

Tailing erosion as water quality forming factor in lake systems arising from abandoned open cast lignite mines

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ABSTRACT

Many East German open cast mines are in the process of reclamation, involving lake formation. Most of these evolving lakes are of poor water quality because of influx of pyrite oxidation products stored in adjacent sediments. Water quality modelling emphasized groundwater induced acidity transport to the lake, whereas water quality impact of tailing erosion was neglected. Formulation of erosion induced mass influxes for a mass balance based water quality prediction model was performed in the presented study. Therefore, sediment transport and chemical sediment properties were quantified. For a chosen test site, the water erosion models RUSLE, EROSION 2D and PEPP were applied and results were compared with field measurements. Annual erosion rates of lake embankment sediments, which lack soil formation and vegetation, were computed. They range from 300 up to 900 tons per hectare. Chemical sediment properties were related to sediment parameters, representing hydrogen ion equivalent release or binding at sediment elutriation. Those parameters are ion composition of pore water and exchange complexes as well as content of minerals, e.g. calcite, jarosite, jurbanite. The parameters were determined in the laboratory. Mineral saturation was calculated by chemical equilibrium modelling of pore water. At two investigated dump slopes, sediments were qualified for an acidity input of 3 mmol (eq) / 100 g dry sediment. For one dump slope sediment, an alkalinity input of 6 mmol (eq) / 100 g dry sediment was obtained. An annual net acidity influx of approximately 0.5 million mol was calculated by relating results of investigated dump slopes to corresponding embankment parts.

INTRODUCTION

With a yearly production of 320 million tons East Germany's lignite production was the first in the world. To produce one ton of lignite, 6 to 7 m³ water and 4 m³ overburden had to be replaced. Recently, many open cast coal mines in Eastern Germany are in process of reclamation. Re-establishment of a natural water balance no longer affected by mining activities involves lake formation in remaining pits. In an area of 2120 km² approximately 45 lakes with a total volume of 4200 million m³ will be established. Some of these lakes will be among the largest lakes in Germany. Figure 1 depicts the geographical position of east German coal mining areas and a more detailed plan of the area south of Leipzig. The indicated lakes evolved in the former mines. Lake Cospuden was chosen as test site for the presented study.

Until now, in the region south of Leipzig about 25 % of the total volume of remaining pits has been filled by groundwater and surface water used to promote flooding. Single lake formation occurs within a period of years. But only after decades a steady state groundwater flow system will be re-established. Lake Cospuden formation started in 1992. In 2000 a final lake volume of approximately 100 million m³ will be reached. Figure 2 shows lake Cospuden in September 1995.

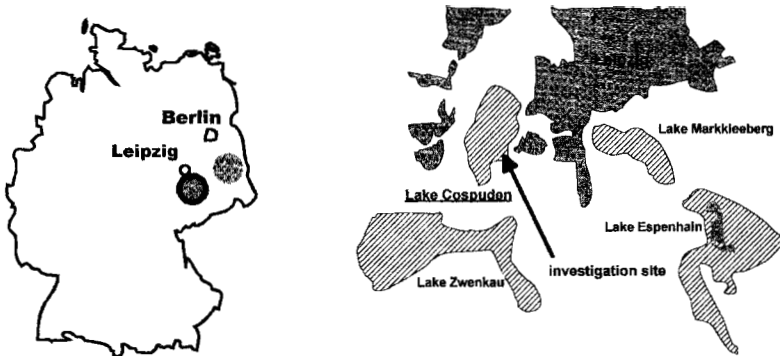


Figure 1: East German lignite mining areas and geographical position of the investigation site, Lake Cospuden.

In the former strip mine, 30 to 40 m thick layered pyrite containing tertiary and quaternary sediments (compare cross section in figure 3) were moved to a dump. Aeration promoted pyrite oxidation in the former groundwater zone and in overburden dumps. Originally, 1 to 8 % pyrite content of aquifer material produced acidity in aerated sediments. It is stored in pore water, secondary formed minerals and at ion exchange complexes. Serious water quality problems arise from influx of those pyrite oxidation products to the evolving lake. Acidity is either eluted from the sediments by groundwater and seepage water and transported to the lake or enters the lake water body by sediment influx due to erosion processes. The aim of the presented study is to quantify erosion induced matter influx, which is essential for predictive modelling of lake water quality.



Figure 2: Lake Cospuden (September 1995).

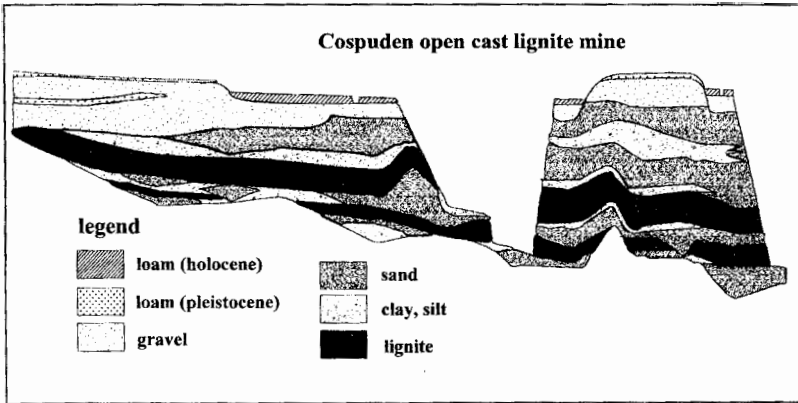


Figure 3: Cross section (W-E) through tertiary sediments adjacent to lake Cospuden.

WATER QUALITY PREDICTION MODEL

For prediction of lake water quality during flooding a coupled model is used. Water and mass balances are calculated for a discrete time step. Those water and mass fluxes are used as input parameters for subsequent geochemical equilibrium modelling of matter transformation processes in the mixed lake water body.

Figure 4 shows a scheme of water and sediment fluxes into the evolving lake.

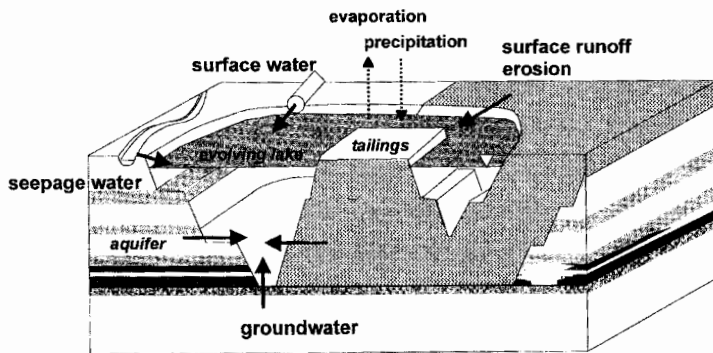


Figure 4: Water and sediment influxes during flooding.

Portions of ground- and surface water inflow to the lake depend on applied flooding techniques and are substantial for lake water quality formation. When groundwater pumping stops, flooding starts due to groundwater inflow, mostly as acidic mine drainage. Surface water of a distinct buffering capacity is used for artificial filling to increase lake water quality. Besides the influence of water and dissolved matter influxes, embankment sediment erosion

has a pronounced effect on water quality. It is supplying either acid mobilizing or acid binding substrates.

Groundwater influx is obtained from results of 3-D-groundwater modelling (PC GEOFIM) and groundwater monitoring data. According to geological properties, water-bearing horizons are divided in 15 sub-domains. Calculated water fluxes are combined with appropriate monitoring data at gauge wells. Monitoring data of surface water quality and quantity are used for calculation of surface water influx. Erosion induced sediment influx is obtained from modelling of sediment transport and from determination of chemical sediment properties as shown below.

The calculated mass influxes are used as input parameters for the geochemical equilibrium model (MINTEQ2A). Lake water quality is modelled as a result of mixing input mass fluxes and lake water. Speciation of solutes is obtained from 0-dimensional calculation of homogenous (e.g complex formation and redox reactions) and heterogenous reactions (e.g. exchange of gases and precipitation of solids). The applied model concept is shown in figure 5.

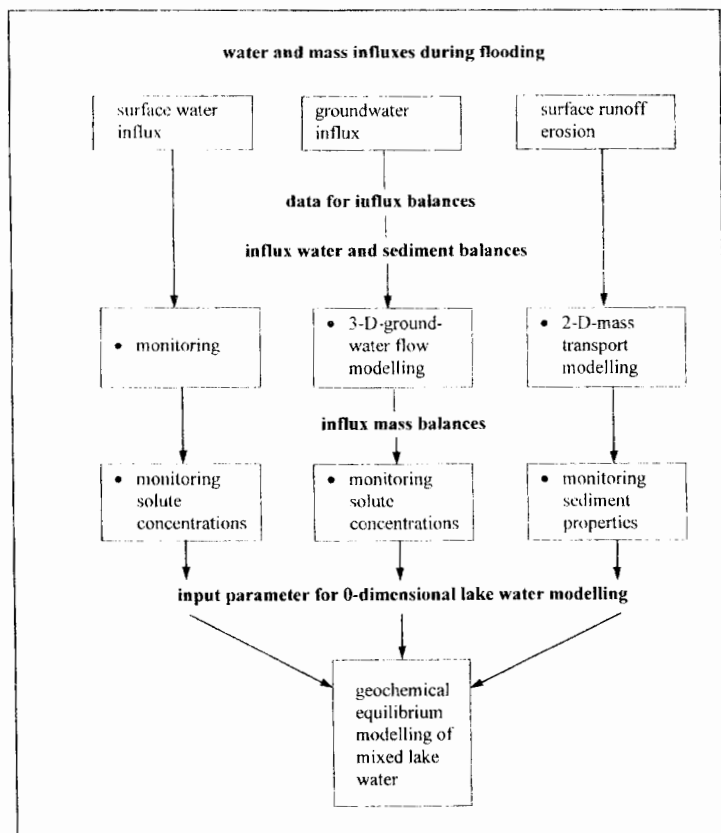


Figure 5: Concept of model based lake water quality prediction.

EROSION INDUCED SEDIMENT INFLUX

Lake Cospuden embankments exhibit distinct water erosion patterns. Surface runoff caused enormous rill and gully formation as shown in figure 6. At banks of 50 to 150 m length, rill profiles of several square centimeters up to some square meters were measured. According to observed erosion patterns, the artificially shaped lake embankment was subdivided and 3 representative dump slopes were chosen as test sites. Whereas gullies of up to 1 m width and 2 m depth dominated at dump slope 1, regular developed rills of 15 to 20 cm width and 10 to 20 cm depth occurred at dump slope 2 and 3. Common features are lack of natural soil formation and vegetation. All sampled embankment sediments are classified as loamy sand.



Figure 6: Water erosion patterns at lake embankment (dump slope 1 and dump slope 2) consisting of overburden materials.

To quantify sediment transport, both soil erosion modelling and field measurements were carried out. Available water erosion models (RUSLE, EROSION 2D, PEPP) were tested with respect to their suitability for these overburden materials. Common soil erosion models developed for agricultural soils base on empirical correlations (e.g. Universal Soil Loss Equation) or are process orientated.

Empirical models like RUSLE (Revised Universal Soil Loss Equation; RENARD et al.) compute an average annual erosion simply by multiplication of parameters representing effects of precipitation and runoff, soil properties, slope length and steepness, cropping management and conversation practices. In addition, determination of all parameters is based on empirical relationships. This implies a limited suitability to adapt these models to conditions very different from those of model calibration.

Process orientated models use physically based erosion and infiltration modules. They are able to compute precipitation event based erosion rates. Whereas erosion in EROSION 2D (SCHMIDT et al.,1996) is treated as interrill erosion, PEPP (process based erosion prediction model, SCHRAMM et al., 1996) integrates a rill erosion module. Input parameters are slope, climate and soil data. Beside slope geometry, duration of precipitation and precipitation intensity, standard soil parameters like grain size distribution, organic matter content, bulk density, initial water content and degree of soil covering are used. Furthermore, the tested models EROSION 2D and PEPP require two soil specific input parameters, roughness and erodibility. They reflect soil sustainability to erosion and are determined by special designed irrigation experiments. These experiments were carried out for the most common soils in saxony by MICHAEL et al. (1996). Results are compiled in a parameter catalogue (appendix to EROSION 2D). Until now, no parameters exist for overburden materials.

Table 1: Slope and soil properties of representative embankment dump slopes.

| | dump slope 1 | dump slope 2 | dump slope 3 |
|-------------------------|--|--------------------------|--------------------------|
| slope length | 150 m | 70 m | 56 m |
| average inclination | 13 % | 19 % | 27 % |
| bulk density | 1.57 g / cm ³ | 1.53 g / cm ³ | 1.53 g / cm ³ |
| organic matter content | 2.9 % C | 1.8 % C | 1.8 % C |
| grain size distribution | | | |
| gravel | 12.6 % | 0.2 % | 13.1 % |
| sand | 67.2 % | 79.2 % | 68.1 % |
| silt | 11.1 % | 10.5 % | 9.7 % |
| clay | 9.1 % | 10.1 % | 9.1 % |
| soil surface | gravel and stone covered | silted-up | gravel and stone covered |
| erosion patterns | gullies, width up to 1 m, depth up to 2m | regular developed rills | regular developed rills |

Calculations with RUSLE, EROSION 2D and PEPP were performed for the chosen representative dump slopes. Field measurement results of slope geometry and laboratory analysis of sediment properties are listed in table 1. Precipitation data obtained by a weather station next to the investigation site were converted into rainfall files by selecting precipitation events with intensities greater than 0.1 mm / min. The annual erosion was calculated as a sum of single event erosion. Roughness and erodibility were estimated from the parameter catalogue.

Table 2 contains annual erosion rates as calculated by the applied models. The values in the last line of the table result from field measurements. Rills and gullies were mapped and the annual erosion was calculated from rillvolumes. The embankment age at the representative hillslopes was about one year.

Table 2: Calculated annual erosion of representative embankment dump slopes.

| | dump slope 1 | dump slope 2 | dump slope 3 |
|--------------------|----------------|----------------|----------------|
| RUSLE | 213 t / ha · a | 191 t / ha · a | 164 t / ha · a |
| EROSION 2D | 339 t / ha · a | 244 t / ha · a | 169 t / ha · a |
| PEPP | no calculation | no calculation | 240 t / ha · a |
| field measurements | 950 t / ha · a | 303 t / ha · a | 373 t / ha · a |

RUSLE calculated the smallest erosion, reflecting limited possibilities for parameter adaption to non-agricultural conditions. Parameter determination was performed using empirical equations from the manual. The rainfall factor was calculated with a modified equation for german conditions (SCHWERTMANN et al., 1987). RUSLE-calculated erosion rates at the three dump slopes do not differ much. EROSION 2D found considerably higher erosion for dump slope 1 than for the other sites. Erosion rates exceed RUSLE-computed ones, but still do not reflect rill and gully formation. Calculations with PEPP were performed for dump slope 3 only. The erosion rate was calculated about 50% larger than the results of RUSLE and EROSION 2D. Since non of the models is able to compute gully erosion, differences between model results and field measurements are highest for dump slope 1. Reasonable estimations of erosion rates at hillslopes 2 and 3 were calculated by EROSION 2D and PEPP. Determination of the soil specific parameter erodibility will improve calculation results and is required for prediction of embankment erosion.

For further calculations of erosion induced mass fluxes, an average erosion rate of 300 tons per hectare and year was used for embankment parts represented by dump slopes 2 and 3. The annual erosion rate for areas represented by dump slope 1 was set to 900 tons per hectare.

EROSION INDUCED MASS INFLUX

Elutriation of sediments within the evolving lake causes acidity or alkalinity influx, induced by chemical reactions with hydrogen ion release or binding respectively. Potential acidity is stored in sediments as hydrogen, iron and aluminium ions in pore water, in secondary formed minerals and in exchange complexes. Potential alkalinity results from

presence of buffer systems (calcite, silica, exchange complex, aluminium and iron buffer as well as sulfate ions).

Table 3: Acidity producing and acidity binding reactions in relation to chemical sediment and elutant properties.

| sediment properties | elutant properties | chemical reaction |
|---|---|---|
| <ul style="list-style-type: none"> pore water composition H⁺ HSO₄⁻ Fe²⁺, Fe³⁺ Al³⁺ CO₃²⁻ HCO₃⁻ SO₄²⁻ | <ul style="list-style-type: none"> pH, ion concentration - pH > 3 pH > 3 pH > 4 pH > 8.2 pH = 4.3 to 8.2 pH < 3 | <ul style="list-style-type: none"> H⁺ release - HSO₄⁻ → H⁺ + SO₄²⁻ Fe³⁺ + H₂O → FeOH²⁺ + H⁺ FeOH²⁺ + H₂O → Fe(OH)₂⁺ + H⁺ Fe(OH)₂⁺ + H₂O → Fe(OH)₃ + H⁺ Al³⁺ + H₂O → AlOH²⁺ + H⁺ AlOH²⁺ + H₂O → Al(OH)₂⁺ + H⁺ Al(OH)₂⁺ + H₂O → Al(OH)₃ + H⁺ H⁺ binding H⁺ + CO₃²⁻ → HCO₃⁻ H⁺ + HCO₃⁻ → H₂CO₃ H⁺ + SO₄²⁻ → HSO₄⁻ |
| <ul style="list-style-type: none"> composition of ion exchange complex e.g. H-R, 1/3 Al-R e.g. 1/2 Ca - R | <ul style="list-style-type: none"> Ca²⁺, Mg²⁺, Na⁺ H⁺, Fe²⁺, Fe³⁺, Al³⁺ | <ul style="list-style-type: none"> H⁺ release e.g. H - R + 1/2 Ca²⁺ → 1/2 Ca - R + H⁺ H⁺ binding e.g. 1/2 Ca - R + H⁺ → H - R + 1/2 Ca²⁺ |
| <ul style="list-style-type: none"> presence of minerals Fe - (OH) - sulfates Al - (OH) - sulfates calcite gibbsite ferrihydrite | <ul style="list-style-type: none"> pH > 3, Fe²⁺, Fe³⁺ pH > 4, Al³⁺ pH = 6.2 to 8.2 pH = 3 to 4 pH < 3 | <ul style="list-style-type: none"> H⁺ release solution and Fe hydrolysis solution and Al hydrolysis H⁺ binding CaCO₃ + H⁺ → Ca²⁺ + HCO₃⁻ e.g. Al(OH)₃ + H⁺ → Al(OH)₂⁺ + H₂O e.g. Fe(OH)₃ + H⁺ → Fe(OH)₂⁺ + H₂O |

Both sediment and eluent properties determine actual H^+ release or binding as a result of sediment elutriation. Acidity producing and acidity binding reactions are indicated and related to chemical sediment and eluent properties in table 3.

Determination of chemical sediment parameters

Whereas standard chemical water analysis yields eluent properties, sediment properties can be determined using characteristic parameters, as:

- pore water composition,
- composition of ion exchange complexes,
- mineralogical composition.

The following methods were applied at the laboratory to determine those characteristic sediment parameters.

Pore water composition:

Preparation of a saturated paste by adding distilled water to 400 grams of air dried sediment while stirring with a spatula. After one night the equilibrium solution was extracted by membran filtration using compressed air and analyzed for its ion composition.

Composition of ion exchange complexes:

Washing of 10 grams air dried sediment with distilled water. After decantation repeated extraction with 1 N $La(NO_3)_3$ solution. The supernatant liquid after centrifugation represents the ion exchange complex composition.

Mineralogical composition:

X- ray diffractometry and calculation of saturation indices of certain minerals by chemical equilibrium modelling (PHREEQC) of pore water revealed mineralogical sediment composition.

Table 4: Pore water composition.

| | <u>dump slope 1</u> | <u>dump slope 2</u> | <u>dump slope 3</u> |
|--|---------------------|---------------------|---------------------|
| pH | 2.51 | 1.82 | 5.28 |
| total ion concentration (mmol (eq) / 100 g dry sediment) | | | |
| c_{total} | 0.98 | 2.05 | 0.41 |
| pore water composition (mmol (eq) %) | | | |
| H | 1.4 | 10.0 | - |
| Fe^{II} | < 1 | < 1 | - |
| Fe^{III} | 6.1 | 24.0 | - |
| Al | 61.7 | 20.5 | - |
| Mn | 1.9 | < 1 | - |
| Ca | 10.2 | 17.0 | 36.5 |
| Mg | 16.8 | 28.0 | 59.6 |
| Na | 1.1 | < 1 | 2.4 |
| K | < 1 | < 1 | 1.5 |

Results of pore water and ion exchange complex composition are summarized for the chosen test sites (dump slope 1, 2 and 3) in table 4 and 5.

Table 5: Ion exchange complex composition.

| | <u>dump slope 1</u> | <u>dump slope 2</u> | <u>dump slope 3</u> |
|--|---------------------|---------------------|---------------------|
| ion exchange capacity (mmol (eq) / 100 g dry sediment) | | | |
| C_{total} | 1.9 | 3.3 | 6.3 |
| ion exchanger composition (mmol (eq) %) | | | |
| H | 40.5 | 33.7 | - |
| Fe ^{II} | 10.8 | 2.2 | - |
| Fe ^{III} | < 1 | < 1 | - |
| Al | 48.0 | 43.5 | 4.5 |
| Mn | < 1 | < 1 | - |
| Ca | < 1 | 19.2 | 84.1 |
| Mg | < 1 | < 1 | 10.9 |
| Na | < 1 | < 1 | < 1 |
| K | < 1 | < 1 | < 1 |

X-ray diffractometry revealed quartz as major mineral component. Sediments of dump slopes 1 and 2 further contain more than 5 % gypsum and muscovite. Chemical equilibrium modelling of pore water of dump slopes 1 and 2 indicated oversaturation of H-jarosite, K-jarosite and gypsum. Furthermore, pore water of dump slope 1 was oversaturated with Jurbanite. Quantification of those minerals by mineralogical methods was not successful because of detection limits. It should be mentioned that pure crystal formations originating from dump slope 2 revealed gypsum with inclusions of ferrihydroxide and pyrite. Therefore, besides jarosite and jurbanite gypsum may be a potential H⁺ source by containing ferrihydroxides like FeOOH, which are able to fully hydrolyze at elutriation. Until now, it was not possible to determine those minerals at low contents in soil samples.

Actual acidity or alkalinity influx

Lake water quality determines actual H⁺ release or H⁺ binding caused by sediment elutriation. At the time of the study, total ion concentration equaled 28 mmol (eq) / l. Major cations were calcium (61 mol (eq) %) and magnesium (28 mol (eq) %). Major anions were sulfate (94 mol (eq) %) and chloride (6 mol (eq) %) and pH = 5.1.

No H⁺ binding reactions from pore water components and minerals can be expected (see table 3). H⁺ buffering of dump slope 3 sediments relies on ion exchange. 95.5 % of total cation exchange capacity (6.0 mmol (eq) / 100 g dry sediment) are available for binding of hydrogen, iron and aluminium ions.

Sediments of dump slopes 1 and 2 release H⁺ from pore waters, ion exchange complexes and minerals. Pore water of dump slope 1 contains 0.68 mmol (eq) / 100 g dry sediment H⁺ equivalents. Pore water of dump slope 2 contains 1.1 mmol (eq) / 100 g dry sediment H⁺ equivalents. Exchange complex bound H⁺ equivalents at dump slope 1 and 2 amount 1.9 mmol (eq) / 100 g dry sediment and 2.6 mmol (eq) / 100 g dry sediment respectively. Actual release or

binding of H^+ equivalents from dump slope sediments are listed in table 6. Since contribution of minerals could not be taken into account, data for acidity release are minimum values.

Table 6: Actual release or binding of H^+ equivalents from dump slope sediments.

| test site | actual release or binding of H^+ equivalents (mmol (eq) /100 g dry sediment) | | | |
|--------------|--|------------|-----------------------------|------------------------|
| | exchange complex | pore water | minerals | sum (without minerals) |
| dump slope 1 | 1.9 | 0.68 | gypsum, jarosite, jurbanite | 2.58 H^+ release |
| dump slope 2 | 2.6 | 1.1 | gypsum, jarosite | 3.7 H^+ release |
| dump slope 3 | 6.0 | - | - | 6.0 H^+ binding |

Formulation of erosion induced mass fluxes to the evolving lake requires combination of sediment transport and sediment properties (see figure 5). The test sites are related to embankment parts they represent. Total acidity or alkalinity fluxes result from combination of total sediment fluxes and actual release or binding of H^+ equivalents as shown in table 7.

Table 7: Erosion induced mass influxes.

| | dump slope 1 | dump slope 2 | dump slope 3 |
|--|----------------------|--------------|--------------|
| represented embankment part (ha) | 10 | 26 | 2 |
| erosion rate (t / ha · a) | 900 | 300 | 300 |
| sediment transport (t / a) | 9 000 | 7 800 | 600 |
| actual release or binding of H^+ equivalents (mmol (eq) /100 g dry sediment) | 2.58 | 3.7 | 6.0 |
| erosion induced mass flux (mol (eq) / a) | 232 200 | 288 600 | 36 000 |
| sum acidity influx | 520 800 mol (eq) / a | | |
| sum alkalinity influx | 36 000 mol (eq) / a | | |
| net acidity influx | 484 800 mol (eq) / a | | |

Figure 7 illustrates the effect of an annual acidity influx of 10 million mol (eq) to the pH development of the lake water. In June 1996 surface water of poor quality was used for flooding. Prolonged acidity input was modeled to cause drop in pH on values around 4 (prediction 2). Measured pH values are indicated as well as a prediction without surface water acidity influx (prediction 1).

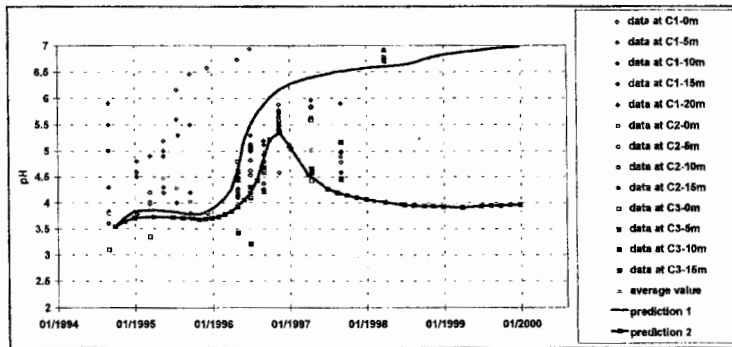


Figure 7: Model prediction of pH development in Lake Cosputen.

The erosion induced net acidity input of approximate 0.5 million mol (eq) / a calculated above is 20 times smaller than the modeled one. Compared to groundwater transported acidity, it is still of a considerable dimension.

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