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EFFECT OF FRYING ON POLYCHLORINATED BIPHENYLS CONTENT IN MUSCLE MEAT OF SELECTED FISH SPECIES

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Polychlorinated biphenyls (PCBs) are xenobiotics of particular toxicological significance. Regarding their high persistence and lipophilicity, the compounds can pose a threat to human and animal health. High PCB bioaccumulation factors were found in the aquatic environment, especially in predatory fish, crustaceans, aquatic birds and their eggs. The compounds are chemical pollutants of biosphere and undesirable substances in food, which is the main source of PCB intake for humans (above 90%).

The aim of the study was to determine concentrations of indicator PCBs (IUPAC numbers: 28, 52, 101, 118, 153, 138, 180) in the raw meat of selected fish species, and to estimate the effect of frying on the compounds concentrations. Chemical analysis was conducted using a GC/MS technique. In the raw fish meat, the mean sum of PCBs ranged from 0.63 μg/kg wet weight in carp to 6.58 μg/kg wet weight in flounder. In the fried fish meat, lower PCB levels in lipids were observed. Mean losses of PCB congeners, on the lipid weight basis, ranged from 32.1% in flounder to 81.1% in cod. The sum of PCBs in the fried fish meat was the highest in flounder, and the lowest in carp. PCB 153 and PCB 138 were the dominant congeners in most of the examined samples, whereas PCB 28 and PCB 52 occurred usually in the smallest amounts. Carp meat, both raw and fried, contained significantly lower levels of congeners PCB 101, PCB 118, PCB 153, PCB 138, than the other fish species examined.

INTRODUCTION

Polychlorinated biphenyls (PCBs) are aromatic compounds formed by two combined phenyl rings, with a varying number of chlorine atoms attached. They are some of the most persistent synthetic compounds, of high chemical stability. PCBs are distinguished by their high thermal conductivity, low electrical conductivity, and good resistance to thermal decomposition. They have low water solubility and high solubility in lipids and organic solvents. Lipophilicity of chlorobiphenyls increases along with the number of chlorine atoms in their molecules [Falandysz, 1999; Mackay *et al*., 2006].

The main sources of PCBs in the environment are solid wastes and wastewaters of both industrial and municipal origin. PCBs may escape from improperly maintained waste sites, penetrating soil and surface waters or leaking to atmosphere. Regarding their slow degradation and problems with their utilization, the compounds are chemical pollutants of biosphere and undesirable substances in food [Flegal *et al*., 2007; Ciereszko & Witczak, 2002].

Food products of aquatic origin, such as fish, contain higher PCB levels in their edible parts than other kinds of food [Falandysz, 1987; Allsopp, 2000].

Food processing usually contributes to reduction of PCB levels in food products. Numerous studies revealed that PCB levels in fish meat decreased as a result of culinary treatment [Zabik *et al*., 1982, 1996; Moya *et al*., 1998; Ciereszko & Witczak, 2003; Witczak & Ciereszko, 2006 a, b].

The aim of this study was to determine concentrations of indicator PCBs (IUPAC numbers: 28, 52, 101, 118, 153, 138, 180) in the raw meat of selected fish species, and to estimate the effect of frying on their concentrations.

MATERIALS AND METHODS

The research material consisted of muscle tissue collected from edible portions of herring (*Clupea harengus*), salmon (*Salmo salar*), common carp (*Cyprinus carpio*), brown trout (*Salmo trutta m. fario*), flounder (*Platichthys flesus*), cod (*Gadus morhua*) and roach (*Rutilus rutilus*). The fish were bought as skinned fillets at retail in Szczecin, except roach which were bought whole.

Sample preparation and determination of polychlorinated biphenyls were carried out according to Polish Standards [PN-EN 1528-1 – PN-EN 1528-4].

Muscle tissue of each fish species was sampled as two combined samples of 5 individuals each. One of the combined samples was examined as raw material, while the other one was pan-fried in plant oil for ten minutes. From each combined sample, three subsamples (10-15 g each) were taken and freeze dried for 36 h in a LyoLab 3000 apparatus. For qualitative and quantitative analyses a solution of 7 PCB congeners (IUPAC numbers: 28, 52, 101, 118, 153, 138, 180) in isooctane

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(NEN 0813, LGC Promochem GmbH D-46 485 Wesel) was used, while recoveries were examined by addition of a known amount of the Pesticides Surrogate Spike Mix (SUPELCO, USA, 4-8460), which was an acetone solution of decachlorobiphenyl and 2,4,5,6tetrachloromxylene. The compounds examined were extracted in lipids (Soxhlet extractor, 8 h) with 150 mL of an acetone/hexane mixture (v/v, 3:1). The extracts obtained were concentrated with a rotary vacuum evaporator to *ca.* 2 mL and fat content was determined gravimetrically. Then the lipids were dissolved in 2 mL of nhexane and purified with 7 mL of oleum (7% SO_3 in concentrated H_2SO_4). Next, the extracts were washed with deionized water, dried over anhydrous sodium sulphate and concentrated to 1 mL using nitrogen evaporation. Analyses were conducted with a GC/MS HP 6890/5973 gas chromatograph (in Selected Ion Monitoring – SIM mode), and operating conditions of the chromatographic analyses were, as follows: carrier gas – helium; pressure: 26 psi (1.77 atm); flow rate: 1.2 mL/min, column: CPSIL8 CB LOW BLEED (60 m x 250 μm x 0.25 μm; Chrompaq); column temperature program: 140°C (hold 0.5 min) → increase rate 10°C/min → 200°C (hold 5 min) → increase rate 5°C/min → 280°C (hold 10 min) → increase rate 30°C/min → 300°C (hold 5 min); single sample analysis time: 43.17 min.

Recoveries of the analysed PCB congeners were determined on the basis of the internal standard recoveries (Pesticides Surrogate Spike Mix), and ranged within 75-91%.

Figure 1 shows a model chromatogram of an analysed sample.

RESULTS AND DISCUSSION

Lipid content in the raw muscle tissue of the examined fish ranged from 0.55% for cod to 13.71% for herring. After frying, lipid content in fillets of all fish species increased significantly and varied from 3.48% for cod to 19.64% for herring (Figure 2).

Residues of PCB congeners were detected in the examined material. The highest level of the sum of PCBs was detected in raw muscles of flounder. In the raw fish meat, the mean sum of PCBs ranged from 0.631±0.440 μg/kg wet weight in carp to 6.584±3.099 μg/kg wet weight in flounder. Lipid normalized levels of the sum of PCBs varied within 10.90±0.27 μg/kg lipids for carp and 182.9±35.90 μg/kg lipids for roach (Table 1).

In the majority of samples examined, the dominant compounds were PCB 153 and PCB 138, with concentrations ranging from 0.918±0.541 and 1.914±1.006 μg/kg w.w. and from 0.049±0.036 to 1.968±1.130 μg/kg w.w., respectively (Table 1).

Of all the fish species examined, the lowest levels of PCB congeners occurred in carp meat, while the highest were found in meat of flounder, salmon and cod. Lipid normalized PCB concentrations were the highest in roach, due to relatively low lipid content (1.22%) in its meat (Figure 2).

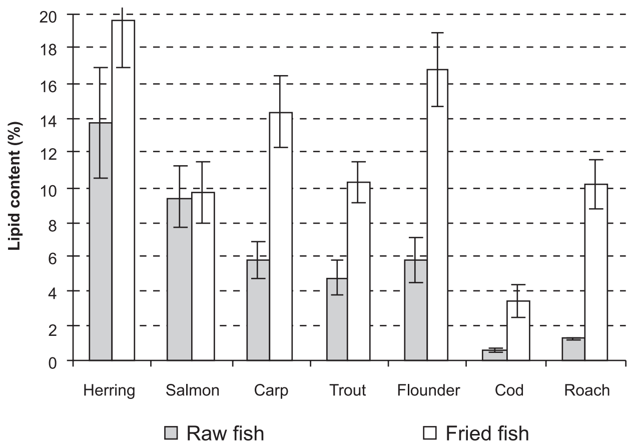

FIGURE 2. Average lipid content in the examined assortment before and after frying.

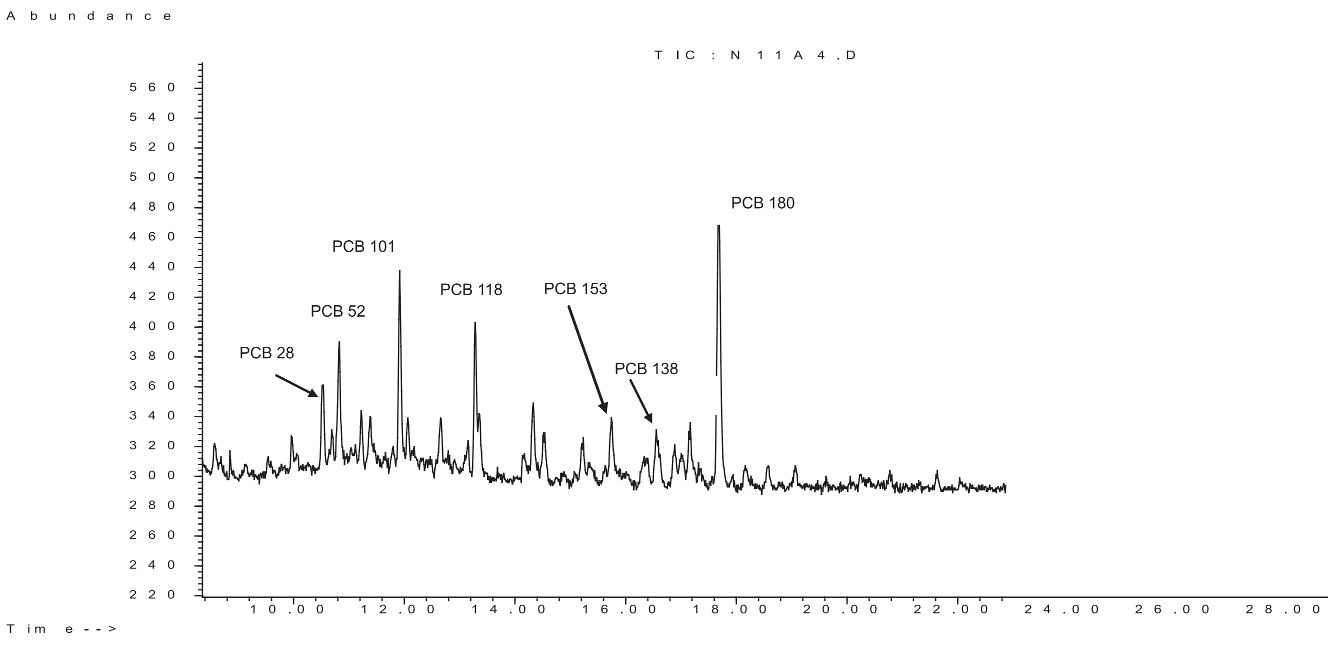

FIGURE 1. A model chromatogram of an analysed sample (herring muscle tissue).

TABLE 1. Levels of indicator PCB congeners in raw fish meat.

ASSORTMENT	Concentration (μg/kg w.w.)							
	PCB 28	PCB 52	PCB 101	PCB 118	PCB 153	PCB 138	PCB 180	ΣPCB
Herring	0.140±0.10	0.171±0.099	0.636±0.220	0.809±0.411	1.264±0.820	1.092±0.540	0.333±0.119	4.445±2.38
Salmon	0.152±0.098	0.105±0.008	0.763±0.301	0.752±0.247	1.732±1.063	1.450±0.940	0.521±0.374	5.474±3.031
Carp	0.080±0.035	0.088±0.032	0.059±0.027	0.065±0.036	0.189±0.117	0.049±0.036	0.091±0.077	0.631±0.440
Trout	0.164±0.102	0.080±0.024	0.470±0.165	0.382±0.111	0.918±0.541	0.846±0.240	0.251±0.118	3.110±1.301
Flounder	0.144±0.100	0.155±0.094	0.741±0.260	0.940±0.230	1.914±1.006	1.968±1.130	0.772±0.279	6.584±3.099
Cod	0.059±0.029	0.033±0.011	0.074±0.050	0.103±0.098	0.286±0.109	0.266±0.197	0.126±0.096	0.950±0.590
Roach	0.188±0.112	0.066±0.018	0.251±0.168	0.185±0.101	0.540±0.123	0.517±0.261	0.485±0.207	2.232±0.990
	Concentration (μg/kg lipids)							
Herring	1.021±0.73	1.247±0.69	4.639±1.21	5.901±2.98	9.220±2.90	7.965±2.34	2.429±1.230	32.42±7.70
Salmon	1.617±0.821	1.117±0.673	8.117±2.970	8.000±1.302	18.43±5.132	15.43±5.921	5.543±1.120	58.23±20.09
Carp	1.382±0.29	1.520±0.07	1.019±0.25	1.123±0.15	3.264±0.22	0.846±0.120	1.572±0.23	10.898±0.27
Trout	3.431±1.08	1.674±0.340	9.833±1.53	7.992±2.40	19.205±3.27	17.698±5.39	5.251±1.15	65.06±13.37
Flounder	2.504±0.04	2.696±1.42	12.887±4.34	16.448±6.34	33.29±5.51	34.23±8.86	13.43±3.57	114.5±93.1
Cod	10.73±3.19	6.000±2.34	13.455±4.67	18.73±5.24	52.00±12.35	48.36±5.67	22.91±8.76	172.7±19.4
Roach	15.41±6.13	5.410±1.98	20.57±5.32	15.164±6.21	44.26±10.01	42.38±12.54	39.75±11.16	182.9±35.90

Of all the PCB congeners analysed, PCB 28 and PCB 52 were the less abundant in the examined filets. Their concentrations ranged from 0.033±0.011 μg/kg w.w. for PCB 52 in cod to 0.188±0.112 μg/kg w.w. for PCB 28 in roach (Table 1).

The environmental fate and behaviour of chlorobiphenyl congeners, as well as their selective accumulation in living organisms, result from physicochemical properties of the compounds. The lower chlorinated congeners, *i.e.* 2,4,4'- T_3CB (PCB 28) and 2,2',5,5'- T_4CB (PCB 52) have higher vapour pressures and water solubility than the more highly chlorinated congeners, such as 2,2',3,4,4',5'-H_6CB (PCB 138) and 2,2',3,4,4',5,5'-H_7CB (PCB 180), which have higher lipophilicity [Falandysz, 2002; Mackay *et al*., 2006].

In the fried fish meat, significant ($p<0.05$) losses of the analysed PCB congeners were usually observed (Figure 3). The lowest losses occurred in the case of PCB 52 in flounder (10.5%), while the greatest losses were found for PCB 138 in herring and carp (100%). Among all the congeners analysed, the highest average losses (72.9%) were observed for PCB 138, and the lowest (44.4%) for PCB 52 (Figure 3).

Changes in concentrations of individual PCB congeners in fish meat resulting from frying were also reflected by reduction in the sum of PCBs. Percentage losses calculated varied from 32.05% in flounder to 81.11% in cod (Figure 4).

Comparing the fried fish, the highest level of the sum of PCBs was detected in flounder – 4.474±3.099 μg/kg w.w., while the lowest level was found in carp – 0.358±0.44 μg/kg w.w. (Table 2).

Reduction of PCB residue levels in processed food usually results from fat leakage (together with dissolved PCBs) during culinary and technological treatments, and partly also from PCBs codistillation with water vapour during thermal processing [Zabik *et al*., 1996; Ciereszko & Witczak, 2003].

We also observed that changes of PCB levels in the fried fish meat were additionaly influenced by fat content increase in the final products which occurred due to absorption of oil used for frying. Simultaneously water content decreased,

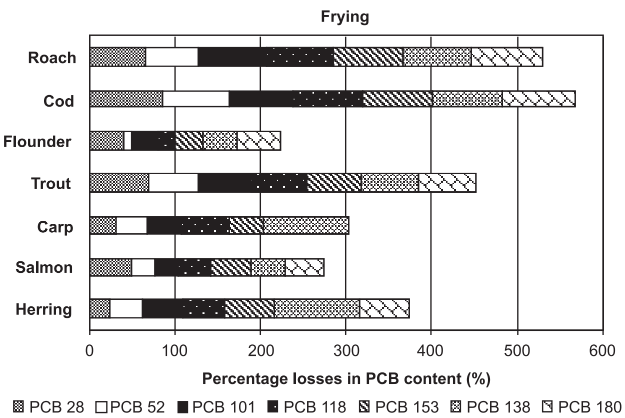

FIGURE 3. Percentage losses of individual PCB congeners in fish meat resulting from frying.

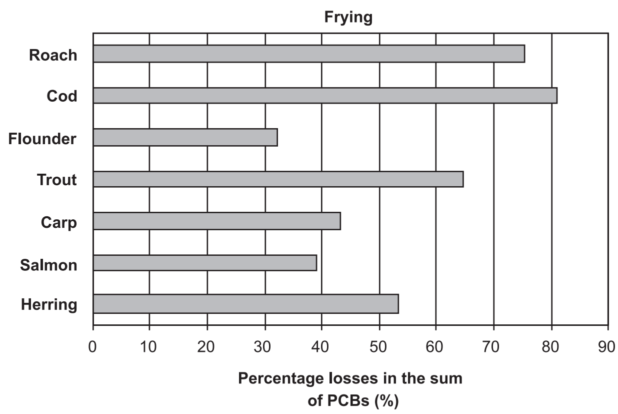

FIGURE 4. Percentage losses of the sum of PCBs in fish meat resulting from frying.

TABLE 2. Levels of indicator PCB congeners in fish meat after frying.

ASSORTMENT	Concentration (μg/kg w.w.)							
	PCB 28	PCB 52	PCB 101	PCB 118	PCB 153	PCB 138	PCB 180	ΣPCB
Herring	0.106±0.002	0.106±0.047	0.339±0.020	0.406±0.011	0.538±0.120	nd	0.147±0.029	2.082±1.008
Salmon	0.077±0.042	0.077±0.008	0.555±0.011	0.458±0.047	0.939±0.081	0.848±0.093	0.294±0.014	3.337±3.031
Carp	0.055±0.015	0.056±0.012	0.036±0.007	0.028±0.010	0.115±0.025	nd	0.091±0.037	0.358±0.440
Trout	0.051±0.011	0.032±0.008	0.189±0.065	0.127±0.011	0.339±0.076	0.281±0.039	0.082±0.038	1.103±1.301
Flounder	0.086±0.035	0.139±0.064	0.518±0.068	0.769±0.130	1.264±0.206	1.204±0.180	0.365±0.079	4.474±3.099
Cod	0.009±0.005	0.007±0.002	0.019±0.005	0.020±0.008	0.054±0.009	0.046±0.009	0.019±0.006	0.179±0.590
Roach	0.064±0.031	0.025±0.010	0.052±0.008	0.043±0.010	0.102±0.026	0.098±0.005	0.084±0.007	0.545±0.990
	Concentration (μg/kg lipids)							
Herring	0.540±1.73	0.540±1.69	1.726±10.2	2.067±8.98	2.739±21.9	nd	0.748±6.00	10.60±70.70
Salmon	0.793±0.821	0.793±0.673	5.716±2.970	4.717±2.302	9.670±5.132	8.733±5.921	3.028±2.120	34.37±20.09
Carp	0.384±0.29	0.391±0.07	0.251±0.25	0.196±0.35	0.803±0.22	nd	0.635±0.63	2.50±0.27
Trout	0.496±1.18	0.311±0.44	1.839±4.53	1.235±2.40	3.298±10.27	2.733±10.39	0.798±2.15	10.73±31.37
Flounder	0.513±0.04	0.830±1.42	3.093±10.34	4.591±16.34	7.546±25.51	7.188±28.86	2.179±10.57	26.71±93.1
Cod	0.259±3.19	0.201±4.34	0.546±9.98	0.575±15.24	1.552±41.35	1.322±35.67	0.546±18.76	5.144±119.4
Roach	0.629±6.13	0.246±1.980	0.511±5.32	0.422±6.21	1.002±10.01	0.963±12.54	0.825±11.16	5.3549±35.9

nd – not detected

which means that the examined compounds co-distillated with water vapour.

Ciereszko & Witczak [2003] reported that PCB residue losses in fried carp meat can be up to 50% of the initial concentration in raw meat, which confirmed previous findings of other researchers [Zabik *et al.,* 1996; Zabik *et al.*, 1995].

Results of this study are also in concordance with previous studies of other researchers which revealed that PCB residue levels decreased in fish tissues subjected to culinary treatment [Smith *et al.,*1973; Falandysz, 1982; Zabik *et al.,* 1996; Moya *et al.,* 1998; Ciereszko & Witczak, 2003; Witczak & Ciereszko, 2006a, b]. Falandysz [1982] reported that food processing caused partial reduction of polychlorinated biphenyl residues. Smith *et al.* [1973] informed that roasting and simmering of salmon meat decreased PCB levels by 2-16%. According to Zabik *et al.* [1982], PCB residues losses in carp meat subjected to grilling, frying or microwave cooking ranged within 10-20%. Whereas in pan and deep fat fried carp harvested from Lakes Erie and Huron PCB losses were up to 30-35% [Zabik *et al.*, 1995]. Moya *et al.* [1998], in their study on the effects of cooking on PCB levels in fillets of winter flounder (*Pseudopleuronectes americanus*), observed even higher PCB losses – up to 47%. The fillets they examined contained 0.8-4.5% of fat and were subjected to deep frying, pan frying with butter and broiling. The highest losses of total PCB resulted from deep frying. The authors attributed this to the fact that the deep frying process created unique cooking conditions that accelerated drying of the fillets compared with broiling and pan frying. They also explained this reduction by evaporation of water and PCBs from the fillets as a result of high temperature of the cooking oil and by transfer of PCBs to the cooking oil which itself could be acting as an extraction solvent.

Deep fried skin-on carp fillets showed lower PCB losses than pan fried skin-off fillets, which indicates that significant reduction of PCB residue levels in the final product may be attained by skin removal [Zabik *et al.,*1995]. Similar conclusion was drawn by Armbruster *et al.* [1987, 1988] who studied effects of thermal processing on PCB levels in bluefish (*Pomatomus saltatrix*) and striped bass (*Morone saxatilis*) harvested near Long Island. Skin removal alone reduced PCB residue levels by 40-50%, and thermal processing – by further 7.5--15%.

Thermal treatment applied in this study significantly influenced changes in PCB residue levels, improving the quality of the examined fish products in terms of organochlorine pollutants content.

CONCLUSIONS

1. PCB 153 and PCB 138 were the dominant congeners in most of the examined samples, whereas PCB 28 and PCB 52 occurred usually in the smallest amounts.

2. Carp meat, both raw and fried, contained significantly lower levels of congeners PCB 101, PCB 118, PCB 153, PCB 138, than the other examined fish species.

3. In the raw fish meat, the mean sum of PCBs ranged from 0.631±0.440 μg/kg wet weight in carp to 6.584±3.099 μg/kg wet weight in flounder.

4. In the fried fish, percentage losses of the sum of PCBs varied within 32.05% for flounder and 81.11% for cod.

5. Comparing losses of individual congeners during frying, losses of PCB 138 residues were the highest (72.97%), and losses of PCB 52 residues – the lowest (44.40%).

6. The main reasons of PCB residues losses in the fried fish meat were probably codistillation with water vapour and extraction to oil.

REFERENCES

1. Allsopp M., Erry B., Stringer R., Johnston P., Santillo D., Recipe for disaster, A review of persistant organic pollutants in food. Greenpeace Research Laboratories. United Kingdom 2000 [http://www.greenpeace.to/publications.asp#2000].
2. Armbruster G., Gerow K.G., Gutenmann W.H., Littman C.B., Lisk D.J., The effects of several methods of fish preparation on residues of polychlorinated biphenyls and sensory characteristics in striped bass. J. Food Saf., 1987, 8, 4, 235–243.
3. Armbruster G., Gall K.L., Gutenmann W.H., Lisk D.J., Effects of trimming and cooking by several methods on polychlorinated biphenyls (PCB) residues in bluefish. J. Food Saf., 1988, 9, 4, 235–244.
4. Ciereszko W., Witczak A., Concentration of PCBs and selected pesticides in bottom sediments, Zebra mussel and in some more important fish species of the Szczecin Lagoon. Acta Ichthyol. Piscat., 2002, 32, 1, 35–40.
5. Ciereszko W., Witczak A., Changes in concentrations of selected PCB congeners in carp meat as a result of cooking. Acta Sci. Pol. Technol. Aliment., 2003, 1, 155–164 (in Polish).
6. Falandysz J., The effect of processing on polychlorinated biphenyl levels in foodstuffs, Roczn. Panstw. Zakl. Hig., 1982, 33, 27–33 (in Polish).
7. Falandysz J., Determination of polychlorinated biphenyl (PCB) residues in seafood products and an attempt to assessing daily intake of the compounds in Poland in years 1970–1982. Studia i Materiały, Seria D, 16, 1987, Wyd. Morskiego Instytutu Rybackiego, Gdynia (in Polish).
8. Falandysz J., Polychlorinated biphenyls (PCBs) in the environment: chemistry, analysis, toxicity, concentrations and risk assessment. 1999, Fundacja Rozwoju Uniwersytetu Gdańskiego, Gdańsk (in Polish).
9. Falandysz J., Wyrzykowska B., Puzyn T., Strandberg L., Rappe C., Polychlorinated biphenyls (PCBs) and their congener – specific accumulation in edible fish from the Gulf of Gdańsk, Baltic Sea. Food Addit. Contam., 2002, 8, 779–795.
10. Flegal A.R., Conaway C.H., Davis J.A., Connor M.S., Introduction. Sources, transport, fate, and toxicity of pollutants in the San Francisco Bay estuary. Environ. Res., 2007, 105, 1–4.
11. Mackay D., Shiu W.Y., Ma K.C., Lee S. C., Halogenated Hydrocarbons. 2006, *in*:, Handbook of Physical-Chemical Properties and Environmental Fate for Organic Chemicals., 2nd edition, vol. 2, (eds. Mackay D., Shiu W.Y., Ma K.C., Lee S. C.). CRC Press, pp. 1570–1931.
12. Moya J., Garrahan K.G., Poston T.M., Durell G.S., Effects of cooking on levels of PCBs in the fillets of winter flounder. Bull. Environ. Contam. Toxicol., 1998, 60, 845–851.
13. Polish Standard PN-EN 1528–1. Fatty food – Determination of pesticides and polychlorinated biphenyls (PCBs) – Part 1: General.
14. Polish Standard PN-EN 1528–2. Fatty food – Determination of pesticides and polychlorinated biphenyls (PCBs) – Part 2: Extraction of fat, pesticides and PCBs, and determination of fat content.
15. Polish Standard PN-EN 1528–3. Fatty food – Determination of pesticides and polychlorinated biphenyls (PCB) – Part 3: Clean-up methods.
16. Polish Standard PN-EN 1528–4. Fatty food – Determination of pesticides and polychlorinated biphenyls (PCBs) – Part 4: Determination, confirmatory test, miscellaneous.
17. Smith W.E., Funk K., Zabik M.E., Effect of cooking on concentrations of PCB and DDT compounds in chinook (Oncorhynchus tshawytscha) and coho (o. kisutch) salmon from Lake Michigan. J. Fish Res. Board Can., 1973, 30, 702.
18. Witczak A., Ciereszko A., Changes in total PCB content in selected fish products during hot- and cold smoking. Acta Ichthyol. Pisc., 2006a, 36, 11–16.
19. Witczak A., Ciereszko A., The effect of smoking process on changes in the content of selected non-*ortho* and mono-*ortho* PCB congeners in mackerel slices. J. Agric. Food Chem., 2006 b, 54, 5664–5671.
20. Zabik M.E., Merrill C., Zabik M.J., PCBs and other xenobiotics in raw and cooked carp. Bull. Environ. Contam. Toxicol., 1982, 28, 710–715.
21. Zabik M.E., Zabik M.J., Booren A.M., Nettles M., Song J.-H., Welch R., Humphrey H., Pesticides and total polychlorinated biphenyls in Chinook Salmon and Carp harvested from the Great Lakes: Effects of skin-on and skin-off processing and selected cooking methods. J. Agric. Food Chem., 1995, 43, 993–1001.
22. Zabik M.E., Booren A., Zabik M.J., Welch R., Humphrey H., Pesticide residues, PCBs and PAHs in baked, charbroiled, salt boiled and smoked Great Lakes lake trout. Food Chem., 1996, 55, 231–237.

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