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# The Influence of Lignite Mining on Water Quality

Barbara Jachimko  
*University of Zielona Góra*  
*Poland*

## 1. Introduction

Mining is one of the oldest human activities in the world. Due to long history and changes in the economic situation there are many abandoned mine areas throughout the world. Mining is an industrial branch which is very destructive to the environment. It influences the environment in a number of ways, such as landscape conversion, changes in the water regime and a negative influence the quality of water. Waters polluted by mining wastes are called AMD (acid mine drainage). AMD, connected to lignite mining are extremely acidic and enriched with iron, sulphate and heavy metals. The pollution was caused by weathering products of different sulphides (mainly pyrite  $\text{FeS}_2$ ), which exposed to water and oxidant weather and release protons, sulphate, ferrous and heavy metals into the water. It threatens to the biota and even completely destroys life for a long, difficult to estimate time. There are two ways of preventing such pollution: prevention of AMD formation (activity by source) and treatment AMD already formed. The aim of treatment techniques is to obtain a long-term effect after the treatment activity has been stopped. Until now no success has been achieved. A very interesting example of the influence of lignite mining on water quality is the “anthropogenic lake district” located in Mużakowski Łuk (Western Poland). It comprises about 100 reservoirs of different origin, size and chemical composition, which have never been reclaimed. Among them there are acidic reservoirs of the meromixic type, whose chemical properties have been monitored almost since the appearance of the youngest one, i.e. 1982. On the basis of almost thirty year long observations some interesting remarks, presented below have been formulated. The most important is the difference between the chemical composition of the upper and lower layer, especially the pH value. The chapter presents short review of the occurrence of post-mining areas, the chemistry of acid mine drainage and the description of the “anthropogenic lake district” as a case study of the influence of lignite mining on water quality.

## 2. Scale of the problem – Global problems with post-mining areas

World lignite deposits are mainly located in Australia, China, some EU countries, Russia, and the USA. China, the world’s largest coal producer now accounts for nearly 40 per cent of the world total. Brown coal (lignite) is burnt in power stations for the production of electricity and is also used for other domestic and industrial purposes (steam coal). In 2007, in Europe, the biggest amounts of lignite were mined in Germany –  $180 \cdot 10^6$  t, Turkey  $74 \cdot 10^6$

t, Greece  $66 \cdot 10^6$  t, Poland –  $57 \cdot 10^6$  t, Czech Republic –  $49 \cdot 10^6$  t, Romania –  $75 \cdot 10^6$  , Bulgaria –  $28 \cdot 10^6$  t. The are three main techniques of lignite mining: underground, open-cast and gasification (on the surface or underground). Nowadays, about 95 to 98% of lignite is mined by the open-cast method and a few percent by the underground one. The underground method was common from the 19<sup>th</sup> c. till the first half of 20<sup>th</sup> c. Then, it was discontinued due to low safety, efficiency and economical aspects. Many accidents and cases of excavation flooding occurred. The youngest mine technique is underground gasification in a temperature of ca.  $1200^\circ\text{C}$ . The method has not become popular due to numerous disadvantages (thermal conversion of the surrounding rocks, toxic gases and fumes, earth surface deformations, changes in the hydrological balance, groundwater pollution by minerals and benzene). Therefore, it can be used in uninhabited areas, such as deserts or sea shelves. (Kasztelewicz et al. 2008).

Europe has a long tradition of mining beginning with flint mines, then metals, such as copper, gold and silver, ending with coal. The Phoenicians mined metals in Spain in the Bronze Age. The first gold coins were produced in Bulgaria, Greece and Turkey around 700 BC. Techniques of underground mining and even dewatering were improved by The Romans. Some fields started to be abandoned in the middle of the nineteenth century and needed control of environmental impact and remediation (Wolkersdorfer & Howell, 2005). In 2008 the 32 EU countries produced 10 per cent of the world's coal (all types) of which approximately half came from Germany and Poland. (Brown et al. 2010) – Table 1. During the last 100 years in Germany 10 lignite mining districts come into being (Niederrheinischer in Nordrhein-Westfalen, Thueringsch-Saechsischer, now Mitteldeutsches, Lausitzer, Niederhessischer, Braunschweig-Magdeburger, Oberpfaelzer, Westerwald, Rhein-Main, Ostdeutcher –after the war in Poland, Schlesischer – after the war in Poland).

In Poland there are five lignite mines: Adamów, Bełchatów, Konin, Turów, and Sieniawa, with a total production of  $56 \cdot 10^6$  tonnes in 2010. In all of them lignite is produced by the open cast method, excluding "Sieniawa". "Sieniawa" located in Ziemia Lubuska, is one of the oldest brown coal fields in Poland. It was discovered in the middle of the 19<sup>th</sup> c. The exploitation started in 1873 in the mine called "Emiliensglück". The mine was destroyed during the Second Word War. Then it was activated in 1950 as the mine "Sieniawa". In 1983 it produced about  $209 \cdot 10^3$  tonnes of coal. More then half of total production, ca. 33 mln Mg per year came from Bełchatów, 20% from Turów, 17% from Konin and 8,5% from Adamów.

### 3. Influence of mine activity on water resources

Mining must be preceded by seam dewatering. It changes the hydrological regime and upsets the water balance. Lowering the water table causes lack of water in domestic wells, it decreases soil humidity and river flow. After closure many pits are filled back with groundwater, which had been earlier polluted by discharge from mining wastes and overburden material. In Poland in 2002, about 361 million  $\text{m}^3$  of water were pumped from open lignite mines, producing cones of depression of  $1200 \text{ km}^2$  (Wilk, 2003). In the "Bełchatów" mine (Poland) the cone of depression was about  $730 \text{ km}^2$ . In Poland the drainage water is generally of good quality and not environmentally hazardous, with the exception of water from Turów with the salinity of over 5 g/l. Some of the serious AMD problems occur in Germany, in the Lausitz and around Leipzig. They come from the high AMD potential and deficit of water in the area. About one third of the total amount of about

500 open-cast pit lakes in Germany were acid with a pH below 3 (Fyson A., Nixdorf B., Kalin M., 2006). 168 pit lakes have been documented in the Lausitz and approximately half of them are extremely acidic (pH 2.4-3.4) (Table 1). Acidic mine lakes in the Lausitz contain huge amount iron (up to 800 mg/dm<sup>3</sup>) and sulphate (up to 4636 mg/dm<sup>3</sup>).

Parameter	unit	minimum	maximum
pH	-	2.4	3.4
Acidity	mmol/dm <sup>3</sup>	0.1	26.6
Total Fe	mg/dm <sup>3</sup>	0.2	800
Sulphate	mg/dm <sup>3</sup>	460	4636
Total organic	mgC/dm <sup>3</sup>	0.8	10.9
Total N	mg/dm <sup>3</sup>	0.9	5.3
Total P	µg/dm <sup>3</sup>	4	26

Table 1. Summary of some chemical parameters for 75 acidic mine lakes in the Lausitz region (Nixdorf et al. 2001).

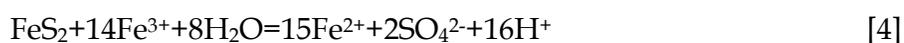
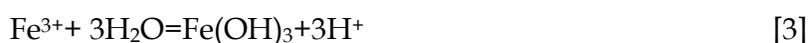
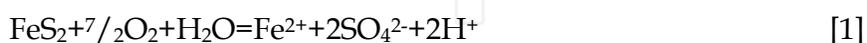
## 4. Chemistry of AMD

### 4.1 Weathering of minerals accompanying coal and metal ores

Mining affects the natural cycles of some elements, mainly sulphur and iron. It changes the natural base acid balance by the increase of the amount of sulphides minerals exposed to water and atmosphere. Protons and other acid ions released due to accelerated dissolution should be neutralised by the reactions of carbonates and silicates weathering. Coal deposits often contain very small quantities of base materials, which inhibits neutralisation. Sulphur-bearing minerals, such as iron sulphides, sphalerite (ZnS), chalcopyrite (CuFeS<sub>2</sub>), galena (PbS), arsenopyrite (FeAsS) often occur with coal and metal ores. The pyrite content in mine wastes was estimated at between 0,1 and 5% (weight) (Blodau, 2006). In Turów (Poland) the pyrite content was in the range of 0,3 to 0,5% (Kwiatkowski, 2006) and in Sieniawa below 0,8%. Iron-disulphides (FeS<sub>2</sub>) exist in two main forms: pyrite and marcasite, which have the same chemical composition and different crystallography. Pyrite is the most stable and prevalent form.

Pyrite oxidation is the main process involved in acidity generation. Two oxidants are involved in pyrite oxidation : oxygen and ferric ion, Fe (III).

The reactions of pyrite weathering under the exposure to air and water are as follows:



According to the reactions [1] and [4] ferrous and sulphate ions are released at the rate of 1:2 (Lenk & Wisotzky, 2001). In the first stage sulphide oxidation one mole of oxidized pyrite gives two moles of protons and two moles of sulphides. In an anoxic milieu, ferrous ions are

inert and can exist in high concentrations. The reaction [4] describes pyrite oxidation by ferric ions (Fe(III)). Ferric ions coming from pyrite weathering supply reactive Fe(III) and allow the process to proceed. The rate constant of the reaction [2] depends on pH, so oxidation of ferrous to ferric in a pH below 3 is extremely slow (Nordstrom, 1982). In the acidic range (pH from 2,5 to 3,5) ferrous oxidation is catalysed by microorganisms. In a neutral to alkaline milieu Fe(III) concentration is limited by the precipitation of hydrous ferric oxides and the importance of the reaction [2] decreases. The reaction of pyrite oxidation is surface controlled (Evangelou, 1995) and the nature of products depends on the origin and shape of pyrite.

The final AMD chemistry depends on the following factors:

- amounts of sulphides exposed to weathering factors,
- weathering reaction rate,
- secondary reactions, such as ferric minerals precipitation and redox reactions,
- availability of base minerals.

#### 4.2 Secondary processes in the aspect of water acidity

After pyrite weathering products, such as protons, sulphates (VI) and ferrous ions are released into water, different secondary processes occur. The processes involve precipitation (mainly of iron, aluminium or calcium minerals), ion exchange, isomorphic substitution, redox reaction (often supported by microorganisms) and the transformation of solids. The processes influence water quality and on the basis of the impact on pH, they are classified as acidity or alkalinity generation. The most important process generating acidity is the precipitation of different ferric compounds (Table 2). Their accumulation in acid reservoirs is an important part of the iron cycle in the environment (Cornell & Schwertmann, 2000).

Mineral	Formula	Reaction
Ferrihydrite	$\text{Fe}(\text{OH})_3$	$\text{Fe}(\text{OH})_3 + 3\text{H}^+ \leftrightarrow \text{Fe}^{3+} + 3\text{H}_2\text{O}$
K-Jarosite	$\text{KFe}_3(\text{SO}_4)_2(\text{OH})_6$	$\text{K}^+ + 3\text{Fe}^{3+} + 2\text{SO}_4^{2-} \leftrightarrow \text{KFe}_3(\text{SO}_4)_2(\text{OH})_6 + 6\text{H}^+$
H-jarosite	$\text{HFe}_3(\text{SO}_4)_2(\text{OH})_6$	$3\text{Fe}^{3+} + 2\text{SO}_4^{2-} + 6\text{H}_2\text{O} \leftrightarrow \text{HFe}_3(\text{SO}_4)_2(\text{OH})_6 + 5\text{H}^+$
Schwertmannite	$\text{Fe}_8\text{O}_8(\text{OH})_x(\text{SO}_4)_y$	$\text{Fe}_8\text{O}_8(\text{OH})_x(\text{SO}_4)_y + (24-2y)\text{H}^+ \leftrightarrow 8\text{Fe}^{3+} + y\text{SO}_4^{2-} + (24-2y+x)/\text{H}_2\text{O}$
Goethite	$\text{FeOOH}$	$\text{FeOOH} + 3\text{H}^+ \leftrightarrow \text{Fe}^{3+} + 2\text{H}_2\text{O}$
melanterite	$\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$	$\text{Fe}^{3+} + \text{SO}_4^{2-} + 7\text{H}_2\text{O} \leftrightarrow \text{FeSO}_4 \cdot 7\text{H}_2\text{O}$
siderite	$\text{FeCO}_3$	$\text{Fe}^{3+} + \text{CO}_2(\text{g}) + \text{H}_2\text{O} \leftrightarrow \text{FeCO}_3 + 2\text{H}^+$

Table 2. Influence of ferric minerals on acidity generation

Goethite (named after Johann Wolfgang von Goethe) is the most thermodynamically stable phase at a pH > 2, but it occurs in minor quantities within precipitates, however, it is common in bottom sludge. Optimum pH conditions for schwertmannite formation were found between 2.8 and 3.2 (Bigham, 1996). Jarosite forms at a pH < 2.5 and ferrihydrite (basic formula  $\text{Fe}_5\text{O}_8\text{H} \cdot \text{H}_2\text{O}$ ) precipitates at a pH > 4.5 (Blodau, 2006).

According to Langmuir (1997) the highest pH value, for the potassium concentration of  $10^{-4}$  and sulphate of  $10^{-2}$  mol/kg the pH value is 3.8. Ferric atoms involved in the crystalline structure of minerals are often replaced by other ions, such as the substitution of aluminium

in goethite. The general formula of goethite incrustated by aluminium is  $Fe_{1-x}Al_x)_y/3O_{1-y}(OH)_{1+y}$ , where  $y$  may reach 0.25 (Wolska & Schwertman 1993).

Aluminium precipitates in the form of gibbsite and kaolinite and in the presence of high sulphate concentrations, in the form of different sulphates such as alunogen, alunite or jurbanite.

According to Nordstrom (1982), the stability for particular minerals, for the sulphate concentration of  $10^{-4}$  mol/kg, ranges as follows:

- 0-3.3 pH - jurbanite  $AlSO_4(OH) \cdot 5H_2O$
- 3.3-4.47 pH - alunite  $KAl_3(SO_4)_2(OH)_6$
- pH > 4.47 gibbsite  $Al(OH)_3$ .

Schwertmannite is metastable to goethite and transforms in solid phase with protons release, according to the reaction:



Another mineral, which often precipitates in post-mine water is gypsum:



The reaction [6] has no direct influence on the acidity balance (except for changing the concentration of  $HSO_4^-$ ), but it considerably limits mineralization.

Processes involved in acidity consumption include the dissolution of base minerals (calcite, dolomite, biotite, plagioclase, orthoclase), cation exchange and redox/precipitation reactions. In the case of lack of base minerals in soils pH may reach an extremely low value, even below 1. It accelerates the rate of silicates weathering and releases aluminium ions into the water. Redox processes with the participation of organic matter usage as a reducing agent result in ferric and sulphate reduction contrary to pyrite weathering. The processes of sulphate and iron reduction are controlled by electron donor availability (Blodau et al., 2000), temperature and pH (Koschorreck et al., 2003).

### 4.3 Acid base balance in AMD

In order to understand AMD chemistry and predicting the results of neutralisation strategies it is crucial to know the acid-base theory by Stumm & Morgan (1996). The theory was adopted to AMD by Uhlmann in Nixdorf & Deneke (2004). On the basis of hydrolysis reactions of weak anions ( $HSO_4^-$ ) or cations ( $Fe^{3+}$ ,  $Al^{3+}$ ) acidity and alkalinity can be described as follows:

$$Aci = [H^+] + 3[Fe^{3+}_{(aq)}] + 3[Al^{3+}_{(aq)}] - [HCO_3^-] - [OH^-] \quad [7]$$

$$Alk = [HCO_3^-] - [OH^-] - [H^+] - 3[Fe^{3+}_{(aq)}] - 3[Al^{3+}_{(aq)}] \quad [8]$$

where:  $[H^+]$ ,  $[Fe^{3+}_{(aq)}]$ ,  $[Al^{3+}_{(aq)}]$ ,  $[HCO_3^-]$ ,  $[OH^-]$  - the activity of the ion.

Calculations of alkalinity and acidity according to the above equations are difficult due to the occurrence of iron and aluminium in different intermediates (hydrolysis), mainly as

hydroxyl complexes. Acidity and alkalinity can also be calculated on the basis of strong anions and cations with the differentiation of oxic conditions. For an anoxic milieu acidity and alkalinity can be described according to the following equations:

$$Aci_{anox} = 2[SO_4^{2-}] + [Cl^-] + [NO_3^-] - 2[Ca^{2+}] - 2[Mg^{2+}] - [Na^+] - [K^+] - [NH_4^+] - 2[Fe^{2+}] \quad [9]$$

$$Alk_{anox} = 2[Ca^{2+}] + 2[Mg^{2+}] + [Na^+] + [K^+] + [NH_4^+] + 2[Fe^{2+}] - 2[SO_4^{2-}] - [Cl^-] - [NO_3^-] \quad [10]$$

Therefore under oxic conditions ferrous ions are oxidised to ferric, which is a weak base. And the above equations can be modified to:

$$Aci_{ox} = 2[SO_4^{2-}] + [Cl^-] + [NO_3^-] - 2[Ca^{2+}] - 2[Mg^{2+}] - [Na^+] - [K^+] - [NH_4^+] \quad [11]$$

$$Alk_{ox} = 2[Ca^{2+}] + 2[Mg^{2+}] + [Na^+] + [K^+] + [NH_4^+] - 2[SO_4^{2-}] - [Cl^-] - [NO_3^-] \quad [12]$$

In order to estimate the acidity of water from copper mines (high copper concentration), Evangelou (1995) formulated the neutralisation potential (NP), which is defined as follows:

$$NP = 2c_{CO_3^{2-}} + c_{HCO_3^-} + c_{OH^-} - c_{HSO_4^-} - c_{H^+} - 2c_{Fe^{2+}} - 3c_{Al^{3+}} - 2c_{Cu^{2+}} \quad [13]$$

c- ion concentration, mmol/dm<sup>3</sup>.

Schoepke (1999) adapted this expression for lignite mine water:

$$NP = 2c_{CO_3^{2-}} + c_{HCO_3^-} + c_{OH^-} - c_{HSO_4^-} - c_{H^+} - 2c_{Fe^{2+}} - 3c_{Al^{3+}} - 2c_{Fe^{3+}} - 2c_{Mn^{2+}} \quad [14]$$

If alkalinity according to Stumm & Morgan (1996) can be described as:

$$Alk = -c_{H^+} - c_{HSO_4^-} - 3c_{Fe^{3+}} + c_{HCO_3^-} \quad [15]$$

Then the NP can be shortened to:

$$NP \approx Alk - 2c_{Fe^{2+}} - 3c_{Al^{3+}} - 2c_{Mn^{3+}} \quad [16]$$

*Alk* means the results of 0.1N NaOH titration to a pH= 4.3.

In acid water NP is below zero and for base has positive values. The goal of remediation strategies is to achieve an NP above 0.2 mmol/dm<sup>3</sup> to lower acidity to the expected level. The choice of a neutralization strategy depends on the neutralization algorithm, which includes the following reactions:

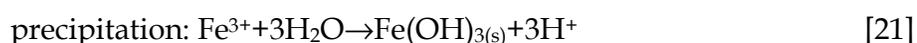


Tootche et al. (2003) studied the buffering mechanisms of acidic mining lakes in the open cast lignite mining lake (Plessa 111) in Lausitz (Germany). On the basis of laboratory investigations, hydrochemical modelling and mineralogical structure he detected four different buffers in four successive sections of the titration curve:

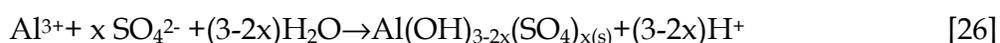
1. hydrogen sulphate buffering with no precipitates :



2. iron buffering with iron hydroxide precipitation and sulphate sorption:



3. aluminium buffering: aluminium hydroxysulphate precipitation:



and also Mn-buffer - manganese oxide precipitation



4. an additional soil phase buffer which was based on:

- ion exchange calcium and magnesium on goethite,
- mineral transformation of schwertmannite into goethite with the release of protons and sulphate:



The first section of the titration curve was in a pH range of 2.5 to 3, the second - 3.5 to 4.5, the third - 4.5 to 5.5 and the last one - above 5.5. The longer the buffer zones are, the more of the neutralization agent must be used.

## 5. Anthropogenic lake district in Łuk Mużakowski – Poland as a case study of the influence of brown coal mining on water resources

### 5.1 Location and origin

The “anthropogenic lake district” is located along the Polish-German border between Trzebiel and Łęknica. It comprises about 100 reservoirs of different sizes and chemical properties with a total surface of about 1 500 000 m<sup>2</sup>. The reservoirs were generated due to lignite mining which started in the second half of the 19-th century and lasted till the seventies of the 20-th century. In 19-th century seven mining companies were operated in this area. The first one was founded in Raszyn in 1883, the next in Tuplice a year later, then “Wiktor” in Trzebiel in 1898 and in Nowe Czaple in 1900. The best known mine in this area was “Babina” in Łęknica. It was founded in 1922 and operated (as the last one) till 1973 (Jędrzak, 1992). Exploitation was done by means of two methods, at the beginning underground and later open cast. Both techniques resulted in unfavourable changes in the landscape, such as pits and sinks, which after being filled with water created pit and hollow lakes.

Reservoirs are divided into three groups depending on their origin and location (Fig. 1.):

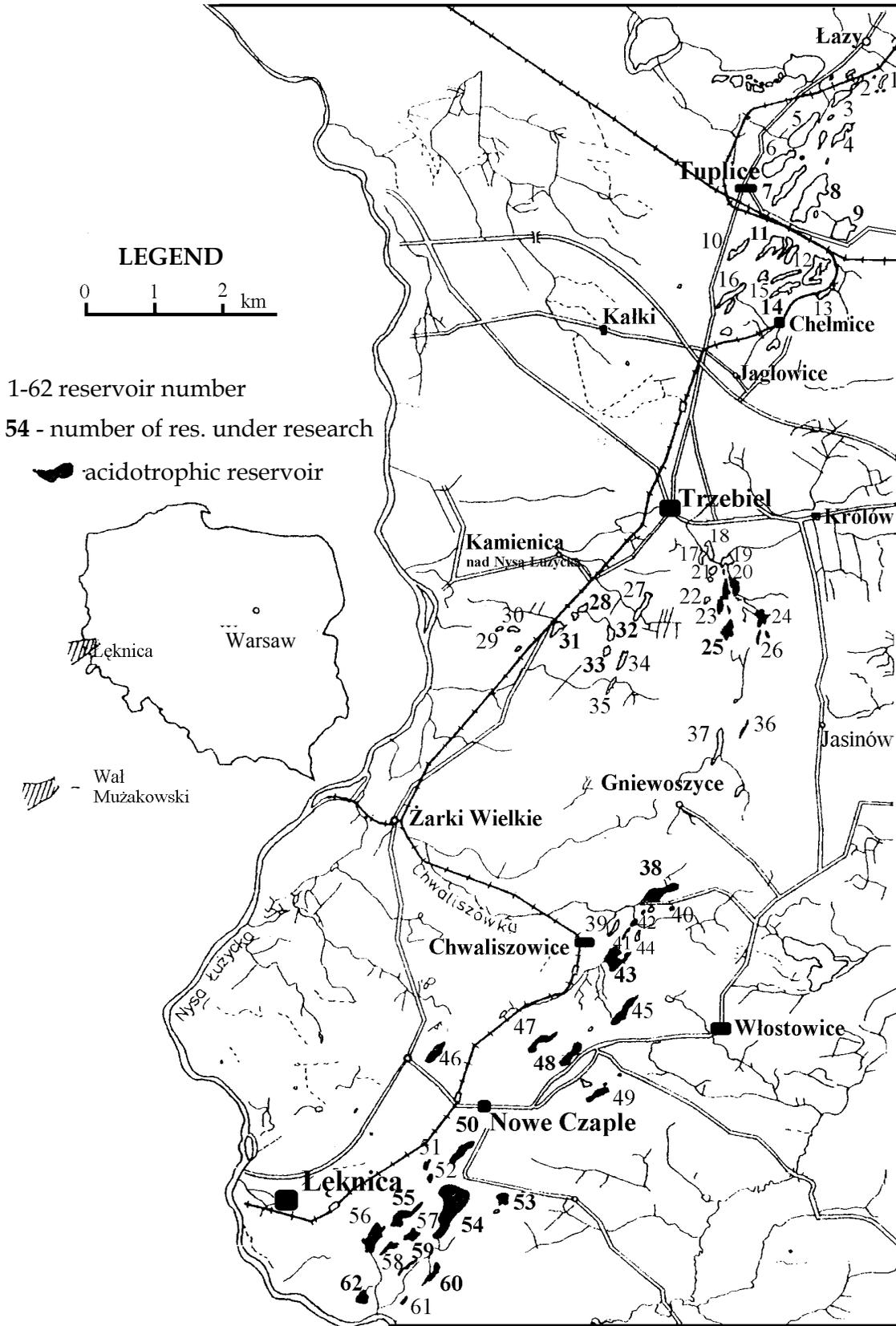


Fig. 1. Map of the "anthropogenic lake district"

1. ca. 30, the oldest hollow reservoirs situated near Tuplice; they have elongated shape with axis NE-SW, their surface varies from <1 to 15 ha and maximal depth from 1.5 to 5.5 m.
2. 33 hollow reservoirs situated between Trzebiel and Kamienica; they are rather small, with a surface lower than 5 ha, but relatively deep to 9.5 m.
3. 33, the youngest and biggest pit reservoirs situated near Łęknica; their surface varies from 1.8 to 22 ha, and the maximal depth from 3.5 to 21.5 m.

The reservoirs have never been remediated. One of them was used as a domestic waste receiver and due to eutrophication it disappeared. Some of the smallest reservoirs are used as fish farms. The rest of them are not exploited economically nor touristically.

## 5.2 Methods

The reservoirs of the “antrophogenic lake district” were monitored irregularly. The first data come from the early eighties, about five years after the youngest reservoir came into being. The chemistry of some reservoirs was described by Jędrzak (1992) and Jachimko (1998). In the years 2006-2009, mainly meromictic reservoirs were investigated (Jachimko, 2007; Jachimko&Kołodziejczyk, 2008). All research was based on an analysis of water samples taken up along the vertical profile (from the surface and then every two meters in depth), from the deepest place of each reservoir. The water analyses were supplemented by sludge chemistry. The hydrogeochemical simulator PHREEQC (Parkhurst&Apello, 1999; Apello&Postma, 2006) was used to calculate saturation indexes with the use of constants included in the program. NP according to the reaction [14] was calculated to evaluate the acidification.

## 5.3 Chemistry of reservoirs

The reservoirs of the “antrophogenic lake district” have different chemical properties (Table 3). According to the pH value and redox potential the reservoirs were classified into three groups:

1. acidotrophic with a pH ranging between 2.6 and 3.7 and redox potential of 605 to 755 mV,
2. intermediate with a pH ranging between 2.6 and 3.7 and redox potential of 605 to 755 mV
3. “others” with a pH ranging between 5.2 and 7.4 and redox potential of 380 to 600 mV.

Acidotrophic reservoirs are located in the south of the district. They are mainly the youngest reservoirs of pit origin. “Others” occur in the north and belong to the oldest group. There were only two intermediate reservoirs, no 11 and no 21, but their chemistry varied from one another. No 21 was similar to the acidotrophic class and no 11 to “others”. The pH value and redox potential were different in the profiles of most reservoirs. The biggest changes of the factors were found in acidotrophic reservoirs Re 25, 48 and 54. Surface waters were very acidic (pH<3.4) and had a high redox potential (>750 mV). However, waters close to the bottom had a pH of above 4 and a redox potential of less than 400 mV. Within the acidotrophic group the changes of the pH value in the vertical profile were insignificant. The pH values in vertical profile in the group of “others” reservoirs were almost constant or increased from the surface to the bottom (with the exception of very shallow reservoir no 8) (Fig. 2).

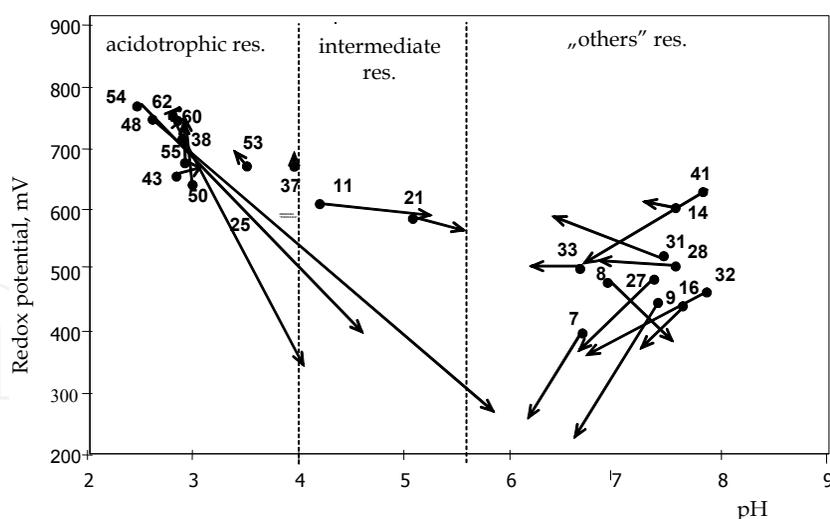


Fig. 2. pH-value versus redox potential in vertical profiles of reservoirs (points represent surface, arrows - bottom layer)

The chemical composition and biocenosis of the “others” reservoirs were similar to the natural lakes. The evidence of mine origin are a high concentration of iron in the deeper part of the water body, the iron to sulphur ratio and a low neutralisation potential. In five reservoirs of this type, (no 7, 9, 27, 33 and 32) iron to the sulphur ratio in the bottom layer meets the values which result from pyrite weathering. The iron concentration in uppers layers is very small, lower than  $0,05 \text{ mmol/dm}^3$ , due to the low solubility of ferric compounds in this pH region. Close to the bottom due to ferric reduction the concentration of iron remained on the primary, high level. The sulphate concentration changed in a very small range with depth and ranged from  $0,15$  to  $2 \text{ mmol/dm}^3$ . In some reservoirs the sulphate concentration decreased with depth, probably as a result of reduction, but in the others it increased as a result of continuous AMD interaction. The bottom layer of reservoirs 25 and 48 contained much more ferrous irons than resulted from pyrite weathering. It was probably connected with meromictic type of the reservoirs.

parameter	unit	“others” reservoirs	intermediate reservoirs	acidotrophic reservoirs
pH	-	5,96 7,85	4,18 5,64	2,41 5,85
calcium	$\text{mg/dm}^3$	10,0 79,6	19,5 50,6	13,9 334
sulphate (VI)	$\text{mg/dm}^3$	10,8 186,3	37 171,6	75,5 3935
tot. iron	$\text{mg/dm}^3$	0,14 39,2	0,87 1,73	0,82 1618
aluminum	$\text{mg/dm}^3$	0,013 0,431	0,114 0,542	0,195 20,59
potassium	$\text{mg/dm}^3$	1,7 11,6	2,3 3,1	2,0 36,9

Table 3. Minimal and maximal values of water quality parameters (own results)

The water of "others" is of the calcium bicarbonate or calcium sulphate type. The percentage of calcium in the "others" reservoirs ranged from 34 to 71% of total cations and decreased with depth. In acidotrophic reservoirs it was smaller and varied from 16 to 59%. The iron content ranged from 0,1 to 30 % of total cations in the "others" reservoirs and reached 68% in the acidotrophic ones. Sulphate averaged from 34 to 72% of anions in the "other" reservoirs and dominated in almost all of them. The percentage of bicarbonate ranged from 9 to 50 % and chloride from 5 to 20% of total anions. Sulphate dominated in all acidotrophic reservoirs and ranged from 81 to 99,6%.

The chemistry of the reservoirs was interpreted with means of SI of ferric, aluminium and calcium minerals. Figure 3 presents the relationship of SI of the selected minerals and pH. The iron content was much higher than calculated from goethite or jarosite K solubility. The reservoirs were oversaturated to goethite in the whole range of pH, and to ferric hydroxides in a pH over 4. Jarosite K was oversaturated in a pH below 3.5 and over 6.5 (with the exclusion of water with very little iron (concentration below 0.2 mg/dm<sup>3</sup>).

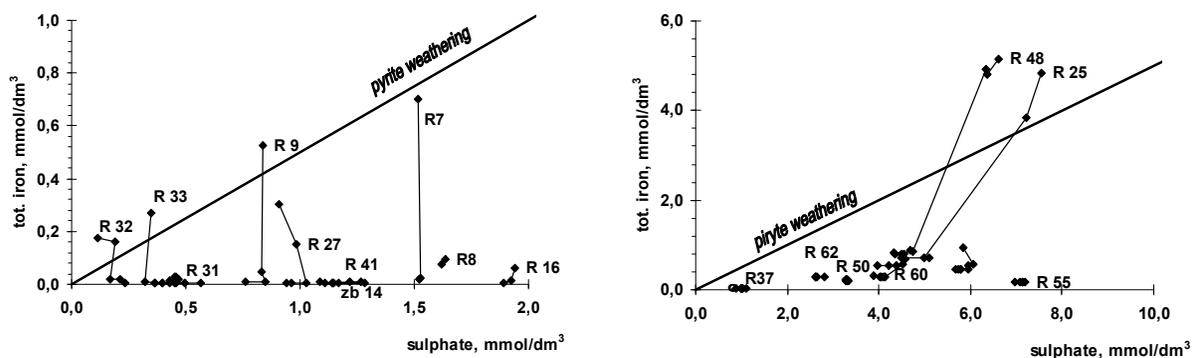


Fig. 3. Coexistence iron and sulphate in "others" (left) and acidotrophic reservoirs of the "antrophogenic lake district".

The saturation of waters by aluminium compounds depended on pH in the following way:

- pH<4.7 - undersaturation to aluminium hydroxide, and below 4 also alunite, SI value increased with pH,
- pH>4.7 - oversaturation to gibbsite, and in the range between 4 to 7.5 also alunite,
- pH>7.5 - undersaturation to aluminium hydroxide and alunite.

Most of the reservoirs, mainly "others" were slightly undersaturated to calcium sulphate, as gypsum or anhydrite. In the "others" reservoirs (no 14 i 16 i 27) and a pH above 7.5 hydroxapatite could precipitate. The saturation state existed only in acidotrophic reservoirs. The content of aluminium was limited by gibbsite in a pH over 4.7.

Based on the correlation between pH and neutralisation potential, sensibility on accidental acidification of "others" reservoirs and reclamation processes of acid reservoirs can be distinguished, as follows (Fig. 4. and 5. ):

1. among the "others" reservoirs no 33 and 7 are the most sensible on acidification,
2. among the acidotrophic reservoirs no 37 and 53 should be reclaimed in first,
3. high pH of the deeper part of reservoirs no 25 and 48 does not indicate their real neutralisation.

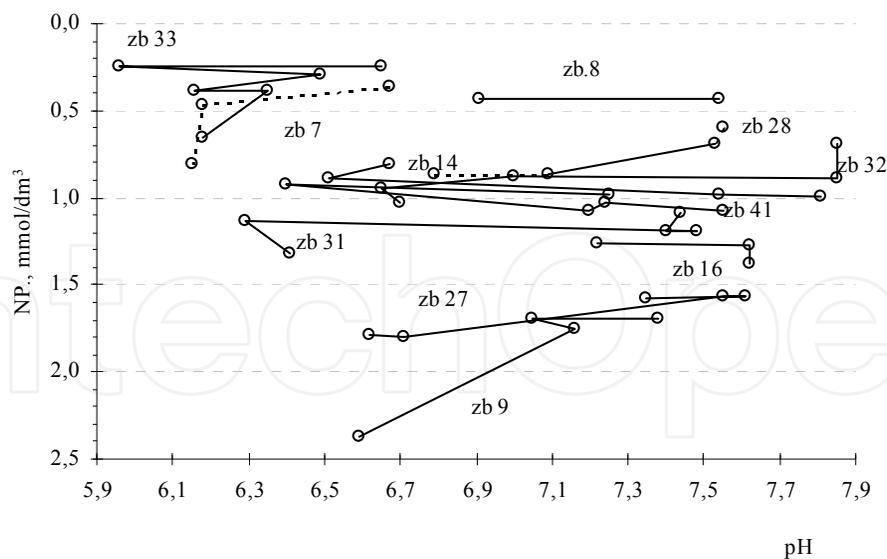


Fig. 4. Correlation between the pH value and the neutralisation potential in vertical profiles of the "other" reservoirs

The reservoirs of "the anthropogenic lake district", despite the different chemical composition and acidity, exhibit features typical of mine waters influenced by pyrite weathering. On the basis of the above analysis it is possible to conclude, that the most crucial factors creating its chemistry is the way of origin. It limits the amount of released pyrite and load of acidity. The inflow of weathered pyrite to the "others" reservoirs was limited by oxygen and ferric accessibility. So, very little amounts of base minerals occurring in the soil were sufficient for neutralisation. In pit lakes, especially in overburden and bottom sludge huge amounts of acidity were released. The following chemistry of acid reservoirs depended on the mixing type. In particular circumstances for meromixis occurred and the water body was divided into two layers of a different chemical composition.

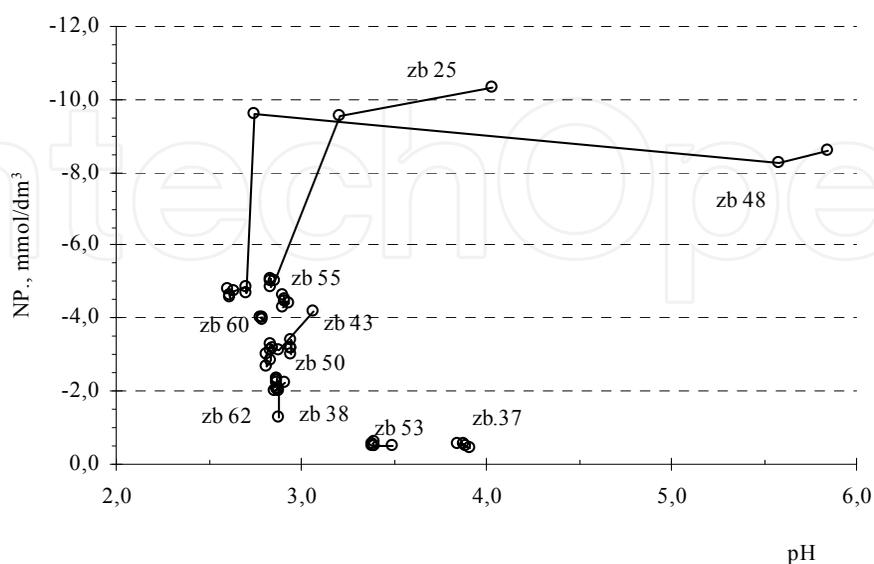


Fig. 5. Correlation between the pH value and the neutralisation potential in vertical profiles of acidotrophic reservoirs

#### 5.4 Influence of the mixing type on the chemistry of lakes – Meromixis reservoirs

Meromictic lakes are lakes which do not mix completely. This phenomenon is not common in natural lakes, but it often occurs in mine lakes. The following mechanism creates meromixis in the development of pit lakes: a) formation of a less mineralized mixolimnion by the inflow of less mineralized water to the lake surface, b) enrichment of iron and sulphate due to the transport of secondary minerals from mixolimnion to monimolimnion, c) influence of sea water d) accumulation of substances in the monimolimnion due to biological decay, e) evaporation f) influence of groundwater with high TDS (Boehrer&Schultze 2006 and references there in). The deepest part of the meromictic lake water body – monimolimnion – has often a very specific chemical composition: strong anoxia, rich in hydrogen sulphide and products of microbial decay. Among the acidotrophic reservoirs of the “anthropogenic lake district” three of them are of the meromictic type with a pH of monimolimnion over 4.6. The pH increase had a great influence on lake chemistry and caused iron and sulphate accumulation in monimolimnion and turned back the stratification of aluminium.

The research presented below focused on the biggest lake - no 54 which comes from 1973 (there are no exact data about the rate and duration of filling up). The reservoir is located about 2 km east of Łęknica, 2.3 km west of Przewoźniki and 2 km south of Nowe Czaple (Fig. 1). The reservoir is surrounded by strongly eroded heaps of excavation. The surrounding area is covered with forest. The morphometry of the reservoir is presented in Table 4.

water level m above see level	surface area m <sup>2</sup>	max length, m	max width, m	extension coefficient	max depth m	shore line length m	shoreline development
132.0	202,000.0	896	468	1.91	21.5	2625	1.65

Table 4. Morphometry of reservoir no 54

The first data about composition of the lake come from February 1981, eight years after coming into existence. From the study it is known that the lake is extremely acidic with a high concentration of iron and sulphur. The reservoir has no surface inflows such as streams or other lakes. It results in the fact that the chemical composition of its waters depends mainly on the composition of groundwater and precipitation, and reactions within the water body.

Chemical composition of reservoir modified significantly during the 27 years of observation (Fig. 6). The pH value changed in different ways in mixo and monimolimnion. In 1981 pH was nearly the same at all depths. A small increase of its value, by about 0.2 units, was observed between 18 and 20 m. Since 1988 a difference between the pH values of the upper and lower strata has been observed. The pH of bottom layer has increased systematically and reached values of about 4.8-4.9 in 2008. Also the thickness of mixolimnion was growing. Nowadays, the region of the greatest pH increase is located between 7 and 8 m below the water surface.

The changes in iron concentration in vertical profile had two patterns. The first one, in the years 1981 and 1988, showed gentle increase in iron concentration from the surface to the bottom. Later a sharp boundary between iron concentration in the upper and lower part of the water body appeared. Iron concentration in the mixolimnion layer was almost steady during the following research period. Iron concentration in monimolimnion increased with depth and ranged from about 100 to more than 800 mg/dm<sup>3</sup> in the years 1981-88 and, since 1993, to more than 1500 mg/dm<sup>3</sup>.

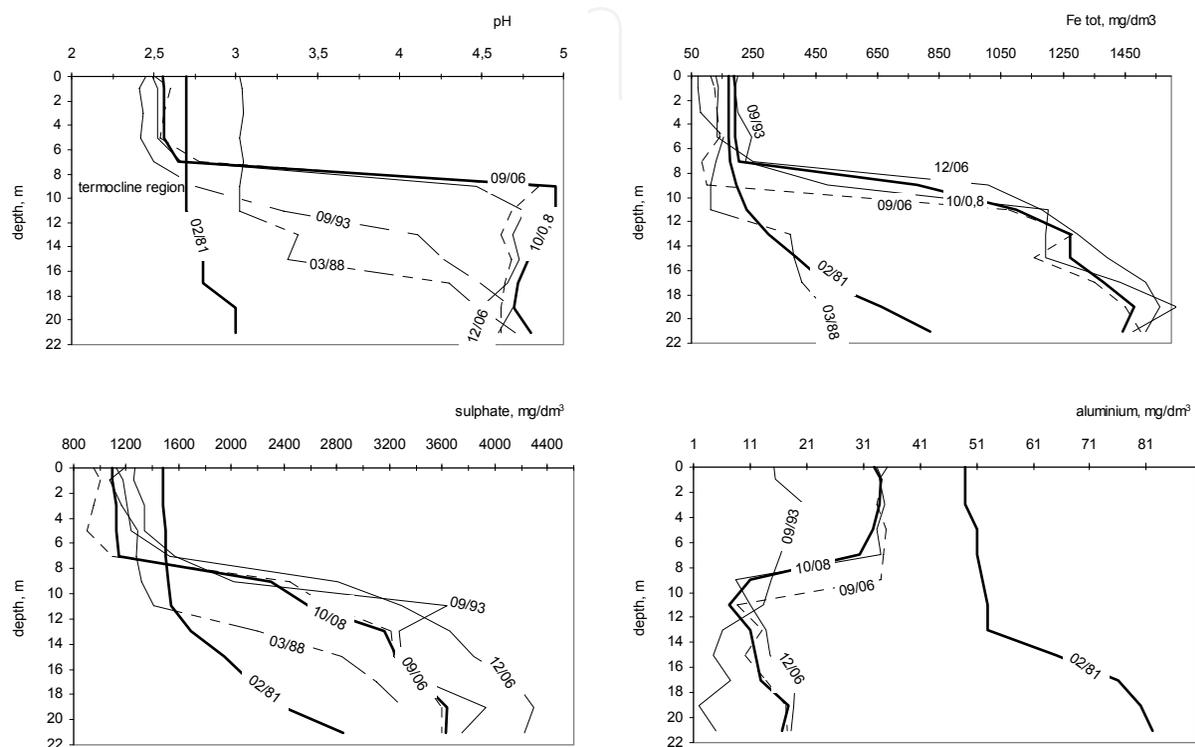


Fig. 6. Chemical composition of reservoir no 54 in the years 1988-2008

Sulphate concentration did not change as rapidly as iron concentration. In the years 1981-88 sulphate concentration was almost steady in the upper part of water body (from the surface to 11 m in depth). Deeper, the sulphur content increased with depth. The research done later, showed a low concentration of sulphate in the layer 0-7 m, a rapid concentration increase between 7 and 11 m to values of about 3000 mg/dm<sup>3</sup>, and then a further gentle increase with depth to a maximum concentration of about 4300 mg/dm<sup>3</sup>.

Aluminium concentration had two different profiles. The first one comes from 1981, when aluminium concentration was the highest and increased with depth. Later the concentration of aluminium decreased significantly with depth from the amount of about 35 to less than 10 mg/dm<sup>3</sup>.

In 1981 potassium concentration was low and varied from 3 in mixolimnion to 7 mg/dm<sup>3</sup> close to the bottom. The potassium amount in mixolimnion did not change significantly during the following period. The maximum concentration in mixolimnion (5.8 mg/dm<sup>3</sup>) was stated in 2008. In monimolimnion potassium concentration increased gradually and exceeded 30 mg/dm<sup>3</sup> in 2008.

Solid name	formula	data	log IS on the depth, m below water surface											
			0	1	3	5	7	9	11	13	15	17	19	21
	Fe(OH) <sub>3a</sub>	1981	-1.15	-1.15	-1.16	-1.17	-1.17	-0.76	-0.72	-0.72	-0.69	-0.67	-0.64	-0.64
		2007	-1,32	-1,34	-1,29	-1,11	-1,18	*)						
goethite	FeOOH	1981	4.50	4.50	4.50	4.40	3.97	3.39	3.39	3.67	3.17	2.85	2.59	3.16
		2007	4.30	4,18	3,99	4,11	4,01							
jarosite K	KFe <sub>3</sub> (SO <sub>4</sub> ) <sub>2</sub> (OH) <sub>6</sub>	1981	2.81	2.66	2.61	2.61	1.7	3.39	3.67	0.75	-0.64	-1.44	-2.14	-0.97
		2007	2.83	2.61	2.28	2.57	2.44							
alunite	KAl <sub>3</sub> (SO <sub>4</sub> ) <sub>2</sub> (OH) <sub>6</sub>	2007							3.33	3.92	2.06	2.68	5.36	6.29
gibbsite	Al(OH) <sub>3</sub>	2007							-0.82	-1.15	-1.59	-1.31	-0.05	0.43

Table 5. SI for selected solids in 1981 and 2007 (lack of value means very low level)

Based on the chemical properties and SI values (table 5) of the lake two periods of its development can be distinguished:

1. "the iron period" with the intensive processes of ferrous precipitation in miksolimnion (thermocline was then located between 9 and 11m below the water surface).

The sedimentation of jarosite K and probably be also goethite caused transport of ferrous, potassium and sulphate to the deeper part of the water body. On the boundary of oxic and anoxic zones dissolution of minerals and the release ions into the water body occurred. We can also suppose that the main reason for the pH change in monimolimnion was ferrous reduction. These processes affected the pH values by consuming protons, but didn't reduce the acidity of water (Schoepke 2001). The potassium stratification confirms the main role of jarosite K precipitation in miksolimnion.

2. "the iron-alum period" - from 1988 up till now, with two buffer zones, ferrous precipitation in miksolimnion and alum in monimolimnion. The processes in the mixolimnion over that period were similar to the "iron period". Also the chemical composition of miksolimnion did not show any significant changes and were similar to the initial composition. The only difference is alum concentration which decreased by about 20 mg/dm<sup>3</sup>, which means almost 50% of initial value. Due to the fact that monimolimnion acted as a sink for ferrous precipitates, the amounts of precipitate dissolution products increased. The concentration of almost all (excluding aluminium) ions analysed increased by orders of magnitude. The pH values increased from 3 to 4.5-4.9. It created the conditions for aluminium salts precipitation and lowered aluminium concentration in monimolimnion.

It seems that the future of reservoir 54 depends on the dynamics of pyrite oxidation products discharge. The total load of iron and sulphate has begun to increase for a few years. It allows the mixolimnion to reach equilibrium with iron compounds. Monimolimnion will still act as a sink for ferrous and sulphate ions. Lack of organic compounds inhibits sulphate reduction and secondary sulphides precipitation. Such a situation will stabilise meromixis for a long time.

## 6. Summary

The “anthropogenic lake district” located in Łuk Mużakowski is a drastic example of the destruction of the environment by lignite mining. The pollution of acid reservoirs is very stable and until now no diminishing tendency has been observed. The chemistry of the group of “others” reservoirs balances between the constant inflow of acid groundwater and neutralising processes as reduction with the use organic matter or the dissolution of base minerals. The composition of acidic reservoirs depends on their mixing pattern. Meromixis, which was created in three of the reservoirs in the area caused changes of chemical properties of monimolimnion. The monimolimnion acted as a sink for products of the dissolution of ferrous salts and affected water density and stabilized meromixis. Intensive precipitation of ferrous salts in the oxic zone and the dissolution of the precipitations resulting from ferrous reduction in monimolimnion cause an increase in pH to a value over 4. It created the next buffering aluminium zone in monimolimnion.

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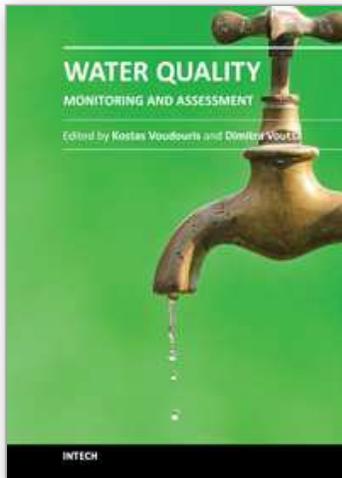
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