The influence of mine water on surface water in the Miłek catchment (Intrasudetic Basin, SW Poland).

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Abstract

The impact of coal mining on the natural environment has been gaining significance since the liquidation of the Nowa Ruda part of the Lower Silesian Mining Region (LSCB). Huge amounts of waste rocks containing various minerals have been disposed on a tailing located in the Miłek catchment. This tailing generates acid mine drainage (AMD). At the foot of the tailing, acid water with high concentration of sulfates, iron, aluminum and other ions has been observed. The main objective of hydrogeochemical examinations carried out from 2003 to 2011 was to recognize hydrochemical changes in surface water caused by water seeping through tailing rocks.

Key words: mine water, acid mine drainage, mine waste dump, Intrasudetic basin, SW Poland

Introduction

The Lower Silesian Coal Mining Region is located in the south-western region of Poland and is divided into two parts: the Wałbrzych region and the Nowa Ruda region. In the Nowa Ruda region, the oldest documented mining operations took place in the 14th century. The mining industry reached the peak of its intensity in the 19th century. At the end of the 20th century the Polish government decided to shut down all Lower Silesian Coal Mining Region mines due to the economic factor. During hundreds years of coal mining a huge amount of waste rocks containing various minerals have been disposed on tailings located in some parts of the Nowa Ruda region.

We have studied a dump which is located in the Miłek stream catchment (near Ludwikowice Kłodzkie, Fig. 1), the latter with the total area of 4.9 km^2. The dumping of the studied mine wastes started probably during the second part of 19th century and lasted until 1970. The waste material is derived from the nearby "Wacław" shafts located in the Ludwikowice Kłodzkie mining area (a part of the Nowa Ruda mine) and comprises mainly Carboniferous sedimentary rocks. According to the grain size of rock material, waste rocks vary from clays to gravel and contain a lot of iron minerals including sulphates, oxides and hydroxides (Chudy, Marszałek, 2008).

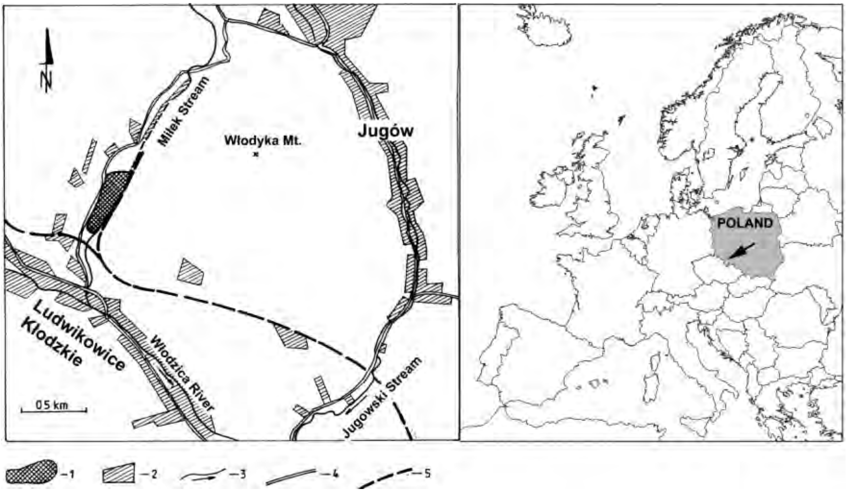

Figure 1. *The location of the study area. 1 – mine tailing, 2 – villages, 3 – stream, river, 4 – roads, 5 - train*

Methods

Water samples were collected between 2003 and 2011 from selected points into HDPE bottles according to the methodology given by Witczak and Adamczyk (2005). An analysis was made by the Physical Geography Department Laboratory of the Wroclaw University: HCO_3^-, Cl^-, SO_4^{2-} using spectrophotometric methods, and Na^+, K^+, Ca^{2+}, Mg^{2+}, SiO_2, PO_4^{3-}, Fe_{tot}, Mn^{2+}, Zn^{2+}, Cu^{2+}, Cd^{2+}, Al^{3+}, Ni^{2+} using atomic absorption spectroscopy (AAS) methods ("Avanta Σ" apparatus by GBC company). pH, EC (specific electric conductance) and water temperature were measured in the field by means of a WTW Multi340i multi-parameter instrument, and SenTix 81, SenTix ORP and TetraCon 325 electrodes.

Kurlov's formula was used to show water composition (Macioszczyk, 1987):

$$SpGM\frac{anions}{cations}T,$$

where:
Sp – specific component/element [mg/L],
G – gas content [g/L],
M – TDS [g/L],
anions and cations – written in descending sequence as % mval/L,
T – water temperature [^{0}C].

Results and Discussion

The water in the upper parts of the Miłek (P1) catchment, located in the gneisses massif of the Sowie Mts., has low mineralization up to 0.19 g/L and is of the 4-ions HCO-SO-Ca-Mg chemical type (Table 1). The predominant ions are bicarbonate (up to 108 mg/L), calcium (from 12 to 27 mg/L), sulphates (up to 30 mg/L) and magnesium (from 5 to 10 mg/L).

Table 1. *The chemical composition of groundwater outflow from a mine tunnel (Kurlov's formula) after the termination of coal mining operations (cf. Chudy 2008, modified).*

Sampling points	**Kurlov formula**
P1	$M^{0,17-0,19} \dfrac{HCO_3^{74-43} \quad SO_4^{50-15}}{Ca^{57-49} \quad Mg^{35-32}} T^{6-8,3}$
P2	$Fe^{23-29} \quad M^{0,954-2,1} \dfrac{SO_4^{98-71}}{Ca^{72-61} \quad Mg^{25-16} \quad Na^{14-8}} T^{11,8-14,8}$
P3	$Zn^{4-6} Al^{9-69} Fe^{7-195} \quad M^{0,9-3,1} \dfrac{SO_4^{99-98}}{Ca^{67-52} \quad Mg^{42-27}} T^{8-11}$
P4	$Zn^{0,5-4,0} Al^{4-22} Fe^{0,2-29} M^{0,4-1,1} \dfrac{SO_4^{98-88}}{Ca^{72-65} \quad Mg^{30-18}} T^{4,2-11,8}$
P4 (1962-1968)	$M^{1,5-1,7} \dfrac{SO_4^{98-96}}{Ca^{65-48} \quad Mg^{42-28}}$

One permanent mine water outflow (P2) runs from the mine shaft, coming probably from a permeable tectonic fault, and the other runs from the foot of the dump (P3). Field studies carried out between 2004 and 2012 revealed very high concentrations of many ions in these outflows.

The mine water from point P2 has TDS up to 889 mg/L, is of the 4-ions SO-Ca-Mg-Na type and has slightly acidic to acidic pH. Sulphates dominate among the anions and their concentrations range from 219 mg/L to 612 mg/L. The highest concentrations of bicarbonates (up to 95 mg/L) and chloride (from 13 to 17 mg/L) are measured in the spring season; the same concerns the amount of calcium (up to 102 mg/L), magnesium (up to 21 mg/L) and sodium (17 mg/L). This water has a high iron (42 mg/L) content and a very high aluminium (28 mg/L) content.

The water from point P3 is of the 3-ions sulphate – calcium – magnesium type (Tab. 1, Fig. 2), low acid (pH 2.6-2.8), without the odour of hydrosulphuric acid. The water temperature is strictly correlated with the air temperature and oscillates from 2.2 to 13.6 degrees Celsius. The predominant ions are sulphate (from 661 mg/L in the autumn-winter season to 2192 mg/L in the spring season), calcium (from 102 to 266 mg/L; the highest concentrations in the spring season) and magnesium (from 29 to 101 mg/L). Ions existing in the smallest concentrations in the water comprise the following: chloride (from 3 to 15.2 mg/L) and sodium (from 6.6 to 14.9 mg/L). A characteristic feature of this water is a high concentration of iron (from 7 to 195 mg/L), an extremely high concentration of aluminium (from 9 mg/L in the summer-winter season to 69 mg/L in the spring season) and a high zinc concentration (from 4 to 6.1 mg/L).

During the whole year in the area below the outflow, the water at the bed of the stream contains a light orange, yellow and cream residue (sulphate and iron ions), which is oily on the touch.

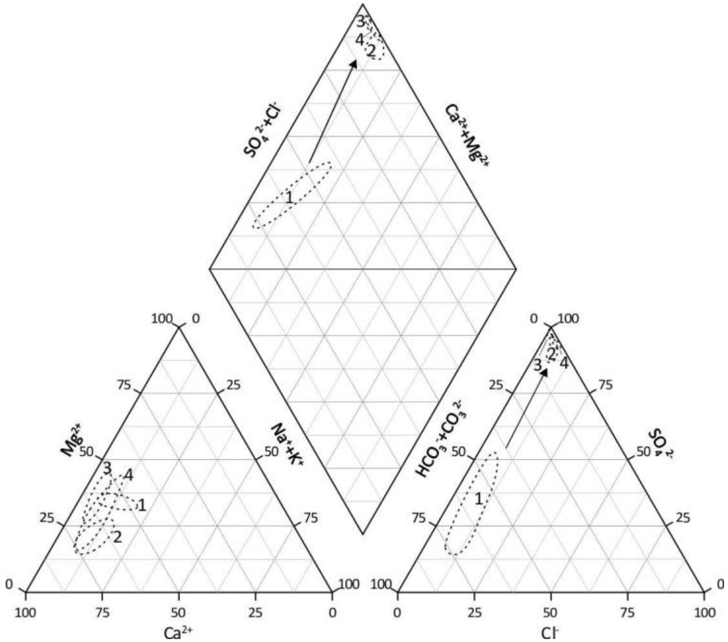

***Figure 2**. The Piper diagram showing the chemical composition of surface and groundwater in the Miłek catchment. 1 – water from the upper parts of Miłek stream (P1),2 – mine water outflow from the mine shaft (P2), 3 – mine water outflow from the foot of the dump (P3),4 – water from the mouth of the Miłek stream (P4).*

For the water at the mouth of the Miłek stream (P4), apart from the present analysis, the authors have two incomplete analyses conducted in the years 1962-1968. At that time the water was acidic (pH), and its mineralization was 2-3 times higher (from 1510 to 1708 mg/L) as compared with the current value. The predominant ions are sulphate (from 735 to 920 mg/L), chloride (from 12 to 20 mg/L), calcium (from 153 to 202 mg/L), potassium (from 10.5 to 19 mg/L) and sodium (from 15.5 to 21.5 mg/L). This water has a high iron (up to 60 mg/L) content, 2-3 times higher as compared with the current value.

The water collected between 2003 and 2011 is of the 3 -ions SO-Ca-Mg type, mainly acidic (pH 3.0), sometimes to slightly acidic (pH 6.8) , has TDS of up to 1253 mg/L. The predominant ions are sulphate (up to 684 mg/L), calcium (up to 142 mg/L), magnesium (up to 36 mg/L) and chloride (up to 24 mg/L). The water is characterized by a high iron (up to 29 mg/L), aluminium (up to 22 mg/L) and zinc (up to 4 mg/L) content. At the bed of the stream, it contains a light orange residue.

Conclusions

The hydrogeochemical examinations carried out in the area of the Miłek catchment confirmed a significant influence of the seepage water infiltrating through the mine waste dump on the surface water, which results in a change in their compositions. The tailing rock containing sulphidic minerals generates acid mine drainage (AMD) in the classical form, which is an exceptional case in former LSCB.

As indicated by previous studies (Chudy, 2008; Chudy, Marszałek 2008), the seepage water from the mine tailing has extremely high concentrations of iron (up to 9500 mg/L) and sulphates (up to 38000 mg/L), which is caused by oxidation reactions of sulphates (mainly pyrite). The oxidation of pyrite could occur with O_2 or Fe^{3+} as an oxidant. The overall process is described by the following reaction (Stumm, Morgan, 1981, Appelo, Postma 2007):

$$FeS_2 + \frac{15}{4}O_2 + \frac{7}{2}H_2O \quad \rightarrow \quad Fe(OH)_3 + 2SO_4^{2-} + 4H^+ \qquad [1]$$

The oxidation of the disulphide to sulphate by O_2 is described by the reaction:

$$FeS_2 + \frac{7}{2}O_2 + H_2O \quad \rightarrow \quad Fe^{2+} + 2SO_4^{2-} + 2H^+ \qquad [2]$$

The second pathway of pyrite oxidation is done by Fe^{3+}:

$$FeS_2 + 14Fe^{3+} + 8H_2O \quad \rightarrow \quad 15Fe^{2+} + 2SO_4^{2-} + 16H^+ \qquad [3]$$

The third [3] reaction is fast and generates much more H^+ ions and lower pH than the second [2] reaction, which is slow. The isotopic composition of ^{18}O in SO_4^{2-} shows that pyrite oxidation is 0.25:0.75 for the O_2 and Fe^{3+} oxidation reactions.

Below the mine tailings, the surface water has a significantly changed chemical composition. Iron compounds are precipitated as an orange precipitate containing mainly iron hydroxides and small amounts of schwertmanite. (Chudy et al., 2010).

With the knowledge of the chemical composition of the water and the amount of water flow in the stream, it became possible to calculate the amount of salt delivered to the main river of the area – the Włodzica. The amount of salt discharged was calculated as the sum of calcium, magnesium, sodium, potassium, chloride, bicarbonate and sulphate ions. The total load of salt discharged through the Miłek stream is about 0.12-0.15 t/d. This load of salt is one of the smallest in the LSCB area as compared to the other mine outflows.

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