2015			EFSA 2012		
		Mean	Median	P 95	Mean	Median	P 95
Egg	Hen	0.75	0.36	2.1	1.62	0.61	5.16
Meat	Bovine	0.78	0.64	1.8	2.34	1.68	5.97
	Poultry	0.53	0.37	1.2	0.99	0.56	2.79
	Pig	0.33	0.32	0.55	0.31	0.11	0.79
	Turkey	0.53	0.50	0.91	--	--	--
	Sheep	0.75	0.51	2.0	1.24	0.84	3.18
	Wild Boar	0.80	0.46	2.3	--	--	--
Milk	Cow	1.0	0.77	2.7	1.91*	0.77*	4.36*
	Sheep	0.93	0.46	2.6			
	Buffalo	0.54	0.45	1.1			
	Goat	1.5	0.64	3.9			
Liver	Sheep	0.88	0.85	2.2	--	--	--
Fish	Trout	0.22	0.17	0.55	1.05	0.94	2.01
	Bass	0.49	0.25	1.5	--	--	--
	Sea Bream	0.35	0.31	0.82	--	--	--
	Sturgeon	0.53	0.51	0.56	--	--	--

\* Referred to milk and dairy products in general