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Prediction of Sulfate Concentrations and Mass Fluxes in Aquifers Influenced by Lignite Mining in the South of Leipzig (Germany)

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Abstract Aquifers in the Central German Mining District are affected to a great extent by open cast mining of lignite. In order to continue the water management objectives, a prediction of the groundwater qualities is necessary. Therefore, the mobilized sulfate potential, as a main component present in the aquifers, is derived from a geological and a geochemical model. Flow and non-reactive transport modelling is carried out by an existing large scale model (HGMS) of the company IBGW Leipzig. The long-term prognosis of the sulfate behaviour in the investigation area for the next 100 years results from the combination of all these models.
Keywords lignite mining, groundwater quality, pyrite oxidation, hydrochemical modeling, 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 affected to a great extent. Environmental objectives of the EU's Water Framework Directive are not achieved within the current management periods for the area south of Leipzig.This region is a main part of the Central German Mining District, which occupies a working area of 2,048 km^2.Accordingly, the University of Freiberg has been instructed 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, because of mobilizing reactions. The main variable of the mobilized potentials is the fraction of oxidized pyrite-sulfur. After the mining process, the groundwater level rises, natural flow systems are reestablished and the migration process of the AMD-waters starts. In order to predict the impact of dump waters on unaffected aquifers a geological model, which includes the regular sequences of geological layers, is generated. Forefield concentrations of total sulfur, sulfide-sulfur and carbonate are balanced from data of older forefield drillings in the geochemical model. Simultaneously, the actual groundwater compositions in the investigation area rerecorded, for assessing the groundwater quality especially for the mining districts. Migration processes in groundwater are simulated by using the existing large scale model (HGMS) of the company IBGW Leipzig, based on the transport code PcGeofim®. Selected "Hotspot zones" are modelled subsequently, by means of a reactive hydro geochemical transport model, based on the program code PHT3D.

Overall, the purpose of this paper is the assessment of actual groundwater qualities based on the hydro geochemical interaction of dump materials with rising groundwater after the mining process. Furthermore, a prediction of groundwater composition for the next decades is established by using one-component transport modelling and reactive transport modelling.

Geological model

To verify the distribution of geological layers in the investigation area, data of petrographic facies are collected. Afterwards, several thousand drillings are used to generate a geological model, which describes the deposits and sequences of overburden layers. Different vertical layers of silt, clay, sand or gravel are 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 *Selected stratigraphic units and a general evaluation of their acidity and buffering effects*

Number	Stratigraphic unit	Lithology	Generation of acidity	Buffering effect
1	Quaternary, cohesive	boulder clay, alluvial clay, banded clay	0	++++
2	Aquifers 2.2, 2.4	sand	+	+
3	Aquifers 2.5	shellsand	+	+
4	Aquifers 2.6, 2.7	silt, clay, sand	+++	0
5	Aquifer 3	fine sand to coarse sand	++(+)	0

Geochemical model

In order to characterize overburden materials related to their chemical and physical properties, results of geochemical forefield investigations of about 200 drilling points are used. During the forefield investigations, for sediments of each geological layer parameters like total sulfur content, sulfate-sulfur content, total carbon content (C_t), carbonate content ($CaCO_3$) and Exchange Capacity (CEC) were determined. The analyzed parameters of each geological layer were now integrated in a laminar interpolation, to find out geochemical potentials of several areas. Therefore, a weighting of geochemical parameters, according to their thickness inside the geological layer, is necessary (Hoth et al. 2000). The interpolation is based on the kriging method, realized with the modeling and visualization software SURFER®. Fig. 1 exemplifies 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 2.7were analyzed. The dark 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 of acidification.

Particle diameter distribution and permeability of overburden material

During mining, especially during the dumping process by conveyor bridge technology, silt and clay layers were mixed with sandy materials. This causes heterogeneous overburden dumps in detail, but homogeneous in a larger scale of view. To estimate an average particle diameter distribution within the dumps, a large number of drilling points with grain size profiles of forefield conditions are considered. The permeability of overburden dumps has an important influence on the hydraulic behavior. To come up with a realistic estimation of dump permeabilities, the balancing of the pelite content is of great importance.

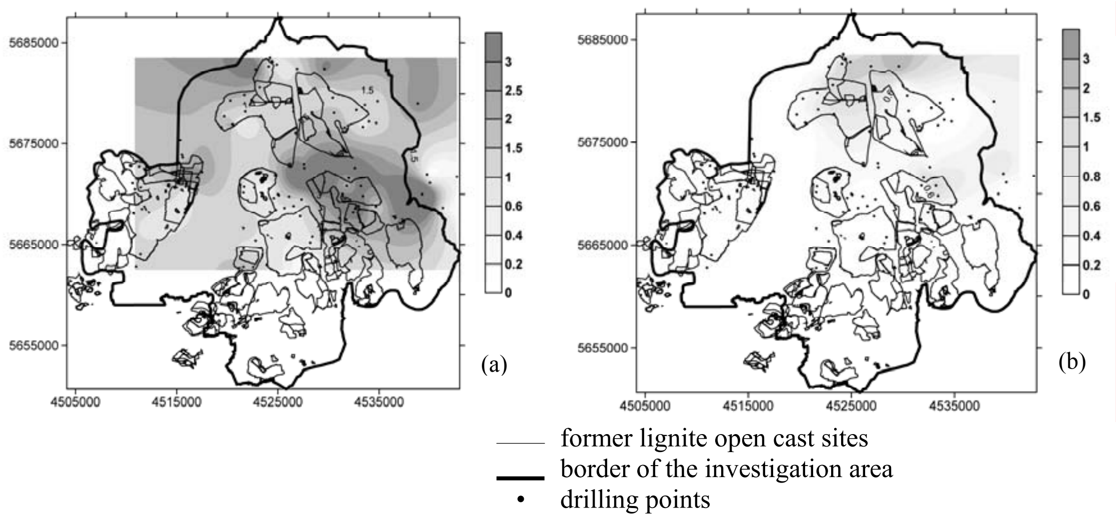

Fig. 1 *Predicted total-sulfur(a) and $CaCO_3$ (b) content (wt%) of forefield materials of unit 4*

Characterization of dumps and prediction of dump 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 in several parts. Geochemical parameters, derived from forefield analyses, are converted into predicted parameters for dumps (Hoth et al. 2000).

The prediction of dump water quality is carried out using the hydro-geochemical equilibrium model PHREEQC (Parkhust and Apello 1999). Therefore, the important initial amounts of minerals, including pyrite, calcite, dolomite, siderite and gypsum, are used. The possibility of precipitation ($Fe(OH)_3$ and Kaolinite) is respected too, considered as modeling minerals. Furthermore, the CO_2 content of the waters and in the gas phase has importance, related to the equilibrium to the carbonatic minerals. To calculate the equilibrium of groundwaters, an initial CO_2 concentration in the gas phase is defined. Also, the equilibrium between water solution and predicted CEC (derived from forefield data) is included in the model, based on standard exchange coefficients of 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. All together, data of 1300 groundwater observation wells are used. To describe the actual sulfate pollution of groundwater, series of measurements from 2009 to now be considered and medians are determined.

The saturation state of the groundwaters, related to relevant mineral phases (gypsum, calcite, siderite) is also determined with 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 ground waters.

Of course dump aquifers show the highest sulfate concentrations (on average 1650 mg/L) in the investigation area. The reason for extreme sulfate concentrations, greater than 3000 mg/L, seems to be the combination of overburden material behavior (high content of pyrite) and very low carbonate contents (no buffer) together with low pelite contents (oxidizing conditions – air access). Furthermore, slopes and edges of active mining and overburden dumps are parts with high sulfate concentrations, because of the oxygen exposition. In some parts of the investigation area, groundwater of unaffected aquifers shows high sulfate concentrations too. This is related to the aeration during the artificial lowering of the groundwater level. Furthermore, the interaction of outflowing of dump ground waters, plays an important role. For modeling sulfate transport in groundwater, initial concentrations of sulfate are defined for every stratigraphic unit.

Mass flow and one-component transport modelling

Results of overburden characterization and groundwater analysis were combined into an existing large scale model for mass flow modeling (called HGMS) of the company IBGW Leipzig. The program code PcGeofim® was used to calculate groundwater flow and migration process. The model based on a geological structure. Natural geological layers, mining dumps and parameters of active mining were included in HGMS (IBGW GmbH 2010). The geological series of layers includes all aquifers of investigation area. Zones between aquifers were regarded as aquitards.

The rise of groundwater level influences the migration process of dumps. For the simulation of the migration process, including advection, dispersion, sorption and relevant chemical reactions, PcGeofim® was used. Furthermore, intended operations in active open cast mines were considered as well as the mass flow inside unsaturated dump material Sulfate concentration of groundwater is impacted by chemical reactions. Accordingly, a concept for consideration of minerals in solid matter was developed. Especially the formation of gypsum, during the genesis of dumps and the rise of groundwater level, was important. Therefore, the additional storage of gypsum was described with isotherms, including the thermodynamic equilibrium between formation and dissolving.

The consideration of mass flux by using HGMS is possible in lateral and vertical direction. By using leakage-factors, a simulation of mass flow is not possible. Therefore, boundary conditions were defined for unaffected aquifers. To find out zones with considerable plumes in future, was the first step. Afterwards, current groundwater conditions were included and the mass flow out of dumps was considered. Furthermore, the future groundwater compositions will be simulated up to the year 2100.

Reactive transport model

The reactive transport modeling is used to look into the three-dimensional reactive processes of mass flow. These work can and will be done only for two or three small scale parts of the whole investigation area. Therefore, a model which describes sulfate formation and sulfate release, according to other substances in aqueous solution or in solid phases, is generated. The large scale model HGMS, including hydraulic parameters and storages of substances, is used as background for reactive transport modeling. Calculations of reactive transport and the regional distribution of sulfate are based on the computer code PHT3D, it combines geochemical modeling (PHREEQC) with three-dimensional groundwater flow.

Conclusions

Calculated geochemical parameters of dump sediments, especially sulfate, carbonate, total carbon and pelite content, describe forefield conditions for every stratigraphic unit and depict "Hotspot zones". In these zones, high sulfate concentrations, as a result of pyrite oxidation, are generated. That causes sulfate migration by groundwater flow to unaffected areas and acid mine drainage effects, if buffering geological materials are not available. The main problems are related mostly to two stratigraphic units - unit 4 (aquifers 2.6, 2.7) and unit 2 (aquifers 2.2, 2.4).High sulfur contents and low carbonate contents in these units, combined with a high permeability of these sediments are responsible for that. The results are integrated in the sulfate transport model.

The current groundwater conditions are determined by analyzing numerous data of observations wells and dump gauges. Thereby, the highest sulfate concentrations, partly more than 6000 mg/L, are detected in dump aquifers. In average, a sulfate concentration of 1650 mg/L is shown for dump groundwaters. For the stratigraphic units 2 and4, high sulfate concentrations in unaffected aquifers are detected, similar to the forefield conditions. The evaluation of saturation states in aquifers, respective gypsum, calcite and siderite shows, that groundwater of dumps and the mentioned zones are predominantly saturated or oversaturated.

Based on the current groundwater conditions and calculated geochemical potentials of the overburden dumps, initial parameters are defined and implemented in sulfate transport model. The first results of sulfate transport modeling show mainly local pollution plumes in neighborhood to the 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. In fact, there are pollution plumes; areas between these plumes will not be affected. The future work will be concentrated on the transport modeling and the reactive transport modeling, for a spatial and temporal differentiated prediction of groundwater condition.

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