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Research Article

Update of Indicator PCB Levels in Food in Southern Italy: Assessment of the Dietary Exposure for Adult and Elderly Population

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The levels of non-dioxin-like PCB indicators (iPCBs 28, 52, 101, 138, 153, and 180) were determined in food samples (seafood, meat and processed meat, milk and dairy products, hen eggs, olive oil, and other fats) to evaluate the exposure of adult and elderly population. iPCB levels in samples were in the following order: fishery products > meat and processed meat > milk and dairy products > olive oil and other fats. None of the samples had concentrations above the maximum permissible limits for human consumption established by the European Union legislation, except for salami samples. The dietary intake for the total population was 12.33 $ng{\cdot}kg^{-1}$ $bw{\cdot}d^{-1}$, while depending on the sex/age groups, exposure was estimated between 9.60 and 12.11 $ng{\cdot}kg^{-1}$ $bw{\cdot}d^{-1}$, with seafood being the major contributor. The exposure scenario indicates that further efforts must still be carried out to protect the consumer from these harmful chemicals.

1. Introduction

Polychlorinated biphenyls (PCBs) are a group of man-made substances comprising 209 congeners classified as persistent organic pollutants (POPs). This complex mixture of isomers differs in chemical structure according to the position of chlorine substituents around the biphenyl backbone. It is well recognized that different chlorination patterns deeply affect the biological activity and toxicological profile of these chemicals. PCBs with non- or mono-ortho chloro substitution, known as dioxin-like PCBs (dl-PCBs), exert their toxicity primarily through binding with the aryl hydrocarbon receptor, while the remaining congeners classified as non-dioxin-like PCBs (ndl-PCBs) appear to act via different modes determining a complex spectrum of adverse effects, such as neurological, neuroendocrinological, and immunological disorders [1]. To this latter group belong six congeners, the so-called indicator PCBs (iPCBs), considered a suitable tool to monitor the contamination degree because their sum constitutes about half of the total ndl-PCBs present in feed and foods [2]. In addition to their higher abundance compared to other congeners, recent studies indicate that ndl-PCBs are not eliminated as easily as dl-PCBs and that low ndl-PCBs doses, such as those similar to background contamination in food, can cause negative effects when exposure is prolonged over time [3]. Consequently, understanding the level, trend and impact of these contaminants on human health remain a salient point for the researchers [2]. Many studies have been carried out to evaluate the dietary exposure to these POPs worldwide lending, in general, greater attention to more toxic dl-PCBs than ndl-PCBs. Also, in Italy, the volume of existing data for iPCBs is limited and predominantly referred to contamination of fishery products [4–8] and milk [3, 9–12], while the other foods have been the subject of very few investigations, although some of them showed levels of contamination often exceeding the established limits for human consumption [13–16]. For example, the results obtained from the analyses of meat and hen egg samples from two regions of Southern Italy, Apulia [13] and Campania [16], revealed

iPCB concentrations above maximum permissible limits set by the European Union. Consequently, this study is placed in the context of the need to acquire further data on the presence of such contaminants also in other food products in Italy. In making the analysis, we ascertain whether the concentrations of six iPCBs are compliant with the European Union food law requirements. Finally, we estimate the exposure level in the adult and elderly population and compare it with the acceptable daily intake of 10 ng·kg^{-1} bw·d^{-1} set for the sum of six ndl-PCB indicators at the national level by various countries.

2. Materials and Methods

2.1. Sample Collection. In May–July 2019, food samples were randomly acquired from five supermarkets in cities (Bari, Lecce, Taranto, Foggia, Brindisi, and Matera) in Southern Italy. A total of 35 types of foods are classified into the following groups: (1) fish and seafood, (2) meat and meat-based products, (3) milk (whole cow milk) and dairy products, (4) eggs from free foraging chickens, and (5) extra-virgin olive oil and fats were acquired from five supermarkets of each city. For each food, 120 individual items were taken and combined in two composite samples. For shellfish and crustaceans, 300 individual units were used to prepare the composite sample. For bluefin tuna, slices ($n = 30$) of about 0.1–0.2 kg of muscle tissue were taken. The composite samples (only edible parts) were homogenized and stored below −20°C.

2.2. Analytical Method and Instrumental Analysis. The concentrations of six indicator PCBs (iPCBs: 28, 52, 101, 138, 153, and 180) were determined. The analytical method has been reported in detail in the previous paper [17]. Homogenized samples (0.5–3.0 g) were mixed with Na_2SO_4 and spiked with PCB 143 used as the internal standard. The mixture was extracted with hexane, and the extracts were evaporated to dryness, permitting the gravimetric determination of the fat content [18]. The milk fat was isolated by using liquid-liquid extraction with diethyl ether and petroleum ether (1 : 1) [19]. An aliquot (about 100 mg) of the extracts was dissolved in hexane and cleaned by passing through 8 g of acid silica (H_2SO_4, 44% w.w.), using 50 mL of a mixture of hexane/dichloromethane (1/1, v/v) for elution of the analytes. The eluates were dried completely by using a stream of nitrogen and redissolved in 100 μL of iso-octane. To control the whole sample preparation process, appropriate C^{13}-labelled standards (Wellington Laboratories Inc., Guelph, Ontario, Canada) were added to the samples before extraction (PCB-LCS-H) and at the clean-up stage (PCB-SCS-H). The identification and quantification of iPCBs were performed on an HRGH/HRMS system composed of a gas chromatograph trace series coupled with a mass spectrometer 2000 MAT 95 XL (Thermo Finnigan, Bremen, Germany). An Rtx-PCB capillary column (60 m × 0.25 mm i.d. × 0.25 μn film thickness; Restek, Cernusco Sul Naviglio, MI, Italy) was used for chromatographic separation of iPCBs. The initial oven temperature was set at 100°C for 2 min, then raised to 180°C, at a rate of 20°C/min, hence increased to 260°C at a rate of 2°C/min, and finally brought up to 300°C with a rate of 5°C/min held for 4 min. The carrier gas used was helium, 6.0 purity degree (99.9999%), in the constant flow mode of 1.0 mL/min. The injection temperature was set at 280°C, and the injections were performed in the splitless mode with a 1 ml vol. (for 1 min. and a split rate of 140 ml/min). The temperatures of the ion source and transfer line were set at 270 and 290°C, respectively. The electron ionization mode (E.I.) was used for mass method operations. Electron energy and detector voltages were 35 eV and 350 V, respectively. The detector resolving power was >10.000 (10% valley definition), and the two most intense ions were monitored for the determination of the single congeners. Six points (0.5, 2.0, 5.0, 10.0, and 20.0 ng·g^{-1}) and calibration curves ($r^2 > 0.999$) in the linear response interval of the detector were created for quantification. To test the accuracy of the method, certified reference material and blank samples were analysed with each batch of samples. Levels of each iPCBs in the procedural blanks were always under the limit of detection. To correctly identify the tested compounds and to check matrix interferences, some food samples were fortified with a known amount of each PCB congener obtaining satisfactory recoveries (PCB 28 = 91%, PCB 52 = 94%, PCB 101 = 96%, PCB 138 = 100%, PCB 153 = 104%, and PCB 180 = 98%). All samples were analysed in triplicate. Precision was tested every day by running 3 replicates of the calibration curves at the beginning and at the end of the analyses (RDS = 15%). Mackerel oil (No 350-Community Bureau of Reference, BCR) from Promochem GmbH was used as reference material. For the replicate, standard reference materials, and recovery of labelled compounds, the relative standard deviations (RSD) were <10% for all the detected compounds. The recovery rates of labelled standards were between 85 and 120%. The limits of detection (LODs) were calculated on the blanks (n.°30) analysed along with unknown samples and were set as three times the signal-to-noise ratio (0.04–1.40 pg·g^{-1} for PCBs). The limits of quantification (LOQs) were between 0.0004 and 0.04 ng·g^{-1} for PCBs and corresponded to ten times the signal-to-noise ratio. Concentrations of ndl-PCBs are

Table 1: Daily intake of food items (gday^{-1}) by the Italian population for two different sex/age classes (adults and elders).

Food categories	Total population	Adults		Elders	
		M	F	M	F
Fish and seafood	38.8	41.2	38.7	42.2	28.6
Veal	42.7	48.7	38.0	51.9	35.2
Pork	12.7	16.4	11.6	11.5	7.2
Poultry	20.8	22.8	18.5	20.5	22.0
Processed meat	27.3	35.4	23.3	24.7	17.6
Milk	119.3	94.3	110.5	119.7	129.9
Yogurt	20.6	16.3	26.8	10.6	18.9
Cheese	57.0	65.6	54.5	57.0	49.9
Hen eggs	20.9	24.4	18.7	23.2	18.8
Olive oil	32.7	36.7	31.4	37.9	29.3
Butter	4.1	4.8	3.9	3.9	3.6
Other fats	0.9	1.3	0.8	0.4	0.4

Table 2: Concentrations of individual iPCBs (ngg^{-1}) and their sum (Σ6 iPCBs).

Food groups	Samples	% Lipid	PCB 28	PCB 52	PCB 101	PCB 138	PCB 153	PCB 180	Σ6 iPCBs
Fish[a]									
Rosefish	120	0.3	≤LOD	≤LOD	0.129	0.591	0.867	0.294	1.883
European hake	120	0.9	0.153	≤LOD	0.225	0.945	1.773	0.900	3.997
Red mullet	120	1.3	0.169	0.195	0.351	1.989	3.445	1.495	7.644
Common sole	120	0.2	≤LOD	≤LOD	0.248	0.856	1.430	0.330	2.866
Atlantic bluefin tuna	30	1.0	0.300	0.240	0.570	2.650	4.860	2.570	11.190
Average fish	—	—	0.125	0.088	0.305	1.406	2.475	1.118	5.516
Seafood[a]									
Cephalopods									
Common octopus	120	0.1	≤LOD	≤LOD	0.007	0.008	0.011	≤LOD	0.029
Common cuttlefish	120	0.4	0.012	0.016	0.015	0.024	0.031	0.016	0.114
European squid	120	0.8	0.008	0.008	0.024	0.016	0.032	0.016	0.104
Average cephalopods	—	—	0.007	0.008	0.015	0.016	0.025	0.014	0.082
Shellfish									
Mediterranean mussel	300	2.5	0.184	0.251	0.378	2.527	4.150	2.201	9.690
Striped Venus clam	300	2.7	0.100	0.150	0.425	0.825	1.070	0.650	3.220
Common scallop	300	0.9	0.036	0.108	0.198	0.621	0.783	0.315	2.061
Average shellfish	—	—	0.107	0.170	0.334	1.324	2.001	1.055	4.990
Crustacean									
Red shrimp	300	0.3	≤LOD	≤LOD	0.006	0.006	0.020	≤LOD	0.036
Spottail mantis shrimp	300	0.4	≤LOD	≤LOD	0.040	0.500	0.928	0.220	1.690
Norway lobster	300	0.3	≤LOD	≤LOD	≤LOD	0.110	0.190	≤LOD	0.304
Average crustacean	—	—	0.001	0.001	0.016	0.205	0.379	0.074	0.677
Average fish and seafood	—	—	0.069	0.070	0.186	0.833	1.399	0.644	3.202
Meat and processed meat[b]									
Veal fillet	120	5.4	6.380	8.260	10.550	10.690	12.350	3.240	**51.470**
Pork loin	120	7.5	4.250	6.000	7.120	7.480	5.560	2.020	32.430
Chicken breast	120	0.9	5.230	5.360	6.080	8.120	9.110	1.780	35.680
Turkey breast	120	1.2	6.750	6.420	5.850	7.960	10.020	2.250	39.250
Average meat	—	—	5.653	6.510	7.400	8.563	9.260	2.323	39.708
Salami	120	37.9	1.675	11.175	6.690	13.570	9.790	3.740	**46.640**
Mortadella	120	9.5	1.950	4.440	6.260	1.980	2.030	0.940	17.600
Raw ham	120	4.5	≤LOD	2.220	2.090	0.970	1.060	≤LOD	6.388
Baked ham	120	19.80	0.840	2.350	3.960	1.120	1.590	0.530	10.390
Average processed meat	—	—	1.120	5.046	4.750	4.410	3.618	1.310	20.255
Milk and dairy products[b]									
Milk	120	3.7	0.600	1.195	1.990	1.080	2.415	2.200	9.480
Hard cheese (cow milk)	120	30.1	3.030	30.380	7.430	0.810	1.560	≤LOD	**43.215**
Hard cheese (sheep milk)	120	31.5	2.870	15.270	3.810	3.820	3.760	2.020	31.550
Yogurt	120	3.6	2.110	1.000	6.230	2.280	4.540	2.420	18.580
Mozzarella	120	21.9	≤LOD	≤LOD	≤LOD	≤LOD	0.540	0.330	0.884
Stracchino	120	19.8	≤LOD	0.100	0.170	0.210	0.650	0.350	1.484
Ricotta	120	16.9	≤LOD	≤LOD	≤LOD	≤LOD	0.700	0.280	0.999
Mascarpone	120	45.7	≤LOD	≤LOD	0.250	0.120	0.540	0.380	1.275
Average milk and dairy products	—	—	1.078	5.995	2.486	1.041	1.836	0.998	13.433
Hen eggs[b]	120	23.1	1.860	0.990	0.760	3.830	1.950	1.330	10.700
Olive oil[b]	120	78.0	1.450	1.370	≤LOD	≤LOD	≤LOD	≤LOD	2.825
Other fats[b]									
Butter	120	77.2	1.580	5.590	2.800	0.740	2.140	1.880	14.730
Margarine	120	72.0	1.430	3.560	3.260	0.930	2.010	0.980	12.170
Mayonnaise	120	69.4	1.740	2.420	3.570	1.680	1.770	0.460	11.640
Average fats	—	—	1.583	3.857	3.210	1.117	1.973	1.107	12.847

Maximum permissible levels (MPLs) set by European Union regulation [18]: 75 ng·g^{-1} wet weight for fish and seafood, 40 ng·g^{-1} lipid weight for meat and processed meat, milk and dairy products, hen eggs, olive oil, and other fats; bold values > MPLs a: ng·g^{-1} wet weight; MPLs b: ng·g^{-1} lipid weight.

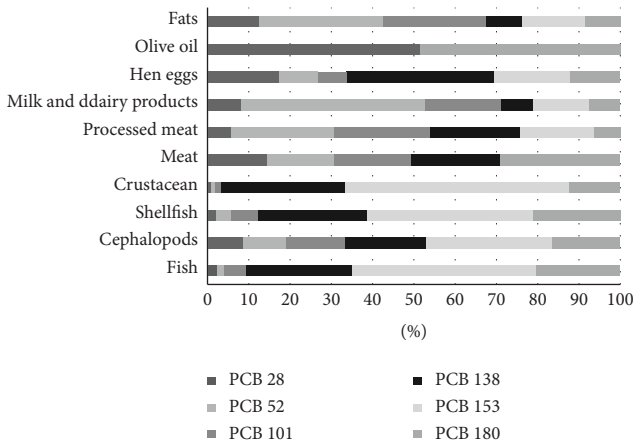

FIGURE 1: Percentage of contribution (%) from each congener to the sum of six iPCBs.

expressed as $ng \cdot g^{-1}$ on wet or lipid weight basis in accordance with the EU legislation [20].

2.3. Exposure Assessment. The estimated dietary intakes (deterministic approach) were calculated by multiplying the food consumption data by the mean concentrations of iPCBs in each food followed by dividing by the body weight. Consumption (Table 1) and biometric data of the total population and of two sex/age classes (male adults: 18–64.9 years, body weight 78.4 kg; female adults: 18–64.9 years, body weight 62.2 kg; male elders: ≥65 years, body weight 78.1 kg; female elders: ≥65 years, body weight 65.0 kg; total population: body weight 70.0 kg) were obtained from the Italian national food consumption survey INRAN-SCAI [21, 22]. To estimate the total intake of the sum of the six iPCBs (Σ6 iPCBs), individual daily intakes of each iPCBs from different food items were added up. For exposure calculations, the contamination level of each sample expressed in lipid weight was converted into wet weight by using the lipid content of the samples. Exposure data were estimated according to both the lower bound and upper bound hypotheses. The Kruskal–Wallis test was undertaken to compare the estimated weekly intake according to the gender for each age group. All p values below 0.05 were considered statistically significant.

3. Results and Discussion

3.1. Levels in Food and Compliance with the EU Regulation. The analytical results of six iPCBs and their sum (Σ6 iPCBs) for each food category, expressed as $ng \cdot g^{-1}$ on a wet or lipid weight basis to facilitate the comparison with the maximum permissible limits (MPLs) set by the European Union (EU) [20], are illustrated in Table 2. The differences between upper and lower bound values are negligible, so the results are expressed in upper bound hypothesis.

Within the fishery products, the highest contamination mean level was in fish, followed by shellfish, crustaceans, and cephalopods that showed moderate concentrations. Because the species are from different trophic guilds, there is the potential for the observed concentration differences. In fact, when analysing the contaminant load, one must take into account the trophic niche in which the species are inserted. Consequently, the increase in concentrations from low-trophic-level invertebrates to high-trophic-level organisms is an expected finding, demonstrating the effect of PCB biomagnification through the food chains. In the similar way, within fish, the top-level carnivore as the Atlantic bluefin tuna showed the highest levels, followed by benthic species rosefish and red mullet, while the lowest value was in the European pelagic hake. The contribution of individual six congeners to total iPCB was generally in the order of PCB 153 > PCB 138 > PCB 180, while the combined contributions of the lower chlorinated congeners to iPCB total strongly varied being 9.4%, 12.2%, 33.3%, and 2.3% in fish, shellfish, cephalopods, and crustaceans, respectively (see Figure 1).

The distribution pattern of congeners is in very good match with results reported in the literature, where PCBs 138, 153, and 180 are described as the predominant congeners in marine biota [23]. The prevalence of these highly chlorinated congeners is the result of their greater resistance to metabolism and elimination than the lower congeners and is also linked to their major abundance in technical mixtures, such as Aroclor 1254 and 1260 [24], the most commonly used in Europe. For fishery products, the EU has set an MPL for the Σ_6 iPCBs of $75\ ng \cdot g^{-1}$ wet weight, and our results in all marine species did not exceed this limit. Within the meat group, the sum of the six congeners on a median basis was $39.71\ ng \cdot g^{-1}$ lipid weight, and the highest concentration was found in veal fillet, followed by poultry composite samples having values in a similar range, whereas the pork meat showed the lowest concentrations. A higher variability level was registered within processed meats with concentrations ranging from $6.34\ ng \cdot g^{-1}$ lipid weight in raw ham to $46.64\ ng \cdot g^{-1}$ lipid weight in salami. Differences in PCB contamination between meat samples could be explained by a multitude of factors such as the environmental quality in which the animal lives, the contamination level in the feed, and the life span of the animals [25]. For processed meats, the different manufacturing processes altering the fat content can be responsible for the observed variability in the levels of these lipophilic chemicals [26]. For example, Kuzukiran and Filazi [27] found in fermented air-dried meat products (salami, soudjouk, and sausage) from a variety of animals a wide concentration range ($0.407–3.936\ ng \cdot g^{-1}$ lipid weight) corroborating the assumption of an influence of meat processing in PCB levels. The study of the distribution profiles of iPCBs highlighted a difference between meats and processed meats (see Figure 1). For meat samples, the profile was dominated by PCBs 153 (23.3%), 138 (21.6%), and 101 (18.6%), followed by PCBs 52 (16.4%) and 28 (14.2%) having a similar percentage contribution and by PCB 180 whose concentrations accounted for 5.8% of all six congeners. Instead, in processed meat, the percentage contribution of PCB 52 to the sum of six indicators was the highest (24.9%), followed by PCB 101 (23.5%) and PCBs 138 (21.8%) and 153 (17.9%), while the lowest percentages were for the congeners 28 (5.5%) and 180 (6.4%). However, regardless of the observed profile differences, the values in

Table 3: Estimated daily intakes of iPCBs (ngkg^{-1} bwd^{-1}) from the consumption of various categories of food for the total population and for adults (18–64.9 years) and elders (≥65 years).

Food categories	Total population				Adults		Elders			
	M		F		M		F			
	LB	UB	LB	UB	LB	UB	LB	UB	LB	UB
Fish	3.057	3.057	2.898	2.899	3.431	3.432	2.980	2.980	2.427	2.427
Seafood	1.061	1.062	1.006	1.007	1.191	1.192	1.035	1.035	0.842	0.843
Meat	0.574	0.574	0.593	0.593	0.573	0.573	0.584	0.584	0.483	0.483
Processed meat	2.337	2.337	2.705	2.705	2.244	2.244	1.895	1.895	1.622	1.622
Milk	0.341	0.341	0.235	0.235	0.355	0.355	0.307	0.307	0.400	0.400
Dairy products	2.840	2.841	2.909	2.911	3.067	3.068	2.533	2.535	2.679	2.680
Hen eggs	0.738	0.738	0.769	0.769	0.743	0.743	0.734	0.734	0.715	0.715
Olive oil and fats	0.478	0.478	0.510	0.511	0.510	0.511	0.430	0.431	0.431	0.432
Total intake	**11.426**	**11.428**	**11.625**	**11.629**	**12.114**	**12.118**	**10.498**	**10.501**	9.599	9.602

Bold values >10 ngkg^{-1} bwd^{-1} (ADI); M = male, F = female; LB = lower bound, UB = upper bound.

meats depicted a contamination rather high with concentrations exceeding (see veal fillet) or close to the MPL of 40 ng·g^{-1} lipid weight imposed by the EU. In contrast, in the processed meats, no sample exceeded the normative value, except for salami samples characterized by the high level of iPCBs (46.64 ng·g^{-1} lipid weight). Within the milk and dairy products, the concentrations varied from a minimum of 0.87 ng·g^{-1} lipid weight in mozzarella to a maximum of 43.21 ng·g^{-1} lipid weight in cow milk cheese samples. With percentages respectively of 44.6%, 18.5%, and 13.7%, PCBs 52, 101, and 153 dominated, whereas the remaining congeners contributed to the total concentration of iPCBs (PCB 28: 8.0%, PCB 138: 7.7%, PCB 180: 7.4%) in quite similar proportions (see Figure 1). However, concentrations exceeding the EU maximum permissible level were found solely in cow milk cheese samples. Concerning hen eggs, the concentrations of iPCBs did not exceed the MPL of 40 ng·g^{-1} lipid weight established by the EU. PCBs 138 (35.8%) and 153 (18.2%) were the major contributors to the Σ_6 iPCBs, followed by PCB 28 which also showed a consistent percentage (17.3%), while the smallest percentage quotas were for PCBs 101 (7.1%), 52 (9.2%), and 180 (12.4%) (see Figure 1). The olive oil samples with concentrations of 2.82 ng·g^{-1} lipid weight due solely to PCBs 28 (51.4%) and 52 (48.6%) met the requirements specified in the EU legislation. In the remaining fats, PCBs levels ranging from 11.64 ng·g^{-1} lipid weight for mayonnaise to 14.73 ng·g^{-1} lipid weight for butter samples were below the MPL.

3.2. Dietary Exposure to iPCBs. Consumer health protection starts with the development of measures to reduce exposure to toxic substances, so reference toxicological doses have been set to ensure that people do not exceed a certain body load. ndl-PCBs account for the majority of total PCBs in the environment and food [2] and thus are responsible for a significant quota of human exposure. Nonetheless, establishing a guide value for ndl-PCB remains complicated due to the simultaneous presence of the much more potent dl-PCBs which makes it difficult to determine the precise contribution of ndl-PCB to consumer exposure. However, an acceptable daily intake (ADI) of 10 ng·kg^{-1} bw·d^{-1} for their sum has been set at the national level in some countries [28, 29]. Taking into account this reference value, dietary intakes in the total population and the adult and elderly groups exceeded the ADI, with a single exception represented by elderly females for whom a value lower but close to the fixed limit was calculated (see Table 3).

Table 4: Percentage of contribution (%) from each food categoriy to the total dietary intake of iPCBs for the total population and for adults (18–64.9 years) and elderly (≥65 years).

Food categories	Total population	Adults		Elders	
		M	F	M	F
Fish and seafood	36.0	33.6	38.2	38.2	34.1
Meat and processed meat	25.5	28.3	23.2	23.5	21.9
Milk and dairy products	27.8	27.1	28.3	27.1	32.1
Hen eggs	6.5	6.6	6.1	7.0	7.4
Olive oil and fats	4.2	4.4	4.2	4.1	4.5

However, the intake values did not reach levels of statistical significance within sex/age groups ($p > 0.05$) suggesting similarity in rates and patterns of consumption for population classes examined. In fact, in adults and elders, fishery products dominated in the exposure with percentages ranging from 34.1% to 38.2%. The food group milk and dairy products was the second most important source of iPCBs intake contributing between 27.1% and 32.2% in male adults and female elders, respectively. Meat and processed meat showed a contribution lower than dairy products, with percentages varying from 21.9% to 28.3%. Hen eggs, olive oil, and other fats played a minor role with contributions ranging between 6.6% and 9.0% and 3.4% and 4.5%, respectively. Similarly, in total population, fish and other seafood categories were the major contributors to total intake (36.0%), followed by milk and dairy products (27.8%), meat and processed meat (25.5%), hen eggs (6.5%), and olive oil and other fats (4.2%) (see Table 4).

An analogous contribution pattern has been observed in earlier dietary intake studies carried out in Italy [14] and in other European countries such as France [30], Spain [31], and Germany [32]. Exceptions have been encountered for the Belgian adult population where dairy products and

Table 5: Estimated intake of iPCBs' comparison with other European surveys.

Food	Country	Total population	Adults	Elders	References
Fish and seafood	Italy	4.12	3.90–4.62	4.01–3.27	This study
	Austria	—	0.60–0.83	—	Mihats et al., [34]
	France	—	1.73	—	Sirot et al., [30]
	Greece	2.02–4.03	—	—	Stagakis et al., [40]
Fish	Italy	3.06	2.90–3.43	2.43–2.98	This study
	Greece	2.15–3.38	—	—	Renieri et al., [39]
Meat and meat products	Italy	2.91	2.81–3.29	2.10–2.47	This study
	France	—	0.28	—	Sirot et al., [30]
Meat	Italy	5.51	—	—	Barone et al., [13]
Chicken meat	Poland	17.7	—	—	Rusin et al., [41]
	Poland	2.1	—	—	Rusin et al., [41]
Milk and dairy products	Italy	3.18	3.15–3.43	2.85–3.09	This study
	Austria	—	1.34–1.74	—	Mihats et al., [34]
	France	—	0.35	—	Sirot et al., [30]
Cow milk	Poland	2.1	—	—	Rusin et al., [41]
	Poland	1.1	—	—	Rusin et al., [41]
Hen eggs	Italy	0.74	0.74–0.77	0.71–0.73	This study
	Italy	0.07–0.36	—	—	Castellani et al., [42]
	Austria	—	0.11–0.14	—	Mihats et al., [34]
	Poland	2.7	—	—	Rusin et al., [41]
	Poland	0.6	—	—	Rusin et al., [41]
Olive oil and fats	Italy	0.48	0.51	0.43	This study
	France	—	0.23	—	Sirot et al., [30]
Total diet study	Italy	11.43	11.63–12.11	9.60–10.50	This study
	Italy	—	10.9	—	Fattore et al., [14]
	Austria	—	2.6–3.2	—	Mihats et al., [34]
	Belgium	5.33	—	—	Cimenci et al., [33]
	France	—	2.78	—	Sirot et al., [30]
	France	—	7.7	—	Arnich et al., [35]
	Germany	—	5.6	—	Fromme et al., [36]
	Denmark	—	12.6	—	Fromberg et al., [37]
	Norway	5.2	—	—	Kvalem et al., [38]
	Europe	4.3–25.7	—	—	EFSA, [22, 43]

fishery products contribute almost equally to the total dietary exposure to the iPCBs [33] and in the Austrian adult population with milk and dairy products being the major contributing food group to the total dietary intake of ndl-PCBs (50–55%) followed by the seafood and meat group [34]. Table 5 illustrates the comparison among iPCBs estimates of dietary exposure in Europe, as described in the literature. In Italy, for the general population in 2008, the mean estimated intake of iPCBs was 10.9 $ng{\cdot}kg^{-1}$ $bw{\cdot}d^{-1}$ for adults [14]. In the case of France, the mean exposure to the six indicators of PCBs was 2.71 $ng{\cdot}kg^{-1}$ $bw{\cdot}d^{-1}$ in the adult population [30], while another French study for the same population group reported a higher exposure level equal to 7.7 $ng{\cdot}kg^{-1}$ $bw{\cdot}^{d-1}$[35]. In a German survey carried out with 20 participants employing the duplicate diet method, Fromme et al. [36] showed an average intake for the sum of iPCB indicators in adults of 5·6 $ng{\cdot}kg^{-1}$ $bw{\cdot}d^{-1}$. The dietary exposure of Belgians to iPCBs ranged from 5.33 $ng{\cdot}kg^{-1}$ $bw{\cdot}d^{-1}$ to 6.05 $ng{\cdot}kg^{-1}$ $bw{\cdot}d^{-1}$ for the lower and upper bound approaches, respectively [33]. In Austria, the mean dietary intake of iPCBs was estimated between 2.64 and 3.19 $ng{\cdot}kg^{-1}$ $bw{\cdot}d^{-1}$ for adult women and men [34]. A dietary intake of 12.6 $ng{\cdot}kg^{-1}$ $bw{\cdot}d^{-1}$ was reported from Denmark [37], while a Norwegian study found a median intake of 4.4 $ng{\cdot}kg^{-1}$ $bw{\cdot}d^{-1}$ and 5.4 $ng{\cdot}kg^{-1}$ $bw{\cdot}d^{-1}$ for adult and elderly consumers, respectively [38]. The most recent studies available in the literature (Table S. 2) present the intakes of these chemicals from specific food products. For example, for fish and fish products from Greece, different authors have reported exposure levels near our results [39, 40]. Higher levels have been reported in Poland for chicken meat and hen eggs [41], whereas in Italy, a lower intake from free-range hen egg consumption was registered [42]. It is obvious that the comparison of the exposure results, already complicated by different methodological choices (sampling strategies, analytical methods, targeted congeners, and the level used to assess the dietary intake), must also consider the population's dietary preferences and the rates of consumption. Consequently, it should be clearly pointed out that the comparison of the different exposure results has to be interpreted with care. However, the estimated total intake of the exposure levels calculated in the present study were in line with data extracted from EFSA [43], reporting estimates of ndl-PCB exposure for the European population from 4.3 to 25.7 $ng{\cdot}kg^{-1}$ $bw{\cdot}day^{-1}$. Nevertheless, the intakes estimated near the higher part of the ADI of 10 $ng{\cdot}kg^{-1}$ $bw{\cdot}day^{-1}$ could

be the result of special consumption habits, as particularly high consumption of seafood plays a fundamental role in consumer exposure.

4. Conclusions

The present results could serve as a set of reference data for exposure to the adult and elderly population in Southern Italy, for which no data are currently available, except for one of our previous surveys [44]. Concentrations found in foodstuffs examined appear to be below the currently recommended European non commercialization values, with the exception of salami samples. The estimation of the dietary intake due to consumption of the entire group of these foods producing values above or near to the amount declared by various countries as tolerable daily intake seems to indicate that efforts must still be carried out to observe a major reduction of human exposure. This is also in consideration of the fact that estimation of the intake does not consider other food items consumed which can be a source of these harmful chemicals. It is hence worth emphasizing the need to monitor the residues of ndl-PCBs in all foods and especially in fishery products which are considered the major contributor to the dietary ndl-PCB intake.

Data Availability

Raw data were generated at the University of Bari, Veterinary Laboratory. Derived data supporting the findings of this study are available from the corresponding author M.M.S. on request.

Conflicts of Interest

The authors declare that they do not have any conflicts of interest.

Authors' Contributions

Conceptualization was conducted by Grazia Barone and Maria M. Storelli; methodology was conducted by Grazia Barone, Arianna Storelli, and Rita Garofalo; software work was carried out by Grazia Barone and Arianna Storelli; validation was conducted by Arianna Storelli and Maria M. Storelli; formal analysis was performed by Arianna Storelli; data curation was carried out by Grazia Barone and Arianna Storelli; writing and original draft preparation were conducted by Arianna Storelli and Maria M. Storelli; writing, review, and editing were performed by Arianna Storelli and Maria M. Storelli. All authors have read and agreed to the published version of the manuscript.

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