

SULPHATES IN GROUNDWATER OF THE KARST-FRACTURED TRIASSIC AQUIFERS IN AREAS OF INTENSIVE MINING DRAINAGE (THE OLKUSZ AND THE BYTOM REGIONS)

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ABSTRACT

The analysis of sulphates content in groundwater has been based on results of the regional and local groundwater quality monitoring performed in the area of the Olkusz and Bytom regions. There are situated still active and abandoned Zn-Pb ores mines. These areas have been subjected to long-lasting intensive drainage, which has caused significant changes of hydrogeochemical conditions in Triassic aquifer. The results of these changes are mainly: high concentrations of sulphates (up to more than 5000 mg/dm³) in groundwater of the Triassic karst-fractured aquifers in both regions. Numerous industrial and communal waste sites were constructed directly on the area without any protective measures. These waste sites, particularly in recharge areas, are an additional source of sulphates noticed in groundwater. So the final contents of sulphates observed in groundwater in these areas is the result of an overlapping negative impact of both above mentioned factors.

INTRODUCTION

Upper Silesia is the most important, and only one, in Poland region Zn-Pb ores exploitation. There have been four major areas of these ores exploitation: Tarnowskie Góry, Bytom, Olkusz and Chrzanów. Contemporary exploitation takes place in Olkusz and Chrzanów areas. Zinc and lead ores occur in dolomites and limestones of the Lower and Middle Triassic. These carbonate Triassic sediments are the most important and resourceful karst-fractured aquifers being the major sources of potable water for the Upper Silesian agglomeration. Intensive drainage of these aquifers by Zn-Pb ore mines and numerous groundwater intakes have caused their overexploitation which has led to the creation of deep

regional cones of depression, changes of flow directions and increase of hydraulic gradients. These long lasting hydrodynamic changes in these aquifers and negative influence of great number of pollution sources situated on the surface of the ground have resulted in general decreasing trend of groundwater quality. Particularly warning significant increase of sulphates content in groundwater of the Triassic aquifers has been observed.

In areas subjected to long lasting mining drainage caused by Zn-Pb ores exploitation the highest concentrations of sulphates in groundwater (up to more than 5000 mg/dm³) have been noticed. Observed increase of sulphates content in some groundwater intakes not subjected to the direct influence of mining activity is alarming.

The analysis of sulphates content in groundwater of the Triassic aquifers in two regions has been performed in this paper (Figure 1):

- Olkusz region – subjected to the direct and indirect impact of still active Zn-Pb ores mines,
- Bytom region – subjected to still continued drainage by abandoned Zn-Pb ores mines.

The analysis of sulphates content in groundwater has been based on results of the regional and local groundwater quality monitoring performed in these both areas.

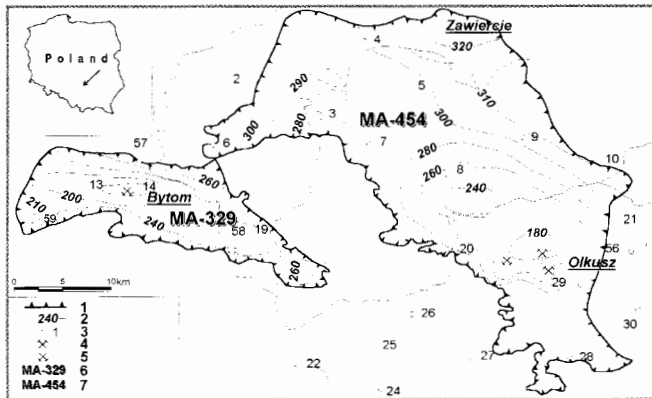


Figure 1. Hydrogeological sketch of the Olkusz and Bytom regions.

- 1- Boundaries of main aquifers,
- 2 – Contour lines of the Triassic carbonate aquifer (in meters above sea level),
- 3 – Sites of the regional groundwater quality monitoring, Zn-Pb ores mines: 4 – Active, 5 – Abandoned, karst-fractured main aquifers: 6 – Bytom, 7 – Olkusz – Zawiercie.

HYDROGEOLOGICAL SETTING

Olkusz and Bytom regions belong to the Silesian-Cracow Monocline built up of the Triassic and Jurassic formations discordantly overlying folded and faulted Palaeozoic basement. Zinc and lead ores occur in dolomites and limestones of the Lower and Middle Triassic. These carbonate rocks are the main aquifers in both regions. In the profile of the Triassic carbonate formations (limestones and dolomites) two water-bearing horizons can be differentiated: the Muschelkalk horizon and the Roetian one. These two horizons are frequently considered jointly as one aquifer known as the complex of the Triassic carbonate series (Rózkowski, ed., 1990). Hydraulic structure of fractured and karstified Triassic rocks consists of three types of spaces: pores, fissures and caverns. Limestones represent fissured-cavernous type of the aquifer while dolomites represent porous-fissured-cavernous type (Motyka, 1998). Fissures and karstic channels are favourable pathways of groundwater flow while the pore space is the main water reservoir.

There are Quaternary, Jurassic, Triassic and Carboniferous-Devonian aquifers in hydrogeological profile in the Olkusz region (Figure 2). The Triassic aquifer is the most resourceful and intensively drained by three Zn-Pb ores mines (Boleslaw – abandoned at the end of 1996, Olkusz and Pomorzany) and numerous groundwater intakes. Mining drainage in this region has been lasted for more than 400 years. Significant

increase of this drainage has been observed from 1975 after starting of exploitation by the Pomorzany mine - third and biggest mine in this region. Average total amount of water pump out by three mines is about 250 m³ per minute. Triassic aquifer is recharged directly in outcrop areas or indirectly through permeable Quaternary and Jurassic sediments (Figure 2). Important, from the point of view of amount of water inflowing into the Zn-Pb ores mines, are hydraulic contacts of the sedimentation-transgressive type between Jurassic – Triassic and Carboniferous/Devonian - Triassic aquifers (Motyka, 1988) (Figure 2). Intensive drainage by mines and great number of groundwater intakes has caused significant lowering of groundwater table up to over 120 m resulted in creation of the extensive regional cone of depression covering an area of about 470 km² (Rózkowski et al., ed. 1997) (Figure 1).

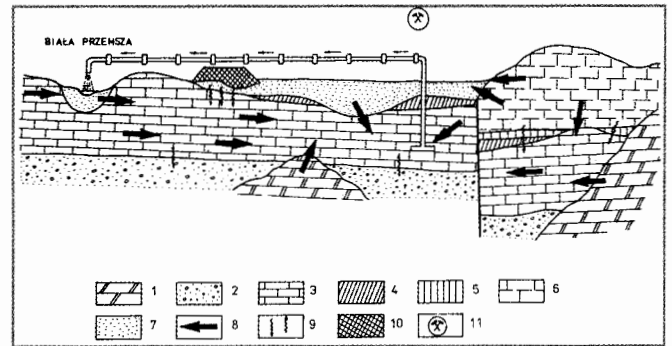


Figure 2. General cross-section – Olkusz region.

- 1- Devonian and Lower Carboniferous carbonate rocks,
- 2- Permian conglomerates,
- 3- Lower and Middle Triassic carbonate rocks,
- 4- Keuper clayey sediments,
- 5- Middle Jurassic marls,
- 6- Upper Jurassic limestones,
- 7- Quaternary sands,
- 8- Direction of intensive groundwater flow,
- 9- Direction of water leakage,
- 10- Landfill,
- 11- Mine (according to Motyka and Wilk, 1980).

There are Quaternary, Triassic and Carboniferous aquifers in hydrogeological profile in the Bytom region (Figure 3). Triassic aquifer is generally uncovered and recharged by direct infiltration in areas of outcrops of the carbonate series or indirectly through Quaternary overburden (Figure 3). The upper part of the Triassic aquifer (Middle Triassic) has been drained by Zn-Pb ores mines for the last hundred years in this region. Mining activity was finally finished in the end of 1989. Unfortunately, because of still performed exploitation of hard coal conducted in Carboniferous rocks situated below Triassic aquifer, pumping of water from this aquifer can not be suspended. Since 1991 after previous closing down of all vertical workings Bolko Shaft has become the central and the only one drainage shaft. Water from other workings is directed towards this shaft by two water galleries, the eastern and western ones, at the average rate about 20 m³ per minute (Figure 3). Intensive mining drainage has led to the creation of a vast (over 100 km²) regional cone of depression in the reach of which the lowering of groundwater table ranges from 30 up to 100 m (Kropka and Witkowski, 1994) (Figure 1).

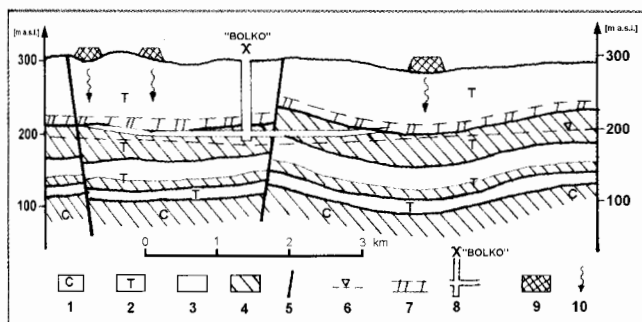


Figure 3. General cross-section – Bytom region.

1- Carboniferous, 2- Triassic, 3- Permeable rocks, 4- Low permeable rocks, 5- Faults, groundwater head of the Roethian aquifer, 6- Ore-mine workings (abandoned), 7- "Bolkó" shaft and two water galleries, 8- Landfills, 9- Seepage of polluted water (according to Kropka and Witkowski, 1994 – modified).

As it was already mentioned the karst-fractured Triassic aquifers are the most important and resourceful in both regions. They form main aquifers called: Bytom (MA 329) and Olkusz-Zawiercie (MA 454) (Kleczkowski, ed., 1990) (Figure 1). Unfortunately because of high groundwater pollution areas subjected to intensive long lasting mining drainage are currently proposed to be excluded from the range of main aquifers (Kropka, 1996, Rózkowski et al., ed., 1997).

CURRENT GROUNDWATER QUALITY

After closing down of the Zn-Pb ore mines the central of the Bytom groundwater basin is still being drained by two water galleries (the eastern and western ones). Presently, in both galleries there are two measuring sites of the regional network of groundwater quality monitoring: site No 13 (western gallery) and No 14 (eastern gallery) (Witkowski, 1997). The chemical composition of water drained from the western part is different from that from the eastern one. It is caused both by natural factors (liability of the mineral composition of the Zn-Pb deposits) and by human ones (differences in the land use and various degrees of the human threat). The TDS of water in the western part varies from 2950 to 4000 mg/dm³ and in the eastern part from 2150 to 2430 mg/dm³. Content of sulphates varies from 1040 to 1520 mg/dm³ and from 930 to 1100 mg/dm³ while chlorides from 370 to 800 mg/dm³ and from 180 to 220 mg/dm³ respectively. Waters occurring in the western part are considerable enriched with the Cl ions. Content of SO₄ in this part is also higher than in the eastern one.

Content of Zn varies from 5,29 up to 12,9 mg/dm³ (in the western gallery) and from 10,7 up to 15,6 mg/dm³ (in eastern gallery). Observed concentrations of Pb in water are similar in both galleries and vary from 0 up to 0,036 mg/dm³.

Generally quality of groundwater of the Triassic aquifer (Muschelkalk) in the central part of the Bytom is very low and concentrations of many pollution indicators (sulphate, chloride, ammonium nitrogen, cadmium, zinc) exceeding standards for potable water.

Natural water inflows to three Zn-Pb ores mines, observation wells as well as springs drained Triassic carbonate rocks situated in the Olkusz area were once sampled in the period of 1996 – 1997. Assessment of groundwater quality in the Olkusz mining area has been based on these results. Generally, the water studied shows considerable variations. The TDS of these waters varies from 400 up to over 8500 mg/dm³. Contents of sulphates vary from 40 up to 5990 mg/dm³, chlorides from 10 up to 30 mg/dm³, calcium from 80 up to 520 mg/dm³ and magnesium from 12,5 up to 1331 mg/dm³. Increase concentrations of metals have been observed too (Table 1).

| Extreme concentrations | Ca | Mg | Na | HCO ₃ | SO ₄ | Cl | Fe | Mn | Zn |
|---------------------------|--------|---------|-------|------------------|-----------------|-------|--------|--------|--------|
| Min [mg/dm ³] | 82,31 | 12,53 | 2,09 | 239,2 | 35,72 | 10,48 | <0,010 | <0,002 | 0,007 |
| Max [mg/dm ³] | 523,80 | 1331,40 | 19,09 | 701,4 | 5986,0 | 27,63 | 1,191 | 6,111 | 10,900 |

Table 1. Minimum and maximum contents of selected pollution indicators in groundwater of the Triassic aquifer in the Olkusz Zn-Pb ores mining area (1996/1997).

Presented above data show that particularly dangerous, from the groundwater quality point of view in both considered regions, are observed high concentrations of sulphates. This decided that analysis of sulphates content in groundwater has been the main subject of presented paper.

OBSERVED CHANGES OF SULPHATES CONCENTRATIONS IN GROUNDWATER

The analysis of sulphates content in groundwater has been based on results of the regional and local groundwater quality monitoring performed in the area of the Olkusz and Bytom regions. There are 6 regional groundwater quality monitoring sites, in the Bytom region, and 3 in the broadly meaning Olkusz region. 7 of them, except previously mentioned two water galleries are situated outside of the direct impact of mining activity (Figure 1). These monitoring sites are a part of regional network established for purposes of the Katowice Regional Water Management Council in 1993. All monitoring sites have been sampled twice a year in the period of 1993-1998. Unfortunately there is a lack of systematic data concerning natural inflows to three Zn-Pb ores mines (Boleslaw, Olkusz and Pomorzany) situated in the Olkusz region. Only systematic monitoring of mine water pumped out by shafts of these mines has been performed. Presented in this paper spatial distribution of sulphate concentration in groundwater in Olkusz mining area is based on the results of single sampling of natural inflows to these mines, existing observation wells and springs done in 1996-1997.

Groundwater of shallow zone of circulation in limestones and dolomites are usually good quality, even when deposits of sulphide ores appear there. Before intensive mining activity TDS of groundwater in carbonate Triassic rocks in Olkusz

region varied between 250 and 500 mg/dm³ and average concentrations of sulphates was 37 mg/dm³, calcium - 56 mg/dm³, zinc - 0,52 mg/dm³, lead - 0,06 mg/dm³.

Currently groundwater quality has been significantly changed in areas of Zn-Pb ores mines activity. Current concentrations of sulphates in groundwater of the Triassic main aquifer (MA 454) in the Olkusz region have been at Figure 4. Highest sulphates concentrations exceeding 5000 mg/dm³, and even over 11000 mg/dm³ in one case, have been noticed in central part of the Pomorzany mine. Contour line of sulphate concentration equal to 200 mg/dm³ (standard for potable water in Poland) encircles almost whole area of these three mines.

Unfortunately, because of the lack of data, reliable analysis of sulphate content and eventual tendencies of changes in groundwater quality, in the areas of mine activities is impossible. Changes, occurring in the analysed Triassic aquifer can be observed solely in the monitoring network observation gaps. Particularly interesting results of the observations of sulphates content changes in the analysed groundwater were obtained in the monitoring site number 8, which is one of wells belonging to a large intake Lazy Bledowski (Figure 1). In the waters of this well there is observed a stable, consequent increase of sulphate content from 63 mg/dm³ (1993) to about 300 mg/dm³ (1997) (Figure 5). Unfortunately, since 1998 when the exploitation of the well was abandoned, the analyses have not been carried any more. Simultaneously, in the monitoring site number 5 it is observed decrease of sulphates content in the analysed groundwater (Figure 5). Reasons for these changes will be discussed in the next chapter.

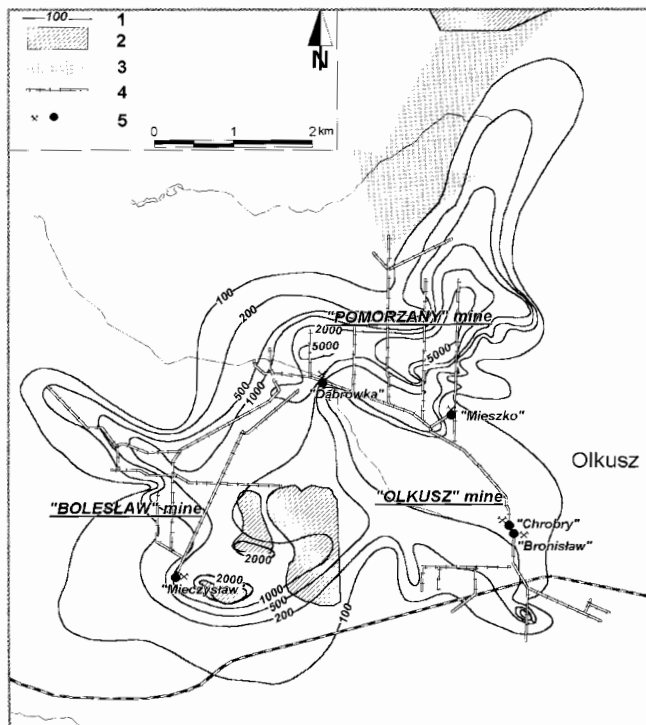


Figure 4. Concentration of sulphates in groundwater of the Triassic carbonate aquifer in the Olkusz region.

- 1- Contour of sulphates concentration in groundwater (mg/dm³).
- 2- Sources of pollution (landfills).
- 3- Flow direction of groundwater polluted by lignosulfonates.
- 4- Zn-Pb ores mines workings.
- 5- Shaft.

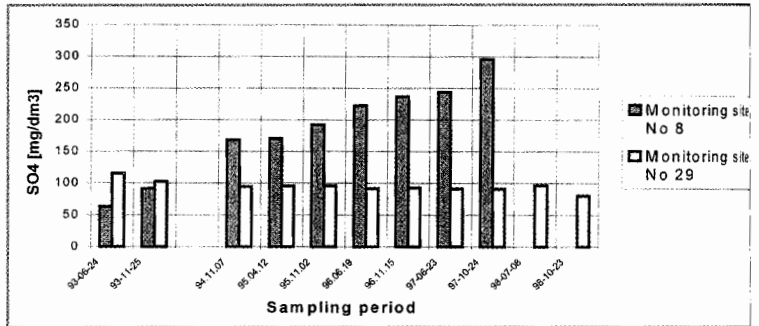


Figure 5. Content of sulphates in groundwater in selected regional monitoring sites in Olkusz region.

Indirectly, the quality of the cumulative mine water pumped out from the mines may testify to the hydrogeochemical changes in the Triassic aquifer in the area of the mines activity. Observations carried in recent years (1993-1998) in the cumulative waters pumped in the shafts of Boleslaw, Olkusz and Pomorzany mines indicate a significant variability of sulphate content. This ion concentrations in the mine water pumped out from the Boleslaw mine via the Mieczyslaw shaft fluctuated in the range of 600-1900 mg/dm³. Sulphate concentrations in the mine water pumped out via the Bronislaw shaft from the Olkusz mine fluctuated from ca. 85 to ca. 190 mg/dm³. Water inflowing to the Pomorzany mine is carried away via 3 shafts (Chrobry, Mieszko, Dąbrowka). High quality water inflowing to the eastern part of the mine is pumped out via the Chrobry shaft. Concentrations of sulphates in this water vary in the range of 60-80 mg/dm³ and only sporadically concentrations of 120-140 mg/dm³ are recorded. Poor quality waters, polluted not only with sulphates but also with lignosulfonates flowing in from the paper mill in Klucze are pumped out via the Mieszko and Dąbrowka shafts (Figure 4). Concentrations of SO₄ in the mine water pumped out via the Mieszko shaft were the most often in the range of 200-400 mg/dm³. In several cases the concentrations exceeded 800 and 1000 mg/dm³. Via the Dąbrowka shaft there is pumped out mine water polluted with the lignosulfonates flowing in from the northern parts of the Pomorzany mine (Fig. 4). Between 1993 and 1998 the sulphate concentrations in these waters ranged between ca. 100 to almost 250 mg/dm³. Periodically, higher concentrations, exceeding 300 mg/dm³ were observed. In the mine water pumped out via four active shafts between 1993 and 1998 a tendency to increase SO₄ concentrations was not observed. This tendency in the recent years was observed solely in the Chrobry shaft (Figure 6).

Analysing the quality of mine water pumped out from the mines it is necessary to be conscious that the mine activity itself deteriorates quality of water. Water inflowing to the mine is usually higher quality than the pumped out one. It is influenced by many factors i.e. mining and disintegration of rocks, transport, using of process water and presence in excavations of miners and machines. Mining and co-related disintegrating of rocks produce suspension. Very large area of contact between

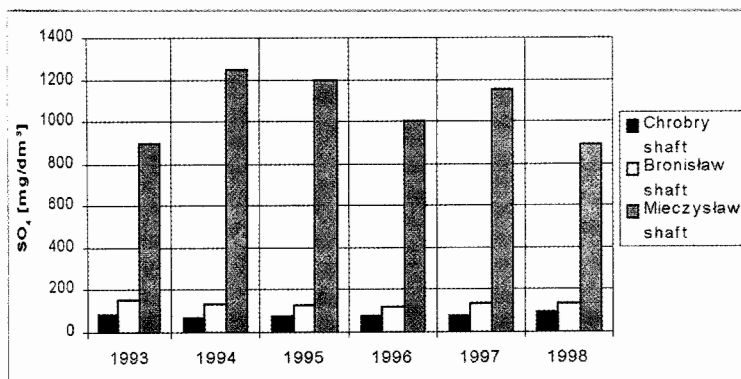


Figure 6. Average sulphate concentrations in mine water pumped out from Zn-Pb ores mines in Olkusz region.

the water and the suspension encourages dissolution of some constituents and increasing its concentrations. Transport of winning and materials from the mine encourages formation of the suspension and its mixing with the water. Using of process water in the excavations (mainly for hydraulic backfilling) has caused dissolution of constituents produced by weathering processes. It has caused changes of mine water quality.

Water pumped out of abandoned mines represents natural inflows. This situation is observed in the area of Bytom. Regularly analysed since 1993 waters exhausted via the eastern and western drain gallery do not exhibit a tendency of sulphates content increase (Figure 7).

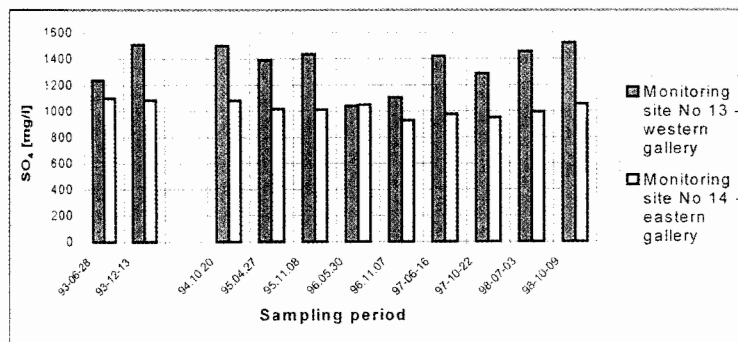


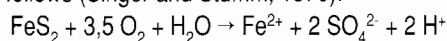
Figure 7. Content of sulphates in groundwater in selected regional monitoring sites – Bytom region.

MAJOR REASONS OF HIGH SULPHATES CONTENT

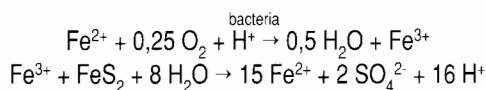
In both analysed areas changes of groundwater quality were caused by the natural processes connected with the changes of the natural hydrogeological conditions and with the negative impact of surface pollution sources. Primary, main reason for some observed disadvantageous changes in the groundwater quality in the analysed Triassic aquifers (MA 329 and 454) is decreasing of the natural level of groundwater table in carbonate Triassic rocks preceding deepening of the unsaturated zone. Saturated zone has been radically reduced especially in the centres of mining exploitation area.

Replacing of the saturation zone by the unsaturated one results in the change of oxidation-reduction conditions and connected chemical reactions changing primary quality of water. Additionally, decreasing of the primary groundwater level causes changes in the directions of surface and groundwater flows. As a consequence, poor quality water both natural (more mineralised) and polluted by a human activity can recharge the aquifer from the surface of the area or from other parts of the aquifer or from other aquifers.

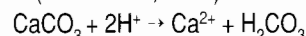
Lowering of the groundwater table and deepening of the unsaturated zone is a reason for the increase of oxygen content in the primary more or less reducing environment. This causes oxidation of sulphide minerals. Especially disadvantageous for the water quality is weathering (oxidation) of pyrites taking place as follows (Singer and Stumm, 1970):



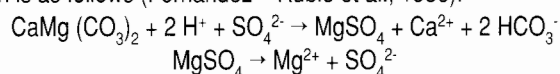
With the assistance of bacteria there takes place oxidation of the bivalent iron to the trivalent iron and then occurs a complicated process of this iron reduction to the bivalent form. The following reactions take place:



Similar reactions take place while other sulphide minerals weathering present in the sphalerite and galena beds. Increase of the hydrogen ions concentration is the result of both of these reactions which means the decrease of the pH reaction and increase of water acidity. There is also observed an increase of sulphate concentration and due to metals higher migration in the acid water environment also their higher concentration. Acidity of water can be fast neutralised (buffering process). Velocity of the process depends on the kind of rocks in which the reactions take place. Among others carbonate rocks (limestones and dolomites) have acid buffering abilities. For instance the buffering process in limestones takes place according to the reaction as follows (Smith et al., 1994):



Dolomite reacts similarly but simultaneously with the increase of calcium ion concentration, concentration of magnesium ions increases as well. The reaction of dolomite dissolution is as follows (Fernández – Rubio et al., 1986):



As a result of buffering reactions increase of calcium and magnesium ions content and as a consequence total water hardness are observed. As stated above acidity of water (decrease of pH reaction) activates metals gathered in the rocks and makes them in their ion forms to get to the water solutions. Buffering process (increase of pH reaction) causes precipitation of metal salts and is often encouraged by bacterial activity. In se of the decrease of metals content in the water solution, concentrations of some of them usually remain much higher than in natural water solutions.

Process of geochemical transformation of the unsaturated zone in the rocks hosted sulphides of metals is much more complicated than one described above. In the drained and oxygenated and simultaneously dried (older parts of the mine) part of the bed there are created complicated mineralogical forms, characteristic for the weathering zone of sulphide beds. There are, the most often, hydrated minerals of calcium, magnesium and iron, containing increased quantities of metals built into their crystal lattice. Solubility of hydrated sulphate of iron (melanterite) and magnesium (epsomite, hexahydrate) is particularly high. TDS of water increases drastically if process water (e.g. filling water) is conducted into the excavations like this or if the excavations are flooded. Increase of concentration of magnesium, calcium, iron and other metals sulphides is the most significant.

The inflow of pollution from the surface of the ground is an additional factor considerably affecting the chemical composition of groundwater in the Bytom and Olkusz regions.

The most important sources of pollution of the human origin deteriorating quality of groundwater in the Triassic carbonate formations in the area of Olkusz are:

- former discharge area of lignosulfonates from the Paper and Cellulose Mill in Klucze which were deposited in the sands of Pustynia Bledowska (Desert of Bledów) (Figure 4);
- heaps of after-flotation waste from which leachates are enriched in sulphates and heavy metals;
- heaps of reach in metals wastes from the furnace bridges situated in the area of ZGH Boleslaw; and
- after-flotation and communal wastes deposited without any protection in the abandoned open pit of Galmei in the area of Boleslaw.

Groundwater in the Triassic aquifer in the area of Olkusz is also influenced by uncontrolled leaks of sewers from leaky septic tanks and manure as well as pouring out these wastes directly onto the ground.

It can be deduced from the disposition of sulphate concentrations (Figure 4) that the processes of sulphate minerals oxidation taking place in the rocks are the most significant factors influencing these concentrations in the Triassic aquifer in the area of Olkusz.

Much more complicated situation has occurred in the Bytom area where exceptional concentrations of different real and potential sources of groundwater pollution had been noticed (Kropka and Witkowski, 1994). In the reach of Triassic aquifer in the Bytom region are situated over active and abandoned landfills. Most of coal mine wastes landfills are situated in the central and western parts of this area. These landfills are important sources of chlorides and sulphates. Observed significant groundwater contamination in this region is a result of this overlapping negative influence of various pollution sources. Higher negative human impact is noticed in the western part of the region and in this part groundwater is more polluted. Dea-

ling with the causes of differences in sulphates content in water in the western and eastern galleries it is necessary to take into consideration the character of the Zn-Pb deposits. In the western and central part of the Bytom area the sulphides minerals are predominant while the farther east the more oxygenic minerals can be found (Kropka and Witkowski, 1994) so oxidation processes can create there potentially greater amount of soluble sulphates. Similarly to the Olkusz region most important source of sulphates in the groundwater of the Triassic aquifer in the Bytom area are oxidation processes of sulphides minerals. Increasing trends of chlorides, sodium and potassium concentrations and quite stable content of sulphates in analysed groundwater suggest significant impact of pollution percolated from the surface of the ground on quality of this water.

Because of intensive drainage and karst-fractured type of the Triassic aquifer with generally fast groundwater flow created in them soluble products of sulphides oxidation are removed relatively quickly with mine water pumped out from the mines. This situation will change when mining drainage will be stopped and gradual flood of mines will happened. In that case decrease of velocity of groundwater flow will occur and longer period of the contact between water and rocks will cause significant increase of concentrations of such constituents as SO_4 , Ca and Mg. It will happen in the area of currently abandoned Boleslaw mine (Adamczyk et al., 1998). A good example of the scale of risk to groundwater quality caused by returning to natural hydrodynamic conditions in the areas previously intensively drained is the case of significant groundwater intake called Lazy Bledowskie. This intake is generally situated North of Olkusz and it pumps out water from carbonate Triassic aquifer. Significant increase of sulphates content in this water has been observed in the last few years. In the same time raising of groundwater table, caused by reduction of water withdrawal and simultaneous significant increase of precipitation, has been observed. This negative trend is particularly visible in observation well No 8 (Figure 5). Observed increase of groundwater contamination caused gradual closing of few wells of this intake.

It should be mentioned that according to the latest results of the groundwater quality monitoring performed in whole Katowice district in areas not subjected to negative impact of mining activity and other local sources of pollution general decrease of sulphates content in monitored groundwater is observed (Figure 5).

SUMMARY

High concentrations of sulphates, heavy metals and locally also lignosulfonates (in the Olkusz area) observed in the discussed areas have been caused by overlapping influence of various sources of pollution situated on the surface of the ground and also by processes of sulphates oxidation connected with the mining activity.

In both discussed areas the processes of oxidation of sulphate minerals taking place in the massif are the most significant reason for the observed high concentrations of sulphates in the groundwater of Triassic aquifers.

Currently it is not possible to discover the quantitative share of sulphates originated from the separate sources (oxidation of sulphates in the unsaturated zone, landfills, communal sewers) in their total quantity recorded in the analysed groundwater. This problem is currently under careful examination and hope is set on the analysis of the isotope composition of sulphur.

As a result of planned closure of some Zn-Pb ore mines and successive ceasing of water pumping leading to reconstruction of the primary hydrodynamic conditions, are expected important changes of groundwater quality and their significant sulphate content increase.

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