

SOIL MOISTURE VARIABILITY AND ACID MINE DRAINAGE IN THE SPOIL DUMP OF PYRITIZED HYDROQUARTZITE IN THE REGION OF BANSKÁ ŠTIAVNICA, SLOVAKIA

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Abstract The problems of acidification of some environmental components by acid mine drainage (AMD) are well-known all over the world, mainly in the regions with developed mining industry and in the regions with abandoned mines. AMD production and its consequences can be found also in the spoil dump of pyritized hydroquartzite in the locality of Malý Šobov in the Banská Štiavnica region (central Slovakia). This paper presents the results of two-year soil moisture research, its distribution, regime in the AMD production in this locality. The seasonal soil moisture variability in the spoil dump corresponds with the AMD production intensity (in spring and autumn the spoil dump moisture of about 20 % and temperature create suitable conditions for the activity of autochthonous microflora). The space distribution of soil moisture identified the permeable zones in the spoil dump (where the primary process of pyrite decomposition is expected) and impermeable zones where water and AMD are retained. This knowledge could be useful in application of other remediation processes into the spoil dump.

Keywords: spoil dump, quarry, soil moisture, AMD, pyritized hydroquartzite

1. INTRODUCTION

Historical and present extraction of coal, gold, polymetallic and other ores has resulted in the exposition of large quantities of sulphides at a surface distributed in mine tailings, tailing dams, open pits, etc. Pyrite, which is the most abundant mineral in these sulphide deposits, is decomposed in water and in the presence of oxygen and it produces effluent - acid rock drainage ARD (created in situ without any mine activity) or acid mine drainage AMD (produced in mine waste) (Sánchez-Espaná, 2005; Druschel, 2004; Naicker, 2002; Holmström, 1999, 2001; Dhahabpour, 2005). Production of AMD is augmented by microorganism consortium Archaea and Bacteria,

especially of the genera *Acidithiobacillus* (Norris, 2007; Tributsch & Rojas-Chapana, 2007; Rawling, 2002; Mahmoud, 2005). AMD is strongly acid sulphate water (pH 1-3) and it contains, except for primary products of the pyrite oxidation (H_2SO_4 , Fe^{3+}), also other solubilized metals (Cu, Pb, Zn, Al, etc.) depending on the composition of deposited waste rock. The leaching study, conducted with *Acidithiobacillus ferrooxidans* and *A. Thiooxidans* of tailing sediments and ore minerals from Sb, Au deposit Pezinok (Malé Karpaty Mts, Slovakia) shows the oxidation and the leaching progression on surface of the following minerals: löllingite, arsenopyrite, stibnite, native Sb, sphalerite, chalcopyrite (Andráš et al. 2008). AMD, mine waste dump, settling pit represents dangerous waste and high risk to the environment, contamination of groundwater, surface water, plants, animals. Several studies show the anomalous cell-wall, occurrence of calluses, resin canal and heavy metal contamination of plant tissue with Fe, Zn, Pb, Cu (Andráš et al., 2007, 2008). Charbonnier (2001) pointed out this danger within the framework of inventory control of mine waste in the EU countries.

All the land components of the mountains Štiavnické vrchy, especially the surroundings of Banská Štiavnica (Križani, 2007), are deeply affected by ore extraction, dressing and compaction. In the period from the 16th to 19th century it was the extraction of precious ores, at the end of the 20th century it was the extraction and dressing of nonferrous ores and open-pit mining of hydroquartzite. Environmentally dangerous locality in this region is the spoil dump where the pyritized hydroquartzite is taken after the hydroquartzite extraction in the locality of Banská Štiavnica – Malý Šobov. Several studies are devoted to the AMD microflora, acidification and contamination of the environment, possibilities of maintenance and passive treatment of this landfill (Šucha et al., 1997, Šottník & Šucha, 2000, Šottník, 2000, Šlauková & Bella, 2006, Kočická, 2007, Marušková, 2008). Križani et al., (2009) investigated the bioavailability of some elements in the locality of Šobov (quarry and dump), Sedem Žien (settling pit), Nová shaft (dump). Sequential analysis indicated high portion of Mn, Cu, Zn, Pb, Cd in the bioavailable forms and low portion of bioavailable bonds in As, Sb, Hg in the selected minerals (pyrite, marcasite, sphalerite, galena). The experiment of static maceration of heavy metals by acid metal-rich sulphate water from Šobov showed different surface degradation of mineral pyrite, marcasite, sphalerite, galena and formation of secondary mineral on their surface (mainly jarosite and gypsum).

Our paper deals with time and space distribution of soil moisture of this spoil dump of pyritized hydroquartzite and its possible relation to the production of AMD. The paper presents the results of soil moisture research from the two - year observation period 2006 - 2007.

2. DESCRIPTION OF THE STUDIED AREA

The studied area is situated on the slope of a mountain called Malý Šobov I, approximately 1 km northwards from Banská Štiavnica (Fig. 1). There is a quarry, where the surface mining of secondary quartzite, rich in silicium dioxide (SiO_2), was carried out during the period 1952-2005. The extracted rock served for the production of the refractory bricks in the factory Dinas, Banská Belá. In the same area, at the

distance of 200 m southwards from the quarry, there is a spoil dump used as a disposal site for waste rocks that are unsuitable as input raw material for the production of bricks.

There are two types of rocks in the quarry:

- the first type consists of monotonous fine-grained quartzite with younger quartz stringers, characterized in the presence of two generations of pyrite,
- the second type of quarry rocks, important from the point of view of acidification, consists of dark rocks rich in pyrite, which, because of their inconvenience for the production of Dinas products, are taken to the spoil dump.

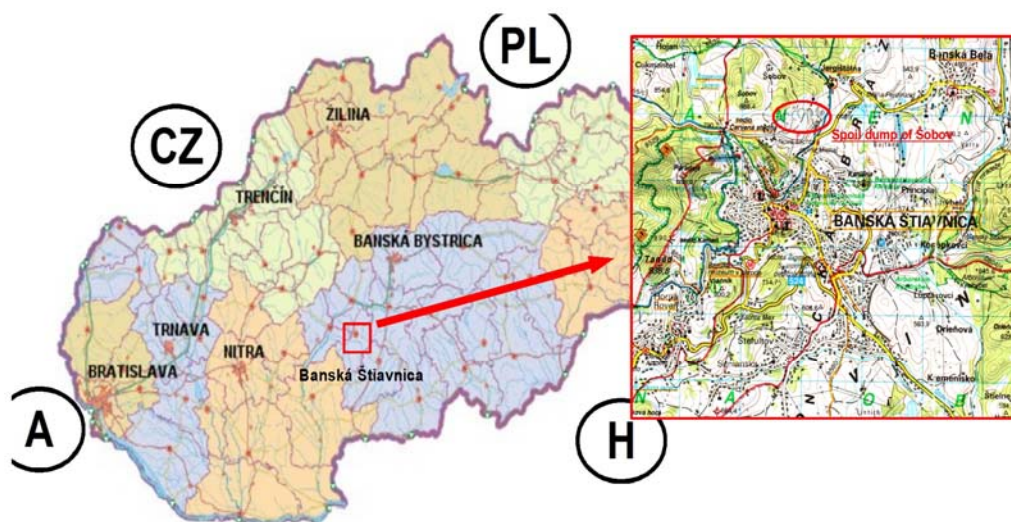


Fig.1 Map of the surroundings of Banská Štiavnica

The spoil dump, rich in impurity of pyrite, is formed by non-sorted unconsolidated material of various grain compositions (substantial part is formed by pyritized quartzite of boulder character). Therefore it is characterized by good permeability for atmospheric oxygen and water, and together with the southern exposure of the slope the dump creates convenient conditions for the oxidation of pyrite by acidophilic bacteria, *Acidithiobacillus ferrooxidans*, *A. thiooxidans*, *Leptospirillum ferrooxidans* (Šlauková, 2006; Šlesárová, 2004). The primary product of biooxidation, sulphuric acid, attacks consequently other exposed minerals and thus the metals mobilization and migration is facilitated. The effluent from the spoil dump (AMD) is acid (pH 1,7-2,5) and contains solubilized metals, mainly Fe, Al, and Mn, Zn, Cu, As, Si in minor abundance (Michalková, 2008). Concentrations of different elements, as a result of the processes of weathering and lixiviation in the spoil dump, are characterized by seasonal variability (Praženicová, 2008).

3. CLIMATIC AND HYDROGEOLOGICAL CONDITIONS

From the point of view of climate the area of quarry and spoil dump of Malý Šobov and its wide surroundings (the Štiavnické Vrchy Mountains) belong to warm

temperate zone with humid climate (Lapin et al., 2002). Bláha & Vitásek (1993) give the following values for some climatic characteristics of the studied area:

- long-term average annual evaporation: 600 mm,
- long-term average of annual temperatures: in January – 4 to – 6°C, in July 16 to 17°C.

Average annual precipitation depth ranges from 750 to 950 mm, while during the warm half-year it is approximately 400 to 500 mm. The most of precipitation falls in May (85 mm) and the least in January (40 mm). The average for the period 1951-1993 was 753 mm/year, and for the period 1951-2005 it decreased to approximately 700 mm/year. The decrease can be attributed to global warming of the Earth, which has been particularly notable in the last 30 years.

The studied area belongs to the river Hron basin. The regime of surface runoff is of pluvial-snowy type with accumulation from December to February. At long-term average the highest rate of streamflow is in March - April and the least in September.

According to the principal hydrogeological regions of Slovakia (Malík & Švasta, 2002) the studied area belongs to the zone 88 – neovolcanites of northern slopes of the mountains Štiavnické vrchy and Javorie. The underground water is created only by atmospheric precipitation. Part of it infiltrates into the rock environment and substantial part forms surface runoff. Very small values of specific groundwater flow were found in the studied area (on average $0,8 - 1,2 \text{ l.s}^{-1} \cdot \text{km}^{-2}$). This implies that atmospheric precipitation is accumulated only in the upper part of the weathering zone. For the total groundwater and surface specific runoff Bláha & Vitásek (1993) indicate $10 \text{ l.s}^{-1} \cdot \text{km}^{-2}$.

In case of storm rainfall, when runoff is faster than weathering process, there is a dilution of AMD. The weathering process on the spoil dump happens during the rainless period, too.

4. MATERIALS AND METHODS

Soil moisture of the spoil dump of Šobov was monitored in three geomorphological levels. By the geomorphological level we generally understand a horizontal line going from the east to the west of the spoil dump. Level 1 corresponds to a base of upper slope of the spoil dump (line made by the points 11, 21, 31), level 2 to the central horizontal line from the spoil dump to waste rock (line going through the points 12, 22, 32) and level 3 – the line creating upper border of the humid slope of the spoil dump – corresponding to the points 13, 23, 33. The spoil dump as a place of deposit of waste rocks has an area of 3000 m^2 (width of 30 m, length of 100 m). For the needs of our study we divided it from the east to the west into three sampling zones. In each zone we fixed the sampling points from the north to the south. The zone 1 is represented by the points 11, 12, 13; the zone 2 by the points 21, 22, 23 and the zone 3 is represented by the points 31, 32, 33.

In each of them we took the soil samples in the depth of 10, 20 and 40 cm. Thus we formed a matrix SSD (i, j, k) where SSD stands for the samples in the spoil dump, i – indicates the zone number, j - number of the sampling point and k – number corresponding to the depth (k=1 for 10 cm, etc.). Check samples come from the control

area, situated in the south and adjacent to the spoil dump. These samples are represented by the vector CS (k) – check sample of the number of the depth k.

Samples of 10 to 15 g were taken by earth auger into LDPE waterproof packets. The samples were then transported to the laboratory. Soil moisture was monitored in 2006 and 2007, in April, May, June, July and October. July and October represent the summer season and the autumn season respectively. Thus we obtained the set of 270 sample data of soil moisture. From the climatic and hydrological points of view the years 2006 and 2007 were proved to be similar. Therefore we worked with the set of values representing the average of these two years, i. e. the set of 135 data. Actual moisture content of the spoil dump was determined by the gravimetric method according to the formula:

$$W=100* (w_b - w_n)/w_n \quad [\%]$$

where

W represents soil moisture in weight percentage of the sample dried at 105°C,

w_b – weight of the sample of raw soil (i. e. containing water),

w_n – net weight of soil dried at 105°C.

5. RESULTS AND DISCUSSION

Due to a lack of studies dealing with soil humidity specific for spoil dumps, we used, for the needs of comparison, results acquired in the spoil dump and in the control area. The values of humidity obtained from the control area are generally higher than the values obtained from the spoil dump. Soil humidity of the control area oscillates between 20 and 40%, while soil humidity of the spoil dump ranges from 15 to 30% (Fig. 2 & 3). This difference is due to low slope angle and to different soil structure of the control area. According to the figure 2 three characteristic periods of the soil humidity regime of the spoil dump and of the control area can be observed: moisture increase from April to June, decrease during summer months and slight increase during autumn months. This corresponds with climatic conditions of the locality, precipitation and temperature.

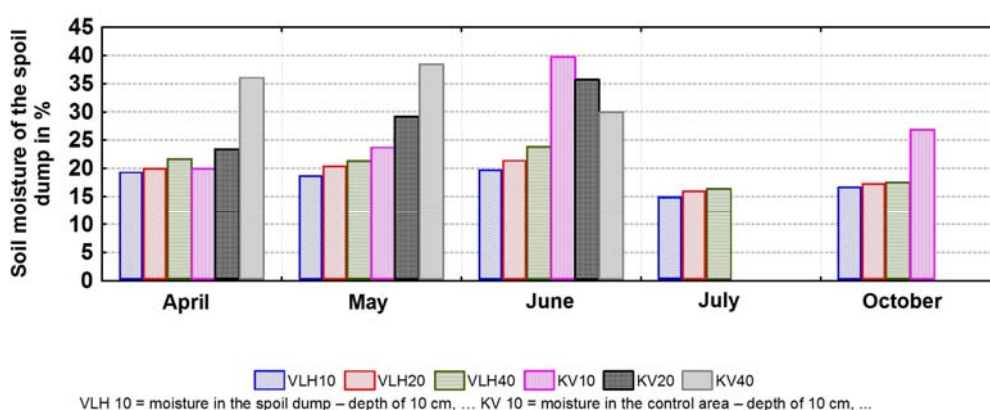


Figure 2, Time distribution of soil moisture of the spoil dump of Šobov, Banská Štiavnica, Slovakia

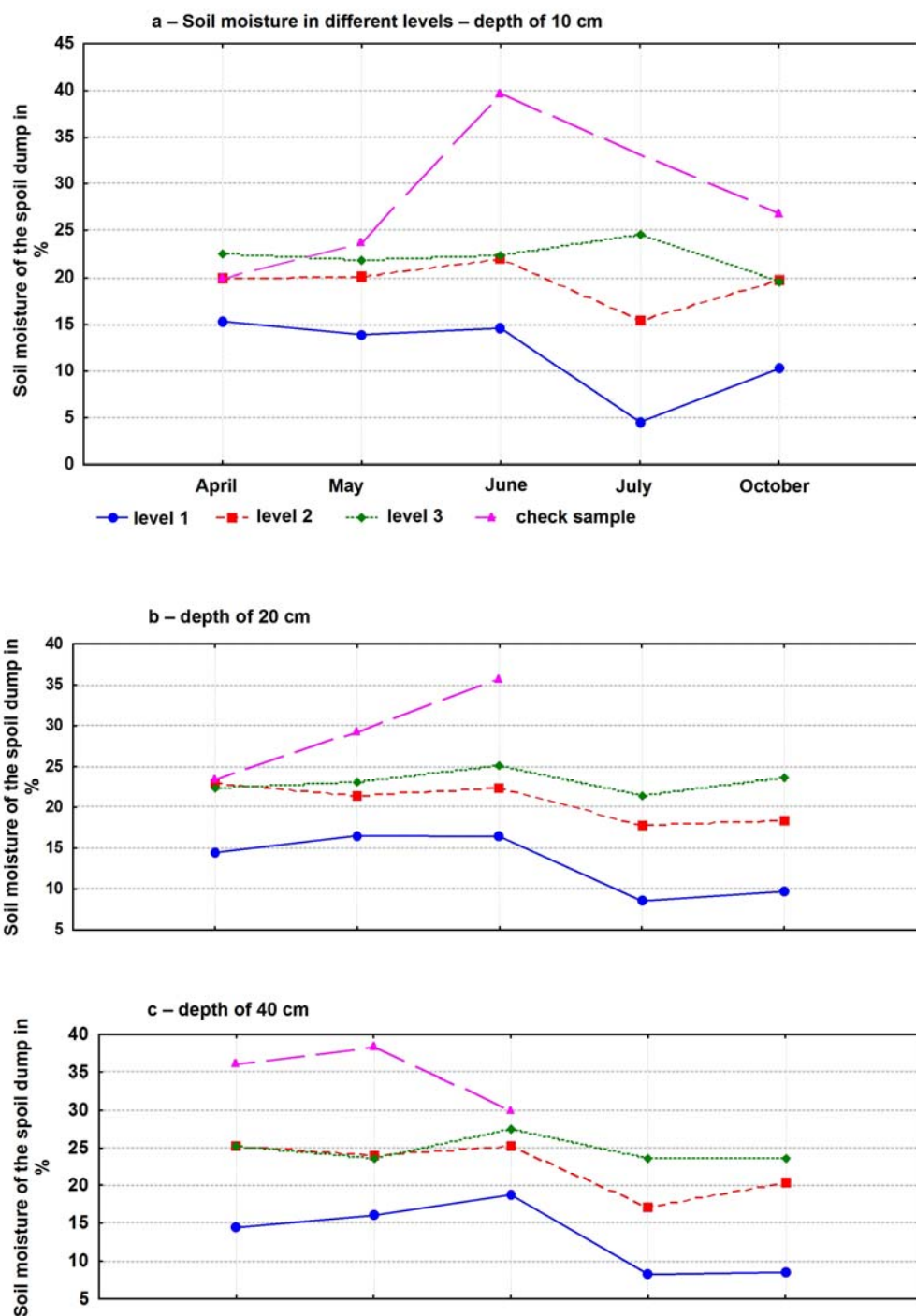


Figure 3, Time distribution of soil in different levels of the spoil dump Šobov, Banská Štiavnica, Slovakia

During the observation period winter months had the most precipitation (cca 80 mm/month) and summer months the least (cca 42 mm/month). The seasonal variability of spoil dump humidity and of precipitation in this locality is reflected in the intensity of AMD production (Praženicová, 2008). In spring AMD with lower concentration (but in higher volume) was produced. In summer it was conversely. From the point of view of materials balance of Fe in AMD (Fe is a dominant solubilized metal) it was proved that in summer four times less Fe was released by the biooxidation processes than in spring. It can be concluded that humidity, saturation of soil with the air in spring create favourable conditions for the activity of autochthonous mesophilic microflora in the spoil dump.

Space distribution of moisture in the subsurface layer of the spoil dump, continuous increase of moisture from the highest level (level 1) to the lowest level (level 3), and this for all considered depths of soil (Fig. 3a - 3c), correspond to the dump geometry (slope gradient 70°). Level 1 shows certain stability in moisture values or slight increase from April to June (15-20 %) and sharp decrease in July when the value falls to 5 %.

Similar tendency is observed for the level 2, but with the respective moisture values of 22, 23, 40 % from April to June for the depth of 10 cm; 22, 25 and 37 % for the depth of 20 cm and 25, 27 and 38 % for the depth of 40 cm.

Space variability of soil moisture of the spoil dump is well visible in figure 4. Analysing this figure, we observe that moisture in the zone 1 varies (increases) not only on the surface (i. e. from the highest sampling point to the lowest one), but also in the vertical direction, in a zone of soil aeration. Expressed in figures, this variability oscillates between 12 and 30 %.

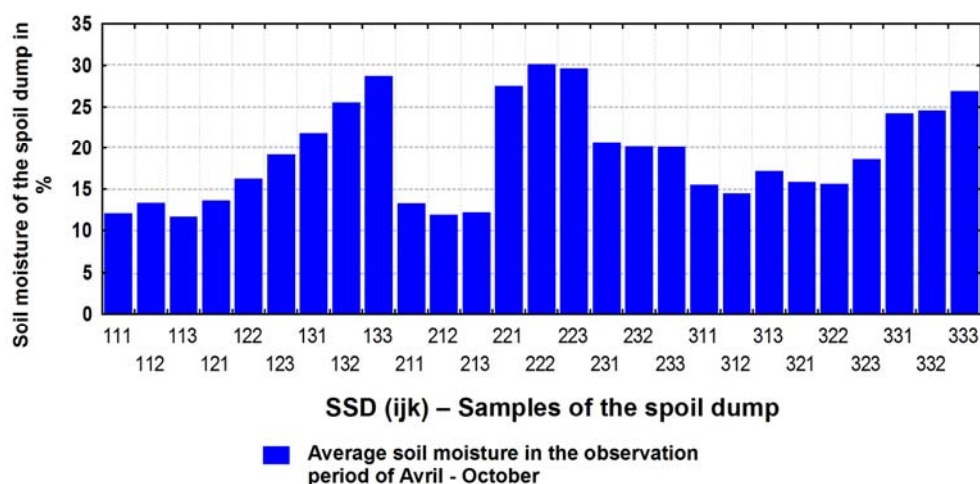
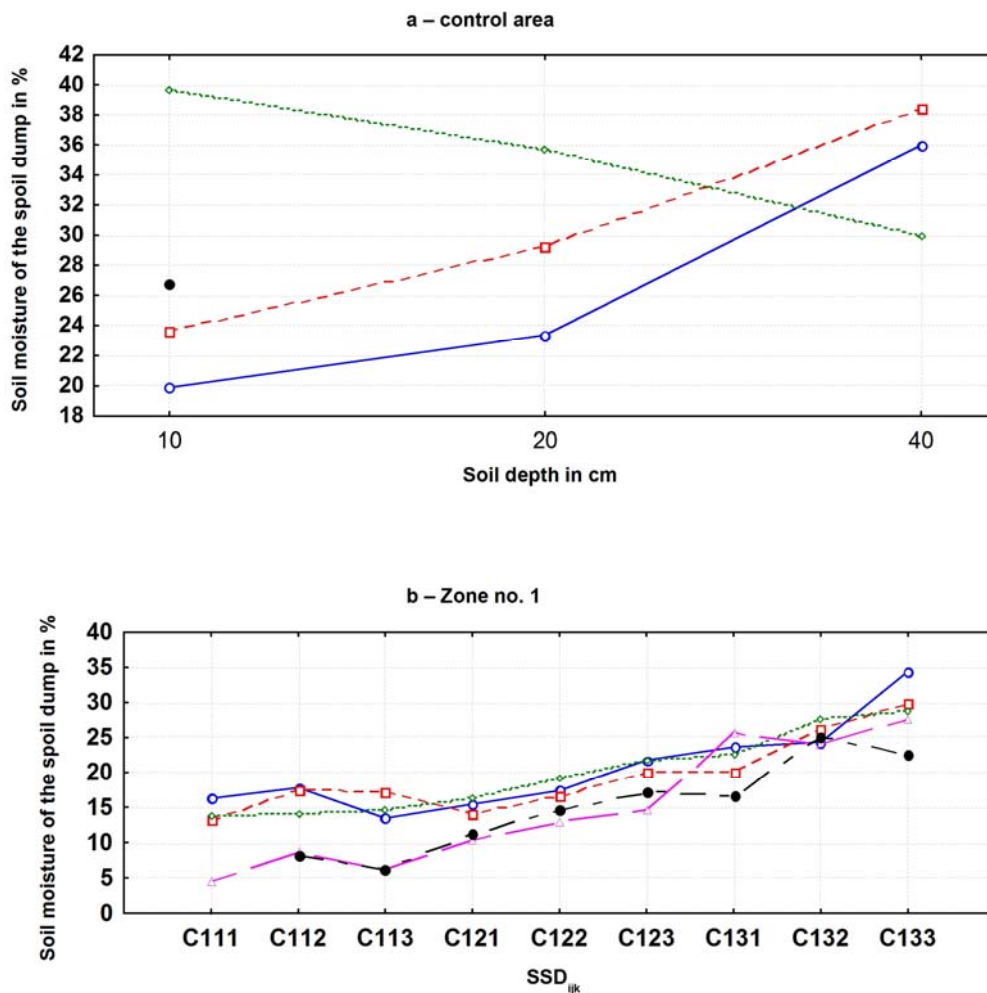


Figure 4. Space distribution of soil moisture of the spoil dump Šobov, Banská Štiavnica, Slovakia

The zone 2 shows variability presenting in the beginning by moisture stability of 11-13 %, then a sharp increase in the middle of the zone reaching the value of 30 % and finally a slight decrease to approximately 20 %.

The zone 3 also shows moisture increase in dependence on the sampling points, but this growth is more moderate in comparison with the zone 1. For example, for the sampling points no. 1 and 2 of the zone 3 moisture is stable at about 15 % and for the sampling point no. 3 of the same zone it is 25 %.

The space and time differences in humidity distribution are probably related to the geomorphology of the spoil dump and underlying rock under the spoil dump. Spoil dump with the high content of pyritized hydroquartzite is deposited in a thick layer on the slope of the underlying rock from quaternary diluvium (permeable horizon) (Rojkovič et al., 2003). Under this permeable layer, there is another horizon created by altered tuff sealing horizon. AMD production in pyritized waste depends on the presence of water and oxygen (Šottnik, 2000). The sharp decrease of humidity in summer, mainly in the depth of 10 cm, as a consequence of higher evaporation and lower precipitation, gave rise to lower intensity of AMD production.



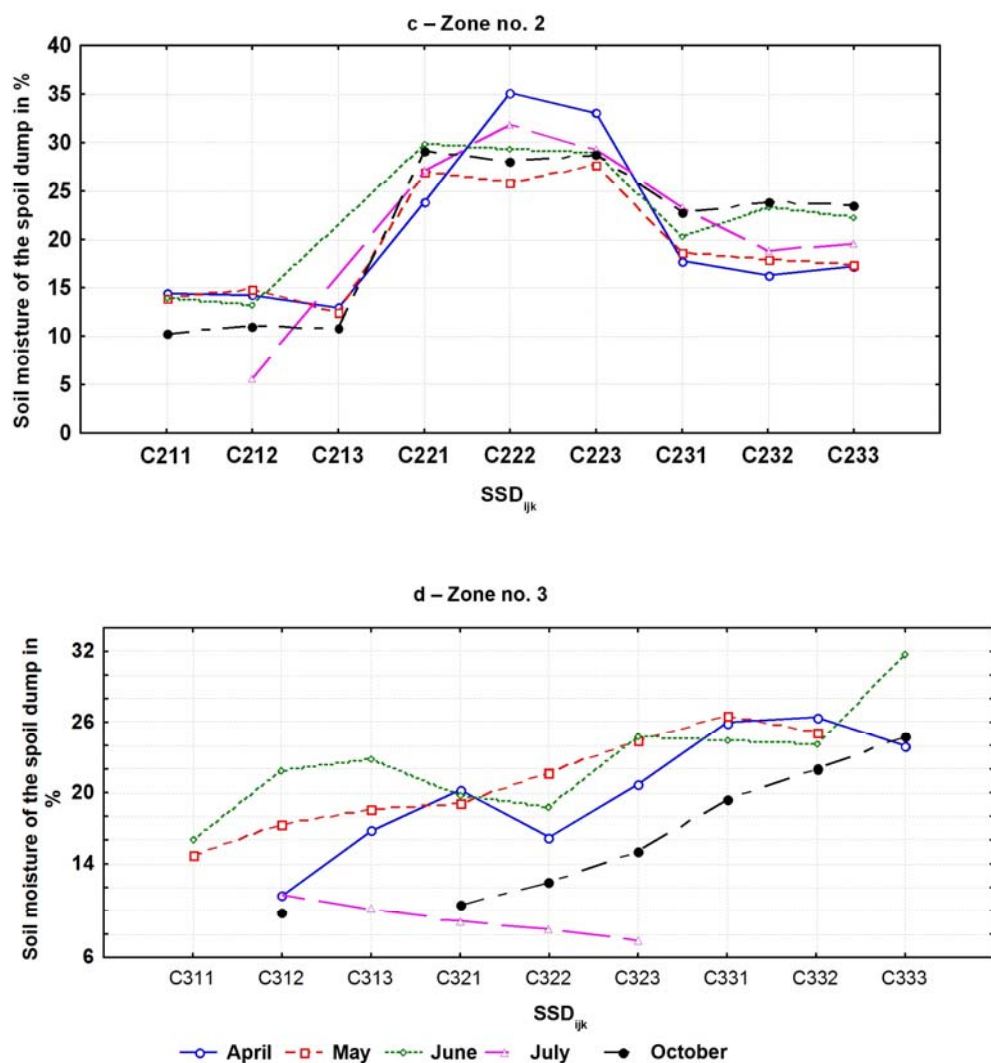


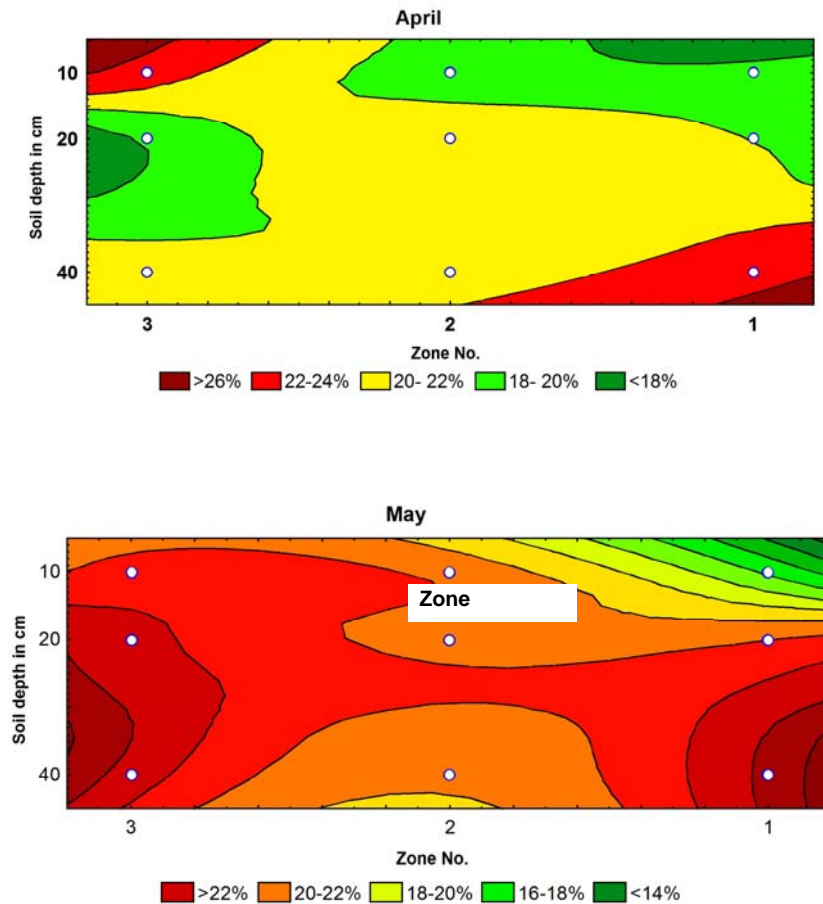
Figure 5, Time and space variability of soil moisture of the spoil dump of Šobov in Banská Štiavnica, Slovakia.

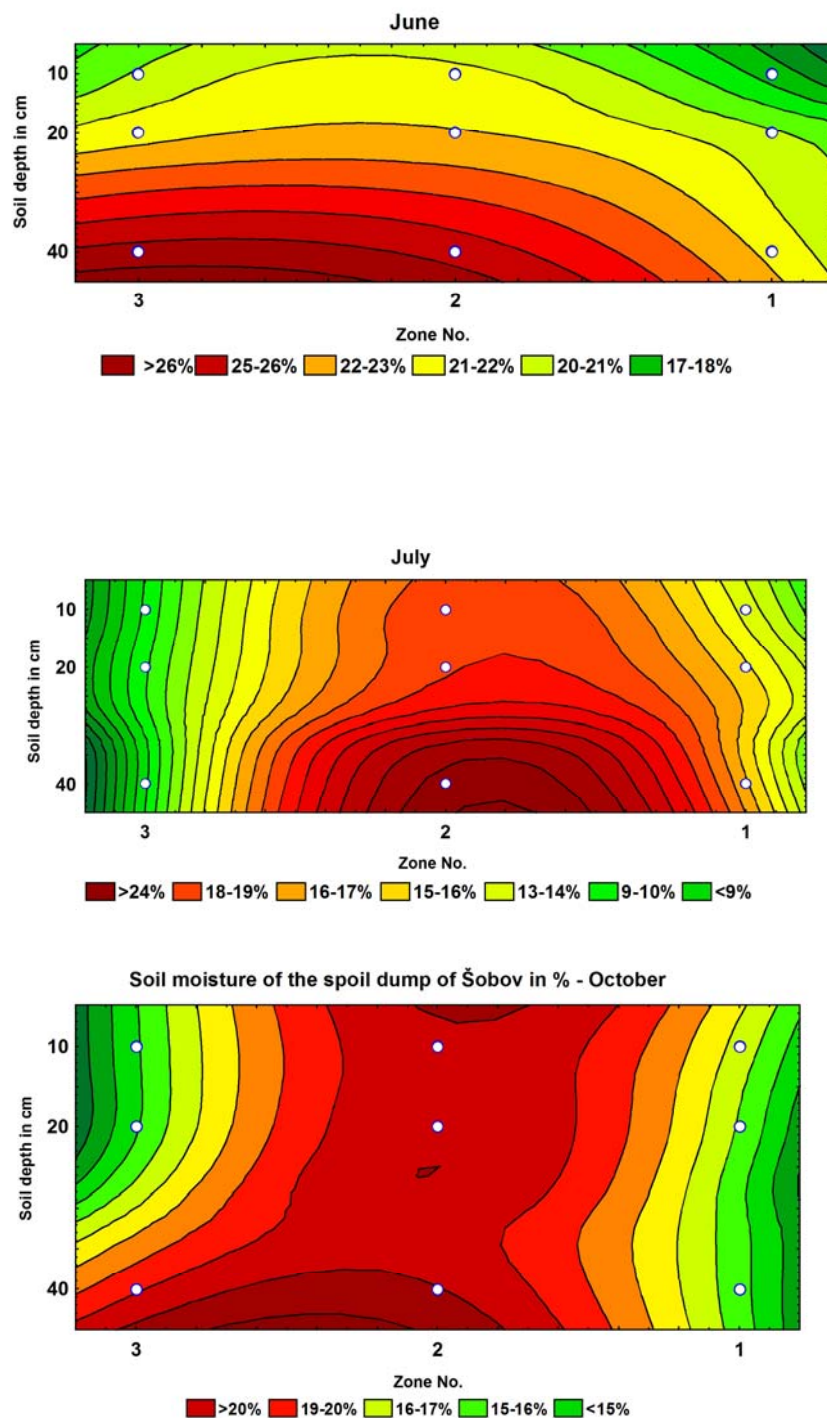
Figures 5 a, b, c, d express dynamics of soil moisture of the spoil dump as a function of two variables: time (months of observation) and sampling points, and this for every zone of the spoil dump. Analysis of zones 1 and 3 indicates that in general moisture increases proportionally with the depth of soil and decreases irregularly from level 1 to level 3 of the spoil dump during the whole observation period (April – October), except for the zone 2, where so-called “moisture wave” is observed in its centre. In that place spoil dump is probably mixed with impermeable clay minerals with significant water retaining capacity and water from higher permeable layers of the spoil dump is retained there. At the same time it can be a proof of the presence of impermeable underlying rock with a layer that still retains drainage water or AMD from higher layers. In this wet zone the intensity of biooxidation processes and AMD

production is minimal, mainly because of atmospheric oxygen and CO₂ limitation.

In the control area, change of moisture variation in dependence on depth is relatively sharp in comparison with the variation observed in different zones of the spoil dump. This moisture increases in April and May and decreases in June. It means that the control area is formed by more permeable material than the spoil dump and soil moisture is created by water from precipitation.

Moisture variability as a function of soil depth and zones for different months of the observation period is a topic of figure 6. Study of this figure shows that for instance in June, July and October soil moisture increases in function of soil depth and it is more important in the middle of the spoil dump. Soil moisture isohypses indicate places and direction of water infiltration from precipitation into the spoil dump. Increased soil moisture (the consequence of precipitation in winter) in the zones 1 and 3 in April (the driest month in the observation period) penetrates into the zone 2 (less permeable).





Figure, 6, Soil moisture isohypses of the spoil dump according to the zones for different months

Moisture penetration into the zone 2 is considerable in following months as a result of intensive precipitation – in May (100 mm/month) and in August (68 mm/month). It seems that peripheral zones of observed spoil dump surface are permeable for precipitation and air, which are favourable conditions for degradation of sulphidic waste and production of AMD. In the central part of the zone 2 (Fig. 5) drainage water, or possibly AMD, are cumulated. During warmer and drier period AMD is concentrated due to increased evaporation and low flow of drainage water in this area and the concentration of dissolved metals are increased three times in comparison to the period with more precipitation (Praženicová, 2008).

According to this study, places with favourable conditions for biodegradation of sulphide materials and production of AMD can be identified on this spoil dump and they would be suitable for application of remediation technologies to limit the AMD production. Wet zone localization in the spoil dump with retained drainage water could help to transfer and regulate the flow of this water.

6. CONCLUSION

This study results can be summarised in these important points:

- 1) Soil moisture of the spoil dump is proved to be sufficient (20%) and its time distribution is suitable from the point of view of releasing soil oxygen and carbon dioxide as one of the components participating in the process of AMD production by biodegradation of present sulphidic waste, mainly pyrite, and also degradation of other spoil dump components and mobilization of other metals.
- 2) The seasonal variability of soil moisture, related to climatic conditions in the area (precipitation, temperature), corresponds with the intensity of AMD production and precipitation in this locality.
- 3) It is evident from the space distribution of soil moisture that the spoil dump has two different zones of the AMD production, or the conditions for AMD production, from sulphidic waste. Peripheral parts, zone 1, zone 3 and partially upper part of the zone 2 are formed by permeable spoil dump and the zone 2 is less permeable and wet part where the drainage water from upper parts is retained.
- 4) This knowledge can be useful in application of other remediation processes into the spoil dump and thus to contribute to the reduction of acidification of the surroundings.

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