Title: The properties of reservoir water in post-mining excavations of Cambrian and Devonian quartzite sandstones (Holy Cross Mountains)

Author: Tadeusz Molenda, Tadeusz Ciupa, Roman Suligowski

ORIGINAL ARTICLE

The properties of reservoir water in post-mining excavations of Cambrian and Devonian quartzite sandstones (Holy Cross Mountains)

Tadeusz Molenda[1] · Tadeusz Ciupa[2] · Roman Suligowski[2]

Abstract

In the Holy Cross Mountains (southern Poland), there are numerous disused quarries. Some of these are filled with water, and some, despite their nearby location, have extremely diverse physicochemical and chemical properties of their waters. One such object is the Wiśniówka Mała reservoir. Its waters contain large amounts of sulfates (> 700 mg/L) and iron (24 mg/L), which are weathering products of rocks containing metal sulfides (mainly pyrite) in the direct drainage zone. As a consequence, there is an increase in the electrolytic conductivity of the water supplying the reservoir, resulting in very low pH values (< 4). This article presents the detailed limnological characteristics of this reservoir and explains the process that led to its water acidification. A control reservoir, Barcza, was also selected for the tests. Although it is also a post-mining excavation, it has a neutral pH and a low concentration of sulfates and iron. The examined reservoirs differ in the hydrochemical type of waters. The Wiśniówka Mała reservoir represents a sulfate–calcium double-ion type (SO_4^{2-}–Ca^{2+}) and the Barcza reservoir, a bicarbonate–calcium type (HCO_3^-–Ca^{2+}). The concentrations of the main ions occurring here are shaped by the lithological features of the surrounding rocks (respectively: Upper Cambrian quartzite sandstones with pyrite mineralization zones, Lower Devonian sandstones cut by mudstones and claystones with tuffite inserts).

Keywords Pit lake · Water geochemistry · Water–rock interactions

Introduction

In recent years, the interest of many researchers has been focused on the water that fills the post-excavation pits in which the excavated materials were magma sedimentary rocks of various ages, including metal ores (Lentz 2002; Ramstedt et al. 2003; Pellicori et al. 2005; Stevens et al. 2005; Balistrieri et al. 2006; Hrdinka et al. 2013; Tucci and Gammons 2015) and rock raw materials (Hrdinka 2007; España et al. 2008; Molenda 2015; Jawecki et al. 2018;

Tadeusz Molenda, Tadeusz Ciupa and Roman Suligowski have contributed equally to this work.

✉ Roman Suligowski
  rsulig@ujk.edu.pl

[1] Faculty of Earth Sciences, University of Silesia, Sosnowiec, Poland

[2] Institute of Geography and Environmental Sciences, Jan Kochanowski University in Kielce, Uniwersytecka 7, 25-406 Kielce, Poland

Pociecha et al. 2018). The research results presented in these papers concern two main aspects, i.e., the occurrence of meromixing in the deeper reservoirs and a recognition of the selected features of water chemistry (Galas 2003; Boehrer and Schultze 2006; Molenda 2014a, b; Kubiak et al. 2018). After the exploitation of the raw materials and the following abandonment of the mines, most of them are flooded and become valuable natural water bodies (Luoma and Rainbow 2008). However, the water in some flooded quarries may be a local threat to the local natural environment (Nordstrom 2011, Migaszewski et al. 2014, 2018b), because it has unfavorable physical and chemical parameters (Migaszewski et al. 2007; España et al. 2008) and a high concentration of trace elements (Williams and Smith 2000; Bozau et al. 2004; Szarek-Gwiazda and Żurek 2006; Triantafyllidis and Skarpelis 2006; Migaszewski et al. 2008, 2016, 2018a, 2019). These include the reservoirs in which the properties of the water are determined by the course of natural geochemical processes that are caused by the presence of metal sulfides, mainly pyrite (the effect is a very low pH value) in the underground and acid mine drainage (AMD) (e.g.,

Johannesson and Zhou 1999; Blowes et al. 2003; Karakas et al. 2003; Migaszewski et al. 2013; Grawunder et al. 2014). This type of water reservoir, which is in the disused quarry of quartzite sandstones, is located in the Wiśniówka mining area in the Holy Cross Mountains (Poland). A few kilometers from this water reservoir, also in a closed sandstone

quarry, but without traces of pyrite mineralization, a water reservoir (called Barcza) was created. Therefore, a comparative analysis of the physicochemical and chemical properties of the waters of these two reservoirs is both desirable and interesting.

Materials and methods

The objects of the research were two flooded quarries—Wiśniówka Mała and Barcza, which were created as a result of the exploitation of various age-old sandstones in the Holy Cross Mountains (Fig. 1). The location of these water reservoirs is unique from a geological point of view. The Holy Cross Mountains are part of one of the most well-known geotectonic structures in Europe—the Trans European Suture Zone (Berthelsen 1992; Nawrocki and Poprawa 2006; Żylińska et al. 2006). The mountains lie at the junction of two or, according to other authors, three great geological units (Stupnicka 1989; Kowalczewski and Dadlez 1996; Lamarche et al. 2002): the East European Precambrian Platform, the West European Platform of Paleozoic Consolidation and the Lesser Poland Bloc—in the rock mass and Alpine basins (Fig. 1a).

Location in the vicinity of the above units resulted in the formation of a complex system of tectonic structures, including the dislocation of the Holy Cross Mountains region and numerous transverse faults. At that time, igneous and hydrothermal phenomena occurred here (Kowalczewski and Dadlez 1996; Mizerski 1998).

The Wiśniówka Mała reservoir (Fig. 2a) is located in the Masłowskie Range within the Paleozoic core of the Łysogóry Block. Quartzitic sandstones from the middle and upper Cambrian (Furongian) with siltstone, clayey
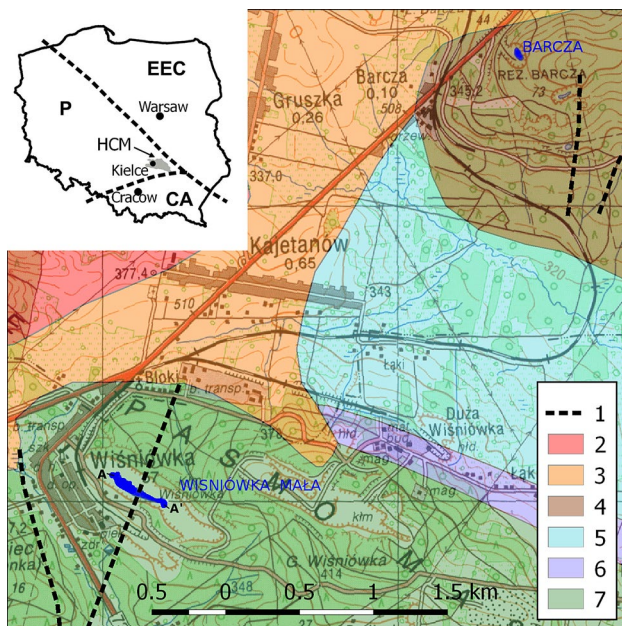
Fig. 1 Location of the water reservoirs Wiśniówka Mała and Barcza against: **a** the large geotectonic structures (Żylińska et al. 2006)—HCM (Holy Cross Mountains), EEC (Precambrian East European Craton), P (Paleozoic, i.e. the Caledonian and Variscan fold belt), CA (Carpathian Alpine orogen) and **b** a simplified uncovered geological map (after Filonowicz 1973). 1—main faults, rock age: 2—Triassic, 3—Permian, 4—Devonian, 5—Silurian, 6—Ordovician, 7—Cambrian, A–A′—longitudinal profile

Fig. 2 Wiśniówka Mała (**a**) and Barcza (**b**) reservoirs
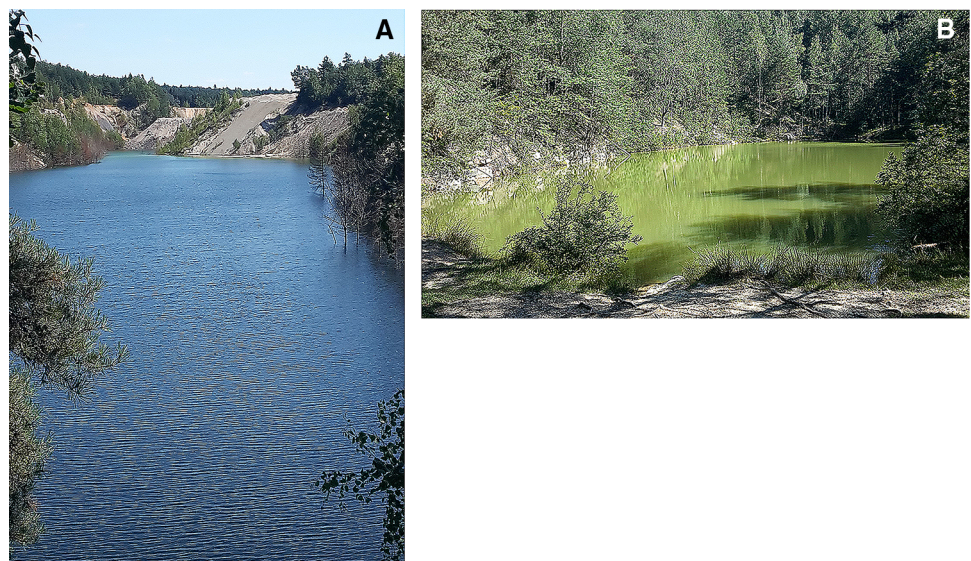
shale (with muscovite and illite), tuffite and bentonite interbeds are found in the area. Together, they form the Wiśniówka Sandstone Formation (Orłowski 1992; Żylińska et al. 2006). Detailed geochemical research conducted in the "Wiśniówka" mining area (4.1 km^2) in recent years by Migaszewski et al. (2007, 2008, 2009, 2013, 2014, 2018a, b) indicate that within these rocks occur pyrite veins, wavellite and variscite accumulation, as well as many supergene minerals (iron oxyhydroxysulfates and oxyhydroxides). The detrital accessory minerals are dominated by zircon, rutile, monazite and tourmaline.

Currently, water from the drainage of the operating mine is used to rinse the extracted aggregate and then is discharged into the Wiśniówka Mała excavation. Its significant depth makes it possible to take post-mining waters of a considerable volume, despite the slope position of the quarry. However, no water leakage or dispersion outside of the boundaries of the mining area was observed (Migaszewski et al. 2016). This water has a very low pH (< 4.0), which is caused by pyrite mineralisation zones in the discussed mining area, which was documented by Migaszewski and Gałuszka (2010) on the basis of detailed mineralogical and petrological studies of the rocks in the disused Podwiśniówka quarry in 2006.

In the Barcza quarry (Fig. 2b), which is located in the Klonowskie Range about four km NE of the Wiśniówka Mała, the extracted rocks were Lower Devonian (Emsian) sandstones and quartzitic sandstones with interbeds of mudstones and claystones, which are classified as the so-called Barczan Formation (Filonowicz 1973; Łobanowski 1990). The quarry has a thin layer of tuffites, which documents the volcanic activity in the Variscan orogeny (Kardymowicz 1960a; Fijałkowska-Mader and Malec 2018) that has a rare mineral called celadonite (Kardymowicz 1960b), which, when dissolved in the water of the reservoir, gives it a dark green colour. This small water reservoir has an almost oval shape. The groundwater inflow into the Barcza excavation is permanent, while the inflow of the surface water is periodic and mainly occurs in spring (thawing) and summer (precipitation). The periodic outflow of excess water from the reservoir is via a ditch. The reservoir, which is located in a forest complex, is part of an inanimate nature reserve that was established in 1984.

In 2016–2018, field hydrogeochemical studies were conducted in the characteristic seasons—spring and autumn (circulation period) as well as in summer and winter (stagnation period). The depths of the lakes were measured from a pontoon using a LOWRANCE HDS 5—Gen 2 echo sounder with a built-in GPS receiver. During the measurements, information about the depth and the coordinates of the point were recorded for each of the signals (impulses) that were sent by the sonar. It is a widely used technique that permits the quick and precise imaging of the bottom of

water reservoirs. Bathymetric plans of the reservoirs were based on the sonar data using ArcMap 10.2.1. The same program allowed the transverse and longitudinal profiles of the examined objects to be drawn.

The basic physicochemical and chemical properties of water (temperature, pH, electrolytic conductivity and turbidity) were measured directly in the field using a YSI multi-parameter EDS 6600 probe. The above-mentioned water parameters were determined in a vertical column in the reservoir every 1 m at the deepest section (Figs. 3, 4). Moreover, the transparency of the water was examined using a Secchi disc.

Water samples for the chemical analyses were taken from the surface layer directly into polyethylene bottles and from the bottom layer (about 0.5 m above the bottom) using an Eijkelkamp pressure gauge. Laboratory analyses of the chemical composition of the water were preceded by its filtration on a 0.45-µm filter (Millipore). The content of the main cations and anions in the water: Ca^{2+}, Mg^{2+}, Na^+, K^+, SO_4^{2-}, Cl^- was determined using a Metrohm 850 Professional IC ion chromatograph, HCO_3^-—titration method with b-r basicity indicator. Additionally, once (19 June 2017), the concentration of iron (Fe_{tot}) and selected trace elements were determined in the vertical column (every 2 m) of the Wiśniówka Mała reservoir using an Inductively Coupled Plasma-Quadropole Mass Spectrometer (ICP-QMS; ELAN DRC II model, Perkin Elmer). A set of Perkin Elmer Multi-element Calibration Standards solutions were used.

Classification of the hydrochemical types of water was based on the classification of Szczukariew-Prikłoński
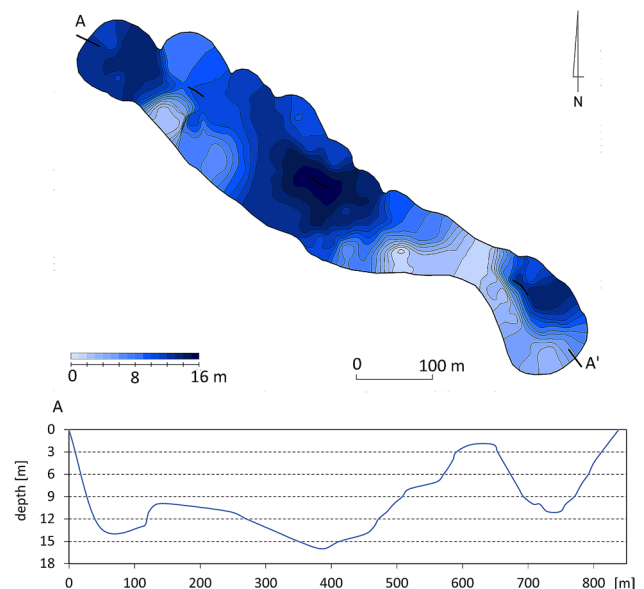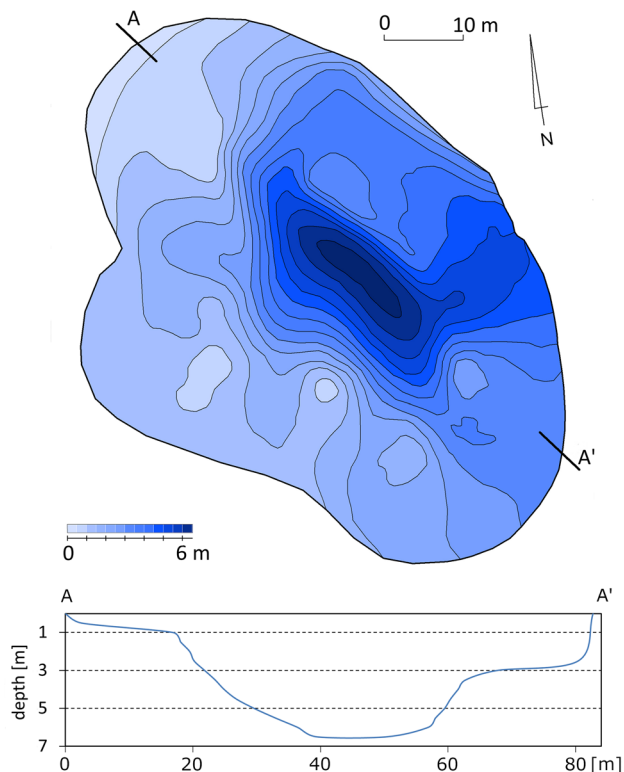

Fig. 3 Bathymetry of the Wiśniówka Mała reservoir. A–A′—longitudinal profile

(Macioszczyk and Dobrzyński 2007). Basic statistical analyses were performed using the R (R Core Team 2014) program.

Results and discussion

The Wiśniówka Mała reservoir, an area of 5.2 ha, has an average depth of 10.9 m, maximum depth of 16 m and the water table at approx. 366 m a.s.l. (Fig. 3).

The shoreline of the Barcza reservoir reflects the shape of the post-mining excavation. Compared with the Wiśniówka Mała, it is a shallow reservoir with an average depth of 2.9 m and a maximum depth of 6.5 m (Fig. 4). It is a reservoir with a small surface area of 0.3 ha, while the water table is at approximately 372 m a.s.l. The water supply of both lakes

Fig. 4 Bathymetry of the Barcza reservoir. A–A′—longitudinal profile

occurs through the groundwater inflow and surface runoff from their direct catchments. In the case of the Wiśniówka Mała reservoir, there is also a forced water circulation that is associated with both the intake and discharge of water into the reservoir, on which there is a pumping station that captures water for the technological purposes of the mining plant. These waters are used to rinse the spoil, after which they are discharged into the reservoir as industrial waste. These waters are very heavily loaded with mineral slurry. The suspension is indicative of their turbidity, which is above 50 NTU. This corresponds to approximately 50 mg/L SiO_2. Therefore, in the place where their water is discharged into the reservoir, an inflow fan has developed. It is very well visible on the bathymetric plan of the reservoir (Fig. 3).

The discharge of the technological waters also affects the transparency of the water in the reservoir. In the drop zone, the visibility of Secchi disk was only a few tens of centimetres and it increased as it moved away from the drop point. In the north-western part of the reservoir, the visibility varied from 1.2 to 4.0 m. In addition to the discharge of the technological waters, in the south-eastern part of the reservoir, gangue rocks from the exploitation of an adjacent quarry are deposited. This caused a change in the original coastline of the reservoir. Within 20 years, the area of the reservoir decreased by about 2 ha. There are also other landfills in the direct basin of the reservoir. Their total area is about 8 ha.
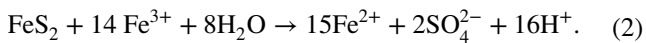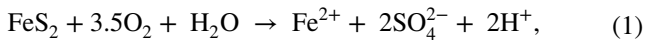
According to the classification of Szczukariew-Prikłoński (Macioszczyk and Dobrzyński 2007), the water in the tested reservoirs is double ion. In the Wiśniówka Mała reservoir, it represents the sulfate–calcium type (SO_4^{2-}–Ca^{2+}), and in the Barcza reservoir, the bicarbonate–calcium type (HCO_3^{-}–Ca^{2+}). The different hydrochemical types are the result of the concentration of the main ions (Table 1).

In the water of the Wiśniówka Mała reservoir, no bicarbonate ions were found. There was a very high concentration of sulfate ions, whose average concentration was 706 mg/L. For comparison, the mean concentration of this ion in the Barcza reservoir was only 7.4 mg/L. The concentrations of the remaining ions (Ca^{2+}, Mg^{2+}, Na^+, K^+, Cl^-, NO_3^-) in the water of the Wiśniówka Mała reservoir were much higher than in the Barcza reservoir. However, no phosphate ions (PO_4^{3-}) were found in the water of either reservoir (Table 1). The water of the Barcza reservoir represented a bicarbonate–calcium hydrochemical type, which is the most

Table 1 Median pH, average EC and concentration of the main ions (mg/L) with the standard deviation (in parentheses) in the near-surface layer of the waters of the tested reservoirs ($n=6$)

Reservoir	pH	EC μS/cm	Ca²⁺ mg/L	Mg²⁺	Na⁺	K⁺	HCO₃⁻	Cl⁻	NO₃⁻	PO₄³⁻	SO₄²⁻
Wiśniówka Mała	2.6 (0.3)	1389 (239)	104.8 (8.2)	14.9 (1.2)	5.6 (0.2)	5.8 (0.7)	< 0.1	7.6 (0.5)	7.6 (0.5)	< 0.1	706.3 (101.4)
Barcza	6.0 (0.3)	66 (12)	8.6 (1.2)	1.2 (0.4)	1.2 (0.1)	1.6 (0.1)	18.0 (1.4)	3.5 (0.2)	3.5 (0.2)	< 0.1	7.4 (1.8)

commonly occurring in nature (Allen 2008). This type of water is determined by the lithology of the basins and their catchments as well as anthropogenic influences. The Cambrian rocks in the Wiśniówka quarry, as was already mentioned, contain metal sulfides, primarily pyrite. The process of their weathering is responsible for the increase in the concentration of sulfate ions; the following chemical reaction describes this phenomenon (Garrels and Thompson 1960):

$$FeS_2 + 3.5O_2 + H_2O \rightarrow Fe^{2+} + 2SO_4^{2-} + 2H^+, \tag{1}$$

$$FeS_2 + 14\,Fe^{3+} + 8H_2O \rightarrow 15Fe^{2+} + 2SO_4^{2-} + 16H^+. \tag{2}$$

The oxidation of pyrite also releases considerable amounts of iron ions. The weathering of pyrite occurs on both the walls of the quarry and on the heaps that are located around the reservoir (Migaszewski et al. 2013). In the depressions in the neighboring quarry in Podwiśniówka, strongly acidic leachate waters (< 2 pH) of very high electrolytic conductivity ($> 10,000$ µS/cm) were found (Migaszewski et al. 2007). A similar pH value was also found in the leachate that was related to the weathering of the sulfide minerals in quarries that are located in other regions of Poland (Molenda 2011). The source of low pH and the slightly increased content of metals and arsenic are also due to the technological waters that are discharged into the reservoir and that originate from rinsing the aggregate that is obtained from the Podwiśniówka quarry. The result is a very high content of As in the water of the Wiśniówka Mała reservoir, which reached 22.8 µg/L at the surface in June 2017, and which increased with depth—in the bottom layer (at a depth of 12 m) it was as much as 62.4 µg/L (Table 2). The concentrations of selected heavy metals and trace elements that were found in the vertical column (every 2 m) in the deepest part of the Wiśniówka Mała reservoir are presented in Table 2. These concentrations were not related to the depth of the reservoir, except for the downward trend in the case of Ba and Cu.

Differences in the physicochemical and chemical properties of the water in the studied reservoirs provide the best evidence of water mineralisation. The determined value of electrolytic conductivity in the Barcza reservoir—66 µS/cm is comparable to that obtained in other post-mining lakes, including those located in crystalline rocks (Molenda 2014a). The electrolytic conductivity of the Wiśniówka Mała reservoir, however, was comparable to those found in the reservoirs that receive leachate from landfill sites that contain metal sulfides (Molenda 2011). It is worth noting that electrolytic conductivity increases with time. In 2005, the average value of the electrolytic conductivity in the surface water layer was 450 µS/cm (Migaszewski et al. 2009), in 2014–2016—1100 µS/cm (Migaszewski et al. 2016), and currently, (2016–2018), it is 1389 µS/cm. In 13 years, it has increased by more than 900 µS/cm. During this period, there was also an increase in the iron concentration ($Fe_{tot.}$) from 0.3 mg/L in 2005 (Migaszewski et al. 2009) to 24 mg/L currently recorded. These changes result from rinsing the aggregate that is enriched with pyrite from the Wiśniówka Duża quarry.

In the vertical profile of the Wiśniówka Mała reservoir, the distribution of electrolytic conductivity in the summer is interesting (Fig. 6). In the thermocline layer, which was identified from the thermal profile, an apparent increase in the electrolytic conductivity of the water was observed. Under the influence of a temperature drop, the density of the water increased at this point, which caused a slowdown of the mineral particles sinking towards the bottom and influenced the increase in the electrolytic conductivity of the water. A similar process has also been observed in other reservoirs with a high water electrolytic conductivity (Molenda 2014b, 2018).

An analogous situation was not found in the case of the Barcza reservoir. The values of the electrolytic conductivity were aligned in the whole vertical profile; only in the zone above the bottom was a small increase observed (Fig. 6). Both in the spring and autumn, there were no apparent differences in the electrolytic conductivity in the vertical column in the case of both reservoirs, which testifies to the full mixing and homogenisation of the waters. This is a typical phenomenon for reservoirs that are located in the mid-latitudes.

Table 2 Concentration of Fe_{tot} and selected trace elements (µg/L) in the water column (every 2 m) of the Wiśniówka Mała reservoir on 19 June 2017

| Depth m	As µg/L	Ba	Cd	Co	Cr	Cu	Fe_{tot}	Mn	Ni	Pb	Tl	U	Zn
0	22.8	11.4	1.05	207	112	828	24000	8739	198	0.73	0.35	16.8	309
2	25.2	11.8	1.11	213	115	870	24800	9192	250	3.49	0.36	16.5	379
4	55.0	8.99	1.05	210	117	811	28750	9285	227	1.66	0.30	16.3	316
6	62.0	8.75	1.03	211	116	797	22000	9202	224	1.13	0.36	17.0	318
8	59.5	8.54	1.01	209	118	793	28200	9435	225	0.64	0.34	16.2	300
10	62.0	8.92	1.26	210	118	798	22550	9382	227	1.35	0.36	17.1	307
12	62.4	8.51	1.06	208	116	788	25200	9288	225	0.64	0.33	16.1	298

A characteristic feature of the Wiśniówka Mała reservoir was also the acid reaction of its water. During the study period, its median was 3.2 pH. For this reason, it was classified as acidotrophic. Similar, or even lower, pH values were found in other acidotrophic reservoirs that fill the bottoms of inactive post-mining excavations (Bednar et al. 2005; España et al. 2008) or those receiving acid leachate from industrial waste dumps (Molenda 2014b). The distribution of water in the vertical column of the Wiśniówka Mała reservoir was also unusual. In the summer, a drop in water pH

in the thermocline was observed (Fig. 5), which is the result of the accumulation of sulphuric acid and pyrite oxidation products in the zone in which the density of water increased under the influence of a temperature drop. The decrease in pH was synchronous with an increase in mineralisation in the thermocline zone (Fig. 5).

In the Barcza reservoir, the pH was also acidic, and its median was 6.0 pH. This weak acid water reaction is characteristic of post-mining lakes whose basins are cut into crystalline rocks (Molenda 2015), which contain no buffering
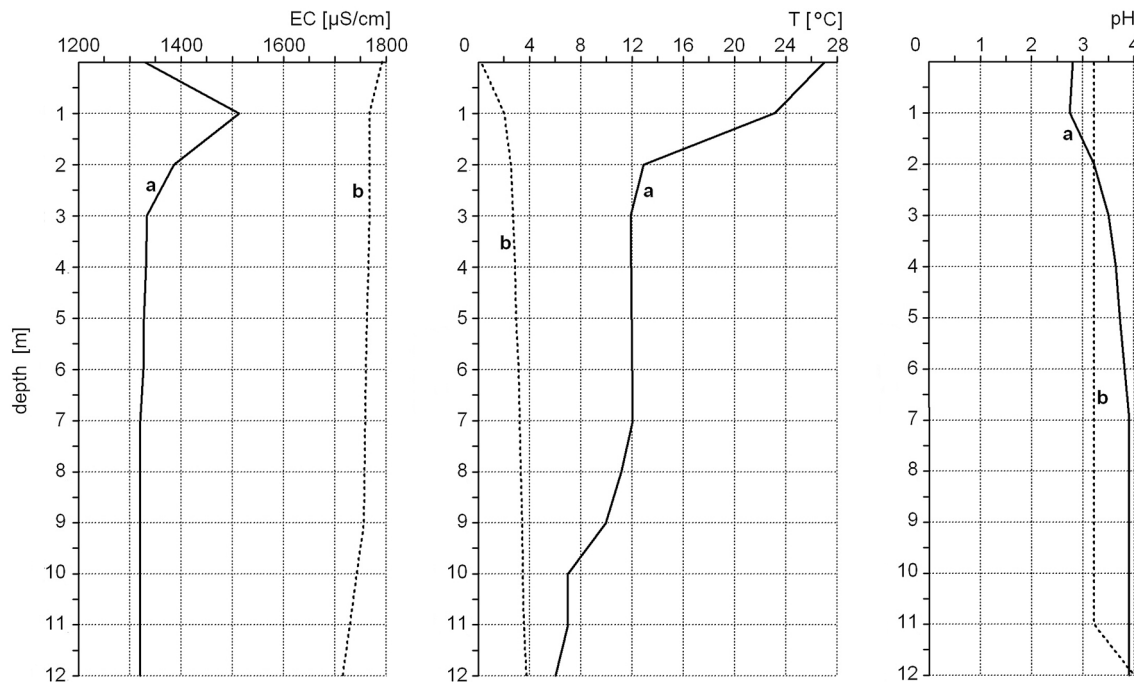
Fig. 5 Distribution of electrolytic conductivity, temperature and pH in the water column of the Wiśniówka Mała reservoir: **a** summer; **b** winter
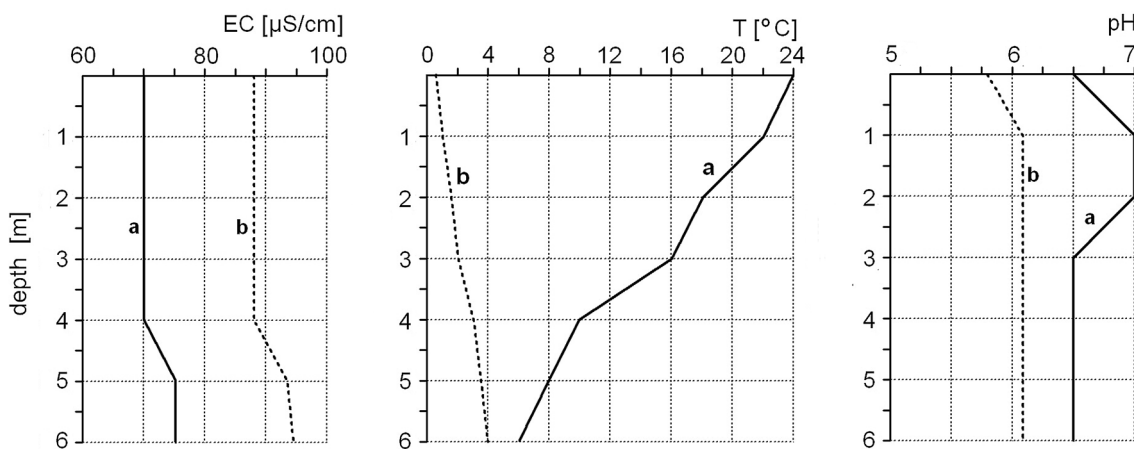
Fig. 6 Distribution of electrolytic conductivity, temperature and pH in the water column of the Barcza reservoir: **a** summer; **b** winter

components, e.g., carbonates, and no mineralisation of metal sulfides, especially pyrites. The pH value of the water in these reservoirs corresponds approximately to the reaction of rainwater. In summer, an increase pH was found at a depth of 2 m (Fig. 6), which was a result of photosynthesis and CO_2 consumption that was also responsible for water acidity.

The distribution of the water temperature in the vertical column of the Wiśniówka Mała reservoir was characteristic for particular seasons. In summer, stratification was found, and in winter—katothermy (Fig. 5) with a full mixing in spring and autumn. An analogous water temperature distribution was found in the Barcza reservoir (Fig. 6).

Conclusions

The chemical composition of the waters in the reservoirs that were formed in the excavations after the exploitation of quartzite sandstones in the Holy Cross Mountains (Poland) is determined by the course of the natural geochemical processes. In the Wiśniówka Mała, which is an endorheic reservoir that is fed with water coming from active mine drainage, there is a specific accumulation of Fe^{2+}, Fe^{3+} and SO_4^{2-} ions. These are the products of pyrite weathering in the zone of the underground surface drainage of the rock formation, which consequently leads to an increase in the electrolytic conductivity of the waters and very low pH values. On the other hand, in the Barcza reservoir, in which pyrite mineralisation does not occur in the surrounding rocks and the chemical composition of water is not modified by human intervention (nature reserve), the most common hydrochemical type of water, bicarbonate–calcium, has been formed.

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