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Toxic elements in free-living freshwater fish, water and sediments in Poland

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Abstract

Samples for analysis were collected from 10 areas, including the major Polish rivers and lakes, with different sources of environmental pollution (industrial, municipal, and farming). The materials was taken from the lakes of Mazury, located in a non-industrialised region, from the Brda River, an area impacted by pig farms, from the lakes of Lipczyno Wielkie/Pomerania, from the Wkra River, an area impacted by poultry farms, from the Dunajec River at the Roznowski Reservoir, from the Vistula River at Cracow and Warsaw, from the Odra River at Wroclaw and the Warta River estuary, and also from Rybnik Power Station Reservoir. Concentrations of Pb, Cd, Hg, and As were analysed in 397 fish muscle and 128 sediment samples using an atomic absorption spectrometry technique. The analytical procedures were covered by a quality assurance programme. It was demonstrated that the average concentrations of lead, cadmium, and arsenic in fish were in the low hundredths and thousandths of a mg/kg and never exceeded permitted limits established for food. Higher values of these elements were found in fish from bodies of water located in the zone of influence of large urban agglomerations, especially the Cracow region. High concentrations of lead and cadmium were also found in Vistula River sediments near Cracow, where the maximum values were 134.10 mg/kg and 21.24 mg/kg dry weight for lead and cadmium respectively. The average concentration of mercury in a predatory fish muscle (0.179 mg/kg) was almost twice as high as in the omnivorous fish (0.103 mg/kg). Only a single fish sample exceeded the maximum limit for this metal (0.50 mg/kg) and did not present a risk to consumers' health.

Keywords: rivers, lakes, fish, sediments, toxic elements, Poland.

Introduction

The problem of presence of toxic elements in fish and other aquatic organisms is connected with water pollution as a result of natural processes and the release of hazardous compounds to soil and air, as well as broader human activity.

Some elements such as zinc, iron, and copper, play an important role in the biological system when present in significant amounts. In contrast, lead, cadmium, mercury, or arsenic can be toxic, even in trace amounts. The concentrations of these elements in fish and sediments are the subject of scientific discussion in many publications (5, 9, 10, 18, 22, 24, 25, 27-29). Lead and cadmium are widespread in the environment. For humans they pose a risk through consumption of contaminated food and inhalation (7, 12, 18, 29, 30).

The predominant form of mercury present in the atmosphere is metallic mercury, which is almost 95% of the mercury emitted into the environment. Biomethylation plays an important role in the movement of mercury, where the products of this process can easily transfer among different compartments of the environment. Both metallic mercury and its various compounds contained in mineral deposits and sediments are biomethylated (1). Methyl mercury compounds are readily soluble in water and become very dangerous for fish and humans (4). High bioaccumulative capacity for mercury in the trophic chain causes the contamination of air, soil, water and food. Fish are a major source of mercury for living organisms, especially for people, and they pose a relevant health risk (3, 13).

In the case of arsenic, inorganic trivalent arsenic compounds are the most toxic to living organisms.

Inorganic arsenic can be methylated after rising up the food chain to humans or animals, thus less toxic organic forms are created. The studies of biological samples identified the occurrence of several tens of organic forms of arsenic. Organic arsenic compounds such as arsenobetaine (AsB) or arsenocholine (AsC) are the dominant forms existing in aquatic organisms (algae, fish, crustaceans, and molluscs). Many authors revealed that the organic arsenic in fish, primarily in the form of arsenobetaine, constitutes 75%-95% of the total arsenic content (8, 16, 17).

The aim of the study was to evaluate the concentrations of lead, cadmium, mercury, and arsenic in selected species of free-living freshwater fish inhabiting different environments (rivers and lakes), and in water and sediments from municipal, industrial, and farming regions.

Material and Methods

Between 2011 and 2013, an analysis was conducted of toxic elements in different species of freshwater fish, sediments, and water obtained from different bodies of water. The samples were collected from 10 areas with different sources of environmental pollution (industrial, municipal, and farming). The material consisted of omnivorous and predatory fish (Table 1), which were taken from locations including Poland's major rivers and lakes (the lakes of Mazury, located in a non-industrialised region, the Brda River, an area impacted by pig farms, the Wkra River, an area impacted by poultry farms, the lakes of Lipczyno Wielkie/Pomerania, the Dunajec River at the Roznowski Reservoir, the Vistula River at Cracow and Warsaw, the Odra River at Wroclaw, the Warta River estuary, and Rybnik Power Station Reservoir).

Table 1. List of examined fish species from rivers, lakes, and reservoirs

	Sampling site	Omnivorous fish	Number of samples	Predatory fish	Number of samples
1	Lake Łańskie Lake Maróz/Mazury	roach (*Rutilus rutilus*) - 15*, bream (*Abramis brama*) - 6	21	pike (*Esox lucius*) - 21*	21
2	Wkra River/Żuromin	roach (*Rutilus rutilus*) – 36, tench (*Tinca tinca*) - 1	37	pike (*Esox lucius*) - 15, perch (*Perca fluviatilis*) - 5, ide (*Leuciscus idus*) - 1	21
3	Brda River/Czluchów	roach (*Rutilus rutilus*) - 5, bream (*Abramis brama*) - 13, tench (*Tinca tinca*) - 9, ablet (*Alburnus alburnus*) - 2	29	pike (*Esox lucius*) - 7, zander (*Sander lucioperca*) - 10, perch (*Perca fluviatilis*) - 9, catfish (*Silurus glanis*) - 3	29
4	Lakes of Lipczyno Wielkie/Pomerania	roach (*Rutilus rutilus*) - 6, bream (*Abramis brama*) - 6	12	pike (*Esox lucius*) - 5, zander (*Sander lucioperca*) - 8	13
5	Roznowski Reservoir	roach (*Rutilus rutilus*) - 8, bream (*Abramis brama*) - 6	14	not determined	0
6	Rybnik Reservoir	roach (*Rutilus rutilus*) - 2, bream (*Abramis brama*) - 19, white bream (*Blicca bjoerkna*) - 4	25	zander (*Sander lucioperca*) - 10, catfish (*Silurus glanis*) - 5, perch (*Perca fluviatilis*) - 2	17
7	Vistula/Cracow	roach (*Rutilus rutilus*) - 18, bream (*Abramis brama*) - 15, white bream (*Abramis brama*) - 9, barbeł (*Barbus barbus* - 1	43	perch (*Perca fluviatilis*) - 4, chub (*Squalius cephalus*) - 4, catfish (*Silurus glanis*) - 3	11
8	Vistula/Warsaw	roach (*Rutilus rutilus*) - 6, bream (*Abramis brama*) - 16, tench (*Tinca tinca*) - 1	23	pike (*Esox lucius*) - 8, zander (*Sander lucioperca*) - 4, asp (*Leuciscus aspius*) - 2, ide (*Leuciscus idus*) - 1, crucian carp (*Carassius carassius*) - 2, perch (*Perca fluviatilis*) - 2, catfish (*Silurus glanis*) - 1	20
9	Odra/Wroclaw	roach (*Rutilus rutilus*) - 9, bream (*Abramis brama*) - 5, white bream (*Blicca bjoerkna*) - 1	15	burbot (*Lota lota*) - 8, perch (*Perca fluviatilis*) - 1, catfish (*Silurus glanis)* - 1, sumik *(Ameiurus nebulosus*) - 4, ide (*Leuciscus idus*) - 2, catfish (*Silurus glanis*) - 1, asp (*Leuciscus aspius*) -1	18
10	Odra/Warta estuary	roach (*Rutilus rutilus*) - 1, bream (*Abramis brama*) - 14, tench (*Tinca tinca*) - 2, white bream (*Blicca bjoerkna*) - 2	19	pike (*Esox lucius*) - 4, perch (*Perca fluviatilis*) - 2, asp (*Leuciscus aspius*) - 2, ide (*Leuciscus idus*) - 1	9
	Total		238		159

*- number of fish analysed

Concentrations of lead, cadmium, mercury, and arsenic were analysed in 397 fish muscle samples (238 omnivorous and 159 predatory) and 128 sediment samples. Fish and sediment samples were collected by the Veterinary Inspectorate and prepared in accordance with the instructions. After delivery to the laboratory, the fish were weighed, measured, and filleted. Then, the samples of muscles were homogenised and stored in plastic bags at -20°C until analysis. Sediment and water samples were stored at 6°C until analysis.

Determination of lead and cadmium in fish was performed by atomic absorption spectroscopy with graphite furnace atomisation (PinAAcle 900T, Perkin Elmer, USA), after dry digestion in an electric furnace at 450°C. Arsenic was determined by atomic absorption spectrometry using hydride generation techniques after digestion with magnesium nitrate in an electric furnace at 550°C. Mercury analysis was performed by atomic absorption spectrometry using a AMA 254 mercury analyser (Altec, Czech Republic). For the digestion of sediments pressure microwave digestion was used. The procedures for determination of lead, cadmium, arsenic, and mercury in biological materials were previously described in details (26). Statistical calculations were performed using Statistica 10 PL (StatSoft, Poland).

An important part of the study was a special quality assurance programme for the analytical methods used. The National Reference Laboratory of the Department of Pharmacology and Toxicology participates in proficiency tests (PT) organised by the European Union Reference Laboratory (EURL-CEFAO, ISS, Rome, Italy) and also organises PT for regional laboratories dealing with the analysis of metals (20). The methods are also tested in the laboratory by analysing certified reference materials (CRM) for trace elements (Table 2).

Results

The results of analysis of lead, cadmium, mercury, and arsenic in the fish muscles are shown in Table 3 and in Figs. 1 and 2.

Lead. The average content of lead in the muscles of omnivorous fish was 0.026 mg/kg. Twofold higher values were found in fish taken from the Roznowski Reservoir, the Vistula at Cracow and surrounding area, and the confluence of the Odra and Warta Rivers (Fig. 1). The highest concentrations of this element in predatory fish were found in fish caught in the Odra River at the mouth of the Warta (Fig. 2). The average concentrations of lead in sediments ranged from 2.05 to 56 mg/kg dry weight. The content of lead in sediment was high, with the maximum value of 163 mg/kg dry weight. It was found in the sediment from the Vistula River near Warsaw (Table 3).

Cadmium. The average concentration of cadmium (Figs. 1 and 2) in omnivorous and predatory fish was low. It was at the level of 0.003 mg/kg for the most regions. Only in samples collected from the Vistula River near Cracow the cadmium concentration was threefold higher at 0.009 mg/kg, with the maximum value being for the roach at 0.030 mg/kg (Table 3). In the sediments, concentrations of cadmium in secondary sludge ranged from 0.023 to 6.141 mg/kg. The highest concentration of this element, as it also was in the case of lead, was found in the sediment from the Vistula near Cracow, where the maximum value reached 21.24 mg/kg dry weight (Table 3).

Mercury. The average concentration of Hg in the omnivorous fish muscles was 0.103 mg/kg, while the highest value was found in samples taken from the Roznowski Reservoir, where the average concentration of this element reached 0.241 mg/kg (Fig. 1). In fish examined from other bodies of water, the average mercury concentration in muscle was at the low-tenths and hundredths of a mg/kg level (Figs. 1 and 2). The average concentration of mercury in the muscle of predatory fish was almost twofold higher than that found in omnivorous fish and was 0.179 mg/kg. The lowest level of this element was found in predatory fish caught in the Rybnik Reservoir (Fig. 2). Table 3 summarises the results of the determination of mercury in the muscles of omnivorous and predatory fish and in bottom sediments. Noteworthy is the high concentration of mercury found in the muscles of omnivorous fish collected from the Vistula near Warsaw and the Odra near Wroclaw, where the average values of Hg were 0.186 and 0.121 mg/kg respectively for roach and 0.118 and 0.181 mg/kg for bream, and were significantly higher than those found in fish of this species from the other examined bodies of water. In the analysed muscles of predatory fish (pike, perch, and zander), the highest average concentration of this element was found in pike (0.292 mg/kg) and the maximum value in the sample from the Vistula River from Warsaw area was nearly double at 0.400 mg/kg (Table 3).

Arsenic. The average and maximum values of arsenic (Fig. 1, Table 3) indicate that a high concentration of this element was in the muscles of omnivorous fish. The highest concentration of this element was found in fish caught in the Odra River near Wroclaw. Arsenic concentrations found in fish from this region were much higher compared with regions outside large urban agglomerations. In sediment the mean concentrations of arsenic ranged from 0.299 to 6.098 mg/kg dry weight (Table 3), while in water the concentration of the elements was low and below the detection limit of the method used (Pb 0.002 mg/kg, Cd 0.001 mg/kg, Hg 0.001 mg/kg, and As 0.002 mg/kg).

Table 2. Concentrations of elements found in proficiency tests and certified reference materials

Material	Pb	Cd	Hg	As
Frozen fish 14th PT, EURL				
Assigned values (mg/kg)	0.370	0.075	0.222	0.926
Measured value (mg/kg)	0.385	0.082	0.211	0.922
Recovery (%)	104	109	95	99.6
Z-scores	-0.1	0.8	-0.5	0.0
Frozen meat 18th PT, EURL				
Assigned values (mg/kg)	0.051	0.015	0.0097	-
Measured value (mg/kg)	0.049	0.015	0.0100	nd
Recovery (%)	96	100	103	-
Z-scores	-0.2	0.0	0.3	-
Frozen fish PIWet-PT2012/ZFT/32				
Assigned values	0.009	0.006	0.116	0.054
Measured value (mg/kg)	0.008	0.006	0.116	0.055
Recovery (%)	89	100	100	102
Z-scores	-0.37	0.0	0.0	0.07
DORM-3				
Certified value (mg/kg)	0.395	0.290	0.382	6.88
Measured value (mg/kg)	0.367	0.276	0.368	6.29
Recovery (%)	93	95	96	91

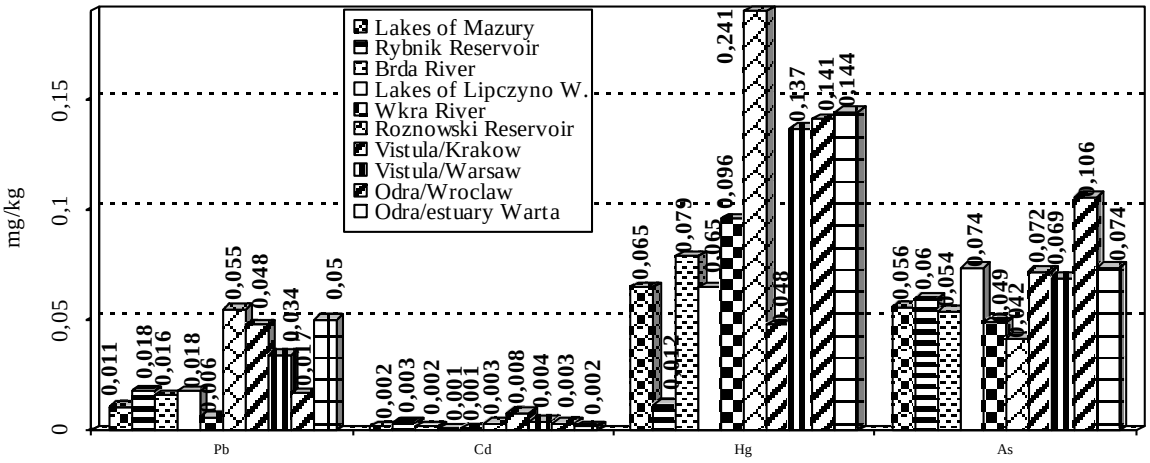


Fig. 1. Mean concentrations of toxic elements in muscles of omnivorous fish

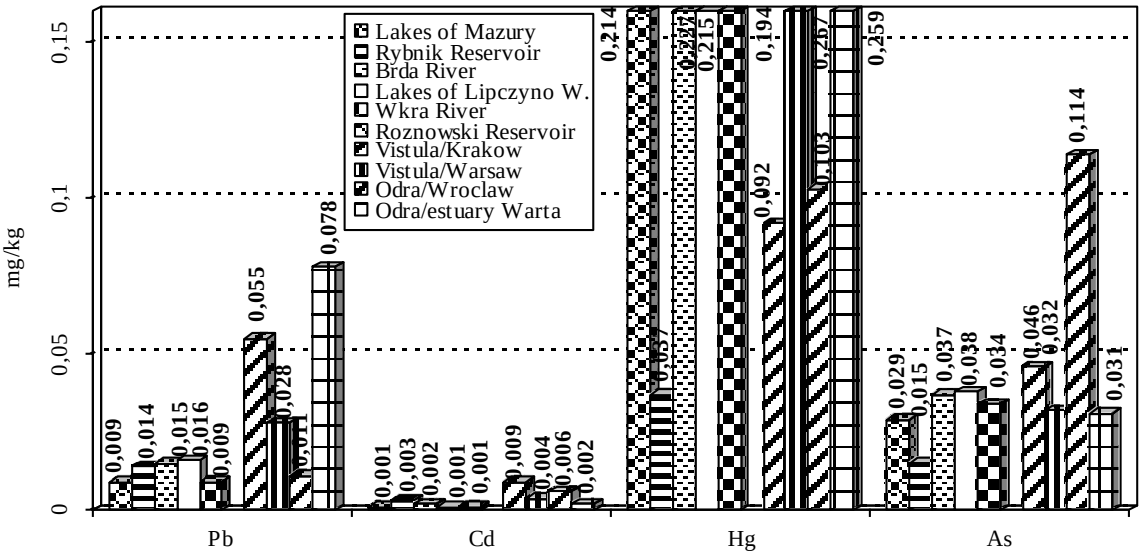


Fig. 2. Mean concentrations of toxic elements in muscles of predatory fish

Table 3. The concentrations of toxic elements in the muscles of selected species of freshwater fish (mg/kg wet weight) and sediments (mg/kg dry weight)

Sampling site	Elements	roach		bream		pike		perch		zander		sediments	
		mean	max	mean	max	mean	max	mean	max	mean	max	mean	max
L. Łańskie L. Maróz/ Mazury	n	15		6		21						5	
	Pb	0.010	0.024	0.014	0.020	0.009	0.049					14.318	25.11
	Cd	0.003	0.006	0.002	0.008	0.001	0.002	nd	nd	nd	nd	0.392	0.721
	Hg	0.081	0.120	0.035	0.120	0.227	0.360					0.037	0.062
	As	0.069	0.100	0.022	0.029	0.029	0.082					3.778	4.134
Wkra River Żuromin	n	36				15		5				10	
	Pb	0.006	0.011			0.010	0.032	0.008	0.012			2.049	2.910
	Cd	0.001	0.005	nd	nd	0.001	0.002	0.002	0.004	nd	nd	0.159	0.451
	Hg	0.096	0.226			0.198	0.350	0.187	0.276			0.003	0.004
	As	0.049	0.111			0.039	0.144	0.020	0.023			0.883	1.546
Brda River Czluchów	n	8		19		5		7		18		15	
	Pb	0.013	0.018	0.017	0.055	0.013	0.019	0.015	0.031	0.016	0.029	2.246	6.288
	Cd	0.002	0.003	0.001	0.002	0.001	0.001	0.001	0.006	0.002	0.007	0.023	0.042
	Hg	0.087	0.181	0.086	0.391	0.247	0.385	0.183	0.291	0.260	0.456	0.002	0.004
	As	0.038	0.073	0.023	0.049	0.021	0.030	0.025	0.039	0.036	0.048	0.299	0.400
L. Lipczyno Wielkie	n	6		6		5				8		10	
	Pb	0.012	0.013	0.027	0.055	0.013	0.019			0.019	0.029	1.343	2.020
	Cd	0.001	0.002	0.001	0.,002	0.001	0.001	nd	nd	0.001	0.001	0.018	0.030
	Hg	0.092	0.181	0.048	0.065	0.247	0.315			0.257	0.322	0.002	0.004
	As	0.027	0.031	0.019	0.032	0.021	0.030			0.031	0.044	0.140	0.275
Roznowski Reservoir	n			19						10		5	
	Pb			0.018	0.047					0.015	0.039	4.555	16.86
	Cd	nd	nd	0.002	0.005	nd	nd	nd	nd	0.003	0.004	0.343	0.419
	Hg			0.012	0.023					0.047	0.068	0.066	0.081
	As			0.050	0.074					0.015	0.022	6.098	6.948
Rybnik Reservoir	n			19						15		15	
	Pb			0.018	0.047					0.014	0.039	2.586	5.821
	Cd	nd	nd	0.002	0.005	nd	nd	nd	nd	0.003	0.012	0.139	0.234
	Hg			0.012	0.023					0.037	0.068	0.005	0.009
	As			0.050	0.074					0.015	0.022	0.481	0.743
Vistula/ Cracow	n	18		15				4				35	
	Pb	0.049	0.122	0.036	0.072			0.068	0.095			55.99	134.1
	Cd	0.011	0.030	0.006	0.010	nd	nd	0.006	0.012	nd	nd	6.141	21.237
	Hg	0.047	0.101	0.050	0.102			0.025	0.036			0.559	2.779
	As	0.104	0.382	0.046	0.061			0.034	0.063			4.601	14.46
Vistula/ Warsaw	n	6		16		8				4		21	
	Pb	0.034	0.050	0.035	0.085	0.033	0.055			0.026	0.036	24.65	163.1
	Cd	0.004	0.005	0.005	0.009	0.008	0.015	nd	nd	0.001	0.002	0.312	0.921
	Hg	0.186	0.239	0.118	0.226	0.292	0.400			0.196	0.207	0.031	0.151
	As	0.134	0.222	0.046	0.065	0.031	0.036			0.017	0.024	2.035	10.52
Odra/ Wroclaw	n	9		5						8		20	
	Pb	0.010	0.015	0.030	0.111					0.007	0.011	7.653	20.36
	Cd	0.003	0.006	0.004	0.005	nd	nd	nd	nd	0.001	0.002	0.342	1.052
	Hg	0.121	0.242	0.181	0.249					0.137	0.241	0.039	0.124
	As	0.117	0.205	0.088	0.129					0.169	0.380	3.868	10.08
Odra/ Warta estuary	n			14		4						2	
	Pb			0.050	0.115	0.036	0.055					7.895	8.427
	Cd	nd	nd	0.002	0.006	0.002	0.003	nd	nd	nd	nd	0.684	0.709
	Hg			0.133	0.309	0.191	0.233					0.033	0.038
	As			0.080	0.296	0.029	0.032					1.066	1.484

nd - not determined

Discussion

Acceptable levels of lead, cadmium, and mercury in freshwater fish are set out in European Commission Regulation (EC) No. 1881/2006, and are as follows: Pb 0.30 mg/kg, Cd 0.05 mg/kg, and Hg 0.50 mg/kg (6). For arsenic however, at European Union maximum permissible level has not been established. It is still under discussion and it is expected that the need will emerge for the dual determination of the presence of this element, both in total and separately in organic and inorganic forms. In view of the high toxicity of arsenic, many EU Member States have set national action levels for this element in fish. In Poland, there are action levels set out in the "National Residue Control Plan" for the interpretation of test results of arsenic in foods of animal origin, and the Plan is annually approved by the Chief Veterinary Officer and European Commission (15). A maximum permissible total arsenic content in the muscles of fish of 4 mg/kg has been adopted in Poland.

The present study demonstrated that the average concentrations of lead, cadmium, and arsenic in fish were in the low hundredths and thousandths of a mg/kg and did not exceed permitted limits in any samples (3, 19). Similar values of lead and cadmium in muscles of

river freshwater fish were detected by other authors (7, 12, 18, 23). By contrast, higher concentrations of lead, often exceeding 1 mg/kg, were found in lake fish (5, 14).

In this study, higher values of these elements were found in fish from bodies of water located in the zone of influence of large urban agglomerations. Concentrations of these elements detected in fish from the Vistula near Cracow, where the observed values were higher compared to other regions, are particularly important.

The average concentration of mercury in the muscles of predatory fish (0.179 mg/kg) was significantly higher than that found in omnivorous fish (0.103 mg/kg). The lowest level of this element was found in predatory fish caught in Rybnik Reservoir (Fig. 2). In individual fish samples (asp 0.533 mg/kg, and ide 0.685 mg/kg), Hg concentrations exceeded the acceptable level, *i.e*. 0.5 mg/kg.

In fish breeding, mercury was found at the level of 0.030 mg/kg (25, 26, 27). In studies of different species of fish caught in the river Vistula, mercury was found in similar concentrations (24). In the Czech Republic, this element was present in fish in concentrations ranging from 0.05 to 0.32 mg/kg (10).

The arsenic concentration in the muscles of freshwater fish-determined in this study seems not to be a major problem, regardless of its place of origin. Both the average and maximum arsenic contents in fish in all the regions studied were low and did not exceed the approved limit, *i.e*. 4 mg/kg (15). Higher levels of this element were found in omnivorous fish, particularly in roach, where the average for this element (0.085 mg/kg) was threefold higher than that found in perch (0.026 mg/kg). In the study conducted in France, arsenic in freshwater fish was found in concentrations ranging from 0.026 to 0.647 mg/kg (8), and in studies conducted in Italy, the results ranged from 0.008 to 0.010 mg/kg (23).

Higher values of the analysed elements were found in the sediments, which were collected from the surface layer to a depth of 20 cm. Evaluated against the criteria for bottom sediments, the level of lead in several samples exceeded the geochemical background level, which is 15 mg/kg dry weight. For other elements studied, *i.e.* cadmium, mercury, and arsenic, geochemical background values are as follows: Cd 0.5 mg/kg dry weight, Hg<0.05 mg/kg dry weight, and As<5 mg/kg dry weight. (2, 11). The highest concentrations of lead, cadmium, and mercury were found in sediments collected from the Vistula near Cracow. In all tested samples from this region the concentrations of these elements exceeded the geochemical background value. In addition, a large number of samples in this region exceeded the probable effects levels (PEL), which specify the concentration of contaminating elements in sediment, over which harmful effects on aquatic organisms are often observed (2, 19). The observed concentrations of mercury in sediments in this region (mean 0.559 and maximum 2.779 mg/kg dry matter) had no great effect on the bioavailability of mercury in fish, because its concentration in the examined fish (Table 3) was significantly lower compared with other regions and lower than the allowable limit (6). In the sediments from other rivers and lakes in Poland geochemical background values were exceeded only in a small percentage of samples.

The PEL was exceeded only for lead (91 mg/kg dry matter) in three sediment samples collected from the Vistula River in Warsaw. The studies conducted by other authors in Poland (9, 21, 28), obtained values exceeding the geochemical background and PEL. In river sediments, the mean values of arsenic were lower than the permissible background (5 mg/kg), but the maximum value of arsenic (14.46 mg/kg) in the sediment of the Vistula near Cracow indicates a higher arsenic contamination in comparison with other regions. Other authors found that arsenic concentration in sediments of the Odra ranged from 6.39 to 192 mg/kg (9).

The study indicated that the concentrations of toxic elements in free-living freshwater fish are rather low and safe. The mean values of lead and cadmium in omnivorous and predatory fish did not differ significantly among them. However, in the case of mercury, the average concentration of this element in predatory fish muscles was almost twofold higher in comparison with omnivorous fish. The studies have also shown that only a single fish sample exceeded the maximum limit for this metal (0.50 mg/kg) (6). In studies on the arsenic concentration, higher values of this element were found in the muscles of omnivorous fish. The highest value of this element was found in fish caught in the Odra and Warta River estuary; however, they do not pose a risk to consumers' health.

High concentrations of lead, cadmium, mercury, and arsenic were found in sediment samples collected from the Vistula near Cracow, where a large number of samples exceeded the value of geochemical background and PEL for these elements.

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