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Abatement of acidification in mining lakes in Germany

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Abstract

Open-cast lignite mining has exposed formerly anaerobic layers to the air. Pyrite and marcasite are oxidized to sulfuric acid and iron sulfate, which later hydrolyses to iron hydroxide. When the pit is refilled with groundwater, its pH is 2–3. These geogenically acidified mining lakes are brown, with high iron content. Biological conditions differ greatly from those in most natural lakes and resemble those in sulfur-acidic lakes impaired by acid rain. The usability of these fish-free lakes is also restricted for recreational purposes.

Neutralization with lime is expensive. The iron-buffered water has a high base buffering capacity stabilizing at pH 2–4. The complex of potential measures for neutralization comprises: the revegetation of the waste heaps, the hydrological regime and coupled transport processes, the selection of suitable filling water and in-lake technologies. Desulfurication takes place with sulfidic binding of iron and natural neutralization under locally anaerobic conditions in the reclaimed overburden heaps, in the deep water of mining lakes, in macrophyte stands and in the sediments.

Anaerobic conditions should be established within the lakes during stratification periods: in the hypolimnion of eutrophic or the monimolimnion of meromictic lakes. Acidic lakes often are not sufficiently productive to achieve deep-water oxygen depletion. Measures to increase the stability of stratification may be necessary, for example by installation of barriers to establish an environment suitable for biological neutralization. Carbon sources for microbial desulfurication may be added artificially or autochthonically produced by primary producers. Therefore, a controlled eutrophication may be useful in the initial filling stages of acidic mining lakes.

Key words

acid mining lakes, biological neutralization, pyrite oxidation.

INTRODUCTION

German unification caused a dramatic decrease in national energy demand and a comparable decrease in brown coal mining. Lignite production in East Germany in 1989 amounted to more than 300 million tons but in 1993 to only around 100 million tons. As the abandoned mines fill with water, many lakes form within a few years. Depending on the kind and origin of the water, the shape (especially the depth) of the basin, its position in the groundwater field and the nature of the geological substratum, the following quality problems have to be taken into account:

(1) Geogenic acidification and coupled metal loading because of oxidation of pyrite and marcasite (FeS_2) during mining operations. Acidification is the most severe problem, especially in the Lusatian lignite region. The report focuses on this problem.

(2) Salinization is important when mining is in close proximity to tertiary coal and Zechstein salt layers. Mining lake Merseburg-East has deep waters with three-fold oceanic concentrations of sodium chloride.

(3) Contamination with hazardous substances may occur when the open-air mine had been used as a dumping site for industrial and communal wastes, when surface water used for filling is polluted, and when geogenically acidified groundwater dissolves heavy metals on its way through the overburden heaps.

(4) Eutrophication is the main problem when the pit becomes filled with nutrient-rich surface waters.

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Algal mass development and turbid waters diminish usability for most purposes. It will be shown that in the case of acidification a controlled addition of phosphate may shorten the succession from acidic to neutral lakes.

(5) Saprobization, that is loading with allochthonous

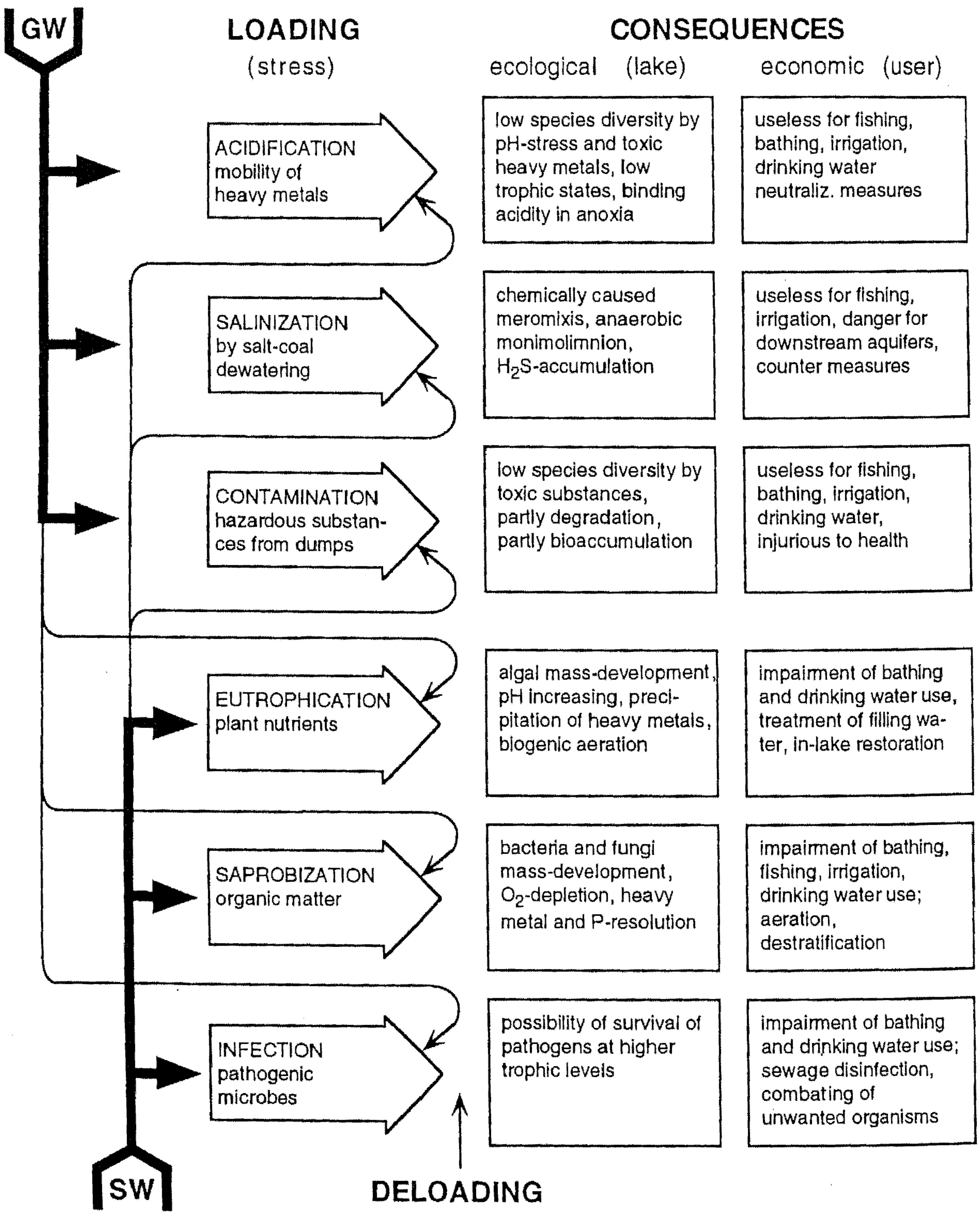

Fig. 1. Types of loading and consequences in mining lakes filled by ground (GW) or surface water (SW).

and autochthonous degradable organic matter, is of decreasing importance. The water quality of the rivers used for filling has improved because of higher standards of sewage treatment. Waste is no longer dumped into mining holes when coal processing industries have been closed.

(6) Infection with pathogens is related to the pollution of filling waters and some kinds of water utilization. Duck feeding on a mining lake precludes bathing because of salmonella infestations of the birds.

Mixing of ground and surface waters allows considerable control of water quality. To abate eutrophication, a high proportion of groundwater is needed. Due to the content of cations such as Fe^{3+} and Al^{3+}, phosphorus will be bound and transported to the sediment. In a similar way contaminants and bacteria may be adsorbed and precipitated. Conversely, with surface water, salty waters may be diluted and acidic waters neutralized by carbonate hardness (Fig. 1).

THE EXTREMELY ACIDIC ENVIRONMENT

Chemical characterization

Geogenic acidification is caused by pyrite and marcasite oxidation. These minerals are commonly associated with coal and all sulfide ores. In this paper the term pyrite is used for simplicity. During the mining process, the geological strata are exposed to the atmosphere and pyrite reacts with oxygen and water to release acidity, iron and sulfate. A secondary effect of the low pH is the release of toxic heavy metals.

The pH values of the investigated mining lakes are distributed around two maxima. Obviously, water chemistry is governed by three different buffering systems: the circumneutral bicarbonate buffer and the acidic aluminium and iron buffer. Alkalinization does not change the pH unless the base-binding capacity is saturated. When this occurs, the pH shifts from one to the other buffering system. The changes are similar to titration curves. With acid rain, the aluminium buffer prevails in a region of pH 3.6–4.2 but in mining lakes the iron buffer is absolutely dominant at pH values <3.8 (Ulrich 1981).

In the Lusatian brown coal region, most lakes were found to be at about pH 2–4, with others between 6 and 8. One exception had a pH of about 5. In this case (the Felix-See), the concentration of aluminium was ten times higher than that of iron. The conditions in this special case resemble those in lakes influenced by acid rain (Fig. 2). From an ecological point of view, the high concentrations of metals in the acidic range are of interest (Fig. 3). The relations between pH and metal content also reveal that an increase of pH by neutralizing measures might be the most promising way to overcome toxic concentrations.

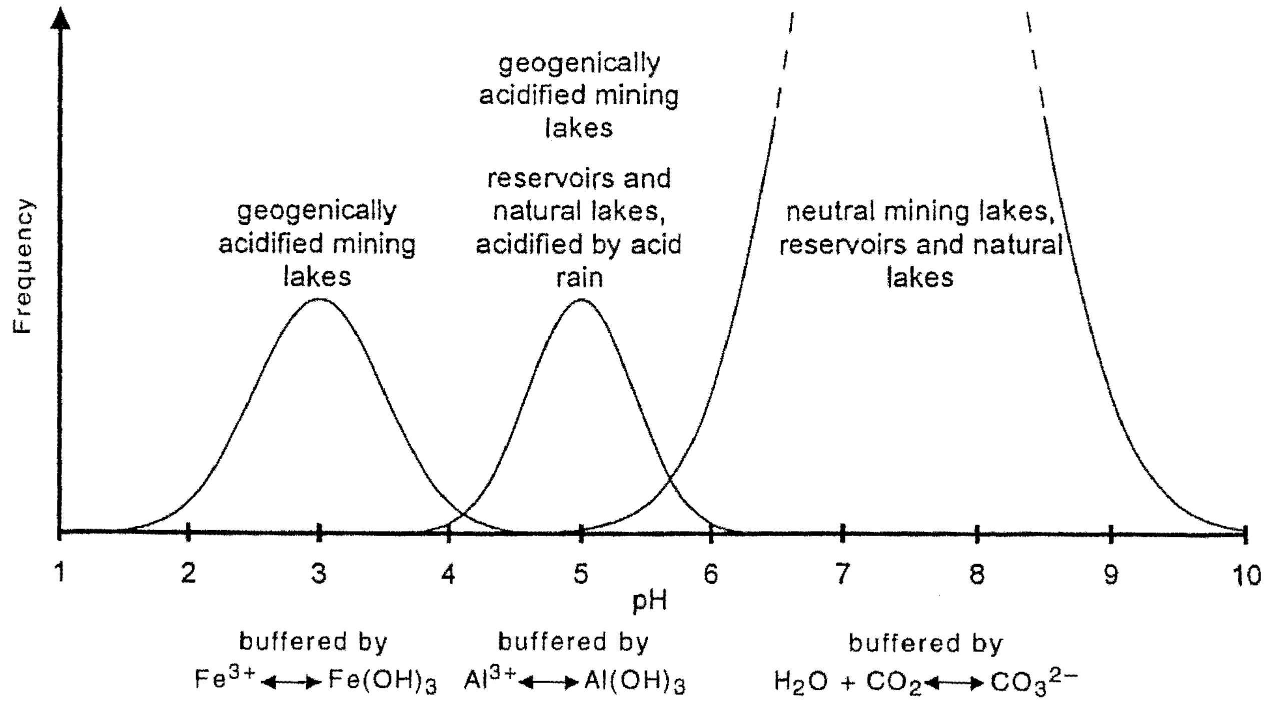

Fig. 2. Generalized frequency distribution of lakes with different acidity in Germany.

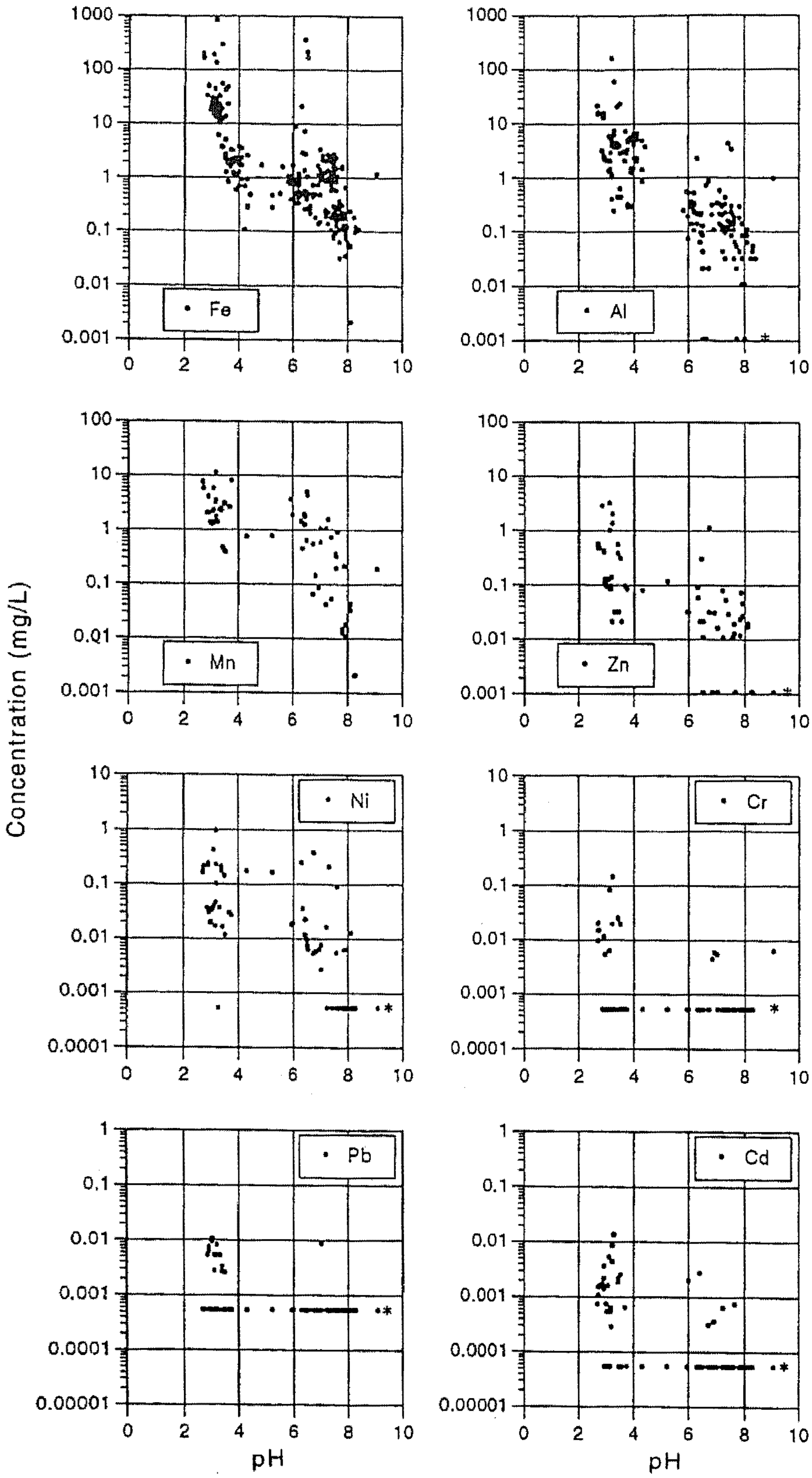

Fig. 3. Metals in the investigated mining lakes show high solubility under acidic conditions. The plotted values are total concentrations (*lower than detectability; from Klapper and Schultze 1995).

Habitat for plants and animals

From the immense literature available on acid rain, it seems that at a pH <4 hardly any life can be expected. Indeed, the youngest stages of geogenically acidified mining lakes with a pH of 2 to 3 are abiotic at first glance. The dark red colour originates from dissolved iron. Nevertheless, the extreme habitat has been shown to be colonized by specialists, some in great abundance because of the lack of competition.

Organisms able to survive in the extremely acidic environment possess mechanisms for detoxification by ion exchange, by oxidation and flocculation of heavy metals, by excretion of chelates, and by transforming the metals into a dissolved form with lower toxicity. Some algae respond with dormancy or concentrate toxicants in terminal cells which they later discard (Lüderitz 1988).

Compared to natural lakes, acid mine lakes generally have low primary productivity. Because of the lack of bicarbonate in acidic lakes, algal groups of the *Scenedesmus* photosynthetic type are absent. The first phytoplankton to appear are from the same groups to be found in bog waters: chloromonads, cryptomonads, dinoflagellates, and so on.

Important processes occur at the level of the pico- and bacterioplankton. The total algal biomass is low, and this criterion alone would accord with a classification as 'oligotrophic' (Nixdorf *et al.* 1995). This low algal production is unexpected, given the relatively high phosphorus supply. Obviously factors other than nutrients limit productivity. Moreover, the seepage waters from overburden heaps with a pH <2 are not really abiotic. At least bacteria occur. The pyrite oxidation itself is described as a chemo-autotrophic microbiological process performed by some species of *Thiobacillus*. Besides algal development, heterotrophic degration is strongly inhibited in the acidic environment. Leaves which have fallen from surrounding trees into the acidic lake remain undegraded for years on the lake bottom. Trees submerged during the filling process persist for decades as undegraded 'acid-preserved'. The low respiration rates and low solubility of CO_2 in acidic waters seem to be the reasons for carbon limitation of bioproductivity (Ohle 1981; Schindler 1994). Macrophytes are lacking only at a very early filling stage.

Upwelling groundwater rich in sulfuric acid, iron and sulphate, a changing shoreline, and an unstable subsurface during the first quick filling phase precludes macrophyte growth for hydrochemical and morphological reasons. Soon after the hydrological regime has stabilized, however, the typical pioneer plant *Juncus bulbosus* occurs. In this 'one-species biocenosis' according to biocenotic laws, *J. bulbosus* is very frequent in the littoral zone and also on the lake bottom if the water is clear enough. Photosynthesis by CO_2 consumption leads to iron flocculation on the stems. Occasionally, the ochre precipitation pulls whole plants down to the bottom. This species, which is mainly vegetatively reproducing, sprouts a second time during summer and appears grass-green in colour.

Pietsch (1993) distinguished some typical communities characterized by the dominant species besides *Juncus* stands: *Potamogeton natans*, *Utricularia minor*, *Sphagnum cuspidatum*, *S. obesum*, *S. inundatum* and the fern *Pilulifera globulifera*. He described a further transition to a stage with neutral water containing bicarbonate and high species diversity. Of the animals, those groups which cannot exist in an acidic environment are those which need calcium carbonate for their skeletons or shells, such as fish, amphibians, snails, mussels and higher crustaceans. The first filling stages in acid lakes are also without zooplankton. The rotifers (especially *Brachyonus urceolaris*) inhabit lakes with pH <3, as does *Chydorus sphaericus*. *Cyclops* was found at pH 3.5 and higher. *Daphnia* occurred only in circumneutral lakes. Whether the acidity excludes them or this is caused by the frequency of invertebrate predators like *Corixa* is not clear (Tittel pers. comm.).

DECISION CRITERIA FOR ACTIVE WATER QUALITY MANAGEMENT

Large mining lakes, about 10 km² or more in area, should be made usable as soon as possible. Many small and shallow lakes have remained acidic for decades, but some deep or organically loaded lakes are neutralized by the internal production of alkalinity. The general question of whether to counteract acidity or not is therefore a question of water policy. The following issues are relevant:

(1) Society at the local and governmental level has an urgent interest in creating jobs. Now that nearly all the lignite industries have been closed, a large proportion of the population in the region is unemployed. Therefore, another form of employment is called for in the post-mining landscape, including those areas covered by water such as the mining lakes.

(2) Water-orientated or water-bound recreation is preferred in most cases by the local citizens. Decision makers and planners, on the other hand, try to concentrate these activities in selected places to meet the

high quality requirements. To be economically efficient, recreation areas have to be well-equipped to attract people after work, on weekends and on holidays. The recreational pressure depends on population density, the distance to large cities, traffic connections, landscape beauty and cultural activities.

(3) The quality criteria for bathing waters are fixed by law. Bathing water has to have a pH of between 6 and 8.5 according to the recommendations of the European Union. Exceptions are allowed under unusual geograpical conditions. Nevertheless, it remains unclear whether acidic water actually endangers health. There are medical spas with acidic moor waters. Bacteriological criteria, commonly used for hygienic control, are generally more favourable in acidic than in neutral waters. People who are not too sensitive tolerate the acidity without any harm to their health.

(4) For water-orientated recreation, one drawback is that acidic lakes are fish-free. All activity related to professional and sport fishing is impossible.

(5) Less dependent on water quality are extractive water uses. Quality standards can be met by water treatment. Acidic raw waters may be neutralized by liming, by addition of sodium hydroxide or by dolomite-filtration. This refers to industrial, irrigation and drinking waters.

(6) Some extreme habitats of acidified lakes should be maintained even though they are created by mining and are the result of human activities. The lakes excluded from active neutralization should include those with an especially high acidification potential. Limnological long-term investigations are needed to elucidate the natural succession process of neutralization as well as the taxonomy and adaptation of organisms in these ecosystems.

Table 1. Inhibitory measures against acidification during the mining process

(1)	Minimization of dewatering before mining in space and time
(2)	Deposition of sulfur-rich overburden in the deepest part of the mine
(3)	Inhibition of acidifying metabolism by bactericides as proposed by Kleinmann *et al.* (1981)
(4)	Refilling of dewatered subsurface as soon as possible
(5)	No elongated mining lakes in the direction of groundwater flow
(6)	End position of the mining should allow filling with groundwater from undisturbed rocks

POSSIBLE WAYS TO CONTROL ACID

Measures to avoid the formation and transportation of acid

At new mines, present knowledge allows pyrite oxidation to be avoided or decreased by taking into account certain rules and recommendations (Table 1).

Pyrite in the lignite and overburden are stable as long as the groundwater is anoxic. To re-establish similar conditions and minimize loading of mining lakes with sulfuric acid, drainage basin measures to be taken into account are those listed in Table 2.

Ecotechnologies for neutralization of acidic mining lakes

The liming of lakes has been a success story in Sweden. Since 1988, about 5000 surface waters have been

Table 2. Abatement of acidification by measures in the drainage basin

(1)	Neutralizing recultivation of the heaps with lime, dolomite, alkaline ashes etc.
(2)	Intensive farming with a high level of fertilization
(3)	Organic fertilizer and harvest residues consume oxygen below ground
(4)	Artificial N-fertilizer in nitrate form to avoid acidification because of nitrification of ammonia
(5)	High proportion of pasture and wetlands
(6)	Mixed forests with high proportion of deciduous trees and with humus rich topsoils
(7)	Long growth periods of trees
(8)	No clear-cutting
(9)	Surface sealing by fish ponds (limed before filling)

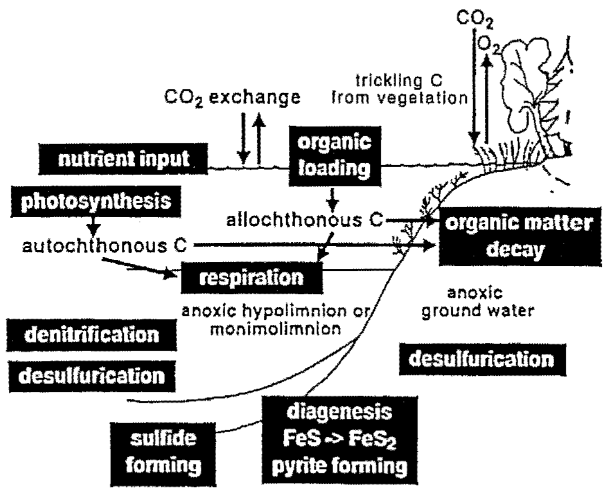

Fig. 4. Processes of alkalinization in a dimictic or meromictic stratified lake.

neutralized in a governmental programme (Olem 1991). These soft-water bodies, impaired by acid rain, are quite different from hard-water mining lakes. The latter are strongly buffered by iron at a far lower pH and demand about ten times higher alkalinization (Schultze & Geller in press). Therefore, the natural process of alkalinization by (denitrification and) desulfurication is at the centre of restoration research. In dimictic eutrophic or meromictic lakes, the anaerobic deep water was found to have been neutralized (Fig. 4 and Table 3). After desulfurication, the resulting neutralized water needs re-aeration to become a fish habitat.

Anoxic technologies are not easily applied at the scale of natural water bodies. However, they are commonly in operation in the fermentation industry and in anaerobic treatment of sewage and sludge. Closed tanks protect the medium against oxygen inputs. In nature, an oxygen deficit is the driving force for oxygen intake into the water. Therefore, it is especially difficult to eliminate the last traces and to enable the obligatory anaerobic desulfuricants to do the desired work of acid binding. The observed better denitrification and desulfurication in nature than in the laboratory is obviously due to variable conditions of oxygen and pH in the microhabitats of natural biofilms.

Table 3. Environmental preconditions for sulfate respiration within an ecotechnology

(1) Exclusion of dissolved (and nitrate-) oxygen
(2) Presence of degradable organic substrate
(3) Inert or degradable materials as substratum for periphyton or biofilm carriers
(4) High content of sulfate and iron
(5) Starting microhabitats with a pH>4 for dusulfurizing bacteria

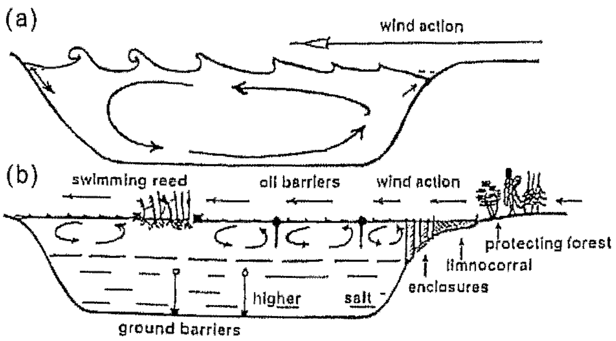

Fig. 5. Circulation and stability without (a) and with (b) barriers.

Whole deep ponds in California were maintained as anaerobic to enable them to become denitrified. Only those with a covered surface and with the addition of methanol as a carbon source achieved the intended denitrification (Brown 1971; Sword 1971; Jones 1974). The cheapest organic substrates are wastes such as liquid manure, pretreated domestic sewage, molasses and some types of industrial waste. In sulfate elimination experiments for drinking water purposes, whey, sucrose, sodium-lactate and ethanol were investigated. The latter performed best. In other substrates the desulfuricants were overgrown by acetogenic bacteria which lowered the pH by the addition of organic acids (Brettschneider & Pöpel 1992).

To make a whole lake anoxic is nearly impossible and also unwanted from an ecological point of view. As shown above, natural desulfurication operates in distinct parts of a lake, in anaerobic deep waters and in the sediments. But bioproductivity is particularly low in acidic lakes. Therefore, only a few mining lakes have anoxic deep waters. However, there are ways to change these conditions, for instance by stabilizing stratification in polymictic lakes. Internal barriers may be installed and wind action thereby decreased (Fig. 5).

A controlled reversal of lake acidification by treatment with phosfate fertilizer has also proven successful (Olofsson *et al.* 1988). It was noted than when nitrate is assimilated it generates base (consumes acid) according to the equation:

$$106\ CO_2 + 138\ H_2O + 16\ NO_3^- \rightarrow (CH_2O)_{106}(NH_3)_{16} + 16\ OH^- + 138\ O_2 \quad (1)$$

Base is also generated through the oxidative decomposition of organic material. If oxygen is the electron acceptor, there is no net generation of base, but if other inorganic oxidants such as nitrate, sulfate or iron oxhydroxides are involved, their dissimilative reduction associated with the decomposition of organic matter generates base. Moreover, dissimilative processes become increasingly significant as deep sediments become more anoxic (Davison *et al.* 1995). Whether it is possible to make better use of the neutralizing forces of the sediments or to increase them is a topic for further research.

Another example of an effective anaerobic ecotechnology was developed for heterotrophic nitrate dissimilation in the reservoir Zeulenroda (Thuringia). A steel cage, 20×60×1.50 m, filled with 13 000 straw packages and equipped with distribution pipes, was positioned on the hypolimnic bottom near the dam.

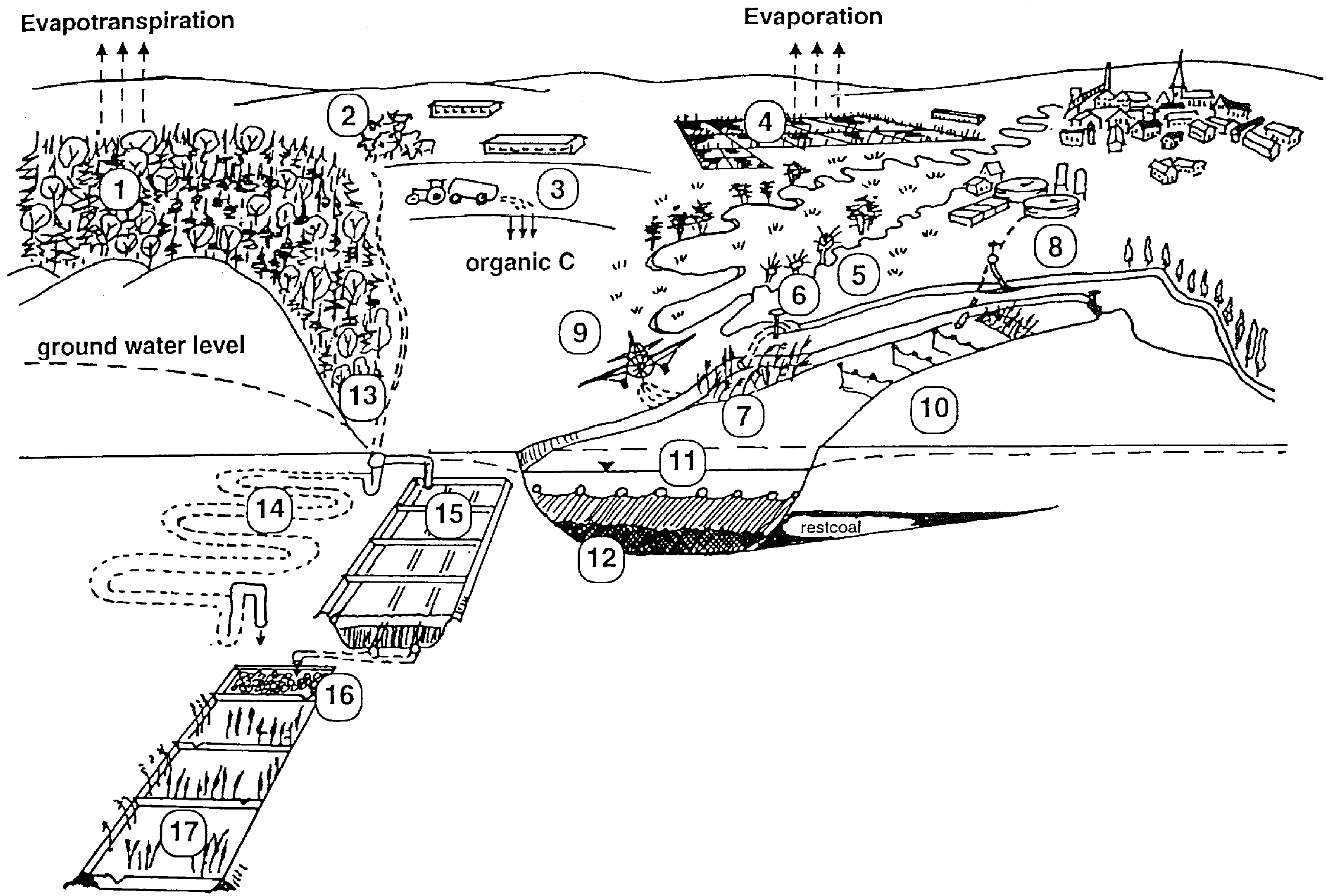

Fig. 6. Neutralization of geogenically acidified water bodies. (1) Recultivation of heaps with mixed forests; (2) grassland for pasturing; (3) organic fertilizing; (4) fish ponds; (5) wetlands; (6) addition of surface water; (7) bank bioplateau; (8) temporary supply of treated sewage (controlled eutrophication); (9) liming; (10,11) chicanes for stabilizing the stratification; (12) deposition of sulfides in the sediment; (13) drain-off of acidic outflow from the heaps; (14) anaerobic limestone drains; (15) infiltration basins with organic substrate overlying the drains (compost, manure, spaghetti, straw, sawdust ...); (16) limestone cascade for re-aeration; (17) constructed wetlands.

Nitrate-rich surface water was pumped through the straw reactor together with fatty acids as a carbon source. The dissolved oxygen was respired, after which the nitrate oxygen was utilized. Nitrogen escaped in molecular form. By this means, the hypolimnion was made anaerobic and nitrate-free within eight weeks. Along a stretch of 5 km of turbulent flow to the terminal reservoir, the water was re-aerated and proved to be a suitable source of raw water for drinking water treatment (Klapper 1991). The in-lake technologies are summarized in Table 4.

Table 4. *In situ* technologies for recovery of acidic mining lakes

(1) Whole water-body anaerobic:
    Only very small water-bodies with high organic load; covered surface
(2) Parts of the lake anaerobic:
    Use of deep water for desulfurication
    Stabilization of the stratification by dividing the surface with barriers
    Addition of organic substrate
    Addition of nutrients, bioproduction of organic matter by controlled eutrophication
    Use of sediments as traps for sulfur and metals
    Use of anaerobic mesocosms for research purposes: enclosures and limnocorrals
(3) In-lake installation of a through-flow reactor:
    Through-flow straw reactor in the deep water and addition of organic substrate
(4) Stimulation of macrophyte growth by preparation of the littoral zone
(5) Water exchange with HCO_3 containing surface waters. The pH rises because of dilution and buffering with bicarbonate

Table 5. *Ex situ* technologies for recovery of acidic mining lakes

(1) Anaerobic treatment
    Anoxic straw-filled trenches (Brown 1971)
    Anoxic limestone drains (Hedin & Watzlaf 1994)
    Closed subsoil reactor, filled with inert or degradable biofilm carrier, addition of C-substrate (Taberham pers. comm. 1995)
    Infiltration ponds with compost bottom and a limestone drainage-layer beneath the organic compost (Kepler & McCleary 1994)
    Mixed microbial mats for bioremediation of metals in acid tailings (Phillips & Bender 1995)
(2) Aerobic treatment
    Reedbed treatment plants for combined purification of tailings and sewage
    Concentrated alkaline recharge pools for acid seep abatement (Nawrot *et al.* 1994)
    Re-aeration channels with limestone overflow barriers
    Various forms of constructed wetlands (Hedin 1989; Dietz *et al.* 1994

In the Lusatian lignite region, especially in the larger mining lakes, the preferred mechanism is filling with surface water. Existing mining water treatment plants may be used to neutralize a part of the filling water through liming.

Ex-situ treatment of acidic waters

Alkalinity production or acidity binding is best accomplished in fully or partly anaerobic systems, while the precipitation of unwanted heavy metals works best in aerobic macrophyte systems after the pH has risen to neutrality. Many proposals comprise combinations of anaerobic with aerobic steps and sometimes with limestone as an inherent constituent of the different steps (Table 5 and Fig. 6).

From the USA and Great Britain, a number of ecotechnologies have been described to abate acid mine drainage (AMD). Canadian activities in this field are called mine environment neutral drainage (MEND). Some of these ideas may be considered suitable for the brown-coal mining regions of Germany. As soon as a self-sustaining water balance is re-established, the neutralizing service ecosystems should be installed at lake inlets or along connecting trenches between two mining lakes.

CONCLUSIONS

Acidification is the most severe water quality problem in lakes which have originated from lignite mining activities in Germany. Only a few smaller bodies of water should be kept acidic for conservation purposes and limnological research. Most should be rehabilitated to become more natural and less acidic environments. The abatement of acidification includes measures at sites of pyrite oxidation, activities to decrease groundwater and acidity transport, as well as *in situ* measures at the mining lakes.

Chemical neutralization is often impracticable because of the large amounts of alkali required and the treatment costs. Large mining basins should preferably be filled with surface water containing bicarbonate. A temporarily higher trophic level has to be tolerated. The most promising alternative for neutralization is the promotion of microbial processes of anaerobic acid binding by denitrification and especially by desulfurication. Aerobic treatment with macrophyte systems such as in constructed wetlands, is suitable for finishing the water treatment by flocculation of the heavy metals as hydroxides.

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