ARCHITECTURE CIVIL ENGINEERING ENVIRONMENT
The Silesian University of Technology
No. 1/2025

doi: 10.2478/ACEE-2025-0014

METAL(LOID)S IN BOTTOM SEDIMENTS ECOLOGICAL AND HUMAN HEALTH RISK ASSESSMENT

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Received: 22.07.2024; Revised: 3.01.2025; Accepted: 9.01.2025

Abstract
The research aimed to assess the ecological and health risks of selected metal(loid)s (Cd, Cr, Cu, Ni, Pb, Zn, and As) in the bottom sediments of four water reservoirs located in various regions of Poland (Central Europe). The results of the ecological assessment indicate that Cd is the primary element posing a risk among analyzed metal(loid)s. Health risks assessment revealed that the analyzed elements enter the human body mainly through ingestion. Moreover, both from a non-carcinogenic and carcinogenic perspective, children are more exposed to the negative impact of these elements. Although the obtained result indicates that carcinogenic risk is acceptable, appropriate actions should still be taken to prevent the possible negative effects of metal(loid)s in the analyzed bottom sediments, particularly regarding Cd, Cr, Ni, and As.

Keywords: **Bottom sediments, Water reservoirs, Metal(loid)s, Ecological Risk Assessment, Health Risk Assessment.**

1. INTRODUCTION

Bottom sediments play a crucial role in the ecological balance of aquatic ecosystems, serving as habitats for various organisms and acting as sinks for contaminants, including metals and metalloids (metal(loid)s) [1, 2]. The sources of metal(loid)s in aquatic ecosystems can be categorized into natural and anthropogenic origins. In the first group, we can distinguish rock weathering, erosion, and atmospheric deposition, while in the second one, municipal and industrial wastewaters, surface runoff from agricultural fields and roads, mining activity, and tourism [3,4]. Metals and metalloids pose significant ecological and health risks due to their non-biodegradability, toxicity, long-term persistence, bioaccumulation, and biomagnification. These properties allow them to exert dangerous consequences even in areas located a long distance from the source of pollution [3, 5].

Nowadays, studies on bottom sediments focus on their contamination with metal(loid)s and the ecological risks these elements pose [1, 2, 6]. Increasingly, research also considers potential hazards to human health [4, 5, 7]. The accumulation of metal(loid)s in bottom sediments profoundly affects benthic organisms and other living organisms through the food chain, including humans [1]. One of the main routes of metals and metalloids penetration into the human body is ingestion. In the food chain, each preceding group provides a food source for the next one. Therefore, metal(loid)s are first taken with food by benthic organisms living at the bottom of the water reservoir. Benthos, in turn, provides fish food, which other animals and humans then eat. Another way elements penetrate the human body is the accidental oral ingestion of bottom sediments. This way is often observed among children, e.g., while playing, especially in the summer. The commonly known path of penetrating these pollutants into the human body is also direct contact with the sediments on the skin (dermal route), which is inevitable when practicing water sports or swimming in a water reservoir. The risk may increase when the skin is damaged [4,8]. Excessive concentrations of metal(loid)s in living organisms may lead to various adverse effects, from allergic and skin diseases to carcinogenesis, cell mutations, immunological defects, morphological abnormalities, and disorders in enzymatic and hormonal activities [9,10]. Long-term exposure to these elements can also lead to organ damage (kidney, respiratory tract, and cardiovascular system damage) and neurological disorders (cognitive impairments, peripheral neuropathy, depression, insomnia) [10].

The chemical composition of bottom sediments is often a direct indicator of anthropogenic impact on the environment [7]. Knowledge about the concentrations of metal(loid)s in bottom sediments is essential and helps to identify and mitigate potential ecological and human health risks, ensuring the safety and sustainability of aquatic ecosystems. Evaluating the risks associated with metal(loid)s in bottom sediments can lead to the development of practical pollution control, remediation, and environmental conservation strategies [8, 11].

The main aim of this study was to assess the potential risk associated with the presence of selected metal(loid)s in the bottom sediments of various water reservoirs (Poland, Central Europe), both in ecological and health dimensions. The primary criterion for selecting water reservoirs was their function, such as recreation, tourism, and water supply, which is essential in the environmental and health context. The considered reservoirs are subject to monitoring by the Chief Inspectorate for Environmental Protection (in Polish: Główny Inspektorat Ochrony Środowiska; GIOŚ). The indices (methods) used to assess ecological and health risks were developed by Müller (1969) [12] and Hakanson (1980) [13] and the United States Environmental Protection Agency (US EPA; 1989, 2001, 2002) [14-16]. These indices constitute a simple and effective tool for assessing the potential ecological and health risks associated with metals and metalloids in the bottom sediments of water reservoirs. So far, few works present a comprehensive approach to risk assessment, both in the ecological and health dimensions, in the context of aquatic ecosystems [8, 17], which constitutes a gap in the existing state of knowledge.

In this paper, we (1) investigate the total concentrations of selected metal(loid)s, i.e., cadmium (Cd), chrome (Cr), copper (Cu), nickel (Ni), lead (Pb), zinc (Zn), and arsenic (As) in bottom sediments from selected water reservoirs (lakes and dam reservoir) in Poland, and (2) assess the ecological and health risks associated with the presence of these selected metal(loid)s in the bottom sediments.

2. METHODS

2.1. Data source

The concentrations of metal(loid)s (Cd, Cr, Cu, Ni, Pb, Zn, and As) in bottom sediments from selected water reservoirs discussed in this article were sourced from a report published on the website of the Chief Inspectorate for Environmental Protection [18]. The report concerns monitoring river and lake bottom sediments in 2020–2021. Based on this data, we assessed the potential ecological and human health risks posed by selected metal(loid) in bottom sediments, which was the main aim of this study.

2.2. Study area

The water reservoir selection was based mainly on the functions they performed. Because our work is concerned with assessing potential ecological and health risks, we decided these would be recreational and tourist functions and water supply. Recreation and tourism are usually related to water sports, swimming, or fishing. However, sometimes they can be limited to, for example, walking or cycling, etc., around the reservoir (but despite this, people still break the prohibition). It happens when a water reservoir serves more specific functions, like water supply or flood control. In such a case, monitoring the characteristics and composition of bottom sediments is essential. This is because sediments in water ecosystems are a sink and a source of potentially toxic pollutants, like metal(loid)s [19, 20]. These pollutants can be released from sediments into the water under reducing conditions, such as a rapid decrease of pH, redox potential value (Eh), or oxygen content in water. In such a situation, secondary aquatic environment contamination with these pollutants may occur [20].

The study area encompasses three lakes and one dam reservoir, specifically:

A) Lake Gardno: located in the Pomeranian

Voivodeship (Northern Poland), on the coast of the South Baltic Sea, within the Słowiński National Park and the boundaries of the Natura 2000 area (54°39′14″N 17°06′43″E).

A distinguishing feature of the Lake Gardno catchment is its coastal location, lack of overland flow, and a large share of sea aerosols in the water cycle [21]. The frontal moraine hills surrounding Lake Gardno, from the west to the south, create a post-glacial landscape. Inside the moraine embankment on the lake's southern shore, a large area is covered by marsh and peat formations. The eastern coast of the lake is formed by the delta of the Łupawa River, and the northern shore is formed by the Gardzieńska Spit [22]. The structure of surface used in the Gardno Lake catchment area is characterized by a dominant share of forests (98.2%), a small share of surface waters (1.2%), and anthropogenic areas, i.e., roads and buildings (0.6%). The lake's catchment area is subject to low anthropogenic pressure, and the only local source of pollution is roads. During the colder period of the year, the environment may also be affected by pollution from the burning of fossil fuels in nearby towns [23]. Moreover, thanks to the mouth section of the River Łupawa, which connects Lake Gardno with the Baltic Sea, also sea waters may source various pollutants in this lake [21].

B) Lake Swarzędzkie: located in the Greater Poland Voivodeship (Western Poland), in the north-western part of the Swarzędz town (52°24′49″N 17°03′54″E). The lake is within the Natura 2000 area (Cybina Valley) of the Wrzesińska Plain.

Lake Swarzędzkie is surrounded by peat deposits, the largest of which is located at the Cybina tributary. To the south and southwest of the lake, there are sands, gravels, and kame silts. The areas east and west of the lake are covered with fluvioglacial sands and gravels from the Poznań phase of glaciation and older boulder clays and uncovered boulder clays from the Leszno phase of glaciation. The soil in question is of medium and poor quality. In the valuation structure of the discussed area, 75% are class III and IV soils [24]. The direct basin of Lake Swarzędzkie is covered by forest (45%), meadows and pastures (31.8%), built-up areas (21.7%), and farmland (1.3%) [25]. The waters feeding the Lake Swarzedzkie are significantly polluted with biogenic substances [26]. Moreover, until 1991, wastewater from Swarzędz was directly discharged into Lake Swarzędzkie [27].

C) Lake Szczutowskie: located in the Masovian Voivodeship (Eastern Poland), in the Szczutowo commune (52°56′30″N 19°34′45″E).

The catchment area of Lake Szczutowskie is located in the eastern part of the Chełmińsko-Dobrzyńskie Lakeland macroregion in the Urszulewska Plain mesoregion [28]. The landscape of this region is monotonous and plain. It comprises Pleistocene outwash sands and gravels of the North Polish Glaciation, with numerous areas of Holocene sands, gravels, river muds, peats, and silts. There are also boulder clays, sands, and glacial gravels. It is characterized by fertile soil [29]. Based on the analysis of GIOŚ INSPIRE maps [30], it can be concluded that agricultural land use prevails in the Lake Szczutowskie catchment area. Forest areas also have a significant share in the land development structure. Residential buildings dominate on the lake's western shore; on the eastern coast, the number of buildings is much smaller.

D) Wisła-Czarne dam Water Reservoir: located in the Silesian Voivodeship (Southern Poland), in the Wisła town (49°36′53″N 18°55′36″E). We considered an artificial dam reservoir instead of a natural lake in this region because the Silesian Voivodeship is more notable for its artificial reservoirs and mining subsidence lakes, which are critical in water supply, flood control, and recreation. Natural lakes are generally smaller and less well-known in this region [20].

The Wsisła-Czarny dam Reservoir was created below the junction of the Biała and Czarna Wisełka streams strands [31]. The reservoir's catchment area is located in the Beskid Śląski mesoregion, which belongs to the flysch Western Carpathians [28]. The city of Wisła is agricultural; as much as 92% of the commune's area is agricultural land, forest land, or wasteland. Urbanized areas constitute approximately 6% of the commune's area, and surface waters constitute approximately 2%. The areas along the banks of the Wisła-Czarne Reservoir are covered with forests. Near the reservoir, there are also numerous meadows and pastures with scattered single-family buildings and local roads [32]. Given the supply streams' good quality, pollutants will likely enter the Wisła-Czarne Reservoir through dry and wet deposition or surface runoff. Moreover, in regions with a history of industrial development, like the Silesian Voivodeship, old contaminants may still be present in the sediments even if these activities have been reduced or ceased [20].

Figure 1 shows the locations of water reservoirs. The outlines of the lakes and one reservoir are from the web maps with ArcGIS Online, Esri's web-based mapping software [33], while the map of Europe

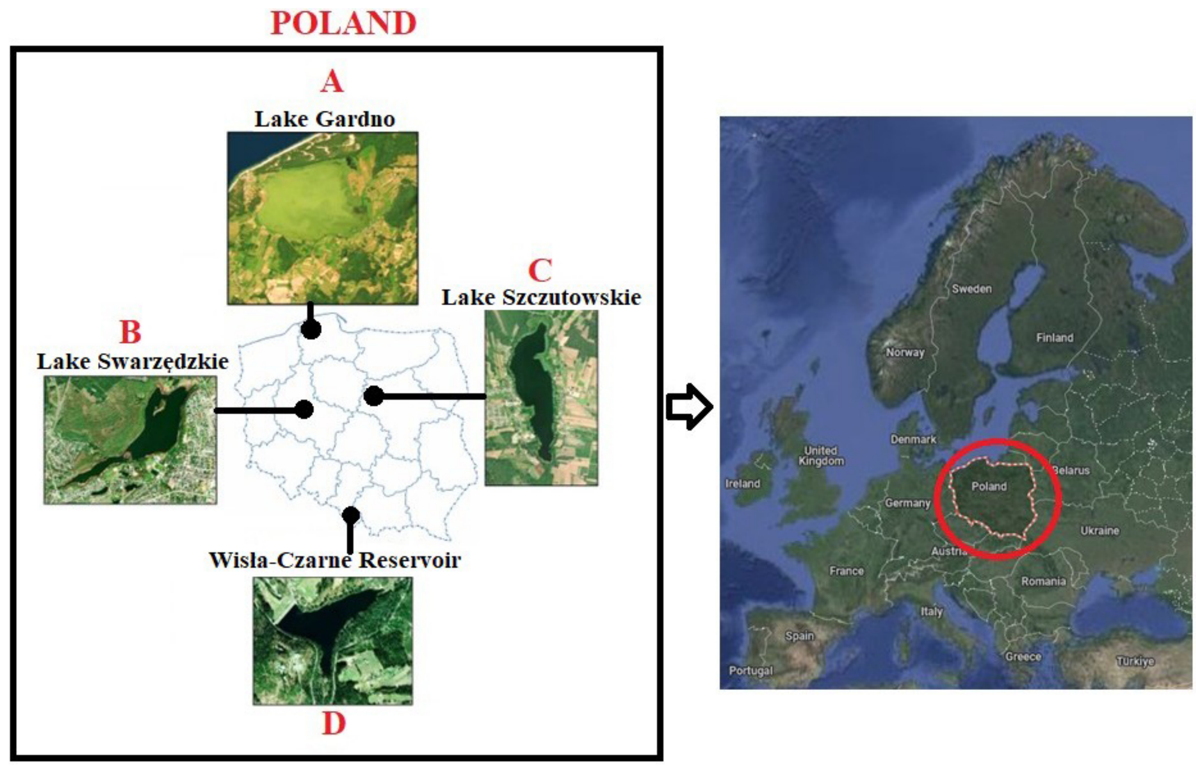

Figure 1.
Location of water reservoirs [33, 34]. Where A) Lake Gardno; B) Lake Swarzędzkie; C) Lake Szczutowskie; D) Wisła-Czarne dam Reservoir

marked Poland is from Google Maps [34]. The morphometric and hydrological parameters of considered water reservoirs are shown in Table 1.

2.3. Sampling

According to the report presented by the Chief Inspectorate for Environmental Protection [18], field research and sediment collection were conducted on PN-ISO 4364:2005 [38]. The number of samples collected depends on the reservoir area. In this study, the number of collected samples was from 1 to 3 per water reservoir. The location of the depths (at which samples were collected) was determined by GIOŚ based on available bathymetric data of the lakes, i.e., determined based on the Hydrographic Map of Poland at a scale of 1:50.000. A stainless Van Veen

Table 1.
Characteristics of water reservoirs [25, 31, 35-37]

Data	Lake Gardno	Lake Swarzędzkie	Lake Szczutowskie	Wisła-Czarne Reservoir
Type of reservoir	polygenetic (coastal)	post-glacial	glacial	dam reservoir (built-in years 1967 - 1973)
Surface area (km^2)	24.7	0.94	0.91	0.40
Length - max (m)	6850	2940	2110	-
Width - max (m)	4730	560	690	-
Depth - max (m)	2.6	7.2	4.4	-
Volume (thousand m^3)	30950.5	2000	1689	5060
Feeder waters	Łupawa, Bagienica Brodna, and Grabownica Rivers	Cybina River and Mielcuch stream	5 drainage ditches; unnamed watercourses	Biała and Czarna Wisełka streams
Functions	tourism and recreation	tourism and recreation	tourism and recreation	water supply, flood control, tourism, and recreation

grab sampler obtained a 5-cm surface layer of bottom sediments. In the case of sampling from several depths, the material collected was averaged by transfer to 1 container and mix. The sediment samples were sieved through a nylon sieve with a mesh of 2 mm, placed in plastic containers, and adequately secured for transport by placing them in a closed cool-box container lined with bubble foil and ice inserts. This procedure was intended to protect the samples from damage and heating. The samples were delivered within approximately 24 hours of collection to the laboratory that performed the analyses [18].

2.4. Determination of metal(loid)s

Determining metal(loid)s in sediment samples was performed by accredited research units commissioned by the Chief Inspectorate for Environmental Protection. Bottom sediment analyses were conducted under the requirements of PN EN ISO/IEC 17025:2018-02 [39]. Sediment samples were dried to constant weight (at 60ºC), ground in a Retsch RM 200 grinder, and subjected to mineralization with aqua regia following the PN-EN 13657:2006 [40]. Metal(loid)s determinations were carried out using inductively coupled plasma atomic emission spectrometry ICP-OES on an Optima 5300 DV Perkin Elmer, according to PN-EN ISO 11885:2009 [41]. To convert the analysis results from $mg \cdot L^{-1}$ into $mg \cdot kg^{-1}$, the sediments were dried (in a separate procedure) at 105 ± 5°C to constant weight by PN-EN 12880:2004 [42]. Sediments were weighed using an analytical balance, Radwag.

2.5. Assessment of potential ecological risk

Various indices (methods) can be utilized to evaluate the potential adverse effects of metal(loid)s on the aquatic environment. In this paper, we used indices referring to one element or group of elements, so-called "single-element indices" and "multi-element indices". Among the first group, we can distinguish the Geoaccumulation Index (I_{geo}) [12] and the Potential Ecological Risk Factor (ER) [13], while in the second, the Risk Index (RI) [13]. The I_{geo} index allows contamination assessment by comparing the current and pre-industrial concentrations of the elements in the relevant geochemical background [19]. In turn, the ER index provides a comprehensive method for assessing ecological risks posed by elements in aquatic environments by incorporating concentration, background levels, and toxicity. Meanwhile, RI refers to the sum of the ER of each considered element [43]. The indices mentioned above are recommended for determining the contaminating effect of metal(loid)s in sediments and the potential ecological risk associated with their toxicity [13]. The methodologies and formulas for these indices are shown in Table 2. Moreover, background values are essential for evaluating contamination levels and potential ecological risks [20]. In this study, the content of the metal(loid)s in the Earth's crust was used as background values, as reported by Kabata-Pendias (2011) [44].

Table 2.
Ecological risk indices

Index	Formula	Scale	Description
Geoaccumulation Index (Igeo) [12]	$I_{geo}=log_2\left(\frac{C_n}{1.5B_n}\right)$ C_n – concentration of element in the sediment sample ($mg \cdot kg^{-1}$), B_n – geochemical background value in the Earth's crust (Cd=0.1; Cr=100; Cu=55; Ni= 20; Pb=15; Zn=70; As=1.8 $mg \cdot kg^{-1}$) [44]	$I_{geo} \leq 0$ $0<I_{geo} \leq 1$ $1<I_{geo} \leq 2$ $2<I_{geo} \leq 3$ $3<I_{geo} \leq 4$ $4<I_{geo} \leq 5$ $I_{geo} >5$	Practically uncontaminated Uncontaminated to moderately contam. Moderately contaminated Moderately to Heavily contam. Heavily contaminated Heavily to Extremely contaminated Extremely contaminated
Potential Ecological Risk Factor (ER) [13]	$ER = T_f^i \cdot CF$ T – toxic response factor of element (Cd=30; Cr=2; Cu, Ni, Pb=5; Zn=1; As=10); CF – Contamination Factor; the ratio between the concentration of the element in sediment and the background value [13, 44]	ER≤40 40<ER≤80 80<ER≤160 160<ER≤320 ER>320	Low risk Moderate risk Considerable risk High risk Very high risk
Risk Index (RI) [13]	$RI= \sum ERi$	RI<150 150<RI<300 300<RI<600 RI>600	Low risk Moderate risk Considerable risk High risk

Table 3.
Methodology for estimation of average daily dose

CALCULATION OF AVERAGE DAILY DOSE FOR METAL(LOID)S VIA INGESTION AND DERMAL CONTACT ($mg \cdot kg^{-1} \cdot day^{-1}$)			
Average daily dose for ingestion (ADD_{ing}) [14]	$ADD_{ing} = \frac{C \times IR_{ing} \times EF \times ED}{BW \times AT} \times CF$		
Average daily dose for dermal contact (ADD_{dermal}) [14]	$ADD_{dermal} = \frac{C \times SA \times AF \times ABS \times EF \times ED}{BW \times AT} \times CF$		
C – element concentration in the sediment sample; IR_{ing} – ingestion rate; EF – exposure frequency; ED – exposure duration; AF – adherence factor; ABS – dermal absorption factor; SA – surface area; BW – body weight; AT – averaging time; CF – conversion factor			
THE PARAMETERS APPLIED TO THE ESTIMATION OF ADD_S			
Parameter	Unit	Parameter value Adults/Children	References
C	$mg \cdot kg^{-1}$	-	This study
IR_{ing}	$mg \cdot day^{-1}$	100 / 200	[16]
EF	$day \cdot year^{-1}$	350	
ED	year	30 / 6	
AF(soil)	$mg \cdot cm^{-2}$	0.07 / 0.2	
ABS	-	0.001	
SA	cm^2	5700 / 2800	
BW	kg	70 / 16	[14]
AT	days	70×365 / carcinogens ED×365 / non-carcinogens	
CF	$kg \cdot mg^{-1}$	10^{-6}	

Table 4.
Methodology for health risk assessment

NON-CARCINOGENIC RISK ASSESSMENT	
Hazard Quotient (HQ) [14, 15]	$HQ_{ij} = \frac{ADD_{ij}}{RfD_{ij}}$
Hazard Index (HI) [14, 15]	$HI = \sum HQ_{ij}$
RfD –reference dose of individual element **If the HQ or HI value > 1 – non-carcinogenic risk is possible; if the HQ or HI value < 1 – non-carcinogenic risk is not possible** [14, 15]	
CARCINOGENIC RISK ASSESSMENT	
Carcinogenic Risk (CR) [14, 15]	$CR_{ij} = ADD_{ij} \times SF$
Total Carcinogenic Risk (TCR) [14, 15]	$TCR = \sum CR_{ij}$
SF – cancer slope factor **If the CR or TCR values > 1×10^{-4} – the carcinogenic risk occurs; if the values are between 1×10^{-6} and 1×10^{-4} – the carcinogenic risk is acceptable; if the values are < 1×10^{-6} – the risk is not carcinogenic** [14, 15]	

THE RFD AND SF VALUES OF ELEMENTS					
Element	**RfD_{ing}**	**RfD_{dermal}**	**SF_{ing}**	**SF_{dermal}**	**References**
	$mg \cdot kg^{-1} \cdot day^{-1}$		$kg \cdot day \cdot mg^{-1}$		
Cd	-	-	6.10	6.10	[45-47]
Cr	-	-	5.00×10^{-1}	20.0	
Cu	4.00×10^{-2}	1.20×10^{-2}	-	-	
Ni	-	-	1.70	42.50	
Pb	-	-	8.50×10^{-3}	nd	
Zn	3.00×10^{-1}	6.00×10^{-2}	-	-	
As	-	-	1.5	3.66	

nd – no data

2.6. Assessment of potential health risk

Bottom sediments can accumulate metal(loid)s over time, posing a potential risk to adults and children through various exposure pathways. In this study, the two most probable routes of exposure were considered: accidental oral ingestion and dermal (skin) contact [14]. Table 3 presents the methodology for estimating the average daily exposure doses (ADD_S) for ingestion and dermal contact.

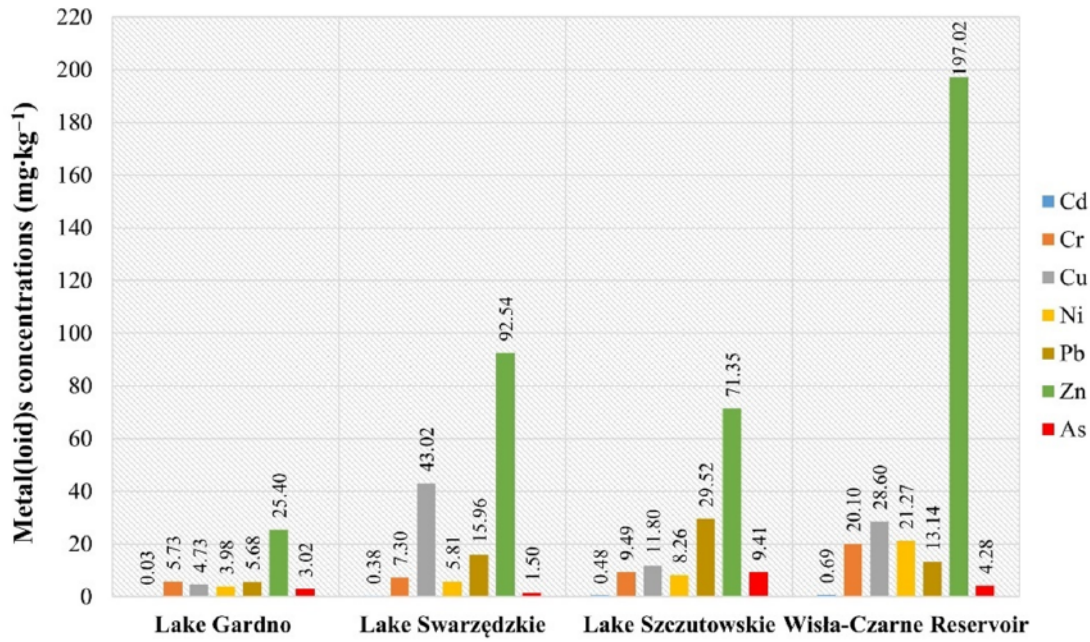

Figure 2.
Metal(loid)s concentrations in bottom sediments

The International Agency for Research on Cancer (IARC) (2012) [45] classifies cadmium, chromium, nickel, and arsenic as Group 1 carcinogens, while lead as Group 2A carcinogens. This indicates their significant risk to human health, particularly in terms of cancer, but not only [9, 43]. Given the above, this paper assessed the health risks associated with cadmium, chromium, nickel, lead, and arsenic in carcinogenic terms, in turn, copper and zinc were considered from a non-carcinogenic perspective. Such an approach provides a balanced view of the potential health risks. Table 4 shows the methods for health risk assessment.

3. RESULT AND DISCUSSION

3.1. Total metal(loid)s concentrations

The mean concentrations of considered metal(loid)s in bottom sediments show a clear pattern where Zn is consistently found in the highest concentration and Cd in the lowest across different water reservoirs. The concentration levels in considered sediments were as follows:

Zn>Cr>Pb>Cu>Ni>As>Cd (Lake Gardno), Zn>Cu>Pb>Cr>Ni>As>Cd (Lake Swarzędzkie), Zn>Pb>Cu>Cr>As>Ni>Cd (Lake Szczutowskie) and Zn>Cu>Cr>Ni>Pb>As>Cd (Wisła-Czarne dam Reservoir). Considering the characteristics of their catchment area, the probable sources of metals and one metalloid in the discussed water reservoirs include municipal wastewater, runoff from agricultural areas, atmospheric deposition, and historical pollution. The concentrations of metal(loid)s in considered bottom sediments are presented in Figure 2.

The data presented by the GIOŚ show that the concentrations of cadmium, chromium, nickel, and zinc in the bottom sediments of the Wisła-Czarne dam Reservoir are several to a dozen times higher than in the lakes in question. The exceptions were copper, arsenic, and lead. In the case of copper and arsenic, higher concentrations of these elements were recorded only in the sediments of Swarzędzkie and Szczutowskie Lakes, respectively. In turn, in the case of lead, its highest concentration was found in the sediments of Lake Szczutowskie. The lowest concentrations of metal(loid)s were indicated in the bottom sediments of Lake Gardno. It is located in a National Park, and the lake's catchment area is almost entirely forested, so the impact of anthropogenic pressure is negligible [48]. This proves that sediments from reservoirs in areas of limited human activity impact are less contaminated. As mentioned, generally, the highest concentrations of elements considered in this paper were recorded in the bottom sediments of a reservoir in Silesia Voivodeship. This region is known for its extensive industrial activities (hard coal mining, metallurgy, coking plants, electric power generation, chemical industries, and metal industries in the country), and it is the most industrialized region in Poland [20,49]. Research in this area confirmed that human activities influence the chemical composition of bottom sediments [8,49-50]. Even though the Wisła-Czarne Reservoir is situated several dozen kilometers from the central part of the Upper Silesian Industrial Region, it stands out from the other water reservoirs considered in this paper. For comparison, the concentrations of Cd, Cr, Cu, Ni, Pb, and Zn in the bottom sediments of water reservoirs in other regions of the country, like the Zemborzycki dam Reservoir located in Lublin Voivodeship (Eastern Poland) or Jutrosin Reservoir in Greater Poland (Western Poland), amounted to 0.5; 5.9; 7.1; 6.1; 54.2 and 43.1 $mg \cdot kg^{-1}$ [51] and 0.2; 3.1; 3.5; 3.7; 6.2, and 23.1 $mg \cdot kg^{-1}$ [52], respectively. It is highly probable, that the source of metal(loid)s in the bottom sediments of the Wisła-Czarne Reservoir is mainly the emission of metal-rich dust by smelters and also a high level of industrial dust that is carried by the wind from the main industrial zone in Silesia and together with atmospheric deposition (dry and wet), entered the aquatic environment. However, these pollutants partly may also be historical. Generally, the Silesian region is characterized by a higher concentration of metal(loid)s in bottom sediments than other regions. This statement was confirmed by various scientific research [49, 50, 53].

3.2. Potential ecological risk

The indices of potential ecological risk used in this paper have both advantages and disadvantages. In the first group, we can distinguish (1) the lack of additional costs associated with the risk evaluation. Risk assessment is based solely on the results of analyses performed as part of routine monitoring. Therefore, the indices constitute a free tool for assessing the quality of bottom sediments; (2) a broader view of the risk evaluation. For example, I_{geo} relates the element concentration in the sample to the background value. In contrast, ER and RI include the toxicity of a particular element; and (3) the universal character of the indices. Therefore, these indices can evaluate various environmental matrices, such as sediment, soil, sewage sludge, etc. The only disadvantage is the problem of choosing the proper background value concentrations for the particular element. The results of the ecological risk assessment are presented in Table 5.

Table 5.
Results of the ecological risk assessment

Metal(loid)s	Lake Gardno	Lake Swarzędzkie	Lake Szczutowskie	Wisła-Czarne Reservoir
I_{geo}				
Cd	-2.6	**1.3**	**1.7**	**2.2**
Cr	-4.7	-4.4	-4.0	-2.9
Cu	-4.1	-0.9	-2.8	-1.5
Ni	-2.9	-2.4	-1.9	-0.5
Pb	-2.0	-0.5	0.4	-0.8
Zn	-2.0	-0.2	-0.6	0.9
As	0.2	-0.8	1.8	0.7
ER				
Cd	7.5	**114.0**	**144.0**	**207.0**
Cr	0.1	0.1	0.2	0.4
Cu	0.4	3.9	1.1	2.6
Ni	1.0	1.5	2.1	5.3
Pb	1.9	5.3	9.8	4.4
Zn	0.4	1.3	1.0	2.8
As	16.8	8.3	52.3	23.8
RI				
Metal(loid)s	28.1	134.5	**210.5**	**246.3**

The I_{geo} values indicate that although cadmium is the metal with the lowest content in the considered bottom sediments, it is still the main cause of their pollution, resulting in moderate and moderate to heavy contamination. At the same time, this also translates into a potential risk in the environmental dimension, which has not been noted about other elements. It should be emphasized here that although Cd occurs in the environment in low concentrations, it is also highly toxic to humans and animals (in excessive amounts), while less harmful to plants [54]. A trend similar to the one presented above was observed in other water reservoirs, where elements occurring in lower concentrations posed a greater risk in the ecological dimension than those with higher concentrations in bottom sediments. As an example, we can cite the results of research conducted for the Dzierżno Duże Reservoir (I_{geo}: Cd>Zn>Pb>As>Cu>Ni>Cr>Fe>Mn) [20] or the Rybnicki Reservoir (I_{geo}: Cu>Cd>Zn>Ni>Cr>Pb) [50], both located in the Silesian Voivodeship and also for the Rzeszów Reservoir (I_{geo}: Cd>Cr>Ni>Cu>Zn>Pb) [49], located in the Podkarpackie Voivodeship. The only water reservoir in which none of the discussed elements, present in bottom sediments posed a threat to the aquatic ecosystem was Lake Gardno, located in the Pomeranian Voivodeship. The lower levels of metal(loid) concentrations in the sediments of Lake Gardno and, therefore, no environmental risk is attributed to a lower degree of industrialization in this region, particularly of heavy industry, compared to other areas in the country.

The ER values reveal that cadmium is a primary element posing a risk among analyzed metal(loid)s (considerable to high risk). This is consistent with findings from the I_{geo} index. The only exception is arsenic in the bottom sediments of Lake Szczutowskie, for which a moderate risk was found. The ER index values for individual elements form the following series: As>Cd>Pb>Ni>Cu=Zn>Cr (Lake Gardno), Cd>As>Pb>Cu>Ni>Zn>Cr (Lake Swarzędzkie), Cd>As>Pb>Ni>Cu>Zn>Cr (Lake Szczutowskie) and Cd>As>Ni>Pb>Zn>Cu>Cr (Wisła-Czarne Reservoir). It is worth mentioning that among the elements considered, cadmium and arsenic have the highest toxicity coefficient, i.e. T=30 and T=10, respectively [13]. For comparison, in the case of the Bardowski Lagoon, the Kozłowa Góra Reservoir, and the Lake Bukwałd, located in the Masovian, Silesian, and Warmian-Masurian Voivodeships, respectively, the ER index values for individual elements formed the following series: Cd>Pb>Zn>Cu>Ni [55], Cd>Pb>Zn>Cu>Cr [56] and Cd>Ni>Pb>Cr>Cu>Zn [57]. The Gardno reservoir stands out as the only water reservoir where no ecological risk was identified due to analyzed metal(loid)s in its bottom sediments. This is perhaps related to the fact that Lake Gardno is located in a protected area of Słowiński National Park and the boundaries of the Natura 2000 area. Therefore, anthropogenic pressure on the environment has a negligible impact here.

Based on the values of the RI index, which expresses the total level of risk caused by a given group of elements, it was shown that the presence of the metal(loid)s in question in the bottom sediments of Lake Szczutowskie and the Wisła-Czarne Reservoir poses moderate ecological threats. No direct ecological risk was identified in the Gardno and Swarzędzkie Lakes case.

Table 6.
Average daily exposure doses of metals in bottom sediments

Metal(loid)s	Adults			Children		
	ADD_{ing}	ADD_{dermal}	ADDs	ADD_{ing}	ADD_{dermal}	ADDs
Lake Gardno						
Cd	1.47×10^{-8}	5.86×10^{-11}	1.47×10^{-8}	2.57×10^{-8}	7.19×10^{-11}	2.58×10^{-8}
Cr	3.36×10^{-6}	1.34×10^{-8}	3.38×10^{-6}	5.89×10^{-6}	1.65×10^{-8}	5.90×10^{-6}
Cu	6.48×10^{-6}	2.59×10^{-8}	6.51×10^{-6}	5.67×10^{-5}	1.59×10^{-7}	5.69×10^{-5}
Ni	2.34×10^{-6}	9.32×10^{-9}	2.35×10^{-6}	4.09×10^{-6}	1.14×10^{-8}	4.10×10^{-6}
Pb	3.33×10^{-6}	1.33×10^{-8}	3.35×10^{-6}	5.84×10^{-6}	1.63×10^{-8}	5.85×10^{-6}
Zn	3.48×10^{-5}	1.39×10^{-7}	3.49×10^{-5}	3.04×10^{-4}	8.52×10^{-7}	3.05×10^{-4}
As	1.77×10^{-6}	2.12×10^{-7}	1.99×10^{-6}	3.10×10^{-6}	2.61×10^{-7}	3.36×10^{-6}
Lake Swarzędzkie						
Cd	2.23×10^{-7}	8.90×10^{-10}	2.24×10^{-7}	3.90×10^{-7}	1.09×10^{-9}	3.92×10^{-7}
Cr	4.29×10^{-6}	1.71×10^{-8}	4.30×10^{-6}	7.50×10^{-6}	2.10×10^{-8}	7.52×10^{-6}
Cu	5.89×10^{-5}	2.35×10^{-7}	5.92×10^{-5}	5.16×10^{-4}	1.44×10^{-6}	5.17×10^{-4}
Ni	3.41×10^{-6}	1.36×10^{-8}	3.42×10^{-6}	5.97×10^{-6}	1.67×10^{-8}	5.99×10^{-6}
Pb	9.37×10^{-6}	3.74×10^{-8}	9.41×10^{-6}	1.64×10^{-5}	4.59×10^{-8}	1.64×10^{-5}
Zn	1.27×10^{-4}	5.06×10^{-7}	1.27×10^{-4}	1.11×10^{-3}	3.11×10^{-6}	1.11×10^{-3}
As	8.81×10^{-7}	1.05×10^{-7}	9.86×10^{-7}	1.54×10^{-6}	1.29×10^{-7}	1.67×10^{-6}
Lake Szczutowskie						
Cd	2.82×10^{-7}	1.12×10^{-9}	2.83×10^{-7}	4.93×10^{-7}	1.38×10^{-9}	4.95×10^{-7}
Cr	5.57×10^{-6}	2.22×10^{-8}	5.59×10^{-6}	9.75×10^{-6}	2.73×10^{-8}	9.78×10^{-6}
Cu	1.62×10^{-5}	6.45×10^{-8}	1.62×10^{-5}	1.41×10^{-4}	3.96×10^{-7}	1.42×10^{-4}
Ni	4.85×10^{-6}	1.93×10^{-8}	4.87×10^{-6}	8.49×10^{-6}	2.38×10^{-8}	8.51×10^{-6}
Pb	1.73×10^{-5}	6.91×10^{-8}	1.74×10^{-5}	3.03×10^{-5}	8.49×10^{-8}	3.04×10^{-5}
Zn	9.77×10^{-5}	3.90×10^{-7}	9.81×10^{-5}	8.55×10^{-4}	2.39×10^{-6}	8.58×10^{-4}
As	5.52×10^{-6}	6.61×10^{-7}	6.19×10^{-6}	9.67×10^{-6}	8.12×10^{-7}	1.05×10^{-5}
Wisła-Czarne Reservoir						
Cd	4.05×10^{-7}	1.62×10^{-9}	4.07×10^{-7}	7.09×10^{-7}	1.98×10^{-9}	7.11×10^{-7}
Cr	1.18×10^{-5}	4.71×10^{-8}	1.18×10^{-5}	2.07×10^{-5}	5.78×10^{-8}	2.07×10^{-5}
Cu	3.92×10^{-5}	1.56×10^{-7}	3.93×10^{-5}	3.43×10^{-4}	9.60×10^{-7}	3.44×10^{-4}
Ni	1.25×10^{-5}	4.98×10^{-8}	1.25×10^{-5}	2.19×10^{-5}	6.12×10^{-8}	2.19×10^{-5}
Pb	7.71×10^{-6}	3.08×10^{-8}	7.75×10^{-6}	1.35×10^{-5}	3.78×10^{-8}	1.35×10^{-5}
Zn	2.70×10^{-4}	1.08×10^{-6}	2.71×10^{-4}	2.36×10^{-3}	6.61×10^{-6}	2.37×10^{-3}
As	2.51×10^{-6}	3.01×10^{-7}	2.81×10^{-6}	4.40×10^{-6}	3.69×10^{-7}	4.77×10^{-6}

ing – through ingestion; dermal – through dermal contact

Table 7.
Results of non-carcinogenic risk assessment

NON-CARCINOGENIC RISK						
Metal	Adults			Children		
	HQ_{ing}	HQ_{dermal}	HI	HQ_{ing}	HQ_{dermal}	HI
Lake Gardno						
Cu	1.62×10^{-4}	2.15×10^{-6}	1.64×10^{-4}	1.42×10^{-3}	1.32×10^{-5}	1.43×10^{-3}
Zn	1.16×10^{-4}	2.31×10^{-6}	1.18×10^{-4}	1.01×10^{-3}	1.42×10^{-5}	1.03×10^{-3}
Lake Swarzędzkie						
Cu	1.47×10^{-3}	1.96×10^{-5}	1.49×10^{-3}	1.29×10^{-2}	1.20×10^{-4}	1.30×10^{-2}
Zn	4.23×10^{-4}	8.43×10^{-6}	4.31×10^{-4}	3.70×10^{-3}	5.18×10^{-5}	3.75×10^{-3}
Lake Szczutowskie						
Cu	4.04×10^{-4}	5.37×10^{-6}	4.09×10^{-4}	3.54×10^{-3}	3.30×10^{-5}	3.57×10^{-3}
Zn	3.26×10^{-4}	6.50×10^{-6}	3.32×10^{-4}	2.85×10^{-3}	3.99×10^{-5}	2.89×10^{-3}
Wisła-Czarne Reservoir						
Cu	9.79×10^{-4}	1.30×10^{-5}	9.92×10^{-4}	8.57×10^{-3}	8.00×10^{-5}	8.65×10^{-3}
Zn	9.00×10^{-4}	1.79×10^{-5}	9.18×10^{-4}	7.87×10^{-3}	1.10×10^{-4}	7.98×10^{-3}

3.3. Potential health risk

3.3.1. Average daily exposure doses

Table 6 presents the values of the average daily exposure doses of selected metal(loid)s in bottom sediments from the discussed water reservoirs.

Prolonged exposure to polluted bottom sediments containing high concentrations of various metals and metalloids may pose a significant health issue [5]. The calculations conducted in this study show that the dominant pathway through which the considered metal(loid)s can enter the human body is oral ingestion, for both adults and children. These results are consistent with those presented by scientists from other countries, such as Colombia [4], Bangladesh [11] and Egypt [58].

The highest values of ADD_{ing} and ADD_{dermal} to considered metal(loid)s were indicated to Zn, for both demographic groups. The mean values of ADDs taken through accidental oral ingestion and skin contact were in the range of 1.47×10^{-8} to 3.49×10^{-5} and 2.58×10^{-8} to 3.05×10^{-4} (Lake Gardno); 2.24×10^{-7} to 1.27×10^{-4} and 3.92×10^{-7} to 1.11×10^{-3} (Lake Swarzędzkie); 2.83×10^{-7} to 9.81×10^{-5} and 4.95×10^{-7} to 8.58×10^{-4} (Lake Szczutowskie); 4.07×10^{-7} to 2.71×10^{-4} and 7.11×10^{-7} to 2.37×10^{-3} (Wisła-Czarne Reservoir), for adults and children, respectively.

The calculations conducted indicate that the values of ADDs for children were higher than for adults. This finding suggests that children are more vulnerable to the risk linked with exposure to various elements in bottom sediments. Other scientists also confirmed the above statement [5, 11, 43].

3.3.2. Non-carcinogenic risk

The results of the non-carcinogenic risk assessment proved that exposure to bottom sediments (from considered water reservoirs) through ingestion and dermal contact did not pose a health risk for Cu and Zn, which means that HQ and HI values were below 1. Thus, while copper and zinc can still be harmful, the risk of adverse health effects from these two exposure routes is relatively low. Similar results were presented by scientists from Turkey, who reported that HQ_{ing} and HQ_{dermal} for Cu in sediments from examined aquatic ecosystem equaled 1.13×10^{-3} and 4.75×10^{-6} for adults and 1.05×10^{-2} and 2.49×10^{-5} for children, respectively. While for Zn: 8.18×10^{-5} and 3.45×10^{-7}, as well as 7.64×10^{-4} and 1.81×10^{-6}, for adults and children, respectively. In turn, HI for adults and children was 1.13×10^{-3} and 1.05×10^{-2} (Cu), as well as 8.22×10^{-5} and 7.65×10^{-4} (Zn), respectively [59].

The results presented in this article also indicate that oral ingestion was the main route impacting the HQ and HI values. This finding is consistent with those reported by scientists from other countries, such as Colombia [4] and Turkey [43]. In general, the highest HQ and HI values were indicated in bottom sediments from the Wisła-Czarne Reservoir and Lake Swarzędzkie, while the lowest in bottom sediment from the Gardno lake (Wisła-Czarne Reservoir; Lake Swarzędzkie>Lake Szczutowskie>Lake Gardno). The obtained results are partly consistent with those obtained by ecological risk assessment. The results of the non-carcinogenic risk assessment are shown in Table 7.

Table 8.
Results of carcinogenic risk assessment.

CARCINOGENIC RISK						
Metal(loid)s	Adults			Children		
	CR_{ing}	CR_{dermal}	TCR	CR_{ing}	CR_{dermal}	TCR
Lake Gardno						
Cd	8.95×10^{-8}	3.57×10^{-10}	8.99×10^{-8}	1.57×10^{-7}	4.39×10^{-10}	1.57×10^{-7}
Cr	1.68×10^{-6}	2.68×10^{-7}	1.95×10^{-6}	2.94×10^{-6}	3.30×10^{-7}	3.27×10^{-6}
Ni	3.97×10^{-6}	3.96×10^{-7}	4.37×10^{-6}	6.95×10^{-6}	4.87×10^{-7}	7.44×10^{-6}
Pb	2.83×10^{-8}	nd	2.83×10^{-8}	4.96×10^{-8}	nd	4.96×10^{-8}
As	2.66×10^{-6}	7.77×10^{-7}	3.44×10^{-6}	4.65×10^{-6}	9.54×10^{-7}	5.61×10^{-6}
Lake Swarzędzkie						
Cd	1.36×10^{-6}	5.43×10^{-9}	1.37×10^{-6}	2.38×10^{-6}	6.67×10^{-9}	2.39×10^{-6}
Cr	2.14×10^{-6}	3.42×10^{-7}	2.48×10^{-6}	3.75×10^{-6}	4.20×10^{-7}	4.17×10^{-6}
Ni	5.80×10^{-6}	5.78×10^{-7}	6.38×10^{-6}	1.01×10^{-5}	7.10×10^{-7}	1.09×10^{-5}
Pb	7.96×10^{-8}	nd	7.96×10^{-8}	1.39×10^{-7}	nd	1.39×10^{-7}
As	1.32×10^{-6}	3.86×10^{-7}	1.71×10^{-6}	2.31×10^{-6}	4.74×10^{-7}	2.79×10^{-6}
Lake Szczutowskie						
Cd	1.72×10^{-6}	6.86×10^{-9}	1.73×10^{-6}	3.01×10^{-6}	8.42×10^{-9}	3.02×10^{-6}
Cr	2.79×10^{-6}	4.45×10^{-7}	3.23×10^{-6}	4.88×10^{-6}	5.46×10^{-7}	5.42×10^{-6}
Ni	8.24×10^{-6}	8.22×10^{-7}	9.07×10^{-6}	1.44×10^{-5}	1.01×10^{-6}	1.54×10^{-5}
Pb	1.47×10^{-7}	nd	1.47×10^{-7}	2.58×10^{-7}	nd	2.58×10^{-7}
As	8.29×10^{-6}	2.42×10^{-6}	1.07×10^{-5}	1.45×10^{-5}	2.97×10^{-6}	1.75×10^{-5}
Wisła-Czarne Reservoir						
Cd	2.47×10^{-6}	9.86×10^{-9}	2.48×10^{-6}	4.32×10^{-6}	1.21×10^{-8}	4.34×10^{-6}
Cr	5.90×10^{-6}	9.42×10^{-7}	6.84×10^{-6}	1.03×10^{-5}	1.16×10^{-6}	1.15×10^{-5}
Ni	2.12×10^{-5}	2.12×10^{-6}	2.33×10^{-5}	3.71×10^{-5}	2.60×10^{-6}	3.98×10^{-5}
Pb	6.56×10^{-8}	nd	6.56×10^{-8}	1.15×10^{-7}	nd	1.15×10^{-7}
As	3.77×10^{-6}	1.10×10^{-6}	4.87×10^{-6}	6.60×10^{-6}	1.35×10^{-6}	7.95×10^{-6}

nd – lack of data (SF) to calculate carcinogenic risk to Pb

3.3.3. Carcinogenic risk

According to the values of the CR and TCR, a higher level of risk, as a result of accidental ingestion or skin contact with metal(loid)s present in bottom sediments, has been reported to children. This confirms that children are more vulnerable to metal(loid)s carcinogenic effects in bottom sediments. Similar observations were also reported by other researchers, who conducted a health risk assessment about selected metals (Cd, Cr, Cu, Ni, Pb, Zn, Fe, and Mn) in the bottom sediments of 11 reservoirs in Poland [49]. Moreover, in this study, the highest TCR mean values were observed in bottom sediment from the Wisła-Czarne Reservoir and the lowest in bottom sediment from the Lake Gardno (Wisła-Czarne Reservoir > Lake Szczutowskie > Lake Swarzędzkie > Lake Gardno). The presented results are consistent with those obtained by the ecological risk assessments. The results of the carcinogenic risk assessment are shown in Table 8.

The CR values through oral ingestion ranged from 2.83×10^{-8} to 3.97×10^{-6} and from 4.96×10^{-8} to 6.95×10^{-6} (Lake Gardno); from 7.96×10^{-8} to 5.80×10^{-6} and from 1.39×10^{-7} to 1.01×10^{-5} (Lake Swarzędzkie); from 1.47×10^{-7} to 8.29×10^{-6} and from 2.58×10^{-7} to 1.45×10^{-5} (Lake Szczutowskie); from 6.56×10^{-8} to 2.12×10^{-5} and from 1.15×10^{-7} to 3.71×10^{-5} (Wisła-Czarne Reservoir), for adults and children, respectively. In tern CR values through dermal contact ranged from 3.57×10^{-10} to 3.96×10^{-7} and 4.39×10^{-10} to 9.54×10^{-7} (Lake Gardno); from 5.43×10^{-9} to 5.78×10^{-7} and 6.67×10^{-9} to 7.10×10^{-7} (Lake Swarzędzkie); from 6.86×10^{-9} to 8.22×10^{-7} and from 8.42×10^{-9} to 2.97×10^{-6} (Lake Szczutowskie); from 9.86×10^{-9} to 2.12×10^{-6} and from 1.21×10^{-8} to 2.60×10^{-6} (Wisła-Czarne Reservoir), for adults and children, respectively. The CR values through the oral ingestion route can be arranged in the following decreasing order:

Ni>As>Cr>Cd>Pb (Lake Gardno),
Ni>Cr>Cd>As>Pb (Lake Swarzędzkie);
As>Ni>Cr>Cd>Pb (Lake Szczutowskie) and
Ni>Cr>As>Cd>Pb (Wisła-Czarne Reservoir), for

adults and children, respectively. In turn through dermal contact: As>Ni>Cr>Cd (Gardno and Szczutowkie Lakes) and Ni>As>Cr>Cd (Lake Swarzędzkie and Wisła-Czarne Reservoir), both for adults and children. In the case of Pb, due to the lack of SF value, we did not calculate the CR_{dermal}. A similar tendency was presented in our previous study, where CR_{ing} and CR_{dermal} of selected metals in the Pławniowice Reservoir (Silesian Voivodeship) decreased in the following order: Ni>Cd>Cr>Pb and Ni>Cr>Cd [8]. The TCR values indicate the same tendency as CR_{ing} and CR_{dermal}.

Almost all CR_{ing} and TCR values for Cd, Cr, Ni, and As were from 1×10^{-6} to 1×10^{-4}. This means that the risk posed by these elements in the bottom sediments of the water reservoirs is acceptable [14, 15]. Similar results were presented by other scientists who also suggested that selected elements (Al, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Cd, Sb, Hg, and Pb) did not pose a significant health risk for adults and children, exposed to bottom sediments in the Çavuşlu stream in Giresun (Turkey) [59]. However, even if the risk is not significant but includes in the range of 1×10^{-6} to 1×10^{-4}, appropriate actions should be taken to prevent the increase in the level of risk in the future, e.g., more frequent monitoring and chemical analysis of bottom sediments. Moreover, the values of the CR_{dermal} to some of the considered metal(loid)s were below 10^{-6}, which means these elements did not pose a carcinogenic risk to humans through skin contact. This was stated about Cd, Cr, Ni, and As in sediments of Lake Gardno and Lake Swarzędzkie, Cd, Cr, and Ni to Lake Szczutkowskie and Cd and Cr to Wisła-Czarne Reservoir. The presented results also indicate that Pb was the only element that did not pose a health risk.

4. CONCLUSIONS

The concentrations of metal(loid)s in analyzed bottom sediments were sourced from a report published by the Chief Inspectorate for Environmental Protection. The level of risk posed by metal(loid)s in aquatic ecosystems depends on several factors, including the origin of bottom sediments, the location of the water body, the sources of pollution (primarily human activities), and the properties of given elements. However, the data used as background values to calculate various ecological indices is also crucial.

The potential ecological risk assessment results revealed that although Cd is the metal with the lowest content in the bottom sediments, it is still the leading cause of their pollution. Cadmium poses moderate and moderate to heavy contamination (I_{geo}), and considerable to high risk (ER).

The results of the non-carcinogenic risk assessment proved that exposure to Cu and Zn in the bottom sediments through ingestion and dermal contact did not pose a health risk. The HQ and HI values for non-carcinogenic elements were below 1. In turn, CR and TCR values for carcinogenic elements were generally in the range of 1×10^{-6} to 1×10^{-4}, posing an acceptable risk. Still, there were some exceptions depending on the demographic group of the water reservoir. However, in this case, appropriate actions should be taken to prevent potential threats in the future. The conducted calculations also revealed that the dominant pathway through which the considered metal(loid)s can enter the human body is oral ingestion, for both adults and children. According to the values of the HQ, HI, CR, and TCR, a higher level of risk, as a result of accidental ingestion or skin contact with elements present in bottom sediments, has been reported for children. This confirms that children are more vulnerable to the non-carcinogenic and carcinogenic effects of metal(loid)s in bottom sediments.

Generally, the highest values of human health risks were indicated in bottom sediment from the Wisła-Czarne Reservoir, while the lowest was from Lake Gardno. The obtained results are consistent with those obtained by ecological risk assessment. Therefore, it is essential to monitor all changes occurring in the aquatic environment to reduce their impact's negative effects. Solving the metal(loid)s pollution problem in the water environment requires dialogue between representatives of various environments, including authorities, entrepreneurs, and society. In the future, the cooperation of these communities will allow for the development of comprehensive approaches to assessing the condition and quality of water reservoirs, an essential aspect of which will be both ecological and health risk assessment.

ACKNOWLEDGEMENTS

The paper has been prepared based on data included in the Report published on the website of the Chief Inspectorate for Environmental Protection (in Polish: Główny Inspektorat Ochrony Środowiska; GIOŚ). The captured data concerned the concentrations of selected metal(loid)s in the bottom sediments of four water reservoirs in Poland.

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