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Impact of residual coal extraction from hard coal mining waste dumps on groundwater salinity

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ABSTRACT

Re-mining of existing dumping sites for the extraction of coal residues (around 10% wt.) with subsequent reuse of a secondary waste in engineering constructions or its re-disposal in the environmentally safe way is an attractive and economically viable way of coal protection as a valuable natural resource and of adequate reduction of dumped waste volume. These methods of bulk waste re-use may though cause also negative consequences, if not properly controlled. In this article, the adverse environmental impacts of technological processes of coal extraction are exemplified in a coal mining waste dump in the Upper Silesia coal basin (USCB) in Poland, where coal waste has been reprocessed since 1998. Both the disturbance of original waste layers in the dump and altered exposure of the processed rock material to the atmospheric conditions during residual coal extraction, and secondary wastes re-disposal/re-use appeared to result in significant increase of groundwater salinity. Therefore, adverse effects of waste re-use and efficient and cost-effective methods of attenuation of salt loads release from this material should be taken into account.

Keywords: Coal; Coal mining waste; Coal re-extraction; Disposal and reuse of secondary wastes; Salt leaching; Groundwater salinization

1. Introduction

Coal provides 26% of global primary energy needs and generates 41% of the world's electricity. The increasing need in energy, in particular in fast developing countries of Asia and Pacific Region resulted in 92% increase of total global hard coal production over the past 25 years and was estimated to 5990 Mt in 2009 [1]. Common use of shearers in coal mining causes generation of a waste rock that accounts for 30–50% of the global coal output, which gives estimation for at least 1840 Mt of extractive waste annually [2]. These bulk wastes are a source of a serious threat to the aquatic environment lasting for decades due to the occurrence and reactivity of geochemically instable sulfides (pyrite, pyrrhotite, marcasite). Physico-chemical weathering transformations that comprise also sulfide oxidation, result in a generation of dissociated sulfuric acid and high sulfate acid leachate (so called ARD — acid rock drainage) with subsequent mobilization of metals — potentially toxic elements (PTE) from waste and subsoil matrix due to acidification. Sulfate and acid generation is described by the overall reaction [3]:

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

These processes, as well as the high chloride salinity of leachates cause a long-term deterioration of ground and surface water resources in the vicinity of dumping sites

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[2,4–7]. Another problem specific for hard coal mining waste is its self-ignition induced by the residues of coal that in older coal mining dumps account for about 10% wt., and by exothermic processes of sulfide oxidation. Such fires cause also serious problems of air pollution due to emission of polycyclic hydrocarbons. The serious environmental consequences of improper management of waste from extractive industries resulted in the recent enactment of the “Mining Waste Directive” (EU Directive 2006/21/EC) [8], which obliges the mining waste facility operators to take the measures necessary to prevent or reduce the generation of waste and its negative impact, to encourage waste recovery through recycling, re-use or reclaiming and to assure safe disposal of waste.

Re-mining of existing dumps for residual coal extraction with a subsequent re-use of a secondary waste in engineering constructions, or alternatively its re-disposal in altered, but environmentally safe conditions has a high potential of the reduction of sulfidic mining wastes volume and of their adverse environmental impact. Remining of coal extractive waste dumps is also a form of post-mining land reclamation in accordance with the principles of advanced land management aimed to transformation of often non-aesthetical and dangerous dumping sites into green or urban areas, and to reduction of the disposed waste volume. It should be though taken into consideration that the re-mining processes cause deep alteration of hydrogeochemical conditions at the dump that may affect the groundwater status.

The major factors that determine the scale of a negative impact of waste on ground and surface waters are: content and kind of soluble compounds, content and reactivity of sulfides, content of buffering constituents and the neutralization potential ratio (buffering capacity), occurrence and content of potentially toxic trace elements (PTE), waste granulation, water balance and conditions of water circulation and air contact/penetration into waste layers [2].

The objective of the presented study is the assessment of the effect of coal residues extraction from hard coal mining waste dumps on the groundwater status. The problem has been exemplified in the Bukow coal mining waste dump, where residual coal extraction is currently at the final stage, thus environmental consequences of the process can be conveniently summarized.

2. Studied object and methods

2.1. Coal mining waste dump and waste characteristics

The Bukow coal mining waste dump of a total area 44.8 ha is located in the Western part of the Upper Silesia coal mining basin (USCB, Poland) in the area of worked out gravel quarry in the Odra River valley (Fig. 1).

Since 1976 up to 2001, discards from coal preparation processes that originated from the coal seams of 600 and 700 group in the carboniferous strata of the Namurian A series mined by the Anna coal mine were disposed at the dump. The disposed discards primarily comprised only coarser waste from heavy liquid separator (59%) and finer discards from jigs (41%). Since 1995 besides discards, small amounts of flotation slurry < 1 mm (4.7%) and other fine waste (1.8%, mostly fly ash and slag from the mine power plant) were disposed off at the dump. The disposed wastes are rather stable with respect to particle size, petrologic and mineralogical composition. Petrologically, they consist mostly of hard mudstones (> 70% wt.), in about 18% of laminated shale with admixture of coal, and of minor amounts of siltstone (up to 9% wt.) and fine-grained sandstone. The major mineral components of wastes are non-swelling clay minerals, predominantly kaolinite, accessory illite and sericite, as trace components chlorite, feldspar and interlayers of illite/chlorite In minor amounts in these wastes occurs quartz (in sandstone, up to 20% wt. in mudstones, and up to 30% wt. in siltstone). Small, but important components of wastes are sulfides (pyrite) responsible for acid potential (AP) and carbonate minerals (dolomite, siderite calcite and ankerite); (Ca + Mg) carbonates constitute neutralization potential (NP) of wastes. Coal and inferior coal occur in amounts up to 10% wt. that makes this material attractive for coal re-extraction. Concerning grain size distribution, a coarse fraction prevails (93.7% wt., in this fraction > 50 mm comprises 43.8% wt.). Wastes are of low water absorbalility (0.78–0.99% wt.), are resistant to weathering decomposition to clay fractions (≤1.22%), show high hydraulic conductivity ($k = 10^{-3}$ cm/s at 25 kPa and 10^{-4} cm/s at 200 kPa) and are permeable to air. Therefore, at this dump, free infiltration of precipitation water, and easy access of air to the internal parts of the dump occurs [9].

Average sulfur content accounted for S_t = 0.82% wt., while the content of sulfide-S responsible for acid generation was as high as 0.68% wt. Prevailing types of sulfide are syngenetic pyrite occurring as weins and coatings, less frequently epigenetic pyrite occupying joints in coal (both types are considered less reactive). A typical paste pH value of a freshly generated waste is alkaline (pH 8.4–8.7)

The material has a relatively high neutralization potential ratio (NPR) = neutralization potential (NP)/ acid potential (AP) = 2.37, which means that the buffering capacity (neutralization potential — NP) of this waste over 2-fold exceeds acid potential (AP) determined by sulfide-S content, thus, based on the British Columbia guidelines the waste is not potentially acid generating and therefore is considered “relatively safe” in this respect [8].

The residual coal extraction started in 1998 at the over 20 year old dump and has lasted for 11 years, gradually covering all the dump area, with a planned closure in 2010. Currently, the extraction operations have been completed at almost all the dump area.

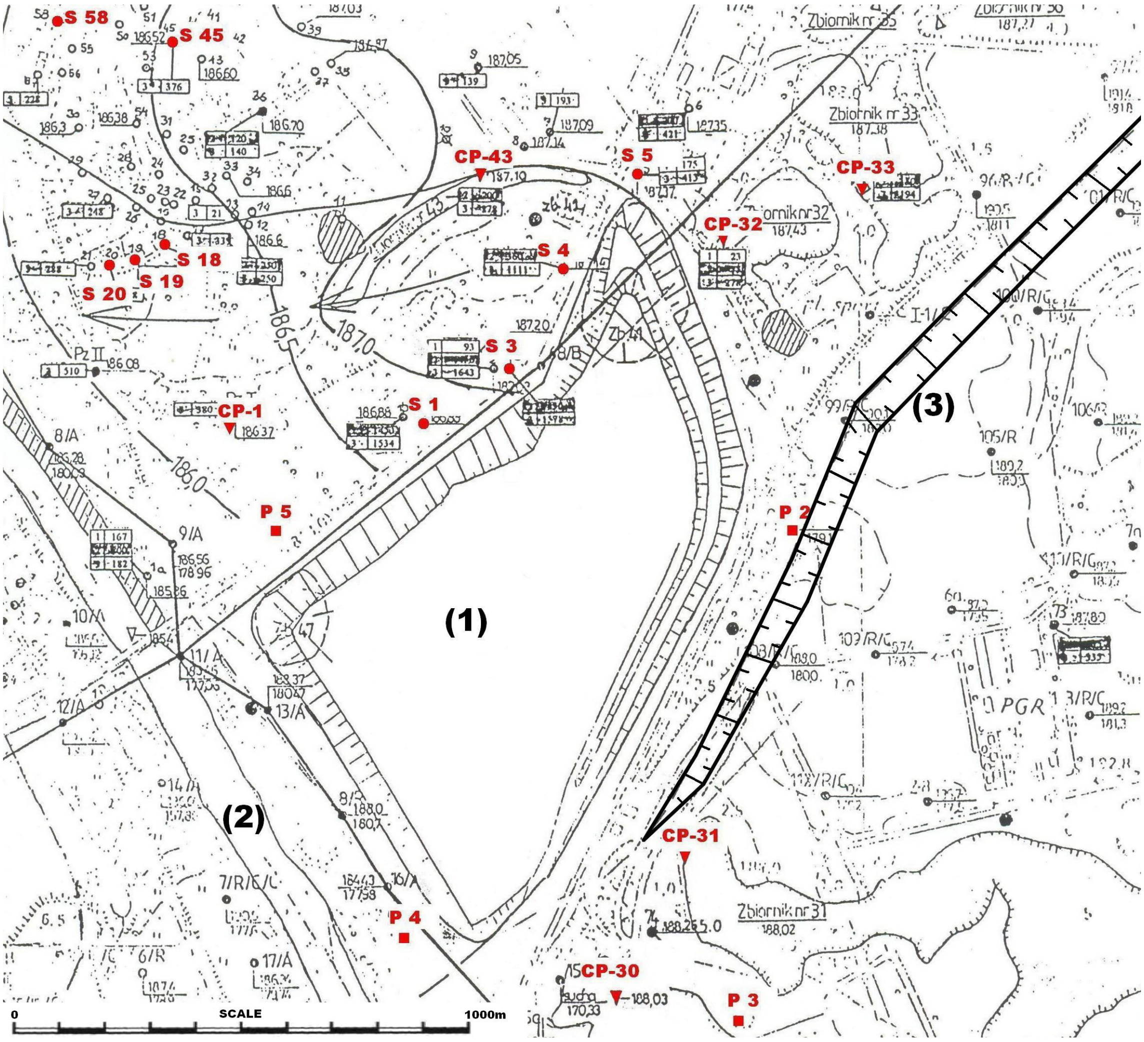


Fig. 1. Location of the coal mining waste dump in Bukow. (1) – coal mining waste dump in Bukow; (2) – the Odra River; (3) – embankments of the Bukow polder; (P5, S1) – sampling points, (CP) – water reservoirs, (→) groundwater flow direction.

2.2. Technology of residual coal extraction

Material is re-mined from old dumping sites, when coal content is estimated to be about 10% wt. The technology of the residual coal re-mining from a dump comprise waste extraction, replacement, transportation, multiple sieving at the shaker screens of different mesh (preliminary screen, 200/40 mm and 25/1 mm screens), washing, elutriation in a settling cone, enrichment in dense media(magnetite) suspension and final re-disposal/reuse of the rejected secondary waste material (Fig. 2). Water in this process is used for transport of the material, its washing and enrichment.

Water circulating in a closed cycle is clarified in a radial thickener with flocculent addition and directed back to the cycle. There are no restrictions with respect to water quality. The mass ratio of water to processed waste is 1:1 Daily water losses that are being replenished from a water reservoir located up-gradient of the re-mined dump (Fig. 1, point CP-31), account for 10% of total water volume circulating in the cycle, i.e. complete replacement of water in the cycle occurs every 10 days.

In the presented process of coal extraction, a number of factors may significantly affect generation and leaching of contaminants. The primary waste layers in a dump become disturbed, waste material is being replaced, admixed with material originating from different layers, sorted, crushed on shaker screens, and extensively exposed to water and air. Furthermore, as a result of coal extraction as a light fraction in the flotation process, and retention of pyrite as a heavy fraction, the secondary waste material rejected from the extraction process

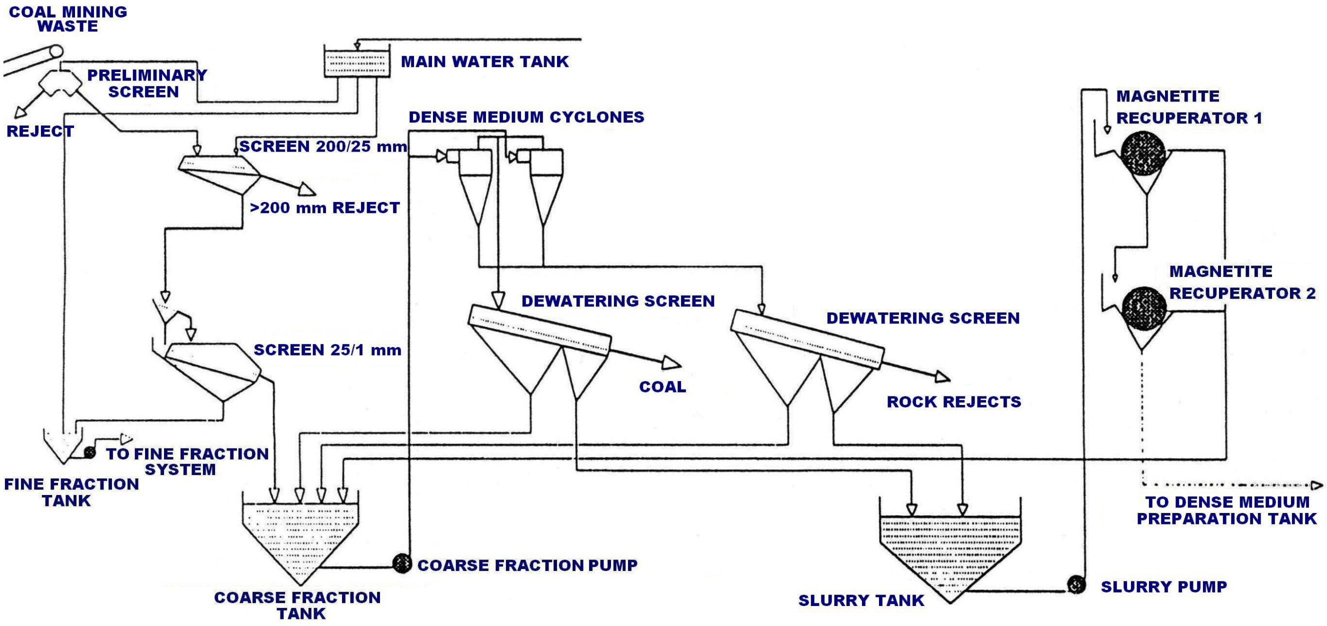


Fig. 2. Flowsheet of the residual coal extraction.

becomes enriched in pyrite at about 10% in comparison to the input coal mining waste. In general, re-disposed waste presents higher water content, higher specific surface and higher pyrite content than the primary waste material disposed at the dump. As a consequence, significant alterations of generation and migration of contaminants in the layers of re-disposed or re-used wastes might be expected.

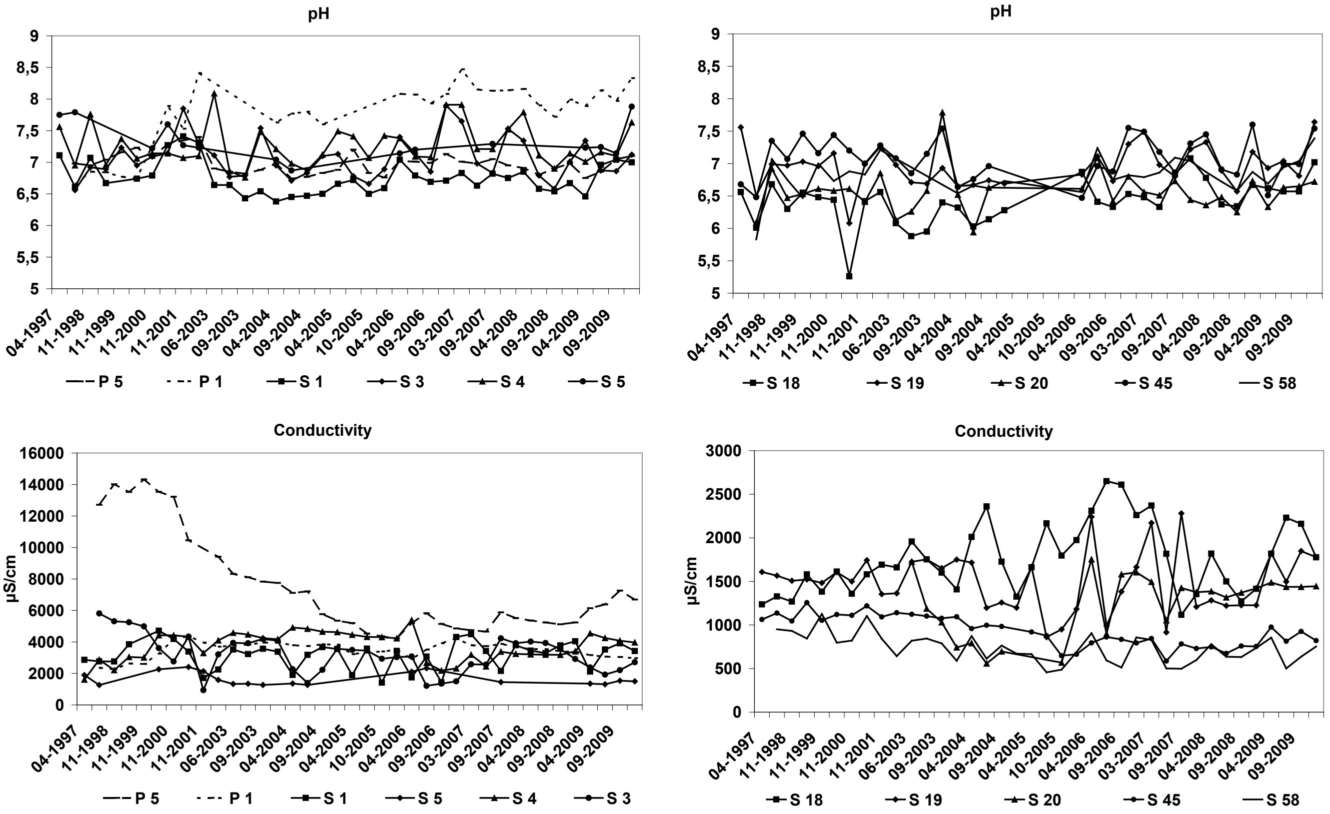
2.3. *Assessment of groundwater status*

Groundwater status in the area has been evaluated on the basis of water monitoring conducted since 1997, i.e. a year before the beginning of the residual coal extraction, within the Local Network for Monitoring Ground and Surface Water Status, designed to assess a potential impact of coal extraction from the Bukow dump on the unprotected sandy-gravel Quaternary aquifer of the riverine origin. The network comprises 14 monitoring wells, of these 3 are located up-gradient of the dump and 5 in the area beyond the impact of the infiltration waters from the dump for monitoring the baseline groundwater quality. Other 6 wells are located down-gradient of the dump in the direction of groundwater flow to the Odra River bed (Fig. 1).

Due to the prevalence of a flat dump surface over the slope area, vertical infiltration of atmospheric precipitation comprises the major part in the natural water balance of the dump.

3. Results and discussion

According to the groundwater flow directions, pollutant loads leached from the dump are flowing in two major streams under the dump in the NE and NW (Bukow community area) and in W-SW directions to the Odra River bed. In 1998, i.e. after 20 years of waste disposal, the leachate from the different parts of the active undisturbed dump contained a large range of dissolved solids. In the groundwater along the W border of the dump, the highest observed conductivity was over 14,000 μS/cm (for groundwater of good chemical status maximum contaminant level (MCL) = 2500 μS/cm) (Fig. 3a), sulfate concentrations reached 6790 mg/L (for groundwater of good chemical status MCL = 250 mg/L), and chloride concentrations over 2000 mg/L (MCL = 250mg/L) (Fig. 4a). At a distance of 450–500 m from the NW and N border conductivity (≤ 1600 μS/cm) (Fig. 3b) and chloride content (≤130 mg/L Cl^-) (Fig. 4b), although elevated, they were lower than the respective reference values, while the values of sulfate concentration in groundwater, despite dilution, were still high (about 500 mg/L SO_4^{2-}) (Fig. 4b), and pH values were between 6.5–8.0 within the range of values posed by the legislation (MCL = pH 6.5–9.5). Concerning metals, permanently high concentrations of Fe and Mn were observed, which is typical for sulfidic waste. The highest salinity and high Cl content of groundwater stream at NW border of the dump (monitored by the P-5 well) was related to the most recent waste disposal directed from the mine before the dump closure. In the following years, chlorides and sulfates occurring in the



(a) (b)

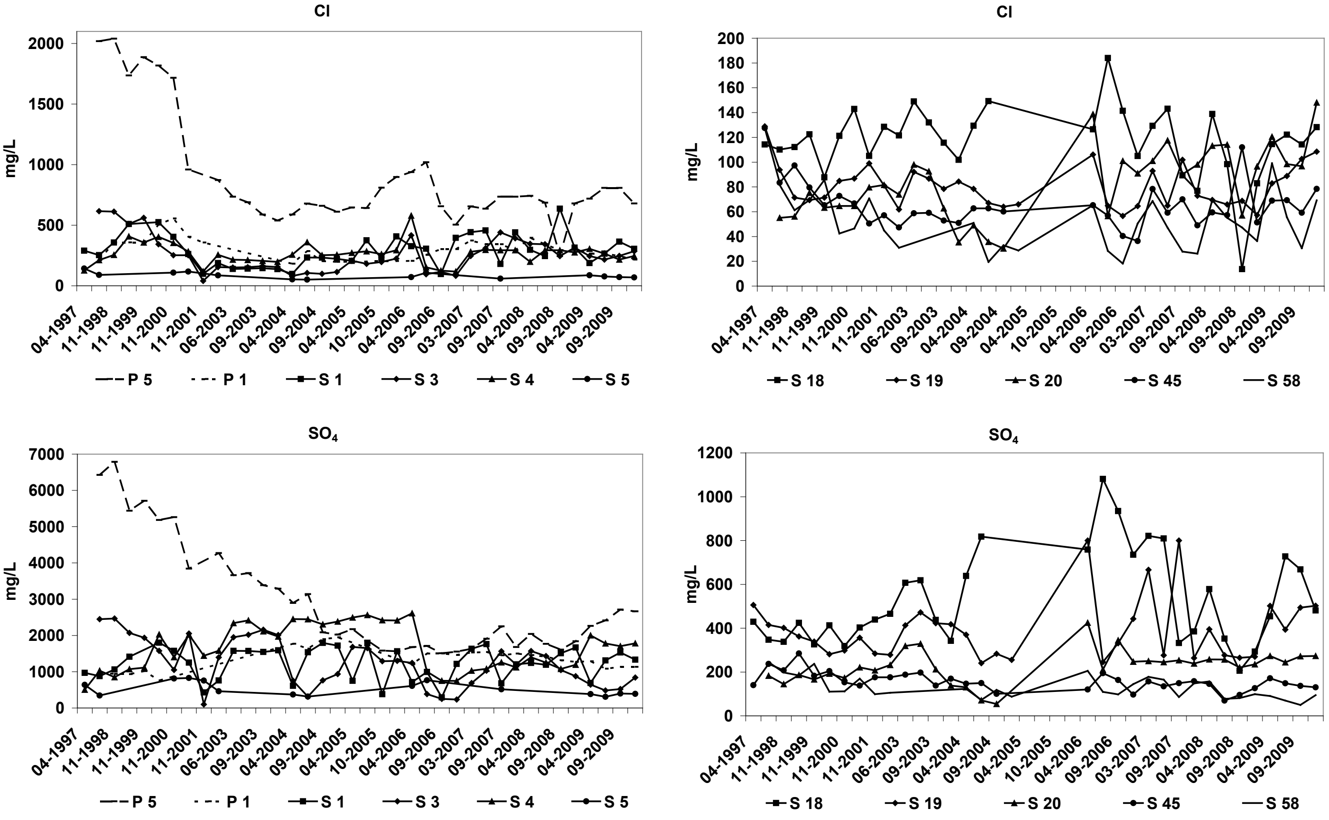
Fig. 3. Temporal alteration of pH and conductivity concentrations: (a) in groundwater along N and NW border of the dump, in the direction of outflow from the dump to the Odra River; (b) in groundwater at a distance of 450-500 m from the dump border in the direction NW and W of the outflow from the dump to the Odra River (S-18, S-19 and S-20) and beyond the dump influence in N direction S-45, S-58).

freshly generated materal, was washed out and the salinity in the receiving groundwater stream was stabilized at the still high level determined by the kinetics of sulfate generation resulted from pyrite oxidation in the waste material at the dump.

Extensive exposure of crushed material to air and water in the technological process of coal separation and subsequent dewatering of re-disposed wastes caused intensification of sulfide oxidation and considerable growth of infiltrating water flow rate. This resulted in increase of generation and leaching of dissolved solid loads discharged from the dump to the groundwater.

Water discharge from the layer of re-disposed waste was assessed at about 700 m^3/d with use of the tool WHI UnSat Suite Plus v.2.2.0.3 comprising computer programs HELP Model and Weather Generator v.1.0. Input data included (a) increase of water content in wastes due to the technological process of coal separation; (b) daily amounts of re-processed and re-disposed waste; (c) actual weather data for the site. Despite the substantial growth of leached dissolved solids load, expansion of water stream and increase of its flow rate resulted in a tendency to a certain equalization of concentrations of leached constituents (expressed by conductivity values) in the outflow along the NE-NW dump border (Figs. 3a, 4a). Simultaneously, an extension of a distance of contaminants (mostly sulfate) transport (Figs. 3b, 4b) was observed. In the subsequent years, significant alterations of groundwater quality downstream of the dump in the N-NW and W directions occurred, which was caused by the extension and changes of the re-mining fronts directions, as well as from construction of a new dump of re-disposed wastes within the same site. Observations conducted within the Local Monitoring Network have showed that waste re-mining strongly affects sulfate salinity (Fig. 4) due to waste "activation", i.e. increase of sulfide oxidation rate resulting in increase of sulfates and heavy metals concentration, drop of pH and enhanced leaching of contaminants.

Currently, along the W-NW-NE borders of the dump in the directions of groundwater outflow to the Odra River, the referred above broadening and joining of separate



(a) (b)

Fig. 4. Temporal alteration of Cl and SO_4 ions concentrations in the groundwater in (a) and (b) locations (see Fig. 3).

streams occur, along with equalization and stabilization of sulfate concentrations in the range of 700–2300 mg/L, mostly 1000–2000 mg/L (Fig. 4a). This is due to the homogenization of the material in the dump, which is by now almost completely filled with a re-disposed carboniferous rock. Therefore, concentration range of leached constituents is determined by concentrations of constituents in the percolating water stream that comprises infiltrating atmospheric precipitation and excessive water from the extraction process in re-disposed wastes.

The major constituent leached from the re-mined waste appeared to be elevated loads of sulfates from enhanced pyrite oxidation due to waste "activation" during coal extraction, and associated with this process release of iron and manganese ions in concentrations 20.5–21.9 mg/L Fe (MCL = 0.3 mg/L) and 2.1–4.1 mg/L Mn (MCL = 0.2 mg/L). Concentrations of chlorides in leachate from the re-mined waste, although considerably increased (> 500 mg/L), were marginal due to re-mining mainly old material, from which these mobile ions were already washed out. Chloride release was nevertheless clearly enhanced by the activation process, mainly by better leaching conditions due to particle size reduction.

The groundwater status in the area, apart from the direct effect of coal extraction processes, was affected also by the alteration of hydrogeological, hydrogeochemical and hydrological conditions resulted from the reuse of secondary waste after coal extraction for engineering constructions. This material shows appropriate geotechnical parameters for use as a crushed stone aggregate in embankments and motorway foundations confirmed by a quality certificate. In this case, the material was used as a common fill for the construction of the anti-flood Bukow polder, up-gradient with respect to the groundwater flow to the dump (Fig. 5).

Construction of a polder embankment from the secondary re-mined waste caused intensification of the adverse weathering processes and sharp deterioration of groundwater quality down-gradient of the polder embankment and up-gradient of the re-mined dump, due to the increase of salinity, mostly sulfate, but to the lesser extent also chloride (Fig. 5). Although pH values of leachate and groundwater continuously remained neutral or slightly alkaline and no ARD generation occurred, the poor groundwater status due to its high salinization precluded its use for any purpose, and Bukow community located down-gradient from the dump (Fig. 1) had to change water supply for a remote source.

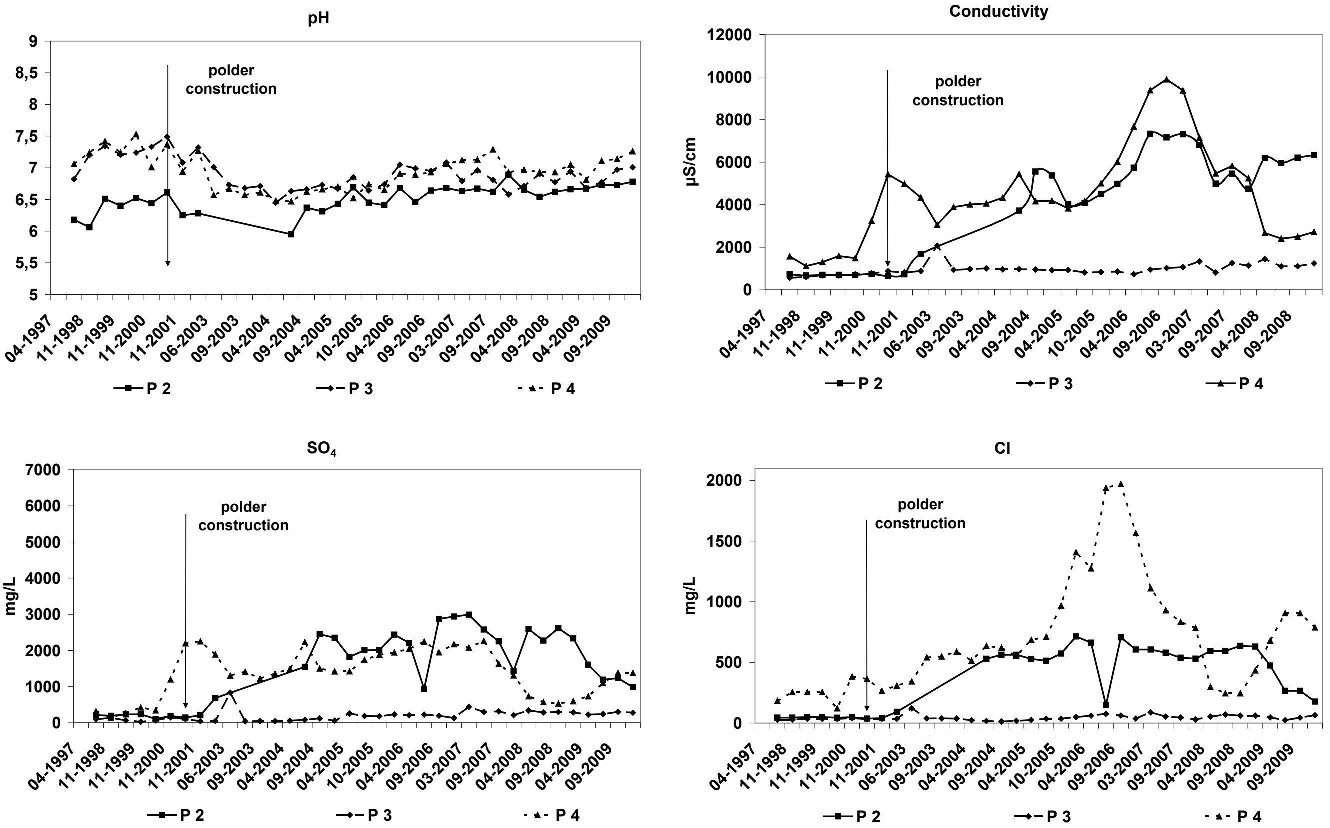


Fig. 5. Temporal alterations of pH, conductivity, SO_4 and Cl ions concentrations in groundwater affected by the leachate from the embankments of the Bukow polder constructed from the re-mined waste rock.

The presented groundwater chemical status in the vicinity of the coal mining dump and polder embankments constructed from the re-mined waste shows that water quality deterioration due to sulfate salinization is the primary negative impact of coal mining waste (and other sulfidic extractive waste) on the aquatic environment, apart from possible ARD generation after depletion of its neutralization potential in specific cases that according to British Columbia Guidelines [10] for NPR range of 2:1–4:1 can occur at "significant preferential exposure of sulfides or extremely reactive sulfides in combination with insufficiently reactive NP".

Extractive Waste Directive 2006/21/EC [8] enforces the waste facility operator to prevent any deterioration of water quality by preventing or minimizing leachate generation and surface water or groundwater and soil from being contaminated by the waste, as well as by collecting and treating contaminated water and leachate from the waste facility to existing effluent discharge limits. Although active methods of leachate treatment are highly efficient and allow to obtain drinking water quality, the high costs of these processes render them viable for use only in arid or highly water deficient zones [e.g. 11–15]. For sulfidic extractive wastes, various alternate solutions have been taken into consideration in order to control sufide [16] or seepage [17], nevertheless passive methods of mitigating long-term generation of highly saline leachate by prevention of pyrite oxidation with use of abundant, cost-effective waste materials seems to be the best approach [18,19].

4. Conclusions

Re-mining of coal extractive waste causes substantial increase of sulfate salinity due to sulfidic waste rock "activation" in the process of coal extraction. Generation and leaching of dissolved solids from these wastes results in an increase of total dissolved solids (TDS) (indicated by the high conductivity), mostly sulfates, as well as of Fe and Mn concentrations in groundwater. These constituents are products of sulfide oxidation and are characteristic components of leachate from sulfidic carboniferous rocks, also from those well buffered and insusceptible to ARD generation. Groundwater deterioration due to salinization caused by sulfide oxidation appears to be high, while its long-term duration is particularly problematic to the environment.

Despite the environmental problems related to the process of re-mining, this way of coal extractive waste reuse should be considered as an attractive alternative of degraded postindustrial lands reclamation due to considerable reduction of disposed waste volume and a possibility of further waste reuse in civil engineering constructions, or re-disposal under environmentally safe conditions. This, though, requires a careful environmental impact assessment, as well as application of precautionary principle and adequate preventive measures in every individual case.

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