DESCRIPTION OF REACTION AND TRANSPORT PROCESSES IN THE ZONE OF AERATION OF MINE DUMPS

Kirsten Pollmer and Michael Eckart

Geocontrol/HPC
Umwelttechnische Beratung Gera GmbH
Heinrich-Laber-Straße 11
D-07548 Gera, Germany
Phone: +49 365 56420, Fax: +49 365 56424

ABSTRACT

The release of contaminants due to oxidation of pyrite in the aeration zone of the waste rock dumps generates nearly half of the contaminant load of the Ronneburg mining district and, if left unremediated, would become a dominant source of contamination. Numerous activities geared toward optimization of the long term contaminant release are presently undertaken. The definition of technical requirements of remediation projects and their economic constraints are carried out by Wismut. The relocation of potentially hazardous mine dump material into the open pit in a sequence according to the NP/AP-ratio of the waste rock material, topped by a well engineered cover system satisfies the main requirements for such an optimization. One of the first requirements in the attempt to minimize (and control) the release processes of pollutants in the cover layer of the backfilled open pit mine is to provide a decryption of the coupled two phase water-gas flow and multiple mass transport as a function of depth and time. To this end the long term effort of Wismut to develop a model for the zone of aeration has been taken up with the following objective:

- *identification of cover parameters leading to consumption of sulphides (thus decreasing the acid generated potential) while keeping the sulphate load low; and*
- *prevention of long-term acidification, in order to minimize the radionuclide release.*

The advantage of the proposed approach is the separation of the gas- and water-phase transport into a convective and diffusive part, the consideration of the seasonal effects and the possibility of plugging-in various geochemical and hydraulic parameters using a discrete numerical model. The modelling results and derived practical conclusions will be presented.

INTRODUCTION

As a consequence of the high volume-ratio between mined out underground works and open pits the remediation of the mine dump-complexes of Wismut near Ronneburg in Eastern Thuringia necessarily implies that most of the mine dump material has to be relocated in the vadose zone. Contrary to wet-deposition, where one can expect a virtually complete long-term-prevention of processes leading to the release of pollutants, access of oxygen to the deposited material in the unsaturated zone cannot be completely prevented. Thus, the following qualitative and quantitative chemical reactions take place according to the presence of reactive components. The essential reactions are:

- oxidation of pyrite, reduction of oxygen, formation of sulphates, decrease in pH;
- tendential neutralization according to availability of basic material (calcite, dolomite);

- *solution* processes; and,
- when reaching the concentration of saturation, the formation of secondary minerals according to the respective mineral paragenesis.

As a result of these processes, highly mineralized water will be formed over a long period of time. According to the specific conditions at the place of reaction, the mineral concentration of surface- and seepage water increases. At Wismut, mine dump-material is either being backfilled into the Lichtenberg open pit or the dumps will remain at their present location and need to be covered (e.g. Halde Beerwalde). In order to assess the efficiency of the remediation process, it is essential to forecast the concentration and calculate the load of the pollutants in the seepage water.

The following statements characterize the complexity of the problem:

- long-time-development of the pollutants without remediation;
- influence of the oxygen-inventory when applying a cover;

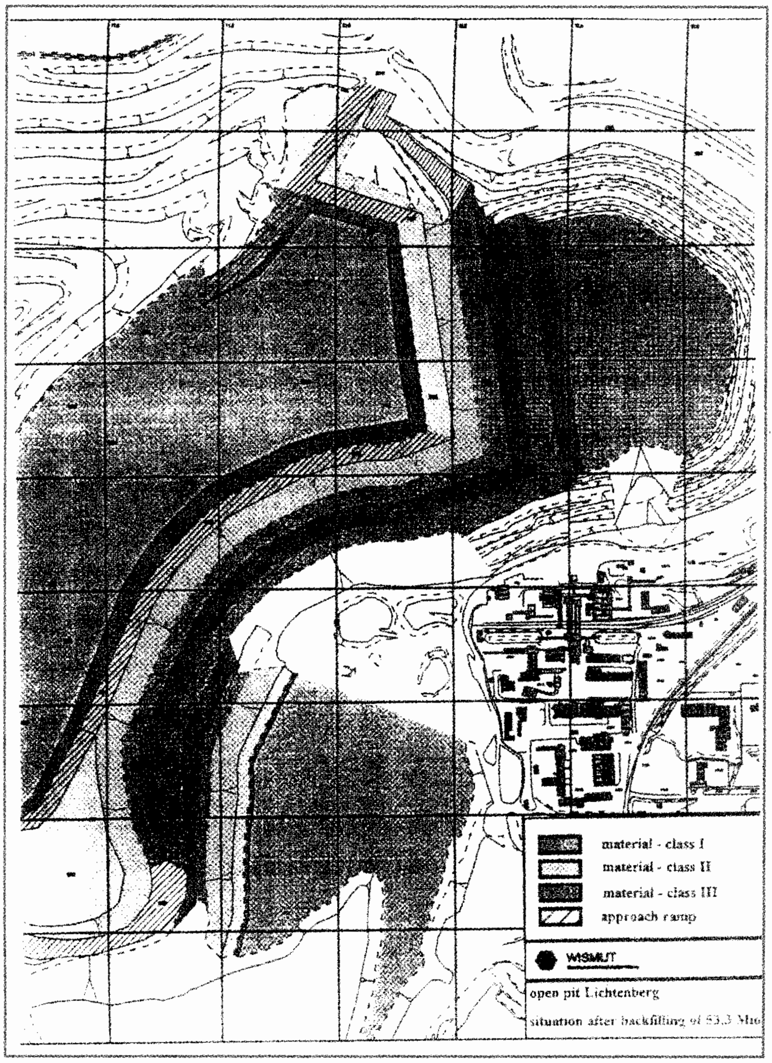


Figure 1. Lichtenberg open pit. Material-classes in the Lichtenberg open pit.

- stabilization of a chemically neutral environment in the upper parts of the mine dumps at minimum oxygen-concentration (prevention of heavy metals leaching); and
- temporal dynamics of the release of pollutants after applying a cover due to a reduced velocity of migration, and the effect of the pollutant-enriched retention-water-phase (two-phase migration water-gas).

In order to find ways to solve these problems, Geocontrol continued some of Wismut's long standing research and development efforts. On the basis of Wismut's model "SICKER" Geocontrol developed the model "TENSIC" (two phase migration/multimigrant transport modelling tool). This tool allows the modelling of the pollutant-load in the unsaturated zone of mine dumps and for the backfill-process of the open pit. The estimation of the effects of certain properties relevant to the migration processes in combination with geochemical interactions represents a linked transport- and reaction process with phase-transitions. So far no programmes are commercially available to simulate these processes to a satisfactory degree of accuracy, and only limited experimental investigations were carried out. In this presentation the results of the modelling of the processes in the aeration zone of the refilled Lichtenberg open pit and in the Beerwalde mine dump will be described.

DESCRIPTION OF THE OBJECTS OF INVESTIGATION

Lichtenberg open pit

Remediation of the Ronneburg mining-area focuses on the concentrated deposition of most of the mine dumps into the Lichtenberg open pit. Potentially acid producing dump material will be safely deposited in the pit in an area, which will be submerged after the flooding of the pit. This safe deposition will be achieved by the fact that the rising groundwater will have lost its oxidizing character and thus the ability to leach sulphides and by minimizing the access of oxygen by aeration by the overlaying overburden. The level of minimisation will depend on the chemical reactions that occur in the upper parts of the zone of aeration (C-zone) and on the conditions of transport for infiltration (fluid phase) and the variable composition of the soil air. The cover (D-zone) overlays the zone of aeration and minimizes the discontinuous transport of the gaseous and aqueous phases.

- Minimizing of migration of oxygen in deep parts of the dump and reduction of sulfatload;
- Consumption of oxygen in the C-zone;
- Fast adjustment of a geochemically stable neutral environment;
- Prevention of channelling;
- Minimization of oxidation processes for the convective mobil oxygen.

Beerwalde mine dump

The Beerwalde mine dump was deposited approximately 25 years ago. The present concept favours a remediation in situ which minimizes the release of pollutants by covering the dump. The purpose of our modelling project is to forecast the development of the composition of the seepage water at the dump's base. In order to achieve this, it is necessary to take into consideration the fact, that the dump has been uncovered for some 25 years. The model will have to take into account the fact, that now and in the following years a covering layer will be applied.

ELEMENTS OF THE MODEL "TENSIC"

Basic parameters for the mine dump cover

Due to the small conductivity and the high retention-saturation of the pore-volume a high saturation is present in the cover material when applying a mine dump cover, e.g. a mixture of clay and silt. When applying a cover over a dump, which has been without a cover until recently (for some 25 years, as an average) the influx of seepage water and the effective oxygen-diffusion-coefficient will be considerably reduced. The time dependent development of the seepage water concentration depends mainly on the ratio of the changes of these two parameters and not on the absolute reduction of one of them.

For all practical purposes a numerical expression for the assessment of the efficiency of the cover is required. As a relatively straight foreward generalized method, wheather a mine dump cover leads to a reduction of concentration of pollutants in the seepage water in the long run, we suggest the combination of the numerical expressions,

D	effective diffusion-coefficient	[m^2/s]	measure of the chemical turn over
M	thickness of the cover	[m]	influences the diffusion gradients
U	Darcy-velocity	[m/s]	influences dilution

to a dimensionless number (number of physical similarities), which we call the POECK-number

POECK-NUMBER = D / (m • u) (Eq. 1)

Regardless of the exact ratio of the parameters included, the development of the pollutants is reduced considerably when applying a dump cover stratum and should thus be considered as a measure for the success of remediation. Which "diffusion-rate to Darcy-velocity"-ratio can realistically be achieved is still controversial. In very many simulations we could demonstrate that the development of the concentrations reacts very sensitively to changes of the POECK-NUMBER, but influences the development of the pollutant-load only marginally.

Model-structure

A two-phase-flow-model "[liquid-gas]" was combined with a multimigrant transport model. The principle structure of the combined model is shown in Figure 2.

Two-Phase-flow WATER + GAS

Precipitation, Temperature, Water content of air — Air-pressure, Change of saturation

Variable: Saturation and Darcy-velocity

Mass-Transport

Solid Phase	Gasous Phase	Liquid Phase z. B.	
$Fe(OH)_3$		ΣSO_4	ΣUO_2
FeS_2		ΣCa	ΣRa
$CaCO_3$		ΣMg	ΣAs
$CaMg(CO_3)_2$		ΣCO_2	
$CaSO_4 * 2H_2O$		$\Sigma FeII / FeIII$	
FeAsS			
UO_2	O_2 (g) —>	O_2 (l)	
$RaSO_4$ u.a.	CO_2 (g) —>	CO_2 (l)	

Reaction

Kinetic of reaction		
r (Pyrit)	=f (O_2, Concentration, FeIII)	Calculation of chemical equilibrium with 44 Species
r (Carbonat)	=f (pH, Concentration)	
r (Gypsum)	=f (Equilibrium) —>	pH-Value
r $(FeOH)_3$	=f (Equilibrium)	
r (UO_2)	=f (r Pyrit) - Sorption - Including	c (Equilibrium)
r (Ra)	=f (rU) - r($XXSO_4$) - Sorption	

Figure 2. Principle structure of the combined model.

In order to solve the differential equation that describes the two-phase flow the subsurface is divided into a finite number of discrete flow-elements and the equation that describes the two-phase flow is transformed into a space- and time discretized form. The boundary conditions of the two-phase-flow-model will be derived from real day to day atmospherical data (rain-fall, relative humidity, atmospherical pressure, temperature). Thus the fundamental transient forces for convective transport of oxygen in the dump (convective transport of oxygen, process of saturation, atmospherical pumping) can be described. The variable "pressure" is calculated according to the temporal *implicit* procedure, whereas the variable "saturation" is calculated according to the temporal *explicit* procedure and corrected by iteration of the pressure. After calculating a solution for the equations of the gaseous- and aqueous phase the phase-saturation and the Darcy-velocity are transferred to the contaminant-transport-model. This in turn calculates for every time step the distribution of the environment inherent concentration for the flow regime.

To quantify the chemical interactions in the electrolyte-solution for each time step and each element of flow, the system of balance of the components has to be resolved. To calculate the activity in these tests the Davis-equation is used. For each proton-concentration the term of solution, e.g. solution of calcite, dolomite and pyrite, is calculated.

Based on the field of velocity, calculated from saturation and pressure, the convection-diffusion-equation is resolved for a freely selectable number of migrants for the fluid and gaseous phase. The interactions between the three phases solid-fluid-gaseous are taken into account. The selection of the migrants depends on the specific material in the considered geological environment. Recently we also appended the carbonate- and sulphate complexes of uranium and radium to the model, which enables us now to describe the development of radionuclides in the seepage of mine dumps

EXPERIMENTAL PROCEDURES

To validate the model and to derive reaction parameters experimental investigations were necessary. Therefore two column-experiments were carried out.

Replica of the uncovered zone of aeration

Since the zone of aeration is still not developed in the Lichtenberg open pit the experimental replication of the zone of aeration can only be simulated in a column experiment. Therefore a depth (thickness) of 10.5 m was simulated in a column experiment, in which in steps of 1.5 m gas-samplers and water-sampling devices were installed. Due to static considerations the columns were separated into 7 interconnected segments. Thus both, the composition of the gaseous phase and the aqueous phase were to be observed during the whole duration of the experiment.

In order to characterize the chemical and physical processes, the following extensive investigations were carried out:

- characterisation of the material at the set up and the end of the experiment;
- grain size distribution;
- chemical composition;
- soil mechanical parameters (water contents, density of dump material);
- semiquantitative mineral-composition;
- microbiological characterisation.

During the experiments the composition of the gaseous phase and the qualitative and quantitative composition of the aqueous phase were determined weekly.

Determination of the relative permeability of mine dump-material

The relative permeability (ratio between saturation and Darcy-velocity) is a fundamental parameter necessary to determine which area of permeability is available for gas-migration and in which proportion the reduction of volume and the saturation of the aqueous phase is changed. This influences directly the retention time after the application of the dump cover and thus the process time after which a change in the concentration in the seepage water at the toe of the dump can be expected.

Due to the importance of this parameter the percolation experiments were carried out at variable flowrates. In this way the curve for the relative transmissivity for the different phases in the dump material were determined.

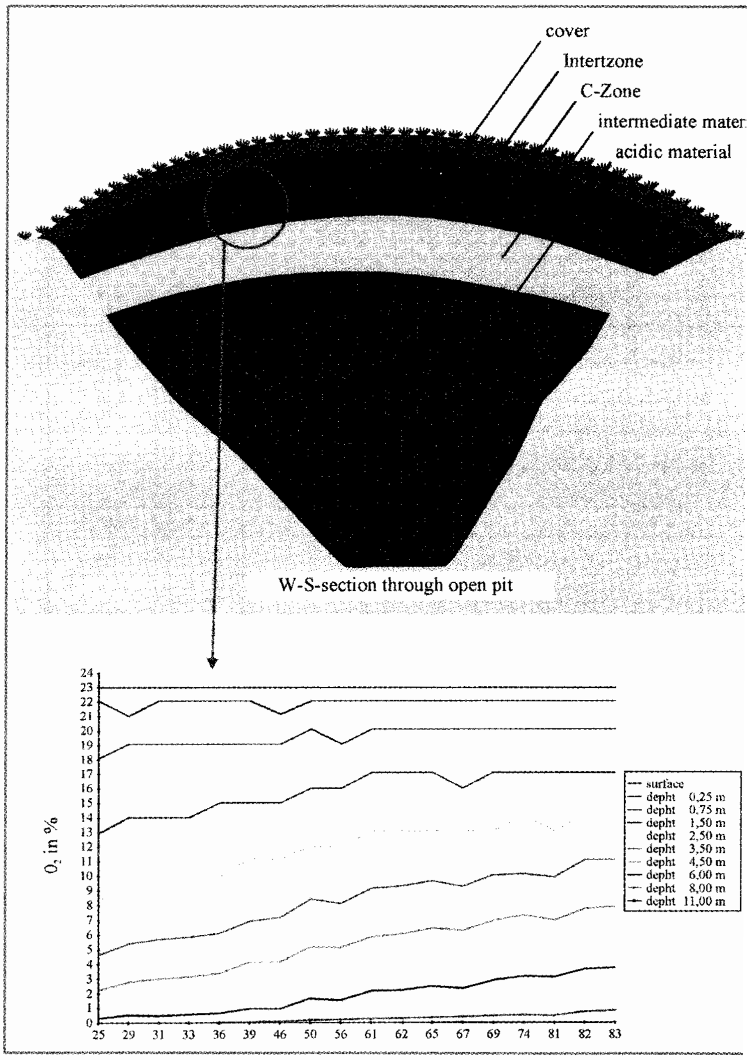


Figure 3. Simplified section trough open pit. Spatial development of oxygen-concentration.

SELECTED EXAMPLES FOR MODELLING

To describe the essential features of the model three examples were chosen. At the first example no acidification occurs, because of the high NP/SP-ratio, although, due to the high concentration of dolomite a very high sulphate load has to be assumed (e.g. Beerwalde mine dump). The second example, "Nordhalde dump", leads to acidification because of the low NP/SP-ratio. Both examples are suitable to document the *undesirable processes* (acidification, high sulphate load) in the upper part of the open pit. In the third example the effects of the mine dump material on the properties of the C-zone of the refilled Lichtenberg open pit will be presented.

Lichtenberg open pit

In order to backfill the C-zone (zone of aeration) of the Lichtenberg open pit material from the Paitzdorf and Reust mine dumps is suggested. In the backfilled Lichtenberg pit the C-zone will act as an additional cover in addition to the actual cover-stratum. Both dumps, earmarked for deposition in this zone, exhibit a relatively high NP/SP-ratio of 3.9 and 4.3 respectively and a considerably lower dolomite-concentration compared to the Beerwalde mine dump.

Without an additional cover the following, depth dependent long term development of the oxygen-concentration in the dump would occur.

Beerwalde mine dump

In this example we would like to simulate the influence of two different cover materials for the Beerwalde mine dump with two different covers placed after an uncovered exposure time of some 25 years. The seepage water from the Beerwalde mine dump has typically a very high magnesium-sulphate-concentration. Due to this fact the reproduction of the presently measured magnesium-sulphate-concentration of 26 g/l is an important calibration criteria for all parameters relevant to the reaction-kinetics. The temporal development of concentration of contaminants primarily depends on storage capacity and at a later stage on the POECK-number.

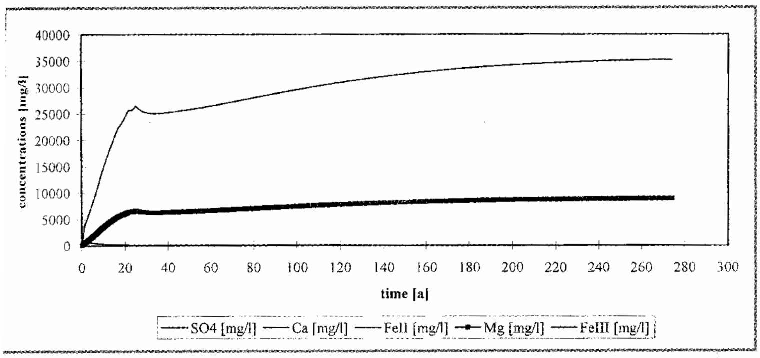


Figure 4. Development of concentrations from dump Beerwalde - covering after 25 years.

Nordhalde dump

This mine dump exhibits an NP/SP-ratio of 1 to 2. This results in the following pH- and contaminant-concentration prior to and after the application of a high quality cover.

SUMMARIZING STATEMENTS

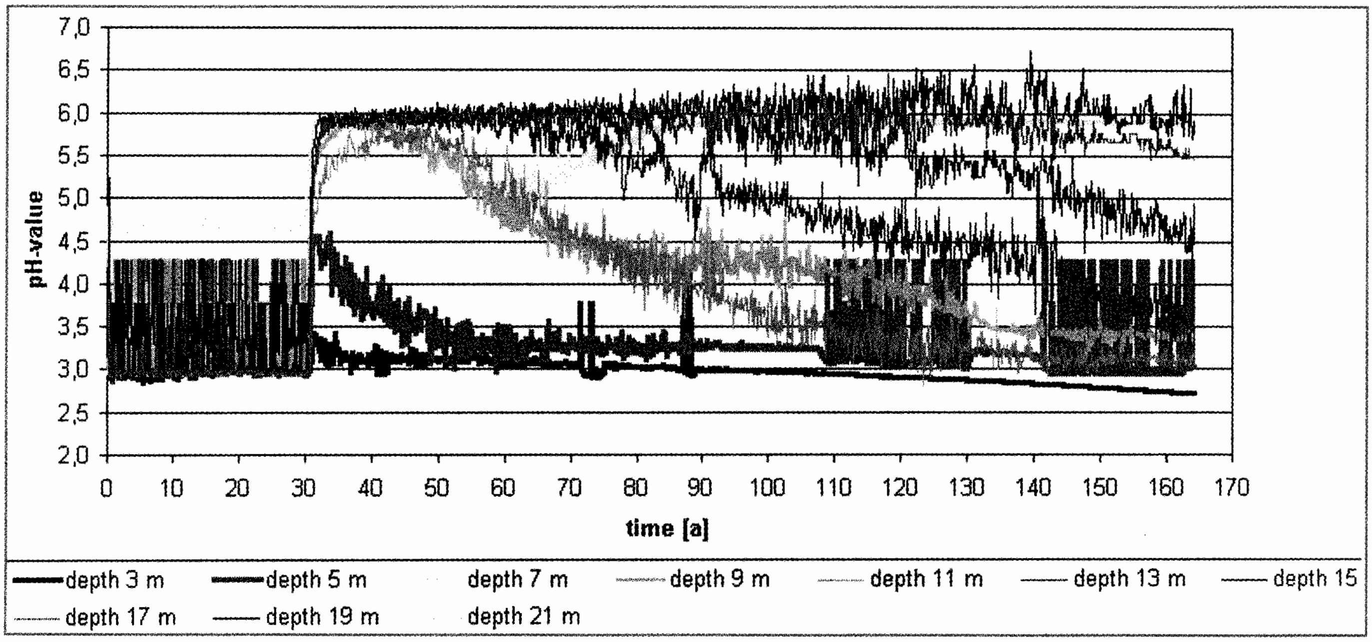
The model TENSIC was developed to characterize reaction- and transport-processes in the zone of aeration of mine dumps and the Lichtenberg open pit. It enables us to illustrate the space-time development of pressure, saturation and concentration in a porous media, taking into account real day to day climatic data.

It is intended to cover certain mine dumps and the refilled Lichtenberg open pit to minimize the environmental impact. Therefore we tried to include the mechanisms of mine dump covers on the contaminant load development in a real 25 years old uncovered mine dump. Thus a continuous modelling from the beginning of the deposition through a 25 year period of uncovered exposure to atmospheric conditions, during which leaching already occurred, up to a 100 to 500 year period in the future of the covered mine dump was carried out. The concentration- and load development was simulated in this model from the available data of tests, carried out on the originally deposited dump material. As a result of this model yield-curves of pollutant load and -concentration were gained for different dump materials and different oxygen- and pH-value profiles.

Starting from the deposition of the dump material up to the present day condition of the dumps these calculations show, that concentrations of the seepage water can be calculated with the concept of modelling presently available to match data for the reaction kinetics for the so far investigated Drosen, Beerwalde, Reust, Paitzdorf and Nordhalde dumps. As initial data for the model-calculation only the solid-concentrations of the leached dump-material (e.g. pyrite, dolomite, calcite) at the time of deposition are required. Follow-up calibration for the specific dumps was not required, so far. Particular prove-criteria are the detailed description of the high magnesium concentrations of the Beerwalde dump and the acidification in the Nordhalde dump. The application of a mine dump cover stratum minimizes the percolation of seepage water and the effective oxygen-diffusion-coefficient. An increase in concentration in the fluid phase is decisively effected by the ratio of change of both parameters, but not by the decrease of one parameter. In order to compare different combinations of parameters, we suggest the characteristic POECK-NUMBER:

$$\text{POECK-NUMBER} = D / (u \cdot M)$$

In covered dumps a reduction in the seepage water-concentration can only expected after decades despite of cover-strata with small POECK-numbers, due to the highly saturated phase of retention water and the low flow rate of the percolating seepage water in the cover stratum. However, a reduction of the contaminant load occurs much earlier, because



[Figure 5. Development of pH-value - with cover after 30 years.

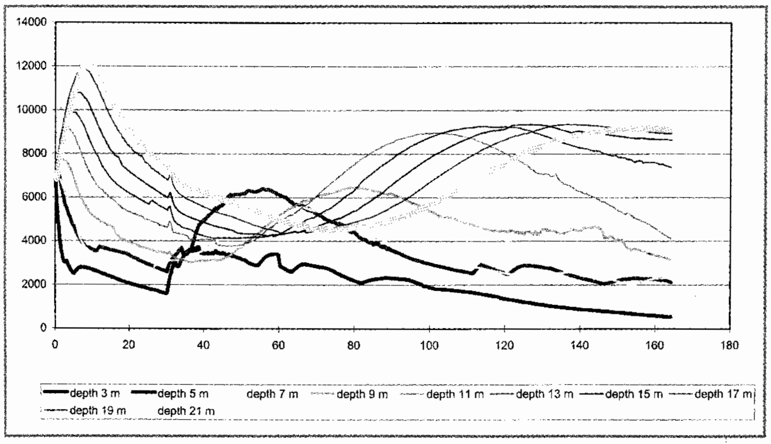


Figure 6. Development of sulphate-concentration - with cover after 30 years.

of the reduced fluid volume. As a result of these investigations we recommend to strictly apply the concept of different zones of deposition (A-, B-, C-zone) for the backfill of the Lichtenberg pit, in order to achieve an approximately 10 m thick zone at the top of the backfilled pit (C-zone), consisting of material from the Paitzdorf and Reust dumps. The tools developed for some problems specific to Wismut can, certainly, also be adopted to other settings were migration of fluids and gases through unsaturated media are of interest. Our numerical models allow:

- improved calibration by use of all available field data (meteorological data, flow, concentration, physical properties of the porous media etc.);
- quantitative forecast of contaminant transport, seepage volume and development of contaminant concentrations for long periods of time;
- cost/benefit analyses by simulating effects of different types of covers on seepage volume, chemical reactions and resulting contaminant concentrations.

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