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ACCUMULATION OF TRACE ELEMENTS IN BOTTOM SEDIMENTS OF THE OTMUCHÓW AND DZIERŻNO DUŻE RESERVOIRS (ODER RIVER BASIN, SOUTHERN POLAND)

Rzętała M. A. **Kumulacja pierwiastków śladowych w osadach dennych zbiorników wodnych Otmuchów i Dzierżno Duże (dorzecze Odry, południowa Polska).** Skład chemiczny osadów dennych zbiorników wodnych określono metodami ICP i INAA. Dowiedziono, że niektóre z mikroskładników (As, Ba, Co, Cr, Cu, Ni, Pb, S, Sr, V, Zn) znacznie odbiegają ilością od poziomów uznawanych za naturalne w osadach wodnych Polski. Osady denne zbiornika Otmuchów reprezentują klasy osadów od praktycznie niezanieczyszczonych do silnie zanieczyszczonych ($-1,26 < I_{geo} < 3,89$), osady zbiornika Dzierżno Duże mieszczą się natomiast w przedziale od niezanieczyszczonych do ekstremalnie zanieczyszczonych ($0,34 < I_{geo} < 5,20$). Wskaźniki zanieczyszczenia świadczą o umiarkowanym zanieczyszczeniu osadów dennych zbiornika Otmuchów ($0,1 < C^i_f < 3,4$) i nawet bardzo wysokim zanieczyszczeniu osadów dennych w zbiorniku Dzierżno Duże ($0,7 < C^i_f < 8,5$). Stopień zanieczyszczenia osadów dennych jest niski w przypadku zbiornika Otmuchów ($C_d = 5,2$), a w przypadku zbiornika Dzierżno Duże ($C_d = 22,6$) świadczy o znacznym zanieczyszczeniu. Wzbogacenie składu chemicznego osadów, świadczy o kumulacji w osadach dennych pierwiastków śladowych, które wobec przekroczenia tła geochemicznego należy utożsamiać z zanieczyszczeniami.

Жентала М. А. **Аккумуляция химических элементов в донных отложениях водохранилищ Отмухов и Дзержно Дуже (бассейн реки Одра, южная Польша).** Химический состав донных осадков водохранилищ определялся методами ICP и INAA. Доказано, что некоторые из микрокомпонентов (As, Ba, Co, Cr, Cu, Ni, Pb, S, Sr, V, Zn) значительно отличаются количественно от уровней, которые считаются натуральными в водных отложениях Польши. Донные отложения водохр. Отмухов представляют классы осадков от практически незагрязненных до сильно загрязненных ($-1,26 < I_{geo} < 3,89$), отложения водохранилища водоема Дзержно Дуже – от незагрязненных до экстремально загрязненных ($0,34 < I_{geo} < 5,20$). Показатели загрязнения свидетельствуют об умеренном загрязнении донных осадков водохранилища Отмухов ($0,1 < C^i_f < 3,4$) и даже очень высоком загрязнении донных осадков в водохранилище Дзержно Дуже ($0,7 < C^i_f < 8,5$). Степень загрязнения донных осадков является низкой в случае водохранилища Отмухов ($C_d = 5,2$), в случае же водохранилища Дзержно Дуже ($C_d = 22,6$) свидетельствует о значительном загрязнении. Расширение спектра химического состава отложений свидетельствует о накоплении в донных осадках остаточных химических элементов, которые в связи с превышением геохимического фона следует отождествлять с загрязнениями.

Key words: bottom sediments, chemical composition, trace elements, heavy metals
Słowa kluczowe: osady denne, skład chemiczny, pierwiastki śladowe, metale ciężkie
Ключевые слова: донные отложения, химический состав, микроэлементы, тяжелые металлы

Abstract

The ICP and INAA methods were used to determine the chemical composition of bottom sediments in the Otmuchów and Dzierżno Duże Reservoirs in southern Poland. It was found that the following components are dominant in the bottom sediments of the Otmuchów Reservoir: SiO_2 (68.20–79.34%), Al_2O_3 (8.31–10.23%), organic matter (2.07–8.88%) and Fe_2O_3 (2.98–3.80%), with different proportions of these components in the sediments found in the Dzierżno Duże Reservoir (organic matter – 44.30–58.12%; SiO_2 – 20.78–28.45%, Al_2O_3 – 7.61–9.22%, Fe_2O3 – 4.39–6.59%); the other oxides also differed in the order of their percentage shares. Most of the trace elements analysed were detected in the bottom sediments of the water bodies examined in amounts corresponding to the concentration ranges found in sedimentary rocks. It was demonstrated that some of the microelements (As, Ba, Co, Cr, Cu, Ni, Pb, S, Sr, V and Zn) deviate significantly from the levels considered natural for aquatic sediments in Poland as evidenced by the ratios of their concentrations to the regional geochemical background, which amounted to $1.10 < I_{RE} < 20.20$ (Otmuchów Reservoir bottom sediments) and $3.44 < I_{RE} < 43.13$ (Dzier-

żno Duże Reservoir bottom sediments). Bottom sediments in the Otmuchów Reservoir range from virtually uncon-taminated to heavily contaminated ($-1.26 < I_{geo} < 3.89$), while those in the Dzierżno Duże Reservoir range from uncontaminated to extremely contaminated ($0.34 < I_{geo} < 5.20$). Contamination indicators point to moderate contamination of bottom sediments in the Otmuchów Reservoir ($0.1 < C^i_f < 3.4$) and very heavy (in some cases) contamination of bottom sediments in the Dzierżno Duże Reservoir ($0.7 < C^i_f < 8.5$). The degree of contamination of bottom sediments is low in the case of the Otmuchów water body ($C_d = 5.2$) and in the case of the Dzierżno Duże water body ($C_d = 22.6$) it points to significant contamination. The bottom sediments studied were enriched with: Pb, Ni, Co, Cr, V, Sr, As and Ba in the case of the Otmuchów Reservoir ($1.09 < I_{AP} < 5.14$) and As, Hg, Cr, Pb, Co, Ni, V, Zn, Cu, Sr, Ba and S in the case of the Dzierżno Duże Reservoir ($1.73 < I_{AP} < 29.75$). The enriched chemical composition of sediments reflects the accumulation of trace elements in bottom sediments; since the geochemical background has been exceeded, this should be equated with contamination.

INTRODUCTION

Basins of artificial water bodies serve as new sedimentary basins. These depressions formed as a response to human-made changes in the erosion base and are places where minerals and organic matter are deposited. The material that accumulates in these water bodies forms their bottom sediments. The bottom sediment cover in artificial water bodies is formed as a result of sedimentation of fluvial debris, the supply of material from the coastal zone, atmospheric deposition, as well as organic matter sedentation processes. Therefore bottom sediments constitute polygenetic material and their chemical composition is largely dependent on the geochemical background of the catchment as well as on the degree of human pressure (PIRRONE and KEELER, 1996; CHEUNG et al., 2003; CABALA and TEPER, 2007; CABALA, ZOGALA, DUBIEL, 2008; NGUESSAN et al., 2009; JOSHI and BALASUBRAMANIAN, 2010; CHRASTNY et al., 2012).

The deposits found within the basins of anthropogenic water bodies are in most cases used in quantitative and qualitative assessments of the rates at which they fill with sediments (VERSTRAETEN et al., 2006). This material is also often used as an indicator of the human impact on denudation rates (FERNEX et al., 2001; BAKOARINIAINA, KUSKY, RAHARIMAHEFA, 2006; DULIAS, 2013; RZETALA et al., 2015). On the other hand, the bottom sediments of anthropogenic lakes are of limited importance in the analysis of the historical evolution of water bodies in view of the young age of such bodies. Another aspect of the bottom sediments in anthropogenic water bodies is their economic utility (RZETALA et al., 2015). Bottom sediments are much more frequently the subject of scientific inquiry concerning the accumulation of pollutants (RZETALA et al., 2013). Thus the characteristics of bottom sediments are used as indicators of both the presence of pollution and environmental change and of the evolutionary dynamics of anthropogenic lakes.

Pollution is usually considered to include emissions that may either be harmful to human health and the environment or may cause damage to property. In the study of bottom sediments, pollution is considered to include the presence of specific substances in the environment in amounts that deviate from its natural content, i.e. geochemical background (MÜLLER, 1979; FÖRSTNER and MÜLLER, 1981). In aquatic sediments, pollutants of very diverse origins are present (PIRRONE, KEELER, 1996; CISZEWSKI, 1998; CHEUNG et al., 2003; NGUESSAN et al., 2009; JOSHI, BALASUBRAMANIAN 2010; SKORBIŁOWICZ E., SKORBIŁOWICZ M., 2011; SKORBIŁOWICZ, 2014; AYTMUKHANOVICH et al., 2014; ILIE et al., 2014; JABŁOŃSKA-CZAPLA, SZOPA, ROSIK-DULEWSKA, 2014). The common feature of pollutants is their sorption in the aquatic environment by mineral suspended matter (e.g. silty minerals) as well as by living and dead organic matter (e.g. algae, humic substances). In this manner, a significant proportion of the trace elements is accumulated in the bottom sediments (LOSKA, WIECHUŁA, 2003; YANG, ROSE, 2005; GHREFAT, YUSUF, 2006; SKORBIŁOWICZ E., SKORBIŁOWICZ M., 2011) and the chemical composition of the bottom sediments of water bodies is of special geoecological significance (RZETALA, 2015b). The presence of metals, non-metals and metalloids in the bottom sediments of water bodies is widely used in studies in order to assess environmental pollution and the role of lentic inland surface waters as places where pollutants accumulate (RADWAN, KOWALIK, KORNIJÓW, 1990; MIHELČIĆ et al., 1996; KOLAK, LONG, BEALS, 1998; RHOTON, BENNETT, 2009; BONANNO, 2011; PALANQUES et al., 2014; STRAKHOVENKO, TARAN, ERMOLAEVA, 2014; MOLLEMA et al., 2015).

The purpose of the study is to determine the basic composition of, and trace element content in, the bottom sediments of two water bodies in the upper section of the Oder River basin. The focus of the research is to identify the scale of anthropogenic enrichment of the chemical composition of bottom sediments in the water bodies situated in catchments that exhibit different types of land use. An additional contribution of this study is the assessment of the trace element content of bottom sediments as indicators of anthropogenic pollution.

STUDY AREA

The study was conducted within two water bodies situated in the upper part of the Oder River basin in southern Poland (fig. 1), i.e. the Otmuchów Reservoir on the Nysa Kłodzka River (left-hand tributary of the Oder) and the Dzierżno Duże Reservoir in the Kłodnica River valley (right-hand tributary of the Oder).

The Otmuchów Reservoir, impounded by a dam, is located in the central, lowland reach of the Nysa Kłodzka River valley; its catchment area as measured at the dam cross section is 2,361 km^2 (*Hydrographic...*, 1983). This is an area clearly divided into two parts in both geological and geomorphological terms – a mountainous landscape prevails in the Sudetes while upland features dominate in the Kłodzko Valley and in the Sudeten Foreland. The catchment consists of diverse geological formations (shale, gneiss, granite gneiss, porphyry, melaphyre, sandstone, marl, gravel, sand, silt, clay, loess). In the south-western (Sudetes) part of the catchment these are strongly metamorphosed Precambrian crystalline, igneous and sedimentary formations as well as sedimentary and metamorphosed Paleozoic and Mesozoic formations (WIŚNIEWSKI et al., 1980). In the north-eastern (Sudetes Foreland) part of the catchment there are mainly postglacial formations, sometimes with Holocene deposits, which overlie an older rock foundation (WIŚNIEWSKI et al., 1980). The presence of surface geological formations and land forms has been conducive to the development of agriculture and forestry and thus agricultural land and forests are the dominant forms of land use in the catchment.

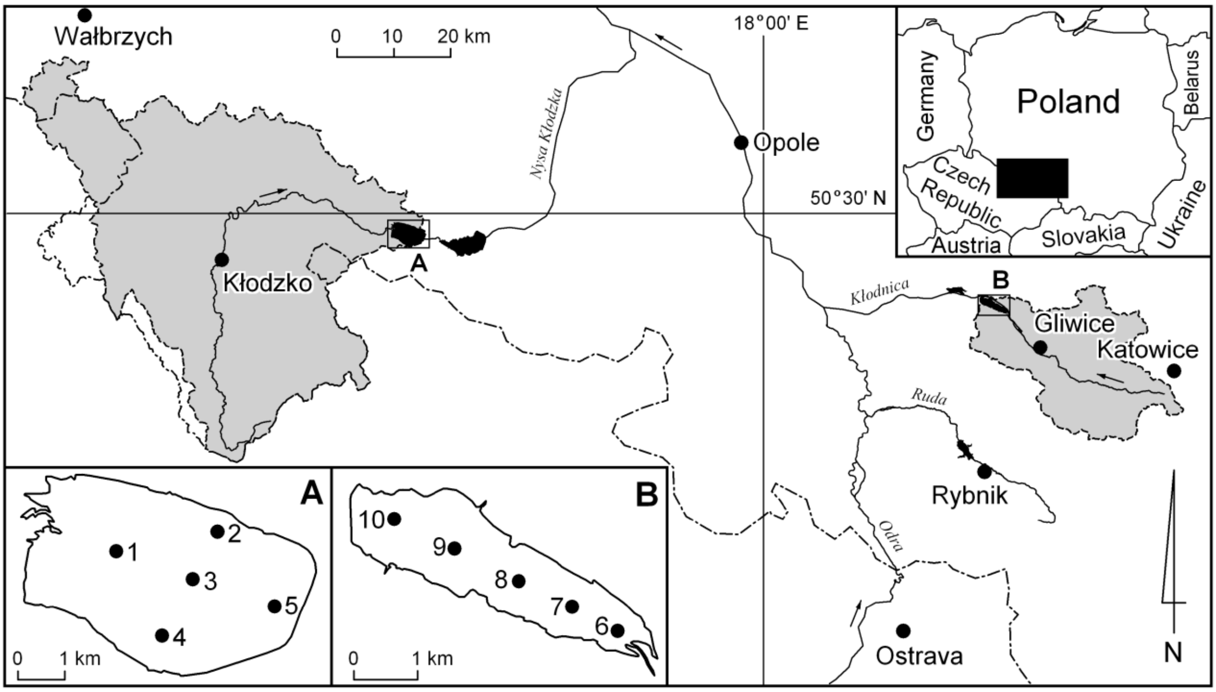

Fig. 1. Location of the bottom sediment sampling sites in the Otmuchów (samples Nos. 1–5) and Dzierżno Duże (samples Nos. 6–10) Reservoirs

Rys. 1. Lokalizacja miejsc opróbowania osadów dennych zbiorników wodnych Otmuchów (próby nr 1–5) i Dzierżno Duże (próby nr 6–10)

Rys. 1. Местонахождение пунктов забора проб донных отложений водохранилищ Отмухов (пробы №№ 1–5) и Дзержно Дуже (пробы №№ 6–10)

The Otmuchów Reservoir is the oldest and largest water body in the Nysa Kłodzka River basin. The reservoir was created by damming the river with a frontal dam with a height of 17 metres and a length of ca. 6.5 km, which allows water to be impounded up to an elevation of 215 m a.s.l. It was commissioned in 1933 with an area of 20.5 km^2, total capacity of 143 hm^3 and average depth of ca. 7.0 metres; its morphometric parameters have since changed significantly as a result of it being filled with sediments (WIŚNIEWSKI et al., 1980). Average annual flows of the Nysa Kłodzka River at reservoir inlet range from a few m^3/s to a few hundred m^3/s with an average of ca. 21 m^3/s (WIŚNIEWSKI et al., 1980). Extreme flows are much greater, e.g. during the flood in July 1997 the flow of the Nysa Kłodzka River reached 2,100 m^3/s (ZIELINSKI, 2003). The Otmuchów Reservoir serves flood control purposes, is used to produce electricity, is a source of water supply and is also suitable for fishing as well as leisure and recreation purposes.

The Dzierżno Duże Reservoir was formed in flooded mineral workings in the central part of the Kłodnica River catchment. At the weir cross section, it has a catchment area of ca. 520 km^2 (*Hydrographic*..., 1983). It lies in a typical upland area. Within the catchment, there are Carboniferous formations (sandstone, shale, coal), which sometimes underlie Triassic (limestone, marl, dolomite) formations as well as Tertiary ones (clay); on the surface, there are Pleistocene (sand, gravel, clay) and Holocene (alluvial soil) deposits. The long-standing exploitation of mineral resources (coal, zinc and lead ores, iron ore, sand, gravel and clay) together with the accompanying development of the processing industry, the emergence of settlements and the subsequent urbanisation processes have resulted in human-made changes in the environment. The urban and industrial nature of the catchment is reflected by the main forms of land use – ca. 32% of the reservoir catchment is covered by industrial areas and industrial wasteland with agricultural land accounting for ca. 43% of the area and forests covering ca. 23% (RZETALA, 2015a).

The Dzierżno Duże Reservoir arose in 1964 when heavily contaminated waters of the Kłodnica River were diverted into former sand workings. The natural hydrological regime of the river was completely disrupted owing, *inter alia*, to water transfers from outside the catchment, the dumping of waste water and water drained from mines and measures implemented to control water circulation (CZAJA et al., 2014). Seasonal fluctuations have been dampened by human activity and flows usually amount to several m^3/s. At the maximum water level (203.5 m a.s.l.), the reservoir had an area of over 6 km^2, capacity of 94.0 hm^3 and average depth of about 15 metres (with a maximum depth of ca. 20 m). The current reservoir retention capability is lower by several million m^3 due to its having been filled with sediments (RZETALA et al., 2015). The Dzierżno Duże Reservoir serves flood control purposes, supplies the Gliwice Canal and the Oder River with water (improving navigation conditions), and is considered a sedimentation tank for the contaminated Kłodnica River waters and also a source of energy resources that are extracted periodically when it is dredged (RZETALA, 2014).

MATERIALS AND STUDY METHODS

During field work, five samples each were collected from the bottom sediment covers in the Otmuchów and Dzierżno Duże Reservoirs. Sediment samples were collected into a polystyrene container by sinking it into surface sediments within the exposed reservoir bottom zone (at a low water stage). Samples within the water-filled depression were collected using the Beeker loose sediment sampler (04.20.S.A version, manufactured by Eijkelkamp) – a sediment core sampler with a transparent polystyrene tube used to collect sediments and a pneumatic closure mechanism. From the material collected, a top sediment layer with a thickness of ca. 1 centimetre was separated according to the guidelines formulated by L. HÅKANSON (1980). The sediment samples collected were dried at a temperature of 105°C in laboratory conditions. The dried material was ground using a mortar and then sieved on chemically neutral sieves to separate the 0.063 mm fraction. The samples prepared in this manner were then subjected to analyses at Activation Laboratories Ltd. (Canada) in accordance with the standards observed by the laboratory.

The main oxides (SiO_2, TiO_2, Al_2O_3, Fe_2O_3, MnO, MgO, CaO, Na_2O, K_2O and P_2O_5), loss on ignition and sulphur content were determined using the ICP method. Using the same method, the content of Ba, Sr, Y, Zr, Be, Ni, Cd, Bi, Cu, Pb and Zn in the sediments was determined. The INAA method was used to determine the content of Ag, Au, As, Br, Co, Cr, Cs, Hf, Ir, Mo, Rb, Sb, Se, Ta, W, La, Ce, Nd, Sm, Eu, Sc, U and Th. Mercury content was determined in sediment samples of 0.5 g each using the Cold Vapour FIMS (Perkins Elmer FIMS 100) method. In the case of main components, the lower detection limit varied and was as follows: for SiO_2, Al_2O_3, Fe_2O_3, MnO, MgO, CaO, Na_2O, K_2O and P_2O_5 – 0.01%; for TiO_2 – 0.005%, and for S – 0.001%. A similar variation was present with respect to the lower limit of detection for trace elements: 5 ppb (Au), 0.1 ppm (Sm, Eu, Sc), 0.2 ppm (Sb, La), 0.5 ppm (Ag, Cd, Cs, Hf), 1.0 ppm (Y, Be, Cu, Zn, Ni, Br, Co, Cr, Ta), 2.0 ppm (Sr, Zr, Bi, As, Mo), 3.0 ppm (Ba, Se, W, Ce), 5.0 ppm (V, Pb, Au, Ir, Nd), 20.0 ppm (Rb) and 5 ppb (Hg). PCB figures, which were necessary for calculating the contamination factor and contamination degree of sediments, were sourced from the results of monitoring of aquatic sediments in the catchments of the Nysa Kłodzka and Kłodnica Rivers conducted by the Chief Inspectorate of Environmental Protection.

In the assessment of the geoecological significance of the content of trace elements in the surface layer of bottom sediments of the Otmuchów and Dzierżno Duże Reservoirs, several indicators were used:

$$I_{RE} = \frac{C_{BS}}{C_{GB}}$$ (Eq. 1) (RZETALA, 2015a, b),

$$I_{AP} = \frac{C_{BS}}{C_{SR}} \quad \text{(Eq. 2) (RZETALA, 2015a, b),}$$

$$I_{geo} = \log_2 \frac{C_n}{1.5B_n} \quad \text{(Eq. 3) (MÜLLER, 1979),}$$

$$C_d = \sum_{i=1}^{8} C_f^i = \sum_{i=1}^{8} \frac{\overline{C}_{0-1}^i}{C_n^i} \quad \text{(Eq. 4) (HÅKANSON, 1980).}$$

The ratio of the values measured to the regional geochemical background (Eq. 1) is the ratio of the average concentration of the element in bottom sediments (C_{BS}) and the regional geochemical background for the element in question in bottom sediments (C_{GB}) (RZETALA, 2015a, b). The ratio of the value measured to the regional geochemical background (I_{RE}) reflects the contamination of sediments when it is greater than unity ($I_{RE} > 1.0$) and the lack of contamination when it is lower than unity ($I_{RE} < 1.0$).

In the study the indicator of anthropogenic enrichment of bottom sediments (Eq. 2) is used to carry out a geoecological comparison of trace element concentrations between the bottom sediments of water bodies and the sediments of flowing surface waters. The indicator of anthropogenic enrichment of sediments (I_{AP}) simply reflects the similarities or differences in the concentrations of the elements analysed in the sediments present at the sites compared (i.e. bottom sediments from a water body and those from a river channel) and is the ratio of the contents of this element in these types of sediments (RZETALA, 2015a, b). An $I_{AP} > 1$ points to bottom sediment enrichment, while an $I_{AP} < 1$ points to the absence of such enrichment.

The geoaccumulation index (I_{geo}) developed by G. MÜLLER (1979) (Eq. 3) was used in order to assess the degree of contamination of bottom sediments. The I_{geo} index value depends on the concentration of the element in question in bottom sediments (C_n), the geochemical background of the element in question (B_n) and the coefficient expressing the natural fluctuation range of the concentration of that element in the environment (equal to 1.5). Adopting the geochemical background at the level indicated for Poland by J. LIS and A. PASIECZNA (1995), the degree of contamination of bottom sediments in the water bodies was assessed in accordance with the guidelines concerning different sediment quality classes (MÜLLER, 1979):

$I_{geo} \leq 0.0$ – practically uncontaminated sediments;
$0.0 < I_{geo} < 1.0$) – uncontaminated to moderately contaminated sediments;
$1.0 < I_{geo} < 2.0$ – moderately contaminated sediments;
$2.0 < I_{geo} < 3.0$ – moderately to heavily contaminated sediments;
$3.0 < I_{geo} < 4.0$ – heavily contaminated sediments;
$4.0 < I_{geo} < 5.0$ – heavily to extremely contaminated sediments;
$I_{geo} > 5.0$ – extremely contaminated sediments.

The degree of sediment contamination (C_d) resulting from the contamination factor (C^i_f) was determined on the basis of the formula (Eq. 4) developed by L. HÅKANSON (1980), where:

C_d – the degree of contamination;
C_f^i – the contamination factor;
$\overline{C}_{0-1}^i$ – the mean content of the substance in question (i) from superficial sediments (0–1 cm) from accumulation areas (at least 5 samples, which provide an even area cover of the lake or basin should be taken);
C_n^i – the standard preindustrial reference level; determined from various European and American lakes to be (in ppm): PCB = 0.01, Hg = 0.25, Cd = 1.0, As = 15.0, Cu = 50.0, Pb = 70.0, Cr = 90.0 and Zn = 175.0).

Contamination factor values (C^i_f) are ratios of the concentrations of individual elements in a 1 cm thick upper layer of bottom sediment surface and pre-industrial concentrations of these substances in sediments. On the basis of the contamination factor (C^i_f) sediments are considered to be uncontaminated, contaminated or enriched:

$C^i_f < 1$ – low sediment contamination;
$1 \leq C^i_f < 3$ – moderate contamination;
$3 \leq C^i_f < 6$ significant contamination;
$C^i_f > 6$ – very heavy contamination.

The degree of sediment contamination (C_d) is in turn based on contamination factor values:

$C_d < 8$ – low degree of contamination,
$8 \leq C_d < 16$ – moderate degree of sediment contamination,
$16 \leq C_d < 32$ – significant degree of contamination,
$C_d > 32$ – very high degree of anthropogenic sediment contamination.

RESULTS

The chemical composition of the bottom sediments of the Otmuchów and Dzierżno Duże Reservoirs exhibits quantitative differences. These differences concern both basic components (table 1) and trace element content (table 2).

The basic components of sediments in the Otmuchów Reservoir can be ordered in the following sequence according to their average percentages by weight: $SiO_2 > Al_2O_3 >$ Loss on ignition (LOI) $> Fe_2O_3 > K_2O > Na_2O > MgO > TiO_2 > CaO > P_2O_5 > MnO$.

Table 1. Basic chemical composition of bottom sediments of the Otmuchów and Dzierżno Duże Reservoirs
Tabela 1. Podstawowy skład chemiczny osadów dennych zbiorników wodnych Otmuchów i Dzierżno Duże
Таблица 1. Основной химический состав донных отложений водохранилищ Отмухов и Дзержно Дуже

Component	Unit	Otmuchów Reservoir			Dzierżno Duże Reservoir		
		minimum	maximum	arithmetic mean	minimum	maximum	arithmetic mean
SiO_2	%	68.20	79.34	73.38	20.78	28.45	25.04
Al_2O_3	%	8.31	10.23	9.61	7.61	9.22	8.76
Fe_2O_3	%	2.98	3.80	3.41	4.39	6.59	5.61
MnO	%	0.06	0.11	0.08	0.05	0.07	0.06
MgO	%	0.58	0.92	0.78	1.19	1.58	1.39
CaO	%	0.50	1.02	0.73	2.17	3.27	2.57
Na_2O	%	0.87	1.18	1.03	0.25	0.34	0.30
K_2O	%	1.98	2.84	2.37	0.99	1.28	1.15
TiO_2	%	0.58	0.98	0.75	0.31	0.41	0.38
P_2O_5	%	0.13	0.28	0.19	0.59	0.72	0.66
Loss of ignition	%	2.07	8.88	5.50	44.30	58.12	51.03

Table 2. Trace elements in the bottom sediments of the Otmuchów and Dzierżno Duże Reservoirs
Tabela 2. Pierwiastki śladowe w osadach dennych zbiorników wodnych Otmuchów i Dzierżno Duże
Таблица 2. Остаточные химические элементы в донных отложениях водохранилищ Отмухов и Дзержно Дуже

Component	Unit	Elements present in the bottom sediments of the Otmuchów Reservoir			Range of element concentrations in Nysa Kłodzka River aquatic sediments [a)]			Elements present in the bottom sediments of the Dzierżno Duże Reservoir			Range of element concentrations in Kłodnica River aquatic sediments [a)]			Geochemical background for Poland [a)]	Range of concentration in sedimentary rocks [b)]
		min.	max.	average	min.	max.	average	min.	max.	average	min.	max.	average		
Ag	ppm	below the lower limit of detection			<1.0	1.0	<1.0	1.6	2.8	2.0	<1.0	3.0	1.0	<1.0	0.05–0.25
As	ppm	39.0	57.0	51.0	<5.0	48.0	11.0	17.0	21.0	19.0	<5.0	12.0	6.0	<5.0	1.0–13.0
Au	ppb	below the lower limit of detection			no data			44.0	296.0	159.6	no data			no data	2.0–7.0
Ba	ppm	488.0	556.0	514.0	41.0	215.0	100.0	1165.0	1863.0	1545.2	85.0	474.0	169.0	54.0	50.0–800.0
Be	ppm	1.0	3.0	2.0	<0.5	2.6	0.9	2.0	2.0	2.0	<0.5	8.0	0.5	<0.5	0.2–6.0
Bi	ppm	below the lower limit of detection			no data			below the lower limit of detection			no data			no data	no data
Br	ppm	5.0	8.0	6.0	no data			12.0	27.0	20.0	no data			no data	1.0–10.0
Cd	ppm	below the lower limit of detection			<0.5	1.4	0.5	6.3	11.4	8.5	<0.5	19.1	3.6	<0.5	0.05–0.35
Ce	ppm	72.0	89.0	81.0	no data			37.0	45.0	42.4	no data			no data	7.0–90.0
Co	ppm	12.0	14.0	13.0	3.0	10.0	7.0	17.0	24.0	20.2	2.0	12.0	6.0	3.0	0.1–20.0
Cr	ppm	88.0	111.0	101.0	8.0	709.0	46.0	88.0	128.0	105.0	4.0	92.0	16.0	5.0	5.0–120.0
Cs	ppm	5.8	7.2	6.4	no data			6.9	8.3	7.9	no data			no data	0.5-10.0
Cu	ppm	15.0	20.0	18.0	9.0	57.0	20.0	102.0	126.0	115.8	10.0	228.0	39.0	7.0	2.0–60.0
Eu	ppm	0.7	1.0	0.9	no data			0.9	1.1	1.0	no data			no data	0.2–2.0
Hf	ppm	17.3	24.4	20.8	no data			3.2	4.3	3.7				no data	no data
Hg	ppb	40.0	188.0	73.0	<0.050	0.190	0.080	95.0	243.0	171.8	<50.0	77.0	20.0	50.0	no data
Ir	ppb	below the lower limit of detection			no data			8.0	13.0	10.0	no data			no data	no data
La	ppm	34.0	53.0	45.9	no data			19.6	24.9	23.2	no data			no data	4.0–90.0
Lu	ppm	0.40	0.62	0.46	no data			0.3	0.3	0.3	no data			no data	no data
Mo	ppm	below the lower limit of detection			no data			2.8	4.5	3.8	no data			no data	no data
Nd	ppm	40.2	54.0	47.0	no data			12.0	18.0	16.2	no data			no data	4.7–41.0
Ni	ppm	24.0	32.0	27.0	7.0	31.0	17.0	48.0	55.0	51.8	7.0	40.0	16.0	6.0	5.0–90.0
Pb	ppm	57.0	68.0	62.0	14.0	234.0	57.0	131.0	199.0	148.8	15.0	242.0	103.0	13.0	3.0–40.0

Rb	ppm	90.0	110.0	100.0	no data			55.0	74.0	65.6	no data			no data	5.0–200.0
S	%	0.025	0.088	0.044	0.024	0.560	0.058	1.5	2.2	1.7	0.025	0.742	0.107	0.040	no data
Sb	ppm	1.4	2.4	1.8	no data			4.4	5.2	4.8	no data			no data	0.03–2.0
Sc	ppm	7.4	9.6	8.4	no data			8.5	9.6	9.3	no data			no data	0.5–15.0
Se	ppm	below the lower limit of detection			no data			below the lower limit of detection			no data			no data	no data
Sm	ppm	5.4	8.4	6.4	no data			3.6	4.4	4.1	no data			no data	1.3–22.1
Sr	ppm	82.0	112.0	94.0	10.0	72.0	23.0	199.0	217.0	212.2	24.0	212.0	66.0	20.0	20.0–600.0
Ta	ppm	below the lower limit of detection			no data			no data			no data			no data	no data
Tb	ppm	0.7	1.6	1.3	no data			0.6	0.7	0.7	no data			no data	no data
Th	ppm	12.6	18.2	15.1	no data			9.6	11.4	10.9	no data			no data	1.7–12.0
U	ppm	4.8	6.8	5.8	no data			3.9	4.7	4.4	no data			no data	0.45–4.00
V	ppm	58.0	76.0	65.0	13.0	26.0	18.0	72.0	93.0	81.8	6.0	25.0	13.0	7.0	10.0–130.0
W	ppm	below the lower limit of detection			no data			3.0	4.0	3.6	no data			no data	no data
Y	ppm	20.0	41.0	32.0	no data			17.0	20.0	18.8	no data			no data	4.0–50.0
Yb	ppm	3.6	4.8	4.2	no data			1.6	2.0	1.8	no data			no data	no data
Zn	ppm	99.0	116.0	110.0	67.0	517.0	181.0	791.0	1019.0	895.6	73.0	1424.0	509.0	62.0	10.0–120.0
Zr	ppm	513.0	535.0	524.0	no data			79.0	125.0	104.0				no data	20.0–220.0

Explanation: [a)] according to: J. LIS and A. PASIECZNA (1995); [b)] according to: A. KABATA-PENDIAS and H. PENDIAS (1993).

This sequence can be considered to represent typical quantitative differences in the concentrations of components which show little variation when individual samples are compared. Among the most important oxides in bottom sediments, silica dominates (68.20–79.34%), followed by Al_2O_3 (8.31–10.23%) with relatively low loss on ignition (2.07–8.88%). The remaining macroelements are represented to a much smaller degree. In the Dzierżno Duże Reservoir, the most common component of sediments in the backflow zone is organic matter (loss on ignition ranges from 44.30% to 58.12%). Other components are also present in a different order: loss on ignition (LOI) > SiO_2 > Al_2O_3 > Fe_2O_3 > CaO > MgO > K_2O > P_2O_5 > TiO_2 > Na_2O > MnO.

Among the trace elements detected in the chemical composition of bottom sediments in the Otmuchów Reservoir, the following are the most prevalent on average: Zr (524 ppm), Ba (514 ppm), Zn (110 ppm), Cr (101 ppm), Rb (100 ppm), Sr (94 ppm), Ce (81 ppm), V (65 ppm), Pb (62 ppm), As (51 ppm), Nd (47 ppm), La (45,9 ppm), Y (32 ppm), Ni (27 ppm) and Hf (20.8 ppm). Cu (18.0 ppm), Th (15.1 ppm) and Co (13.0 ppm) are present at several ppm on average. The remaining trace elements are present in reservoir sediments in average amounts of up to a few ppm: Sc (8.4 ppm), Cs and Sm (6.4 ppm), Br (6 ppm), U (5.8 ppm), Yb (4.2 ppm), Be (2.0 ppm), Sb (1.8 ppm) and Tb (1.3 ppm). Eu and Lu were determined at 0.9 ppm and 0.46 ppm respectively. The average sulphur content is 0.044%. Mercury is present at the level of 73 ppb on average. Several elements were present in amounts below the lower limit of detection of the methods used: Au and Ir (<5.0 ppm), Se and W (<3.0 ppm), Bi and Mo (<2.0 ppm), Ta (<1.0 ppm) and Ag and Cd (<0.5 ppm).

In samples of bottom sediments from the Dzierżno Duże Reservoir, Ba dominates – 1,545.2 ppm on average. The next six elements have average concentrations in sediments of the order of hundreds of ppm: Zn (895.6 ppm), Sr (212.2 ppm), Pb (148.8 ppm), Cu (115.8 ppm), Cr (105.0 ppm) and Zr (104 ppm). The next 12 elements are present in concentrations from a dozen to several dozen ppm: V (81.8 ppm), Rb (65.6 ppm), Ni (51.8 ppm), Ce (42.4 ppm), La (23.2 ppm), Co (20.2 ppm), Br (20.0 ppm), As (19.0 ppm), Y (18.8 ppm), Nd (16.2 ppm), Th (10.9 ppm) and Ir (10.0 ppm). The remaining trace elements are present in average amounts of up to a few ppm: Sc (9.3 ppm), Cd (8.5 ppm), Cs (7.9 ppm), Sb (4.8 ppm), U (4.4 ppm), Sm (4.1 ppm), Mo (3.8 ppm), Hf (3.7 ppm), W (3.6 ppm), Ag (2.0 ppm), Be (2.0 ppm), Yb (1.8 ppm), Ta (1.4 ppm), Eu (1.0 ppm), Tb (0.7 ppm) and Lu (0.3 ppm). The average S content in sediments is 1.725%, Hg content – 171.8 ppb, and Au content – 159.6 ppb. Only two elements were present in amounts below the lower limit of detection: Se < 3.0 ppm and Bi < 2.0 ppm.

DISCUSSION OF STUDY RESULTS

The chemical composition of sediments determined on the basis of the samples collected within the two water bodies studied differs in terms of both basic components and trace elements.

The characteristics of the basic composition of bottom sediments differ for individual water bodies depending on their catchment conditions and the presence of human impact. This is reflected in quantitative differences in the concentrations of the components found in the samples from the water bodies examined. In sediments from the Otmuchów Reservoir, SiO_2 dominates (68.20–79.34%), followed by Al_2O_3 (8.31–10.23%), organic matter (loss on ignition: 2.07–8.88%) and Fe_2O_3 (2.98–3.80%). In sediments from the backflow zone of the Dzierżno Duże Reservoir, the primary component is organic matter (loss on ignition: 44.30%–58.12%), followed by SiO_2 (20.78%–28.45%), Al_2O_3 (7.61%–9.22%) and Fe_2O_3 (4.39%–6.59%). The dominant factor in shaping the relationship between SiO_2 content and loss on ignition in the Dzierżno Duże Reservoir appears to be the supply of anthropogenic pollutants from its catchment area. In this respect, the catchment of the Otmuchów Reservoir is an area from which primarily mineral substances are supplied to sediments. Apart from the components listed as basic building materials, bottom sediments also include (in the form of basic minerals or their components) compounds of the following elements: manganese, magnesium, calcium, sodium, potassium, titanium and phosphorus. Their percentage shares are also dependent on catchment lithology and the nature of the human impact. The CaO found in sediment samples indicates a significant amount of calcium in their composition. This is probably a result of the occurrence of carbonate formations within the geological structure of the catchment; it may also be caused by human agricultural activity and the use of fertilisers containing Ca (this could also explain the presence of potassium in sediments). The phosphorus present in the sediments may be attributed to natural processes (e.g. the leaching of bioelements from the rocks present within the catchment) as well as to anthropogenic sources (e.g. run-off from agricultural land) (RZETALA, 2015b).

The amounts of trace elements present in the bottom sediments of the water bodies studied are even more varied than in the case of the basic components, although the samples collected within individual water bodies are similar in terms of microelement concentrations. Differences in chemical composition are of considerable geoecological significance, the more so that the presence of microelements in the natural environment results not only from natural processes (e.g. the weathering of rocks), but also from their supply from anthropogenic sources (e.g. industry, transport). So-called toxic metals are particularly dangerous to human health and life and to the development of flora and fauna (RZETALA, 2015b). Although some trace elements (e.g. zinc, copper) are considered microelements that organisms require in certain amounts in order to thrive, the same elements in inappropriate amounts as well as others (e.g. lead, cadmium, chromium, nickel) are considered unnecessary and even harmful (KABATA-PENDIAS, PENDIAS, 1979).

Concentrations of some trace elements in bottom sediments are similar to the ranges in which they occur naturally in sedimentary rocks as stated by A. KABATA-PENDIAS and H. PENDIAS (1993) (table 2). In the case of sediment samples from the Otmuchów Reservoir these are: Au, Be, Br, Cd, Ce, Co, Cr, Cs, Cu, Eu, La, Nd, Ni, Rb, Sb, Sc, Sm, Sr, Th, V, Y and Zn. In the sediment samples from the Dzierżno Duże Reservoir, the elements that are within the range specified as natural for their occurrence in sedimentary rocks are: Be, Co, Cr, Cs, Eu, La, Mo, Nd, Ni, Rb, Sb, Sc, Sm, Th, U, V, Y and Zr.

Among all the elements identified, the geochemical background for Poland was determined by J. LIS and A. PASIECZNA (1995) for only twelve (As, Ba, Co, Cr, Cu, Hg, Ni, Pb, S, Sr, V and Zn); virtually in each case, this background value was exceeded in

Table 3. Ratios of the concentrations measured to the geochemical background (I_{RE}) and values of the anthropogenic sediment enrichment indicator (I_{AP})

Tabela 3. Wartości wskaźnika krotności przekroczenia tła geochemicznego (I_{RE}) i wskaźnika antropogenicznego wzbogacenia osadów (I_{AP})

Таблица 3. Значения показателя кратности превышения геохимического фона (I_{RE}) и показателя антропогенного расширения спектра осадков (I_{AP})

Component	Bottom sediments of the Otmuchów Reservoir		Bottom sediments of the Dzierżno Duże Reservoir	
	I_{RE}	I_{AP}	I_{RE}	I_{AP}
As	10.20	4.64	3.80	1.73
Ba	9.52	5.14	28.61	15.45
Co	4.33	1.86	6.73	2.89
Cr	20.20	2.20	21.00	2.28
Cu	2.57	0.90	16.54	5.79
Hg	1.46	0.91	3.44	2.15
Ni	4.50	1.59	8.63	3.05
Pb	4.77	1.09	11.45	2.61
S	1.10	0.76	43.13	29.75
Sr	4.70	4.09	10.61	9.23
V	9.29	3.61	11.69	4.54
Zn	1.77	0.61	14.45	4.95

the bottom sediments of the water bodies studied. Only the amounts of S and Hg found in the sediments of the Otmuchów Reservoir were similar to the geochemical background for Poland or slightly higher. The extent of human-made changes in the chemical composition of the bottom sediments of the water bodies studied is evidenced by the ratios of the values found to the geochemical background (I_{RE}). For sediments in the Otmuchów Reservoir, I_{RE} values range from 1.10 to 20.20, while sediments in the Dzierżno Duże Reservoir exhibit much higher ratios, ranging from 3.44 to 43.13 (table 3). This means that the concentration of trace elements in bottom sediments is considerably higher than the geochemical background and indicates contamination with virtually all of the elements listed. The relationship between the concentrations of elements in bottom sediments and the geochemical background for Poland is especially important from the geoecological point of view since the Oder River basin drains towards the north and there is a real threat of pollution (and contamination) being transported to areas that have been free of pollutants to date.

The contents of trace elements found in bottom sediments in the Otmuchów and Dzierżno Duże Reservoirs are in many cases higher than the ranges in which these elements are present in the aquatic sediments of the rivers flowing through these water bodies (the Nysa Kłodzka and Kłodnica Rivers respectively); the latter ranges were examined along the entire lengths of these rivers by J. LIS and A. PASIECZNA (1995). This points to the enrichment of sediments in water bodies with trace elements when compared to the bottom sediments of the rivers that flow through these reservoirs. The bottom sediments of the Otmuchów Reservoir are enriched with Ba, As, Sr, V, Czr, Co, Ni and Pb, and the sediments of the Dzierżno Duże Reservoir are enriched with all the trace elements analysed at levels ranging from two to several dozen times (table 3). This reflects the considerable accumulation potential of various substances in water body bottom sediments; since the geochemical background has been exceeded, the presence of these substances should be equated with contamination.

The differences between the bottom sediments present in the Otmuchów and Dzierżno Duże Reservoirs are reflected by the values of the geoaccumulation index (table 4) whose calculations take into account the geochemical background for Poland determined by J. LIS and A. PASIECZNA (1995). I_{geo} values indicate that sediments in the Otmuchów Reservoir are either uncontaminated or moderately or heavily contaminated with individual elements. On the other hand, bottom sediments in the backflow zone of the Dzierżno Duże Reservoir contain no Hg in individual samples; apart from this element, they exhibit contamination ranging from moderate (e.g. As) through heavy (e.g. Cu, Zn) to extreme (e.g. S).

Table 4. Geoaccumulation index (I_{geo}) values.
Tabela 4. Wartości indeksu geoakumulacyjnego (I_{geo}).
Таблица 4. Значения геоаккумулятивного индекса (I_{geo}).

Component	Bottom sediments of the Otmuchów Reservoir			Bottom sediments of the Dzierżno Duże Reservoir		
	minimum	maximum	average	minimum	maximum	mean
As	2.38	2.93	2.77	1.18	1.49	1.34
Ba	2.59	2.78	2.67	3.85	4.52	4.25
Co	1.42	1.64	1.53	1.92	2.42	2.17
Cr	3.55	3.89	3.75	3.55	4.09	3.81
Cu	0.51	0.93	0.78	3.28	3.58	3.46
Hg	-0.91	1.33	-0.04	0.34	1.70	1.20
Ni	1.42	1.83	1.58	2.42	2.61	2.52
Pb	1.55	1.80	1.67	2.75	3.35	2.93
S	-1.26	0.55	-0.45	4.61	5.20	4.85
Sr	1.45	1.90	1.65	2.73	2.85	2.82
V	2.47	2.86	2.63	2.78	3.15	2.96
Zn	0.09	0.32	0.24	3.09	3.45	3.27

Sediment contamination factor values (C^i_f) indicate that the levels of sediment contamination in the water bodies studied differ, thus reflecting different degrees of human pressure (table 5). Sediments in the Otmuchów Reservoir are significantly contaminated with arsenic, slightly contaminated with chromium and not contaminated with Pb, Zn, Cd, Cu, Hg or PCBs. Sediments in the Dzierżno Duże Reservoir are much more contaminated. They exhibit very high Cd contamination levels, significant Zn contamination levels, moderate Cu, Pb, PCB, As and Cr contamination levels and the absence of Hg contamination.

Table 5. Values of the sediment contamination factor (C^i_f) and degree (C_d)
Tabela 5. Wartości wskaźnika (C^i_f) i stopnia (C_d) zanieczyszczenia osadów
Таблица 5. Значения показателя (C^i_f) и степени (C_d) загрязнения осадков

Component	Geochemical background [a]	Bottom sediments of the Otmuchów Reservoir		Bottom sediments of the Dzierżno Duże Reservoir	
		C^i_f	C_d	C^i_f	C_d
As	15.0 ppm	3.4	7.3	1.3	22.6
Cd	1.0 ppm	0.5		8.5	
Cr	90.0 ppm	1.1		1.2	
Cu	50.0 ppm	0.4		2.3	
Hg	250.0 ppb	0.3		0.7	
Pb	70.0 ppm	0.9		2.1	
PCB	0.01 ppm	0.1		1.4	
Zn	0.6 ppm	0.6		5.1	

Explanation: [a] according to: L. HÅKANSON (1980)

The degree of contamination of sediments is low in the case of the Otmuchów Reservoir (C_d = 7.3) and in the case of the Dzierżno Duże Reservoir it points to significant contamination (C_d = 22.6).

CONCLUSIONS

1. The chemical composition of bottom sediments in the Otmuchów and Dzierżno Duże Reservoirs exhibits differences in terms of the presence of basic components. The differences in geological structure and land use within the catchment are reflected by the dominance of the following substances in the bottom sediments of the Otmuchów Reservoir: SiO_2 (68.20–79.34%), Al_2O_3 (8.31–10.23%), organic matter (2.07–8.88%) and Fe_2O_3 (2.98–3.80%), with different proportions of these components in the sediments found in the Dzierżno Duże Reservoir (organic matter – 44.30–58.12%; SiO_2 – 20.78–28.45%, Al_2O_3 – 7.61–9.22%, Fe_2O_3 – 4.39–6.59%); the other oxides also differed in the order of their percentage shares.
2. Most of the trace elements analysed were detected in the bottom sediments of the water bodies examined in amounts corresponding to the concentration ranges found in sedimentary rocks. Although the ratio of the values found to the geochemical background for aquatic sediments in Poland was not determined for many elements owing to the absence of relevant data, some of the microelements (As, Ba, Co, Cr, Cu, Ni, Pb, S, Sr, V and Zn) deviate significantly from background levels as evidenced by the ratios of their concentrations to the regional geochemical background, which amounted to $1.10 < I_{RE} < 20.20$ (Otmuchów bottom sediments) and $3.44 < I_{RE} < 43.13$ (Dzierżno Duże bottom sediments).
3. Using the geoaccumulation index, it was found that bottom sediments in the Otmuchów Reservoir range from virtually uncontaminated to heavily contaminated ($-1.26 < I_{geo} < 3.89$), while those in the Dzierżno Duże Reservoir range from uncontaminated to extremely contaminated ($0.34 < I_{geo} < 5.20$). Contamination indicators point to moderate contamination of the bottom sediments in the Otmuchów Reservoir with As and Cr and the absence of contamination with Pb, Zn, Cd, Cu, Hg and PCBs; on the other hand, sediments in the Dzierżno Duże Reservoir exhibit very high Cd contamination levels, significant Zn contamination levels, moderate Cu, Pb, PCB, As and Cr contamination levels and the absence of Hg contamination. The degree of contamination of bottom sediments is low in the case of the Otmuchów water body (C_d = 5.2) and points to significant contamination in the case of the Dzierżno Duże water body (C_d = 22.6).
4. The bottom sediments of the water bodies studied were enriched with: Pb, Ni, Co, Cr, V, Sr, As and Ba in the Otmuchów Reservoir ($1.09 < I_{AP} < 5.14$) and As, Hg, Cr, Pb, Co, Ni, V, Zn, Cu, Sr, Ba and S in the Dzierżno Duże Reservoir ($1.73 < I_{AP} < 29.75$). The enriched chemical composition of sediments reflects the accumulation of trace elements in bottom sediments; since the geochemical background has been exceeded, this should be equated with contamination.

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