Final Outcome of Sulfate Transport Modelling and a Prediction of Ground and Surface Water Quality in Aquifers Influenced by Lignite Mining in Germany

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ABSTRACT

Lignite mining in central Germany has played an important role for more than 100 years. Former and present mining activities, as well as remediation processes, affect the aquifers. The investigation area, south of the city Leipzig, occupies 205 km^2. For this area, the environmental objectives of the European Union (EU)'s Water Framework Directive are currently not achieved. A prediction of ground and surface water qualities has been established focussing on pyritic sulfuroxidation and the generation of sulfate .

To predict the sulfate migration out of overburden dumps into unaffected aquifers, as well as the influence on surface waters, several steps were necessary. The first step involved characterization of the un-disturbed overburden material to predict the geochemical behaviour of the dumps. Therefore, a geological model, generated from several thousand drillings, was formed. Furthermore the concentrations of sulfur and carbonate, within the different geological layers, were examined in the geochemical model. The average dump compositions were based on a blending of the geochemical parameters with the thickness of the different geological layers.

The prediction of dump water quality was carried out using PhreeqC (Parkhust & Apello, 1999) modelling. Predicted amounts of weathering products were used to calculate the equilibrium between groundwater, secondary minerals (gypsum, siderite) and CO_2 concentration of the gas phase by consideration of redox reactions, mineral dissolution, precipitation and cationic exchange. The flow and one component transport modelling for the investigation area was carried out by the existing large scale model (HGMS) of the company IBGW Leipzig and was based on the flow and transport code PCGEOFIM®. Selected "Hotspot zones" were subsequently modelled using a reactive hydro-geochemical transport model, based on the program code PHT3D.

The results describe an expansion of dump water plumes into unaffected aquifers within the stratigraphic sequence from the upper to the lower layers.

Keywords: lignite mining, groundwater quality, pyrite oxidation, hydro-geochemical modelling, prediction

INTRODUCTION

The Central German Mining District has been influenced by lignite mining for more than 100 years. Due to former and present mining activities, aquifers in this area are significantly affected and the environmental objectives of the EU's Water Framework Directive are currently not achieved for the area south of Leipzig. This region is a main part of the Central German Mining District, which occupies a working area of 205 km^2. The University of Freiberg has been commissioned by operators of remediation (LMBV) and active mining (MIBRAG) to predict the future mass fluxes of sulfate in the groundwater bodies and the impact on surface waters. Overburden of tertiary and quaternary age contains distinct proportions of pyrite. During the mining process, the aeration of reduced sulfur compounds in overburden material causes high concentrations of sulfate, iron and trace metals in pore water, which can subsequently be mobilised.

After the mining process has been completed, groundwater levels rise, natural flow systems are re-established and Acid and Metalliferous Drainage (AMD) may occur. In order to predict the potential impact of overburden dump pore waters on unaffected aquifers a geological model, which includes the regular sequences of geological layers, was generated. Concentrations of total sulfur, sulfide-sulfur and carbonate were balanced from existing drilling data in the geochemical model. Simultaneously, the actual groundwater compositions in the investigation area were recorded, to assess the groundwater quality for the mining districts. Migration processes in groundwater were simulated using the existing large scale model (HGMS) of IBGW Leipzig, based on the transport code PCGEOFIM® and sulfate distribution in 2100 main pollution plumes out of overburden dump bodies were assessed. Furthermore, the trend of sulfate concentration was presented on several virtual water gauges. To estimate the overburden dump water impact on surface water bodies, the sulfate mass flow in selected rivers and lakes was calculated. Selected "Hotspot zones" were modelled subsequently, by means of a reactive hydro-geochemical transport model, based on the program code PHT3D.

METHODOLOGY

Determination of the dump groundwater chemistry

Initial concentrations of the main dissolved constituents in overburden dump pore water are a key input into transport modelling and the prediction of future groundwater pollution. Predictions are based on the geological structure of the investigation area (geological model) and the chemical behaviour of the groundwater observed in the field (geochemical model).

Geological model

To verify the distribution of geological layers in the investigation area, petrographic data were collected. Afterwards, several thousand drillings were used to generate a geological model, which describes the deposits and sequences of the overburden layers. Different vertical layers of silt, clay, sand or gravel were detected. According to their chemical properties, these layers are combined to stratigraphic units. Table 1 shows the stratigraphic units with high acidity potential or buffering potential.

Table 1 chart of geological layers in the investigation area

Unit-nr.	Stratigraphy		Lithology	Acidity potential	Buffering effect
1	Quaternary	Quaternary, cohesionless	gravel, sand	0	0
2		Quaternary, cohesive	alluvial clay glacial till banded clay	0	++++
3		Late pleistocene	rubble	0	0
4	Tertiary (Paleogene)	"Thierbach" layers, cohesionless	gravel, sand	(+)	0
5		"Thierbach" layers, cohesive	silt	(+)	0
6		Aquifers 2.2, 2.4	sand	+	0 – (+)
7		Aquifer 2.5	shell sand, shell silt	+	(+) - +
8		Aquifers 2.6, 2.7	silty sand, glauconite silt	+++	0
9		Aquifer 3	fine and coarse sand	+++	+
10		Aquifer 4	Sand, silty sand	++	0
11		Aquifer 5	sand	+	0
12+13+14		Cohesive intermediate material	clay, silt	(+) - +	0
			Occurrence of the characteristic:	0	not
				(+)	very low
				+	low
				++	high
				+++/ ++++	very high

Geochemical model and balancing

In order to characterize overburden materials related to their chemical and physical properties, results of geochemical investigations of about 200 drilling locations were used. During the investigation, parameters such as total sulfur content, sulfate-sulfur content, total carbon content (Ct), carbonate content ($CaCO_3$) and exchange capacity (CEC) were determined for sediments in each geological layer. The analyzed parameters were then integrated using a laminar interpolation, to determine the geochemical potentials of several areas. A weighting of geochemical parameters, according to their thickness inside the geological layer, was found to be necessary (Hoth et al., 2000). The interpolation is based on a kriging method, realized with the modeling and visualization software SURFER®. Fig. 1 illustrates the results of interpolation for Unit 4. Total-sulfur contents in sediments of this unit are shown in Fig. 1(a). In summary, 58 drillings with 160 soil samples located into aquifer 2.6 or 2.7 were analyzed. The red parts of this map illustrate very high total sulfur contents up to 3 wt%. The calculated buffering potential in materials of this unit is shown in Fig.1(b). Beside some northern parts, Unit 4 is nearly free of carbonate. This causes a very high potential for acidification.

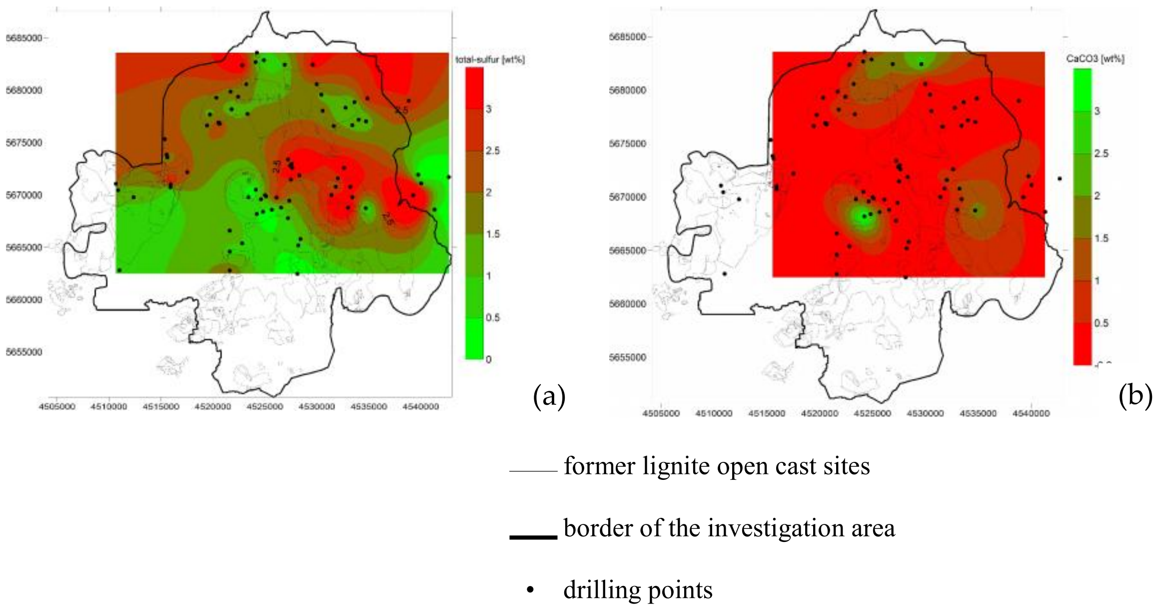


Figure 1 Predicted total-sulfur (a) and $CaCO_3$ (b) contents (wt%) in unit 4

Predicted dump pore water quality

Determination of average dump compositions is based on a "blending" of geochemical parameters with the thickness of the stratigraphic units. The investigation area is rasterized with a spacing of 125 x 125 meter. Geochemical parameters, derived from analyses, are converted into predicted parameters for overburden dumps (Hoth et al., 2000).

The prediction of overburden dump pore water quality is carried out using the hydro-geochemical equilibrium model PHREEQC (Parkhust & Appelo, 1999) and the important initial amounts of minerals including pyrite, calcite, dolomite, siderite and gypsum are used. The potential of precipitation of ($Fe(OH)_3$ and kaolinite) is respected as modelling minerals. Furthermore, the CO_2 content of the pore waters and in the gas phase is also considered and is related to the equilibrium of carbonic minerals. To calculate the equilibrium of groundwaters, an initial CO_2 concentration in the gas phase was defined. Also, the equilibrium between water solution and predicted CEC was included in the model, based on standard exchange coefficients of the PHREEQC database.

Analysis of current groundwater conditions

Characterization of the groundwater conditions of the whole investigation area is based on an intensive data analysis for generating a fundamental database. Data total of 1300 groundwater observation wells were used. To describe the actual sulfate pollution of groundwater, a series of measurements from 2009 to 2014 were considered and medians determined.

The saturation state of the groundwaters, related to relevant mineral phases (gypsum, calcite, siderite) was also determined using PHREEQC. At sulfate concentrations around 1600 mg/l a reduction of calcium is observed, because of precipitation of gypsum.

Saturation indexes of calcite and siderite depend mainly on pH values. Groundwater samples of dumps with pH values according to buffer range of calcite and siderite are often characterized as saturated groundwaters.

Overburden dump aquifers show the highest sulfate concentrations (on average 1650 mg/l) in the investigation area. The reason for elevated sulfate concentrations, greater than 3000 mg/l, appears to be the combination of overburden material behavior (high content of pyrite) and very low carbonate contents (no buffering) together with low clay contents (allowing oxidizing conditions via air access). Furthermore, slopes and edges of active mining and overburden dumps have high sulfate concentrations, due to exposure to oxygen in air. In some parts of the investigation area, groundwater of unaffected aquifers also shows high sulfate concentrations related to aeration during the artificial lowering of the groundwater level. The interaction with outflowing of dump pore waters also plays an important role. For modelling sulfate transport in groundwater, initial concentrations of sulfate were defined for every stratigraphic unit.

Mass flow and one-component transport modelling

Groundwater flow model

Results of overburden characterization and groundwater analysis were combined into an existing large scale model for mass flow modelling (called HGMS) (IBGW Leipzig). The program code PCGEOFIM® was used to calculate groundwater flow and migration processes. The model was based on the expected geological structure. Natural geological layers, overburden dumps and parameters of active mining were included in HGMS (see IBGW GmbH, 2010). The geological series of layers included all aquifers of the investigation area and zones between aquifers were regarded as aquitards. The model was adjusted for hydraulic parameters such as flooding concepts or lake-water levels using water levels from 11.000 observation wells within the calculations.

Concept and parameterization of sulfate transport

The rise of groundwater level influences the solute migration process of overburden dumps. For the simulation of the migration process, including advection, dispersion and relevant chemical reactions, PCGEOFIM® was used. Intended operations in active open cast mines were also considered alongside the mass flow inside unsaturated dump material. The sulfate concentration in groundwater was impacted by chemical reactions. Accordingly, a concept for consideration of minerals in solid matter was developed. The formation of gypsum, during the genesis of dumps and the rise of groundwater level, was found to be important. Therefore, the additional storage of gypsum was described with isotherms, including the thermodynamic equilibrium between formation and dissolving. In summary, nine classes of isotherms were defined (Figure 2).

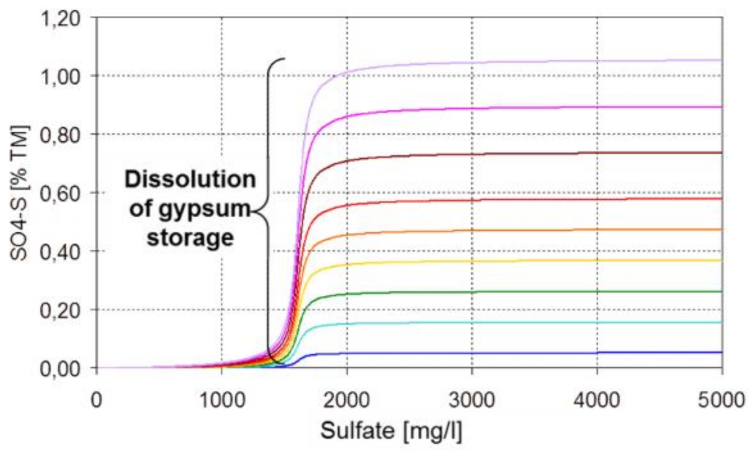


Figure 2 Dissolution of gypsum, considered as isotherms

The consideration of mass flux by using HGMS was possible in lateral and vertical direction. The first step was to investigate zones likely to have considerable plumes in future. Thereafter, current groundwater conditions were included and the mass flow out of overburden dumps was considered and future groundwater compositions simulated up to the year 2100.

Reactive transport model

A reactive transport model was used to investigate the three-dimensional reactive processes of mass flow, for two small scale parts of the whole investigation area. Using this approach, a model which describes sulfate formation and sulfate release, according to other substances in aqueous solution or in solid phases, was generated. The large scale model HGMS, including hydraulic parameters and storages of substances, was used as a background for reactive transport modelling. Calculations of reactive transport and the regional distribution of sulfate were based on the computer code PHT3D, it combines geochemical modeling (PHREEQC) with three-dimensional groundwater flow.

RESULTS AND DISCUSSION

Predicted dump compositions and hazard potentials

After the blending of geological structures and geochemical conditions, predicted dump compositions were derived. Figure 3 presents the predicted total-sulfur (a) and carbonate (b) contents for two selected overburden dumps. Red coloured areas in figures (a) and (b), depict high total-sulfur contents and very low carbonate contents and are described as hazard areas, because of high acidity potential and low buffering effect.

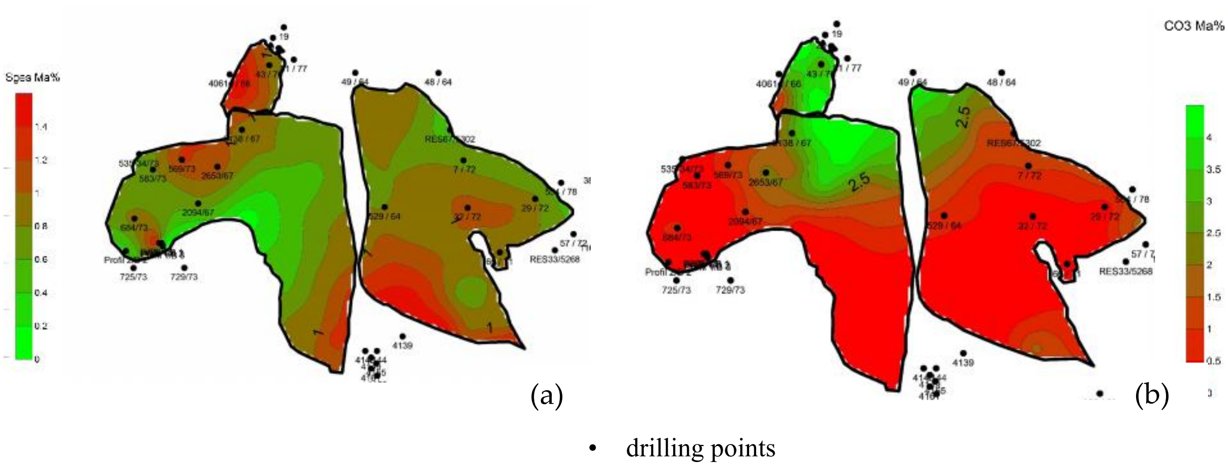


Figure 3 Predicted dump pore water composition (a) total-sulfur and (b)carbonate content in two overburden dumps

Predicted overburden dump pore water qualities

The predicted sulfate concentrations for one part of the investigation area are presented in Figure 4, along with monitoring measurements from the observation wells (coloured points). In the northern dump (called dump Witznitz), very high sulfate concentrations (up to 6000 mg/l) were calculated. Overall, not every measuring point could be reproduced by hydro-geochemical modelling. In these overburden dumps (especially Witznitz) the displacement of overburden because of the mining technology was not considered in the modelling, therefore the red area could be shifted. Furthermore, the measurements represents the part of the filter (around 2 m) and therefore weathering zones were not considered. In summary, the main groundwater condition could be simulated by the model.

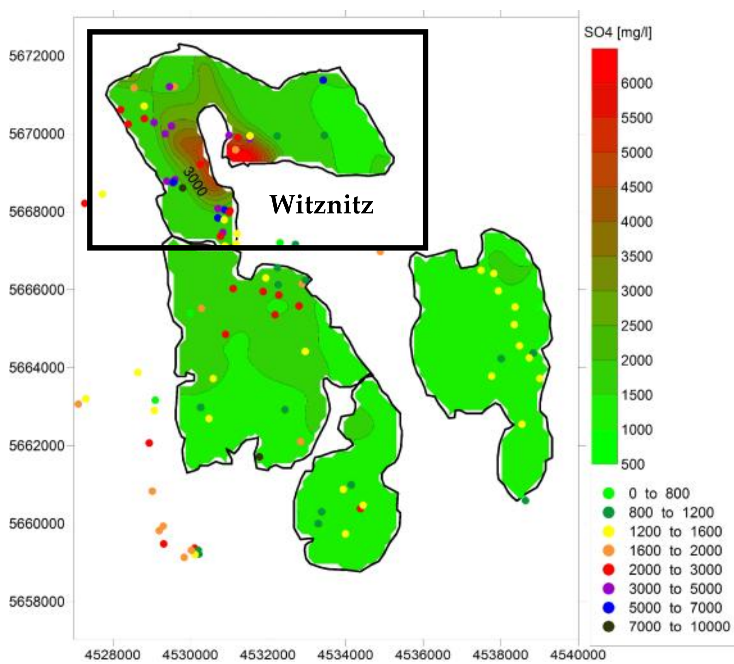
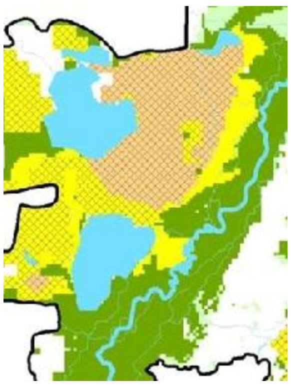


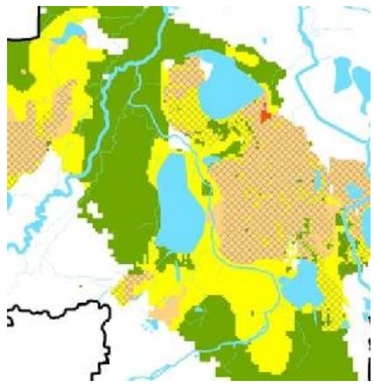
Figure 4 Predicted sulfate concentrations compared with measurements in observation wells

Results of sulfate transport modelling

Figures 5 and 6 present the sulfate concentrations in the outflow of the overburden dumps for selected stratigraphic units in the year 2100. Yellow, orange and red coloured areas suggest groundwater pollution because of dump pore water migration. In unaffected areas the sulfate concentration is around 300 mg/l. According to hydro-dynamical circumstances, stationary conditions are expected in the year 2100.

Overall, the pollution plumes with sulfate concentrations higher than 600 mg/l widen out within the stratigraphy. Local plumes around the dump boarders appeared in the upper layers, because of dilution effects (groundwater recharge) and groundwater seepage into watercourses.





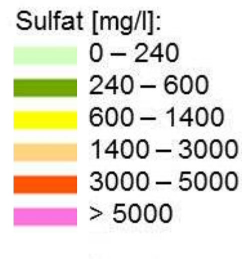
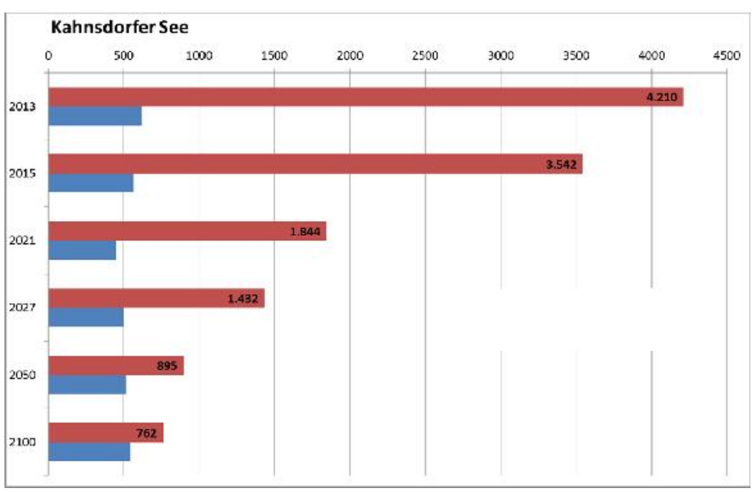


Figure 5 sulfate concentrations in 2100 after transport modelling, aquifers 1.1-1.7

Figure 6 sulfate concentrations in 2100 after transport modelling, aquifer 4

The development of the sulfate mass flow from groundwater to lakes is shown in Figure 7, and is illustrated for one selected lake (Kahnsdorfer See) in the investigation area. This relic lake is next to Witznitz, one of the overburden dumps with the highest sulfate concentrations in pore water. According to Figure 7, the sulfate mass flow into the lake will be reduced from 4210 m^3/d in 2013 to 762 m^3/d in 2100, by nearly constant groundwater influxes.



sulfate mass flow from groundwater to lake [kg/d]

groundwater influx [m^3/d]

Figure 7 sulfate balance till 2100 for one lake in investigation area

CONCLUSION

Calculated geochemical parameters from overburden dump sediments, especially sulfate, carbonate and total carbon, have been used to describe conditions for every stratigraphic unit and depict hazard potentials. In these zones, high sulfate concentrations, as a result of pyrite oxidation, were generated. Sulfate migration into groundwater and flow to unaffected areas has resulted in AMD effects, as buffering geological materials are generally not available. The main AMD problems were related to two stratigraphic units - unit 8 (aquifers 2.6, 2.7) and unit 6 (aquifers 2.2, 2.4), with high sulfur and low carbonate contents, combined with the high permeability. The results were integrated into a sulfate transport model and the current groundwater conditions determined by analysing numerous data of observations wells and dump gauges. The highest sulfate concentrations, of more than 6000 mg/L, were detected in dump aquifers. An average sulfate concentration of 1650 mg/L was shown for dump pore waters. For stratigraphic units 6 and 8, high sulfate concentrations in unaffected aquifers were also detected. The evaluation of saturation states in aquifers, and respective gypsum, calcite and siderite contents indicate that pore water in overburden dumps and the zones discussed are predominantly saturated or oversaturated. Based on the current groundwater conditions and calculated geochemical potentials of the overburden dumps, initial parameters were defined and implemented in the sulfate transport model. The first results of sulfate transport modelling indicate mainly local pollution plumes in areas adjacent to the overburden dumps.

In summary, the investigations show, that the downstream area of the polluted aquifers will be impacted by the inflow of dump groundwater for several decades. Whilst there are identified pollution plumes; areas between these plumes will not be affected.

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