THE LIGNITE PIT LAKES OF LUSATIA, GERMANY-CHEMISTRY, ECOLOGY AND ACIDITY REMOVAL STRATEGIES[1]

A. Fyson[2] and B. Nixdorf

Abstract: Pit lakes in the Lusatia lignite mining district (Germany) are diverse in size and morphometries. Most are extremely acidic with pH ≤ 3 and high in iron concentrations. But a few, with pH 3.5 to 4.5, buffered by aluminum, have low iron concentrations. Productivity in most of the mining lakes is generally low and they support only simple food webs; a few of the acidic lakes, with high nutrient concentrations, are highly productive. State planners hope to develop the lake region for recreation. To that end, various methods are being investigated to remove acidity from the lake waters. The research described here employs controlled eutrophication to enhance element cycling and sediment-bound alkalinity generating processes.

Additional Key Words: aquatic chemistry, acid mine drainage, primary productivity, eutrophication

Introduction

In Germany, there are some 500 flooded, open-cast lignite mines (Tagebauseen). The morphometry, history and chemistry of these lakes are described in Nixdorf et al. (2001a). Many of these lakes are extremely acidic due to high concentrations of dissolved metals (mostly iron and aluminium) in addition to protons (low pH), and also have high sulphate concentrations. During mining operations, water was pumped from the open cast pits. Following closure, many pits filled with groundwater and acidified to pH around 3 through exposure and oxidation of marcasite and pyrite within the mining wastes and overburden

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[2] Andrew Fyson, Boojum Research Ltd. 139 Amelia Street, Toronto, Ontario, M4X1E6, Canada e-mail:afyson@boojumresearch.com (will present the paper) Nixdorf, B. Professor, Department of Water Conservation, Brandenburg Technical University, Cottbus, Seestrasse 45, D-15546 Bad Saarow, Germany e-mail:b.nixdorf@t-online.de

material. The location of the lignite mining regions is shown in Figure 1 and the size of some of these lakes together with those of natural German lakes in Figure 2.

Figure 1: Distribution of lignite mine areas in Germany (dark). The Lusatia region is marked as Lausitzer Revier

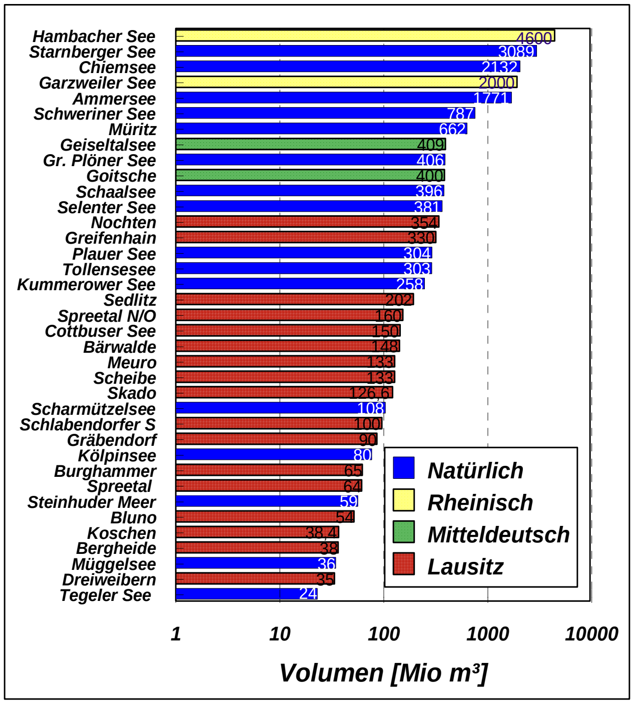

Figure 2. Volume of the largest German natural (Natürlich) and lignite mining lakes.

This emphasises the importance of these lakes in the German landscape. In the Lusatia region of north-eastern Germany, 168 Tagebauseen have been documented. Approximately half of these are extremely acidic (pH 2.4 – 3.4) (Nixdorf et al., 2001a). The chemistry of some of these waters is summarised in Table 1 and discussed in Uhlmann et al. (2004). Lake Grünewald from which water and sediment was used for the experiment described here is shown in Figure 3.

Figure 3. Lake Grünewald (RL 117) from which water and sediments were used in the microcosm experiments. Mean values of chemistry parameters (from Beulker and Nixdorf, 2004) are as follows: pH 3.0, Fe 14 mg L^{-1}, acidity ($KB_{8,3}$) 2 mmol L^{-1} P 3 µg L^{-1} NH_4^+-N 1.5-2.0 mg L^{-1} dissolved inorganic carbon (DIC) 0.2-0.3 mg L^{-1})

Table 1. Summary of some chemical parameters for 75 acidic mining lakes (pH 2.4-3.4) in the Lusatia region. Data are from Nixdorf et al. (2001a).

Parameter	Units	Concentration (mean)	Range
pH		2.9	2.4-3.4
Acidity ($KB_{4,3}$)	mmol L^{-1}	4.9	0.1-26.6
Fe	mg L^{-1}	95	0.2-800.0
Sulphate	mg L^{-1}	1448	460-4636
TOC	mg L^{-1}	3.4	0.8-10.9
Total-N	mg L^{-1}	3.5	0.9-5.3
Total-P	µg L^{-1}	14.3	4-26
chlorophyll a	µg L^{-1}	2	0.5-5.0

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Economically and environmentally acceptable methods are being sought to sustainably remove the acidity of these lakes to assist in their development, mainly for recreational use (Totsche and Steinberg, 2004; Uhlmann et al. 2004). Neutral, neutrient-rich river water has been diverted into some of the larger lakes, but there is insufficient river water to similarly reduce the acidity for all lakes and for many smaller and already filled lakes, this method is impractical. Another method is to enhance *in-situ* alkalinity generating processes through addition of nutrients/or wastes and/or the sustainable enhancement of productivity to feed such processes. This controlled eutrophication is the basis of the study reported here.

Although biodiversity in extremely acidic ecosystems is generally low (Wollmann and Deneke, 2004), productivity may be high in local situations where availability of nutrients, particularly phosphorus and carbon are high (Fyson and Ruecker, 1998; Nixdorf et al., 2004). Bacterial production in the acidic, Lusatian mining lakes is typically in the range found in eutrophic lakes in the region (Nixdorf et al., 2003). However, primary productivity in such ecosystems is generally low (Nixdorf et al., 2003; Beulker and Nixdorf, 2004) and likely limited either by the intrinsically low DIC concentrations at low pH values and extremely low P concentrations attributable to the propensity of this element to coprecipitate with iron(III) oxyhydroxides (Stumm and Morgen, 1996). Such limitations have been found for the Lusatia acidic mining lakes (Krumbeck et al., 1998). The productivity of lakes as reflected in chlorophyll concentrations is closely tied to phosphorus concentration (Fig. 4). If production of these ecosystems can be enhanced, the supply of carbon and nutrients to sediment-bound, alkalinity generating processes can also be increased. The continued enhanced cycling of nutrients and carbon may provide the basis of a sustained acidity removal system.

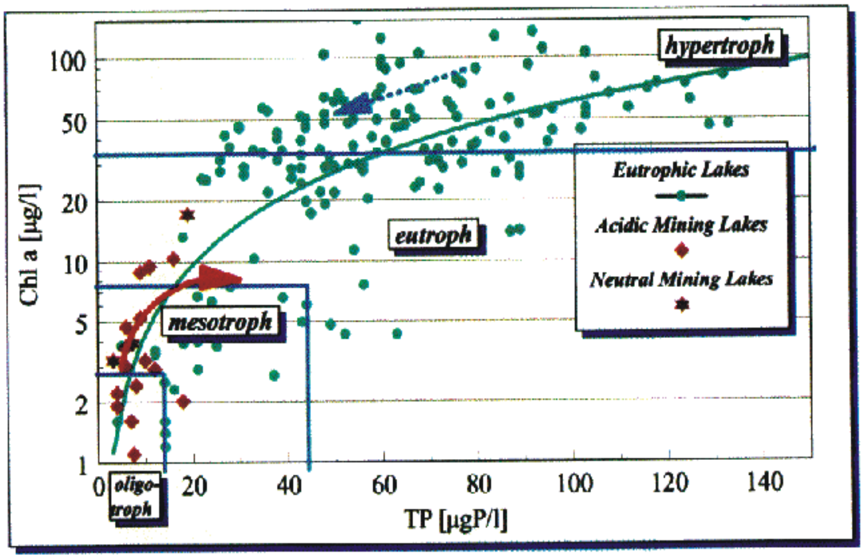

Figure 4. The relationship between chlorophyll concentration and total phosphorus concentrations in lakes. The arrows indicate how these systems will likely develop over time.

With funding from the German government and the lignite industry, a multidisciplinary project, involving researchers from several universities and institutes in the region has investigated the chemistry and ecology of the Lusatian acidic mining lakes in relation to the use of controlled eutrophication to increase lake productivity in the water column and sustainably remove acidity through sediment bound biologically mediated processes in the sediment and water column. The results of this project are described in Nixdorf and Deneke (2004). It has included basic research on particle transport in streams and lakes, pelagic food web interactions and submerged macrophyte metabolism as well as investigations on the roles of wetlands, bacterial interactions at the water-sediment interface (Wendt-Potthoff and Koschorrek, 2002, 2004) and modelling (Nixdorf and Uhlmann, 2002; Uhlmann and Nixdorf, 2002). This contribution focuses on the laboratory experiments on controlled eutrophication, described in more detail in Fyson and Gelbrecht (2004)

Materials and Methods

Laboratory microcosm experiments were carried out with clear PVC columns 2 m high and with an internal diameter of 0.2 m These were filled with 60 L of lake water from Lake Grünewald (pH 3.0, Fe 15 $mg.l^{-1}$, total phosphorus (TP) 5-7 $\mu g.l^{-1}$) and with or without a 20 cm layer of sediment from the lake. The experimental set-up is shown in Figure 5. Microcosms were incubated at room temperature (18-22 °C) under artificial lighting (True Lite fluorescent tubes) with a near to sunlight light spectrum. Light intensity is around 50 Wm-2.s-1 at the top of the water column. Amendments were added as shown in Table 2. Three microcosms were set up for each treatment. Chemical parameters were determined according to standard German methods (DEV) modified according to Zwirnmann et al. (1999). Primary production (PP) was determined by the ^{14}C method as modified by Kapfer et al. (1997) and bacterial production (BP) by a modified ^{3}H thymidine method (Nixdorf and Jander, 2003).

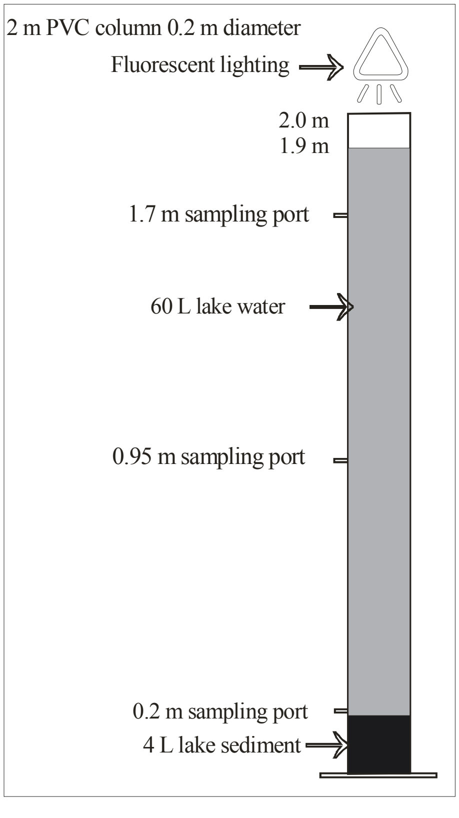

Figure 5. Microocosm set-up

Table 2: Microcosm experiment, treatments and amendments added (3 microcosms for each treatment). All microcosms consisted of 60 L of Lake Grünewald and, except for treatment 1, 4 L of sediment from the same lake

Treatment	Amendment
Control without sediment	None
Complex organic	120 g fresh weight of potato added to the sediment surface in a net bag with a stone to provide weight
Simple organic and phosphate	Addition of Na_2HPO_4 and acetic acid to give concentrations of 200 $\mu g\ L^{-1}$ and 0.1 mM respectively
Control with sediment	Water from and sediment
NaOH	NaOH added to raise pH to 3.5 (precipitate iron)

Results

Microcosm experiments (60 L) were carried out to investigate the effect of phosphorus and organic carbon additions on the water chemistry and biology of acidic lake water in the presence or absence of sediment. Chemical and biological changes were studied in 60 L microcosms with water from Lake Grünewald with or without sediment to determine the effects of added nutrients on primary production and water chemistry. An initial experiment was set up with lake water and amendments, but without sediment to determine the effects of changes in conditions on water chemistry (Fyson et al. (2003) and Fyson and Gelbrecht (2004). Addition of phosphate and organic C (acetic acid) together resulted in higher primary production rates than in controls or with addition of phosphate alone. Phosphate concentrations declined rapidly coupled to removal of Fe. None of the treatments resulted in substantial changes in pH or acidity over a 230 day period.

A second experiment described here included a sediment layer to determine the role of sediment-water interactions on changes in water chemistry and primary production. Additional treatments were applied with the addition of NaOH to raise pH to 3.5 in order to precipitate iron, and of potatoes to generate conditions favourable for anaerobic alkalinity generating processes. The results are shown in Figure 6 and Table 3.

Table 3. Changes in various parameters from set-up to the end of observations (245 days) in the microcosm experiment. Mean values for three microcosms.

Parameter	Water only		Control		P/Acetic acid		NaOH		Potatoes	
	Days		Days		Days		Days		Days	
	0	245	0	245	0	245	0	245	0	245
Fe [mg/L]	4.0	1.3	5.7	<0.2	4.4	<0.2	2.8	<0.2	5.9	<0.2
SRP [µg/L]	4.8	6.1	5.1	3.5	143	3.1	3.2	3.6	3.0	6.6
TP [µg/L]	2.9	3.9	4.1	4.7	169	3.8	5.2	3.8	2.5	24.0
TIC [mg/L]	0.04	0.2	0.25	1.5	0.3	1.9	0.1	31.6	0.1	
637.3										
DOC [mg/L]	1.0	0.8	1.0	2.2	6.2	0.8	1.0	1.3	0.9	5.1
NO_3-N [mg/L]	0.08	0.35	0.06	0.3	0.06	0.2	0.07	0.3	0.06	0.2
NH_4-N [mg/L]	1.1	1.3	1.1	4.0	0.8	3.37	0.8	3.80	0.8	5.1
SO_4 [mg/L]	345	343	346	278	346	256	345	292	344	116
Si [mg/L]	6.8	4.9	7.3	6.5	7.0	3.9	7.2	6.0	7.3	8.8
pH	3.0	3.0	3.0	3.8	3.0	3.8	3.5	4.6	3.0	7.4
Acidity [mmol/L]	2.1	1.8	2.1	0.47	2.1	0.54	2.1	0.23	2.1	0.29
Cond. [µS/cm]	1055	1169	1044	689	1055	671	848	683	1033	576
O_2 [mg/L]	8.0	8.4	8.7	7.1	9.2	7.3	7.9	7.5	3.4	0.8

In the microcosms with lake water alone, there were only small changes in measured parameters (P, Fe, pH, conductivity) over time. Some iron was lost as hydroxide precipitates. The pH increased slightly (Fig 6a). Primary production rates remained very low (Fig 6f). Algal populations (biovolume) declined following the first sampling (19-21 days) as shown in Table 4. The algae were dominated by diatoms (*Eunotia exigua)* and green algae (*Chlamydomonas* spp.)

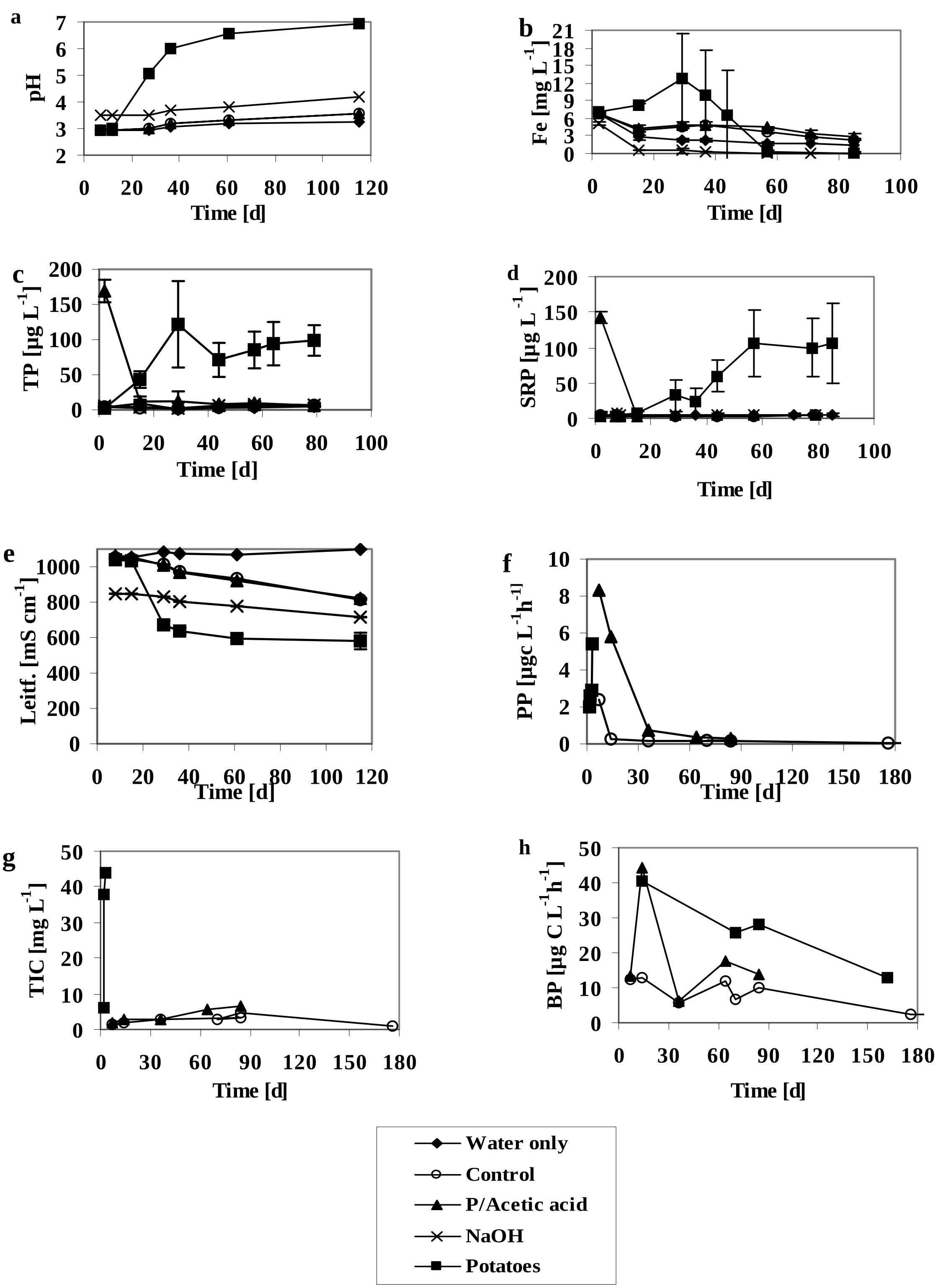

Figure 6. Changes in microcosm water-column chemistry and productivity: a, pH; b, Total dissolved iron; c, Total phosphorus (TP); d, Soluble reactive phosphorus (SRP); e, Electrical conductivity; f, Primary production (PP); g, Total inorganic carbon (TIC); h, Bacterial production (BP); a-e Mean ± SD (n = 3), f-h Single microcosms.

Table. 4. Algal biovolumes in second microcosm experiment after 19-25 and 246-256 days. N = *Nanochlorum* sp., O = *Ochromonas* sp., E = *Eunotia exigua*, C = *Chlamydomonas* sp., X = unidentified green alga.

Amendment	Days	Taxa present	Biovolume $[\mu m^3/mL]$	*Eunotia* $[\mu m^3/mL]$	[%]	*Chlamydomonas* $[\mu m^3/mL]$	[%]
L. Grünewald	water	N O E C	1614	287	18	1274	79
Water only a	21	N O E X	14125	13782	98	0	0
	256	N	10	0	0	0	0
b	19	N O E C	37867	17982	48	19710	52
	256	N O E C	3453	1160	34	2251	65
No a	21	N O E C X	23010	19933	87	1578	7
amendment	246	N O E C X	2818	2278	81	421	15
b	21	N O C	1747	0	0	1714	98
	256	N O C	1707	0	0	1687	99
Phosphate a	21	N O E C X	164926	135547	82	28770	17
/acetic acid	246	N O E C X	2997	2278	76	631	21
b	21	N O E C X	227106	222684	98	1893	1
	256	N E C	8054	5801	72	2142	27
NaOH a	19	N O E C	45409	38728	85	631	1.4
	246	N O	32	0	0	0	0
b	25	N O E C	44242	43853	99	210	0.5
	256	N O E C	2070	1709	83	210	10
Potato a	21	N O E	7164	6834	95	0	0
	246	N O	22	0	0	0	0
b	21	N O E C	24610	9589	39	14756	60
	256	N O C	4354	0	0	4132	95

In microcosms with lake-water and sediment and no further additions, conductivity declined substantially during the course of the experiment largely attributable to the decline in sulphate concentration from 346 mg L^{-1} to 278 mg L^{-1} This was presumably due to sulphate reduction in the sediment and precipitation of iron hydroxy sulphates (such as schwertmannite as described in Blodau and Peiffer, 2003). It is noteworthy that the Fe-concentration in microcosms with sediment fell more rapidly than in the water-only microcosms (Fig 6b). The pH climbed to 3.8 by the end of the measurement period and titratable acidity ($KB_{8,2}$) declined from 2.1 to 0.47 mmol L^{-1}. In contrast, the pH values and acidity changed very little in the water-only microoocosms (Table 4). Clearly, the sediment is able to generate alkalinity. With both these treatments, phosphate concentrations remained very low (Fig 6c, d). Detectable nitrate disappeared. In contrast, in the presence of sediment, ammonium-N concentrations increased. Total inorganic carbon (TIC) and dissolved organic carbon (DOC) concentrations also increased in these microcosms indicating net release or carbon from the sediment from decomposition processes. Other parameters (Mn, Ca, Mg, Al, K, Na, Cl, Si) exhibited little change in concentrations (data not shown). In the microcosms with sediment but no other additions, primary production (Fig 6f) and algal biomass (Table 4) remained low and similar to values of water-only microcosms and was similarly dominated by *Eunotia exigua* and *Chlamydomonas* spp. The increase in TIC was associated with high rates of bacterial production (Fig. 6h).

With the addition of phosphorus and acetate, soluble reactive phosphorus (SRP) concentrations declined from a mean of 143 μg L^{-1} two days after addition to only 6 μg L^{-1} after 79 days, likely due to coprecipitation with iron hydroxides (Fig 6c, d; Table 3). The decrease in sulphate concentration was similar to that of microcosms with sediment alone (Table 3). At the beginning of the experiment, the P and acetate additions led to a dramatic increase in primary production (Fig 6f) but subsequently declined with sinking phosphorus concentration. However, values remained well above those of water only and water/sediment microcosms. Bacterial production, in contrast, remained high (Fig. 6h). The low DOC concentration indicates the uptake and or breakdown (respiration) of the added acetate. Other parameters (N, TIC, metals) exhibited only small changes in microcosms with added phosphorus and acetic acid (Table 3). Si concentrations declined rapidly in association with the build up of the diatom *E. exigua* (Table 4) which uses this element to construct cell walls.

NaOH was added to some microcosms to increase the pH to around 3.5 to rapidly precipitate iron. During the course of the experiment, the pH climbed further to 4.6 (Fig 6a, Table 3) and much of the titratable acidity was removed. With the exception of Na (added to the microcosms), the concentration of other parameters remained similar to those of microcosms with water and sediment alone. The higher pH values did not result in retention of higher P-concentrations in the water column, nor of high primary production rates. There was a greater development of *E. exigua* and higher overall algal biovolume in these microcosms (Table 4).

With the addition of potatoes to the sediment surface and the resulting decomposition, oxygen concentrations decreased to below the detection limit (0.1 mg L^{-1}) within 4 weeks. In these anoxic conditions, the pH values in the water column rose to more than 7 and remained circum-neutral for the remainder of the experiment (Fig 6a, Table 3). At the same time, Fe concentrations increased for a short period with the release of Fe (II) from the sediment. The anoxic conditions were short lived with the subsequent slow recovery of oxygen concentrations. Iron concentration declined to less than 0.2 mg L^{-1} (Fig 6b). There was a dramatic decline in sulphate concentrations from 344 mg L^{-1} to 124 mg L^{-1} (Table 4) presumably due to enhanced sulphate reduction. Conductivity decreased substantially to less than 600 µS cm^{-1}.

The analyses of the sediments (Table 5a) show that the sulphur content of sediments amended with potatoes was clearly higher than with other treatments indicating the net deposit of sulphur. This is best explained by the production of iron sulphides. The low redox potentials and high Fe concentrations in the sediment porewater (Table 5b) indicate iron reduction and release of Fe(II). There was a substantial increase in TP concentration in the water column of these microcosms in the early part of the experiment (149 µg L^{-1} after 49 days). Although subsequently declining, it remained well above that of other treatments (24 µg L^{-1}) at the end of the observation period. In contrast, SRP concentrations were similar to those of the controls probably indicating uptake and rapid recycling within the water column. A further consequence of the anoxic conditions in the potato microcosms was the increase in concentrations of ammonium-N, TIC and DOC. The overall release of nutrients resulted in very high primary reduction rates (Fig 6h) due to the proliferation of green sulphur bacteria. The algal biovolume remained low (Table 4).

Table 5. Sediment porewater analysis and composition of sediments from the microcosm experiment 245 days after set-up. Mean values from amalgamated samples from 3 microcosms.

a) Sediment pore-water

Amendment	Fe [mg/L]	Mn [mg/L]	TP [µg/L]	pH	Redox [mV]
Control	186	0.8	207	6.7	-147
PO_4/Acetic acid	145	0.7	215	6.7	-141
NaOH	148	0.9	251	6.7	-133
Potato	144	0.7	256	6.9	-128

b) Sediment carbon, hydrogen, nitrogen and sulfur (% dry weight)

Amendment	Carbon	Hydrogen	Nitrogen	Sulfur
Control	10.8	1.49	0.20	0.46
PO_4/Acetic acid	11.1	1.51	0.19	0.47
NaOH	10.8	1.49	0.20	0.45
Potato	11.0	1.54	0.21	0.54

Discussion

Acidic mine drainage poses a major environmental problem in many countries. Remediation through constructed wetlands and anoxic limestone drains has been extensively developed, particularly in North America. However, long term, self-sustaining treatment of extremely acidic waters has not been demonstrated. Although the treatment systems have been described as passive, they are not self-sustaining and will require the periodic removal of precipitates and recharging of substrates. A truly passive system (would require no ongoing management.

The water chemistry of the acidic Lusatian mining lakes is essentially the same as the intensely studied acidic coal mine-drainages of the eastern United States with high concentrations of dissolved Fe (III) and sulphate as well as low pH values (2-4). Most attention has been paid to acidity removal through promotion of sulphate reduction in sediments in anaerobic wetlands and other so called "passive" treatment systems (see Kalin et al. (1995) and Skousen et al.(1998) for review of these systems). Other processes, Fe (III) reduction and assimilation coupled to uptake of nitrate can also play a role. However as nitrate concentrations in such waters are generally low, sulphate reduction coupled to Fe (III) reduction and the sequestration of Fe and S as sulphides in the sediments remains the best prospect for acidity removal without the addition of expensive and only short-term effective direct addition of neutralising materials (lime etc).

Sulphate reduction can be enhanced through increasing the supply of organic substrates through addition of wastes or from dead organisms produced within the water body. Increasing autochthonous primary production through controlled eutrophication (enhanced production through enhancement of nutrient supply) is therefore an attractive strategy for the long-term neutralisation of these lakes through sustained supply of organic material and nutrients to sediments. The acidic mining lakes of Lusatia are characterised by extremely low phosphorus and dissolved inorganic carbon concentrations because of the high iron (III) content and low pH respectively. The iron (III) precipitates as iron (III) oxyhydroxides with the coprecipitation of phosphorus. Subsequent reduction of these precipitates in anoxic conditions can re-release the phosphorus.

The experiments described here have shown that an increase in the supply of nutrients can fuel increases in primary production in microcosm experiments. However, only in the presence of sediment and with the addition of readily degradable organic material (potatoes) did neutral conditions develop in the water column and were iron and sulphur sequestered in the sediments. This neutralisation requires the generation of anoxic conditions in and above the sediment. Increased phosphorus and carbon supply greatly enhances primary production and maintains carbon supply for the sediment-bound alkalinity generating processes. The experiment described here and others published elsewhere (Fyson et al., 1998; Fyson and Gelbrecht, 2004) show that once the acidity is removed, pH remains high and an enhanced supply of phosphorus to the water column is maintained. Whether this can sustain treatment of incoming acidic groundwater in the mining lakes has yet to be established. Enclosure experiments in Lake Grünewald (Fyson and Gelbrecht, 2004) have shown enhanced productivity with addition of potatoes. However, as yet only short term experiments have been carried out and long-term experiments of whole lakes are necessary to establish this treatment technology.

The results presented here for Lake Grünewald confirm earlier observations made in 20 L microcosm experiments with lake water and sediment from Koschen (pH 3.1, Fe 21 $mg.L^{-1}$), another lake in the area. In this previous study, anoxic conditions were temporary but neutral conditions with enhanced P concentrations were maintained until the end of the observation period (2 years after set-up) (Fyson et al., 1998; Fyson and Steinberg, 1999). Biological acidity removal clearly works in laboratory microcosms and enhanced P and C supply can greatly enhance primary production, support enhanced algal growth and sustain neutral conditions in the water column.

An understanding of the overall functioning of these ecosystems is essential for the development of eco-technological remediation measures which optimise the role of biological alkalinity-generating processes for sustainable, environmentally acceptable acidity removal. Food webs in these lakes are relatively simple (e.g. top predators are water bugs (coroxids) (Wollmann et al., 2000) and these relatively simple ecosystems provide model system for studies of trophic interactions. With the acquisition of further knowledge, both of ecosystem functioning and the geohydrological situation, integrated development strategies, incorporating long-term, self-sustaining acidity removal may be developed (for these ecosystems?) .

Together with modelling of the chemical processes in relation to lake morphometry (Nixdof and Uhlmann, 2002; Uhlmann and Nixdorf, 2002), the results of the studies described here and those on other aspects of the acidic, flooded lignite pit ecosystems will assist in the development of biologically based remediation strategies (Nixdorf and Deneke, 2004).

There is great interest in developing sustainable treatments for AMD. Most of the "passive" systems developed up to now (see Skousen (1998) for a review of these technologies) are not sustainable. Although they embrace the role of microbial alkalinity generating processes, especially sulphate reduction, they will require ongoing maintenance such as substrate replacement. In addition, it has not been clearly established that the metals and sulfides removed are in the form or stable sulfides in sediments. In order to develop truly sustainable technologies for AMD treatment, ecosystems must be designed and engineered to optimise the role of primary producers (macrophytes or algae) through enhanced biogeochemical cycling to maintain a supply of carbon for the anaerobic, alkalinity-generating processes. The ARUM process (Fyson et al., 1995; Kalin et al., 1995) employs such ecological engineering approach to treat AMD. In these systems, floating vegetation mats generate carbon to sustain anaerobic conditions in and above the sediments below and provide fuel for the alkalinity generating processes. Such systems have been shown to remove acidity for at least 5 years (Kalin and Smith, 1996). Further research and long-term funding is clearly needed to understand how AMD treatment can be sustained indefinitely.

Summary

Microcosm experiments with water and sediment from extremely acidic flooded lignite pits in Germany have demonstrated that supplementary supply of phosphorus and carbon to the water column (controlled eutrophication) can enhance primary production and maintain sediment-bound, alkalinity generating processes. Ongoing studies will develop treatment strategies which are environmentally acceptable and provide a sustainable alternative to expensive chemical treatments for neutralisation of AMD.

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