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REMOVING CONTAMINANTS FROM GROUNDWATER POLLUTED BY THE TRZEBIONKA MINE SETTLING POND LOCATED IN UPPER SILESIA (POLAND)

Tomasz SUPONIK

Politechnika Śląska, Wydział Górnictwa i Geologii, Akademicka 2, 44-100 Gliwice, tomasz.suponik@polsl.pl

Abstract. The Trzebionka Mine settling pond is located in south Poland and has a significant impact on quality of surrounding soil and groundwater. This is mainly due to toxic elements present in wastes disposed in the pond. The wastes are mainly flotation tailings after processing of zinc and lead ores. In the paper, chemical composition of the wastes is presented. Problems with surface water contamination by the Trzebionka Mine settling pond are also presented in the article. It was shown that most problems result from contaminants flowing down in the unsaturated zone, reaching ground waters and then flowing horizontally leading to pollution of the surface water. As a result, the canal surrounding the post-flotation pond as well as Luszowka and Wodna streams and finally the Chechlo river are contaminated. In order to protect these waters a permeable reactive barrier (PRB) technology is proposed to be used in the vicinity of the considered dumping site. The PRB technology is a technique of groundwater remediation in which the contaminants are removed from an aquifer by flowing through a PRB filled with a special material called reactive material. The processes which can be used to treat groundwater contaminated by the Trzebionka Mine settling pond are redox reactions, adsorption and biochemical reactions. At the end of the article a proper selection of reactive materials, and thus treating processes, for efficient PRB application in the considered dumping site area is suggested.

keywords: flotation waste of Pb-Zn production, metals and non-metals in groundwater and in surface water, remediation, PRB Technology

1. Introduction

The Silesia and Cracow province has been developing for hundreds of years. There are mining, power and metallurgical industries in this region. As a result of heavy industry expansion, many dumping sites exist in the area. Some of them are old and forgotten, some are still used, and new are being created. The dumping sites affect the quality of ground and surface water in this region (Cabala, 2005, 2009; Girczys and Sobik-Szołtysek, 2002; Labus, 1999).

The flotation wastes from Trzebionka Mining Company plant processing Zn and Pb ores had been located in the Trzebionka Mine settling pond for over half a century. Rainfall water either flows through the dumping site or forms a surface run-off, and consequently contains different contaminants. Water flows down in the unsaturated zone, reaches the groundwater and then flows horizontally polluting surface water. The leachate from the dumping site may contain different kinds of chemicals like inorganic anions, oxyanions and cations. To remove them from the groundwater, and thus to protect surface water, PRB (Permeable Reactive Barrier) technology may be used. This technology is a technique of groundwater remediation where contaminants are removed from an aquifer by the groundwater flow through a PRB filled with a special material called reactive material. The PRB technique of groundwater remediation is a passive one. Contaminants treatment can be accomplished may occur with the reactive material through physicochemical, chemical and/or biochemical processes. The PRB technology has the potential to effectively remediate subsurface contamination at many types of sites with significant cost savings compared to other approaches. The economic benefits of the PRB technique results from the fact that relatively little energy or labour input is necessary.

The primary goals of this study were to present the impact of the Trzebionka Mine settling pond on groundwater and then on surface water, as well as to present the processes and reactive materials that can be used in the PRB technology for treating groundwater contaminated with metals and non-metals by the Trzebionka Mine settling pond.

2. The impact of settling pond the Trzebionka Mine on water environment

The Trzebionka Mine settling pond (Fig. 1) is located between Chrzanow and Trzebinia towns in the southern part of the Silesia-Cracow Upland. It belongs to Trzebionka Mining Company (presently under liquidation) and occupies ca. 0,65 km² (Nowak, 2008). In the dumping site the flotation waste of zinc and lead ores had been stored in the form of suspension containing dolomite, quartz mixed with zinc sulphides (sphalerite ZnS) and lead carbonates (cerussite PbCO₃) (Jarosiński et al., 2006; Nowak, 2008). The chemical composition of the Zn-Pb waste is presented in Table 1.

The contaminants present in the Zn-Pb waste pollute nearby Luszowka and Wodna streams. Both streams discharge their waters into the Chechlo river (Fig. 1). To confirm that the ground and surface water are polluted by the dumping site leaching tests for wastes taken from the considered dumping site were made. The experiments were performed according to EU Standard (2006). The liquid/solid ratio (L/S) in the leaching tests was 10.

Eight samples of wastes (ca. 1 kg each) were taken from the dumping site. Each sample was made homogeneous before the test. The locations of samples collection are shown in Fig. 1. The depth of sampling points amounted to ca. 0.5 m below ground surface. The wastes were sampled with the use of a paddle.

The concentrations of following elements As, Cd, Ni, Pb, Zn in wastes (for each sample) were measured with the help of Mass Spectrometry (ICP-AES). The results are presented in Table 2.

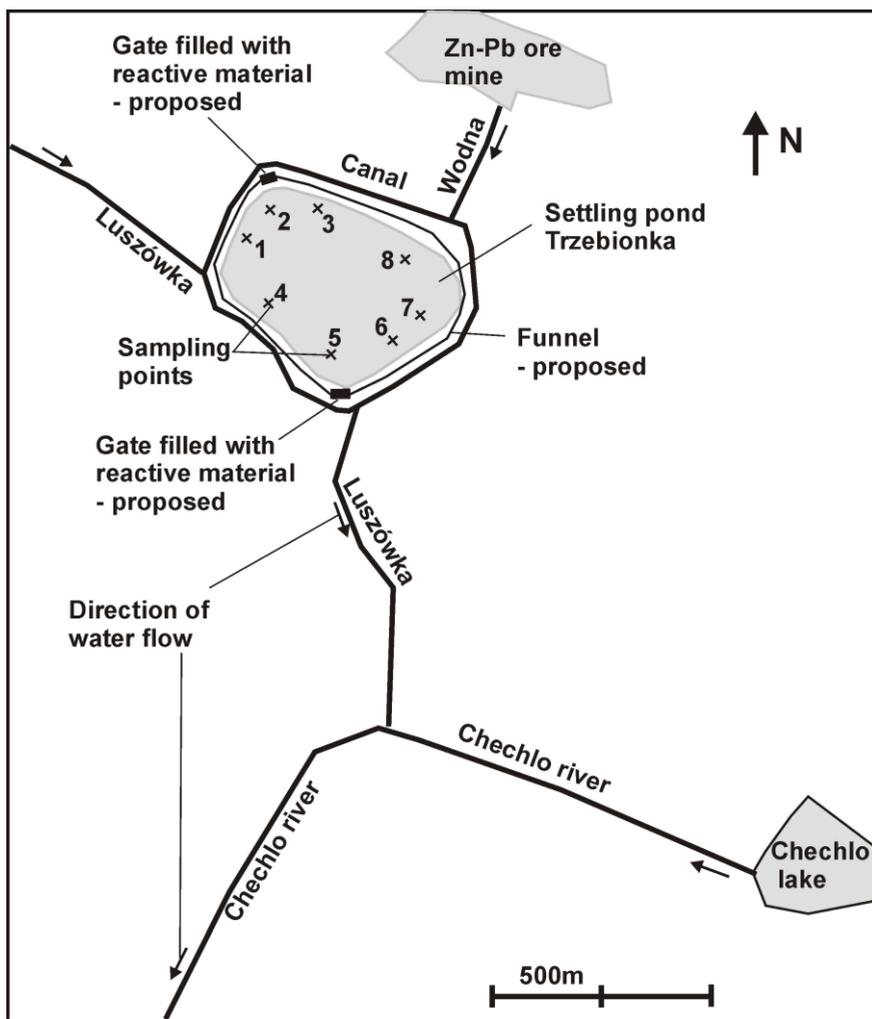


Fig. 1. Surface water in the region of the Trzebieńka Mine settling pond plotted using own data and Mapa Turystyczna (2011)

The arithmetic average and maximum concentration of chosen elements leached from wastes amounted to: $As_{av} = 94 \text{ mg/kg}$; $As_{max} = 155 \text{ mg/kg}$; $Cd_{av} = 48 \text{ mg/kg}$; $Cd_{max} = 81 \text{ mg/kg}$; $Ni_{av} = 42 \text{ mg/kg}$; $Ni_{max} = 68 \text{ mg/kg}$; $Pb_{av} = 129 \text{ mg/kg}$; $Pb_{max} = 192 \text{ mg/kg}$; $Zn_{av} = 936 \text{ mg/kg}$; $Zn_{max} = 1232 \text{ mg/kg}$, whereas $pH = 7.31$ and $EC = 4.820 \text{ mS/cm}$. The results show that there is high probability that investigated elements can be leached from the Trzebieńka Mine settling pond under prevalent pH conditions.

Table 1. Chemical composition of flotation waste from Trzebionka Mining Company (Jarosiński et al., 2006)

Component	mean value [%]	Component	mean value [%]
CaO	30.08	S	0.76
MgO	17.50	Sn	0.042
CO ₂	39.2	As	0.039
Al ₂ O ₃	2.10	Cu	0.021
SiO ₂	2.07	Mn	0.02
Fe	2.03	Cl	0.015
Zn	0.89	Cd	0.009
Pb	0.40	Sb	0.0014
ZnO	0.49	Ag	0.0003
PbO	0.33		

Table 2. Concentration of As, Cd, Ni, Pb, Zn leached from wastes taken from the investigated dumping site (Fig. 1)

Element	Concentration of elements leached from wastes taken from the indicated sampling point							
	[in mg/kg]							
	1	2	3	4	5	6	7	8
As	95	118	155	45	88	71	81	99
Cd	37	81	70	41	15	24	73	44
Ni	17	61	59	44	55	68	22	10
Pb	88	118	104	75	192	126	155	174
Zn	472	988	1174	658	1064	893	1007	1232

High concentration of metals and non-metals in surface water (Table 3) located nearby the settling pond confirms also the surface water pollution by the dumping site. Water in Luszowka stream and in the draining canal area of the post-flotation settling pond is slightly alkaline (pH 8) (Pasiczna et al., 2008). Electric conductivity (EC) in these waters amounted to 0.79 mS/cm and 3.96 mS/cm (median values), respectively (Pasiczna et al., 2008). Since an electric conductivity value above 2 mS/cm indicates substandard water, such a high value of EC proves strong contamination of water in the canal.

The measurements, which results are presented in Table 3, were made in the years of 2003-2005, that is before liquidation of Trzebionka Mining Company. During this time the settling pond was not the only source of surface water contamination. Some wastewater (slightly contaminated with Zn, Pb, Cd) from the ore mine and from ore concentration plant was poured in the Wodna stream (Fig. 1) and in the Trzebionka Mine settling pond (with flotation waste) (Jarosiński et al., 2006; Pasiczna et al.,

2008). Nevertheless, it can be said that the presented in Fig. 1 dumping site was one of the main sources of groundwater and because of that surface water contamination.

Due to toxicity of chemical components of the waste (Table 1) and their high concentration in the surface water (Table 3) and in eluates (Table 2), arsenic, cadmium, nickel, lead, thallium, uranium, and zinc create hazards for the aqueous environment.

Table 3. Concentration of contaminants (metals and non-metals and other selected compounds) in surface water contaminated by the Trzebieonka Mine settling pond (flotation waste of Zn-Pb ores) considered by Pasieczna et al. (2008) as a presumable source of contamination (on the bases of the Chrzanów map sheet by Pasieczna et al. (2008))

Contaminant	Concentration of contaminants in:	
	Luszwonka stream	canal surrounding post-flotation pond
As	max 10 µg/dm ³ ; median 2 µg/dm ³	max 6 µg/dm ³ ; median 3 µg/dm ³
Ba	max 83.41 µg/dm ³ ; median 58,17 µg/dm ³	max 10 µg/dm ³ ; median 2 µg/dm ³
B	max 627µg/dm ³ ; median 200 µg/dm ³	max 131 µg/dm ³ ; median 107 µg/dm ³
Cd	max 10.5 µg/dm ³ ; median <0.2 µg/dm ³	max 3.8 µg/dm ³ ; median 1.0 µg/dm ³
Pb	max 21.0 µg/dm ³ ; median 1.1 µg/dm ³	max 31.9 µg/dm ³ ; median 9.4 µg/dm ³
Li	max 10 µg/dm ³ ; median 7 µg/dm ³	max 26 µg/dm ³ ; median 9 µg/dm ³
Mo	max 3.86 µg/dm ³ ; median 2.21 µg/dm ³	max 8.41 µg/dm ³ ; median 4.68 µg/dm ³
Ni	max 7 µg/dm ³ ; median 1 µg/dm ³	max 48 µg/dm ³ ; median 6 µg/dm ³
Rb	max 12.4 µg/dm ³ ; median 5.0 µg/dm ³	max 16 µg/dm ³ ; median 7.6 µg/dm ³
Sr	max 970.5 µg/dm ³ ; median 424.5 µg/dm ³	max 1018.4 µg/dm ³ ; median 854.1 µg/dm ³
Tl	max 0.75 µg/dm ³ ; median 0.09 µg/dm ³	max 10.79 µg/dm ³ ; median 0.74 µg/dm ³
U	max 1.03 µg/dm ³ ; median 0.60 µg/dm ³	max 1.25 µg/dm ³ ; median 0.82 µg/dm ³
SO ₄ ²⁻	max 1347 mg/dm ³ ; median 181 mg/dm ³	max 4593 mg/dm ³ ; median 3181 mg/dm ³
Zn	max 1491 µg/dm ³ ; median 140 µg/dm ³	max 6602 µg/dm ³ ; median 1681 µg/dm ³

3. Selection of processes and reactive materials for treating groundwater contaminated by the Trzebieonka Mine settling pond

According to future plans (Gazeta Krakowska, 2011) 70-m-high the Trzebieonka Mine settling pond will be a place for viewing and admiring the landscape. Although the wastewater from the ore concentration plant and the mine will not be directed (after liquidation) to the settling pond and to surface water any longer, the problem connected with leaching elements from the waste to groundwater and to surface water will remain. Therefore, in order to protect these waters, the PRB technology can be used in the vicinity of the dumping sites.

The PRB technology is a technique of groundwater remediation. With the use of it, many toxic contaminants can be removed from groundwater. This technique is a passive one, in which contaminants are removed from an aquifer by flowing through a reactive barrier filled with a reactive material (Gavaskar et al., 2000; ITRC, 2005;

Meggyes et al., 1998; Puls et al., 1998; Suponik, 2010). The processes applied to treat groundwater contaminated with metals and non-metals are (Suponik, 2008) include: redox reactions which lead to precipitation of metals, pH control (precipitation), adsorption, and biochemical reactions which lead to precipitation of metals by sulphate-reducing bacteria.

Table 4. Reactive materials used in laboratory or field test for different kind of inorganic contaminants (Gavaskar et al., 2000; ITRC, 2005; Koziol-Komosinska and Kukulka, 2008; Koziol, 2002; Puls et al., 1998; Suponik and Lutynski, 2009; U.S. Department of Energy, 1998; Wilkin and Puls, 2003)

inorganic contaminants			reactive material	type of process
metal	non-metal	other compounds		
Al, Sb, Ba, Cd, Cr ⁶⁺ , Co, Cu, Pb, Mn, Mo, Ni, Ag, ⁹⁰ Sr, Tl, Tc, U	As, Se	NO ₃ ⁻ , SO ₄ ²⁻	zero-valent metals (iron)	precipitation barriers - redox reactions
Cr, Mo, Tc, U	As		ferrous hydroxide, ferrous carbonate, ferrous sulphide	
Cd, Mo, U	As, Se	SO ₄ ²⁻	limestone	precipitation barriers - pH control
	As, Se		activated alumina	
Sb, Bi, Cs, Cr, Co, Hg, Mo, Ag, Tc, tin Sn, U, Zr	As		activated carbon	adsorption barriers
Tc, U			exchange resins	
Cr, Pb, Mo, U, Cu, Cd, Zn, Ni	As	SO ₄ ²⁻	peat, lignite, brown coal	
Al, Ba, Cd, Co, Cs, Cr, Cu, Pb, Mn, Hg, Ni, ⁹⁰ Sr, U, Zn	As, Se		zeolite	
Cd, Co, Cu, Pb, Mn, Hg, Ni, Sn, Zn		SO ₄ ²⁻	coastal hay, wood chips, saw dust, livestock manure, sludge, compost	precipitation barriers – biochemical reactions

The crucial question in the case of contaminated groundwater is whether it is possible to use the PRB technology in an effective way. In the paper by Suponik and Lutynski (2009) factors that limit PRB application were discussed. One of the most important factor during reactive material (and thus process) selection is the type of contaminants. A compilation of laboratory and field research into chemicals treated

with reactive material is provided in Table 4 (Gavaskar et al., 2000; ITRC, 2005; Kyziol-Komosinska and Kukułka, 2008; Kyziol, 2002; Puls et al., 1998; Suponik and Lutynski, 2009; U.S. Department of Energy, 1998; Wilkin and Puls, 2003). It is classified into the type of inorganic contaminant, reactive material type, and finally in the type of the principal process which allows to remove contaminants from groundwater.

On the basis of Table 4 it can be claimed, that the possible processes and reactive materials for use with PRB to remove As, Cd, Ni, Pb, Tl, U, Zn from groundwater are:

1. redox reactions with the use of zero-valent iron – there is no evidence that this barrier removes Zn,
2. biochemical reactions with the use of biomaterial presented in Table 4 – there is no evidence that this barrier removes As, Tl, U,
3. adsorption with the use of zeolite – there is no evidence that this barrier removes Tl.

These processes are shortly discussed below.

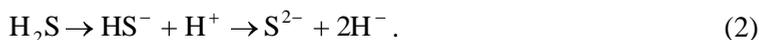
Zero-valent iron Fe(0) is an efficient, easy to use and cheap reactive material for removing metals and non-metals by redox reactions. It is applied when groundwater contains positively charged inorganic cations such as Cd²⁺, Co²⁺, Cu²⁺, Ni²⁺, Pb²⁺, and negatively charged anions and oxyanions containing As(III), As(V), Cr(VI), Se(VI), Tc(VII).

Reactive barriers filled with Fe(0) have several advantages over others ones. Zero-valent iron can work effectively and long (without replacement the material). So, the maintenance costs are rather low. Moreover, scrap iron is relatively cheap and can be obtained in a granular form in large quantities.

Heavy metals can be treated through biochemical reactions in the PRB (ITRC, 2005). In accordance with many studies (e.g. Canova, 2006) the primary removal mechanisms for the metals are sulphate-reducing bacteria (SRB). Bacteria obtain their energy by oxidizing simple organic compounds or molecular hydrogen H₂ while reducing sulphates SO₄²⁻ to sulphides, especially to hydrogen sulphide H₂S (Schulze and Mooney, 1994). An example of a biomaterial that encourages growth of sulphate-reducing bacteria in PRB are coastal hay, wood chips, saw dust, livestock manure (Canova, 2006), sludge and compost. The SRB obligates anaerobes, which prefer the following conditions: pH between 5 and 8; Eh ca. -200mV. A typical overall conversion equation (neglecting the small amount of organic material required to produce biomass) is:



The reduction product of reaction (1), hydrogen sulphide, is a volatile gas. The form in which sulphide occurs depends on the pH. HS⁻ and S²⁻, which occur at neutral and high pH respectively, are both water soluble. H₂S is the predominant form at low pH <6 (Cohen, 2006) because:



The sulphides react with metal ions to precipitate them as metal sulphides, many of which are stable under anaerobic conditions of the treatment system (Cohen, 2006). The heavy metal ions react with dissolved sulphide according to the following reaction:



Bio-barriers are considered a unique type of PRB. Due to delivering amendments into the subsurface (e.g. compost, correction of pH) this PRB works less passively than filled with Fe(0) and may incur greater operation and maintenance costs.

Adsorption is a process that occurs when a liquid solute accumulates on the surface of a solid adsorbent, forming a molecular or atomic film (the adsorbate). The manner and strength of fixation is of great importance in relation to the possibility of remobilisation, and is strongly influenced by parameters such as concentration, solubility, and speciation of the contaminants and co-solvents, and the prevalent pH, oxidation-reduction potential and temperature conditions (Roehl et al., 2000). Sorption material must meet the following conditions: high sorption capacity, high selectivity for the target contaminants, fast reaction kinetics, high hydraulic permeability, long-term effectiveness. It should be non-harmful to the environment, available at reasonable costs (Kowal and Swiderska-Broz, 1996; Roehl et al., 2000), insoluble, not biodegradable, and easy to apply. Unfortunately, sorption material should be replaced and regenerated frequently due to the effects of potential desorption or reversed ion-exchange. This aspect makes adsorption barrier rather expensive and not attractive comparing to other types.

Since the bio-barrier works less passively than reactive barrier filled with Fe(0) and adsorption material (zeolite) needs to be replaced and regenerated frequently, the zero-valent iron seems to be the best reactive material for treating groundwater contaminated with As, Cd, Ni, Pb, Tl, U. It is effective, relatively cheap and works long.

Unfortunately, zinc is the only toxic element (among contaminants presented in Table 3) that is not treatable by Fe(0). With regard to this element, it can be removed from groundwater by adsorption on zeolite, peat or lignite (Table 4), or by biochemical reactions with the use of the SRB (Table 4). Both processes are able to remove also organic and inorganic chemicals from groundwater.

In general, granulated zero-valent iron might be used in the first segment of the reactive barrier, whereas adsorbent or bio-material might be used as a second segment. Since most chemicals can be removed with Fe(0), the material applied in the second segment would not be used completely and it could treat residual contaminants.

To take a decision whether the PRB can be accepted or not in the initial stage of the assessment of possibilities of using the PRB technology, various additional data are needed to be either obtained or measured (Suponik and Lutyński, 2009). They are:

hydrogeological, geological and site, and contaminated groundwater characteristics. Furthermore, in order to assess the effectiveness of suggested processes, laboratory tests with the use of contaminated groundwater and chosen reactive materials need to be performed.

The PRB is currently built in two basic configurations: Continuous Reactive Barrier and Funnel-and-Gate System (Puls et al., 1998; Roehl et al., 2005) divided into: Funnel-and-Gate Open System and Funnel-and-Gate Closed System. The Funnel-and-Gate Closed System was proposed for the area of The Trzebionka Mine settling pond, because it uses impermeable walls (funnel) to closed contaminated region (Fig. 1). The funnel directs the contaminant plume to a “gate(s)” containing the reactive material. Thanks to this configuration it uses less reactive material. In this case the gate(s) should be located in a place which is characterized by the highest discharge of the water flux and the highest concentrations of contaminants. Therefore, to properly locate gate(s) in the vicinity of dumping sites the flow direction of contaminated groundwater should be assessed (location of gates presented in Fig. 1 is only a proposal). The configuration of the PRB proposed in Fig. 1 consists of two gates, each ca. 25 m long and approximately 1.8 km of impermeable walls.

Estimating the cost of the PRB is a difficult task in this stage of consideration, since a number of factors need to be evaluated, including:

- site characterization costs: complete vertical and horizontal delineation of the groundwater plume and characterization of hydrogeologic, geochemical, geotechnical, and microbiological conditions,
- design costs: treatability studies and modelling,
- construction costs: purchase and installation of reactive material and impermeable wall,
- operation and maintenance costs: monitoring and reporting costs and replacement of reactive material.

The total costs of the PRBs vary widely depending on the site type and PRB characteristics. In general, the depth and length of the PRB continue to drive the costs of PRB application (Gavaskar et al., 2000). For example (in the USA the costs is different from that in Poland) the cost was:

- from \$100 000 in Mountain View, California. Fe(0) was used in Continuous Reactive Barrier. Treatment zone depth ca. 5 m; reactive zone length ca. 14 m, total mass reactant 90 Mg (Puls et al., 1998),
- to \$1 000 000 in Lakewood, Colorado. Fe(0) was used in the Funnel-and-Gate Open System. Funnel material: Sealable Joint Sheet Piles, funnel length 320 m, No. of gates 4, treatment zone depth ca. 7 m, reactive zone length 4x12 m=48 m, total mass reactant - no information available, total system length 368 m (Puls et al., 1998).

Since the second segment of the reactive barrier consisted of adsorbent or bio-material and length of the proposed (in the area of The Trzebionka Mine settling pond) PRB is high, the total costs may be high. Originally, the cost of a PRB was compared to a pump-and-treat system (alternative method). According to Gavaskar et al. (2000),

ITRC (2005), Puls et al. (1998) this system is generally more expensive than PRB. Therefore, today, pump-and-treat systems are not being used at the same number of sites as in past years. This reduction is partially due to issues involving overall effectiveness and the high cost of operation and maintenance for the pump-and-treat systems. Therefore, it may turned out that the PRB with Fe(0) in the first segment of the reactive barrier and adsorbent or bio-material in the second segment is the cheapest and the most effective method for protecting Luszowka and Wodna streams, and, as a result of it, the Chechlo river.

4. Conclusions

The Trzebionka Mine settling pond has a significant impact on the quality of groundwater, and thereby on the surface water, Luszowka and Wodna streams and Chechlo river. The chemical composition of flotation waste located in it shows high concentration of various metals and non-metals. Leaching tests confirm that they migrate to water. The metals and non-metals flow down (in the form of dissolved in water) in the unsaturated zone, reach the groundwater and then flow horizontally and pollute surface water (with arsenic, cadmium, nickel, lead, thallium, uranium, and zinc). There are three processes in the PRB technology that are able to remove metals and non-metals from groundwater contaminated by the Trzebionka Mine settling pond. They are: redox and biochemical reactions and adsorption. In order to keep the operation and maintenance cost of the PRB relatively low and to achieve high effectiveness of the treatment process the redox reaction with zero-valent iron was chosen and proposed to be used as a first segment of the reactive barrier, and biochemical reactions with SRB or adsorption process with zeolite, peat or lignite were proposed to be used as a second segment - to remove zinc from groundwater and to treat residual contaminants (remained after first segment).

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