Gypsum treatment as a restoration method for sediments of eutrophied lakes – experiments from southern Finland

V.-P. Salonen · E. Varjo

Abstract The internal load of phosphorus can be treated with chemicals improving a sediment's phosphorus binding capacity. Gypsum ($CaSO_4*2H_2O$) is a novel material to be applied for this purpose. In the hypereutrophic Lake Enäjärvi, southern Finland, sediments were treated with gypsum in test basins. The basins were isolated from open water to create artificial anoxia, which allowed the effects of the treatment on the internal load to be measured. The results indicate that gypsum treatment decreases the sediment's release of nutrients under anoxic conditions. This was demonstrated as less increased total-P and total-N concentrations and decreased chlorophyll-a concentrations in the water column of treated basins as compared with the control basins. Because the gypsum treatment prevents internal load, it has potential in lake restoration in cases where the internal load of phosphorus is accelerated by anoxic conditions at the sediment/water interface.

Key words Gypsum · Internal loading · Lake restoration · Sediment treatment · Sediment chemistry · Water chemistry · Finland

Introduction

Eutrophication of lakes is a challenge for water management in Finland. Problems such as blue–green algae blooms, oxygen deficiency, increased sedimentation rates, and fish-kill reduce the value of the lake as a recreational, water or fishing resource. Eutrophication is directly related to the increased availability of phosphorus, a critical nutrient in the basic productivity of most Finnish lakes (Lappalainen and Matinvesi 1990).

Numerous lakes in southern Finland eutrified between the 1950s and 1980s due to sewage waste and intensive use of fertilisers in their catchment area. Phosphorus then accumulated in large amounts in the sediments at the bottom of the lakes, later to be released as part of the nutrient cycle involving the water/sediment interface. The release of phosphorus is enhanced by oxygen deficit in hypolimnion (Mortimer 1941, 1942), methane ebullition, resuspension by wind or bioturbation (Boström and others 1988). Thus nutrient-rich sediments can maintain a high concentration of nutrients in the water mass and create a problematic internal load.

There have been numerous attempts and various strategies to restore lakes, targeted specifically at reducing external loading from point sources or from agriculture. In addition, a number of other restoration techniques have been attempted aimed especially at reducing or eliminating the internal load, based largely on improving the oxygen state of benthic water or on reducing bioturbation. These restoration projects used one or a combination of the following strategies: hypolimnetic aeration, food web management, field erosion reduction and lowering of associated diffusive load, water level stabilisation, vegetation harvesting, chemical precipitation of phosphorus, removal of the surface layer of the sediments by dredging (Eiseltová 1994), and finally covering and sealing of nutrient-rich sediments at the bottom.

Attempts to physically cover the sediment have been part of lake management experiments since the 1970s. Materials such as lime, fly ash, sand, silt, clay, gravel, plastic, polyethylene and fibreglass have been tested and used (Dunst and others 1974; Cooke 1980; Cooke and others 1986; Baker and others 1993). In Lake Tuusulanjärvi an experiment using clay has been going on since 1993, with positive results and improvements of the quality of the lake water (Pekkarinen 1994; Sommarlund 1996). Clay and phosphorus-precipiting chemicals were used in order to seal off the bottom sediment both physically and chemically from the overlying water mass.

In this paper we report on an experiment using gypsum ($CaSO_4*2H_2O$) as a means of lake restoration. Gypsum is a sulphate mineral that is naturally occurring or is formed in large amounts as an industrial by-product. To our knowledge, gypsum has never been reported for application to lake restoration projects.

In vitro experiments show that gypsum leads to chemical bonding of phosphorus in the sediment with the formation of Ca-P compounds, namely hydroapatite, and therefore reduces phosphorus release from the sediments into

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the water mass (Haapamäki and Salonen 1995). In addition, it was believed that a gypsum layer could act as a low-density physical barrier reducing exchanges at the sediment/water interface. Based on this, a new "gypsum treatment" strategy of lake restoration was conceived and tested. An experiment was conducted in order to test the hypothesis that gypsum addition would lead to a decrease in the release of nutrients from anoxic lake sediments by mechanically stabilising the sediment, by chemically binding the phosphorus into Ca-P compounds into the sediment column, and by inhibiting methanogenesis and hence methane gas release causing sediment disturbance.

The effects of gypsum on lake sediments were tested in in situ experiments in Lake Enäjärvi during the summer of 1996. As part of the experiment we created anoxic conditions in the hypolimnion and at the sediment/water interface in closed test basins within the lake in order to accelerate internal loading. The nutrient release to the hypolimnion in the experimental in situ basin was then measured and compared with the control basin, and with open water conditions in the lake.

The experiment was planned to cover, during a full year cycle, the change in the test basin from oxic to anoxic conditions, and from ice-free to ice-covered conditions. Only the first objective could be met however, since the experiment had to be interrupted after 90 days following the disruption of the basin sets under stormy conditions. Consequently, ice-covered conditions could not be monitored, and the data set is limited to the record of change to anoxic conditions. Because of the untimely abortion of the experiment, the short record does not allow an elaborate statistical analysis of the data. However, the record indicates a clear and remarkable improvement of the water quality and it is believed that the data set presented here is a valuable indicator of the short-term magnitude and direction of change under ice-free anoxic conditions after the addition of gypsum.

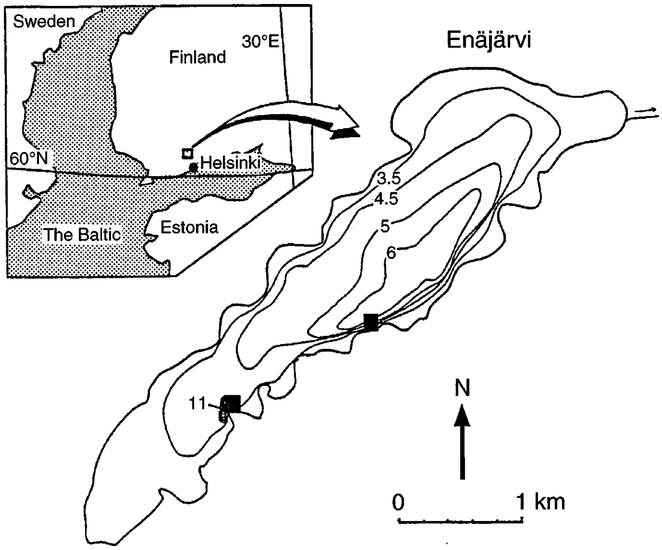

Fig. 1
Lake Enäjärvi and the experimental sites

Materials and methods

Lake Enäjärvi

Lake Enäjärvi is a shallow, hypertrophic lake located in southern Finland (Fig. 1). The lake area is 5.2 km^2 with a catchment area of clayey and silty soils covering 34 km^2. Its mean and maximum depths are 3.5 and 11 m respectively. Lake volume is ca 18 Mm^3 and theoretical residence time 520 d. The period of ice cover is ca. 150 d (Kettunen 1981). The mean total phosphorus concentration in the water mass during the ice-free period is 140 $\mu g\ l^{-1}$ and total nitrogen 2000 $\mu g\ l^{-1}$ (Kettunen and Stenmark 1982). The lake was loaded by sewage waters of the hamlet Nummela from 1950 to 1976 with 900–4000 kg P a^{-1}. Since 1977, the waste waters have been diverted away from the Lake Enäjärvi drainage basin.

According to Alhonen (1982), Lake Enäjärvi has been productive throughout its history. Its eutrophication started when the water level was lowered in 1928 by 1 m (Salonen and others 1993) and was later accelerated due to waste water loading. In 1977, although the external loading was reduced drastically, nutrients in the water mass remained high and increased due to high internal loading from phosphorus release at the sediment/water interface. The highest values of sedimentary phosphorus (total phosphorus >2 mg g^{-1} DW, for the sediment depth 0–10 cm) were measured in the deepest areas of the lake (Alhonen 1983; Salonen and others 1993). Blooms of blue-green algae and oxygen depletion in the hypolimnion have been the most visible consequences of the high productivity.

Lake Enäjärvi is a relatively shallow lake and the dominant wind direction is along the longitudinal axis of the lake. The internal phosphorus loading is enhanced by wind-driven agitation of the water, which is felt at the bottom, leading to a resuspension of particles (Kettunen 1981). Any restoration strategy of Lake Enäjärvi should therefore be based on stabilising the sediment/water interface, isolating the sediment surface from the overlying water mass and improving the phosphorus-binding capacity of the sediment.

Test basins and analytical methods

Two sets of 3 basins, ca. 5 m in diameter, were constructed and placed in the part of the lake where sediments are thick (Fig. 2). The basins were emplaced in May 1996, one set at 5.5 m and the other set at 7.5 m water depth. Each basin isolates the entire water column water and sediment interface from each other and from the surrounding water area. In each set, two basins, here referred to as "test basins", were treated by the addition of gypsum, while the third one was used as a "control basin". Two different available types of gypsum were used: phosphogypsum (easily leachable phosphorus

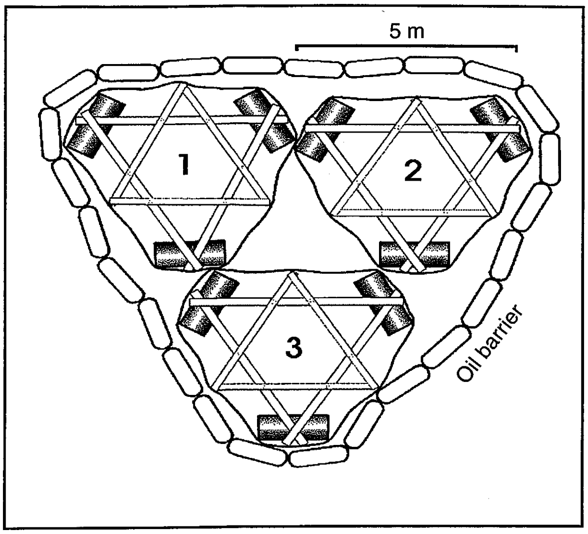

Fig. 2
The experimental basin sets in Lake Enäjärvi. The basins *1* and *2* are treated by gypsum and *3* is the control basin

0.03%), a by-product of phosphate acid production, and ferrogypsum (Fe 4%), a by-product of neutralising acid process waters. Phosphogypsum was used in the 5.5-m-deep basin and ferrogypsum in the 7.5 m basins. Gypsum was added in an amount that would correspond to a uniform layer, 1-cm thick, at the bottom of the test basins. The conditions in the water mass were monitored weekly (oxygen, pH, conductivity and temperature) by YSI 610-Multi-Parameter Water Quality Monitor (YSI-Incorporated). Weather data at the site (average temperature, precipitation) were collected daily from 2 June to 30 September 1996.
Water samples of epilimnion (0-2 m water depth) and hypolimnion were collected five times during the observation period with the subsequent measurements of (10 cm above the bottom) total phosphorus (Finnish standard SFS 3026-digestion with peroxodisulfate), total nitrogen (QuickChem Method 30-107-04-1-B-oxidation with peroxodisulfate) and chlorophyll-a concentration (Finnish standard SFS 5772-extraction with ethanol). These analyses were made at the chemical laboratory of S-W Finland Regional Environment Centre controlled by authorities.
When gypsum was shovelled into the test basins, it sank through the water column within a few hours and accumulated on the loose bottom. Coring of the sediment at the end of the experiment showed that the gypsum had formed a sticky layer, about 1-cm thick at a depth of ca. 2–3 cm below the sediment/water interface. Above the gypsum layer, the uppermost 2 cm are loose sediment, while below, the sediment is amygdaloidal and coloured by sulphides. At the end of the experiment, sediments were sampled and analysed for P-fractions; samples were taken from the uppermost 2 cm, above the gypsum layer, the interval constituting the gypsum layer, and the 2 cm interval below the layer. Samples from corresponding depths from the control basins and open water were analysed for comparative purposes. For the sediments, total phosphorus was analysed by the method of Bengtsson and Enell (1990); the sequential extraction method (Hieltes and Lijklema 1980) was applied to determine phosphorus fractions: labile P (in interstitial water), P bound to Fe- and Al- compounds (released when redox-potential decreases), apatitic P (stabile fraction) and P bound to organic compounds.

Results

In 1996, the early part of the summer was cool and rather rainy. It was followed by a warm period, which persisted from 22 July to 2 September (Fig. 3). At the end of this period conditions of anoxicity in the basins' hypolimnion had been reached (Table 1, Fig. 4) and therefore the effects of gypsum under anoxic conditions are recorded by samples taken in September (Table 2). The warm, dry period was followed by cool and windy days during which time the basin sets suffered damages and the experiment had to be interrupted (Fig. 3).
The trends of total phosphorus, total nitrogen and chlorophyll-a in water all indicated that gypsum treatment markedly affected the water quality. The comparison between test basins, control basins and open water is shown in Fig. 5, where changes in phosphorus, nitrogen and chlorophyll-a concentrations from oxygenated to anoxic conditions are shown as bars.
Under anoxic conditions of the control basin, the release of nutrients was higher than in open water as shown by the chlorophyll-a concentrations. By contrast chlorophyll-a decreased in the test basins. Labile sedimentary phosphorus (Table 3) in the pore water is depleted in test basins as compared to the control basins, most probably as a result of phosphorus bound into the gypsum treated sediment.
There was no notable difference in pH between treated and untreated basins. The values remained between 7

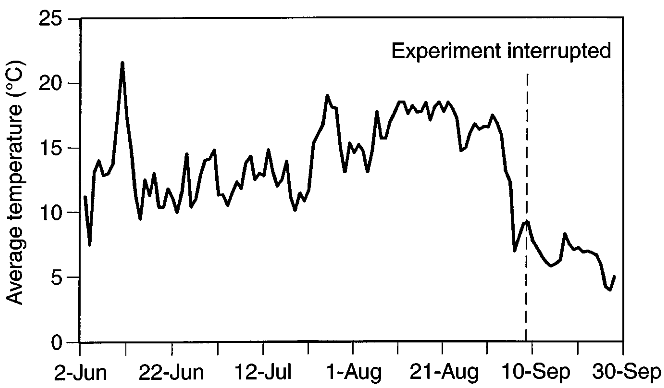

Fig. 3
Average daily temperature in Lake Enäjärvi area during the experiment

Table 1
Oxygen concentrations (mg/l) in test basins and open water during the experiment

Date		2.6	8.6	17.6	24.6	30.6	7.7	17.7	26.7	7.8	16.8	1.9
Epilimnion of 5.5 m basins	treated 1	11.31	11.82	10.13	10.21	11.05	8.47	9.03	7.66	6.57	3.65	3.98
	treated 2	10.95	11.97	9.67	10.14	11.25	8.66	9.44	9.73	7.72	5.63	4.02
	control	10.81	11.14	8.40	7.08	8.51	8.30	8.14	7.12	5.44	3.89	2.62
	open	10.83	11.70	10.60	10.87	11.90	9.33	10.19	9.98	9.84	9.88	8.62
Epilimnion of 7.5 m basins	treated 1	11.20	12.24	10.16	10.01	12.20	8.43	7.90	7.25	9.65	8.23	1.70
	treated 2	10.24	10.49	10.45	9.88	12.08	8.28	7.91	8.89	9.41	6.77	2.36
	control	9.19	9.49	9.25	8.98	11.97	7.40	7.03	5.73	6.93	5.14	2.38
	open	10.67	11.50	10.74	10.96	11.30	9.32	9.87	9.38	10.2	9.79	8.39
Hypolimnion of 5.5 m basins	treated 1	10.17	9.95	8.41	6.90	5.39	8.53	8.81	3.52	4.07	0.54	1.70
	treated 2	10.59	9.65	8.48	7.26	6.89	8.51	9.16	3.58	4.90	0.50	1.34
	control	9.82	9.47	7.07	5.24	4.85	8.25	7.91	3.31	2.37	0.21	0.48
	open	10.56	9.85	10.32	6.98	3.67	9.38	10.07	3.58	6.28	0.41	7.34
Hypolimnion of 7.5 m basins	treated 1	10.67	10.64	8.03	5.40	3.54	8.29	7.00	1.59	4.65	0.42	1.15
	treated 2	9.42	8.96	8.55	6.45	4.79	8.23	7.08	3.11	4.42	0.57	0.12
	control	6.63	6.68	7.63	4.69	4.72	7.21	5.95	2.21	3.52	0.22	0.06
	open	10.35	8.20	10.08	6.81	2.54	8.94	9.71	3.40	3.75	0.42	7.87

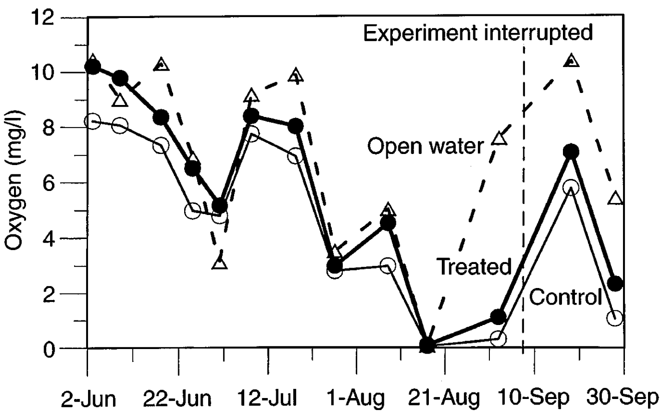

Fig. 4
Oxygen conditions in studied water columns of Lake Enäjärvi during the experiment

and 8 throughout the experiment. Conductivity in epilimnion of the test basins varied from 0.100 to 0.170 mS/cm and in control basins from 0.090 to 0.112 mS/cm with no measurable difference in the hypolimnion. Gypsum increases sulphate (SO_4^{2-}) concentration in sediment, but not in the water column as concentrations measured in all water masses were alike.

Discussion

Methanobacterial ebullition may be the main agent transporting phosphate trough the sediment/water interface (Matinvesi 1995). As the productivity of an aquatic system increases, methanogenesis usually becomes a more important terminal diagenetic process because sulphate reduction is limited by the rate of sulphate flux into the

Table 2
Nutrient- and chlorophyll-a-concentrations (μg/l) in test basins and open water during the experiment

Date			2.6	17.6	7.7	7.8	1.9
Total P	5.5 m epilimnion	treated 1	86	99	110	130	220
		treated 2	80	92	110	110	130
		control	43	59	100	130	240
		open water	57	76	110	110	110
	5.5 m hypolimnion	treated 1	80	110	130	120	190
		treated 2	68	85	140	89	120
		control	51	100	140	130	250
		open water	150	78	110	160	110
	7.5 m epilimnion	treated 1	42	78	100	140	110
		treated 2	60	61	110	100	110
		control	59	92	110	140	110
		open water	53	82	120	120	110
	7.5 m hypolimnion	treated 1	46	93	140	130	350
		treated 2	46	110	120	120	220
		control	68	110	150	100	320
		open water	240	290	170	120	110
Total N	5.5 m epilimnion	treated 1	1400	1100	1200	1100	1500
		treated 2	1400	1100	990	1200	1100
		control	1300	1200	910	1300	1400
		open water	1300	1000	1100	1200	790
	5.5 m hypolimnion	treated 1	1200	1000	1000	990	1100
		treated 2	1200	1100	1100	1100	940
		control	1400	1100	970	1100	1400
		open water	1700	1100	970	1100	800
	7.5 m epilimnion	treated 1	1400	1100	1100	1000	1000
		treated 2	1300	1200	960	960	1000
		control	1400	1100	1100	850	1100
		open water	1300	1900	1100	990	810
	7.5 m hypolimnion	treated 1	1400	1100	970	970	1500
		treated 2	1400	1000	910	1400	1200
		control	1400	1100	1000	1200	1400
		open water	1400	1100	1100	1300	760
Chl-a	5.5 m	treated 1	20	42	25	11	9.4
		treated 2	21	27	28	19	11
		control	18	16	33	8.4	14
		open water	20	28	30	16	27
	7.5 m	treated 1	12	32	30	17	9
		treated 2	22	20	24	15	13
		control	13	25	28	8.7	30
		open water	23	28	24	16	25

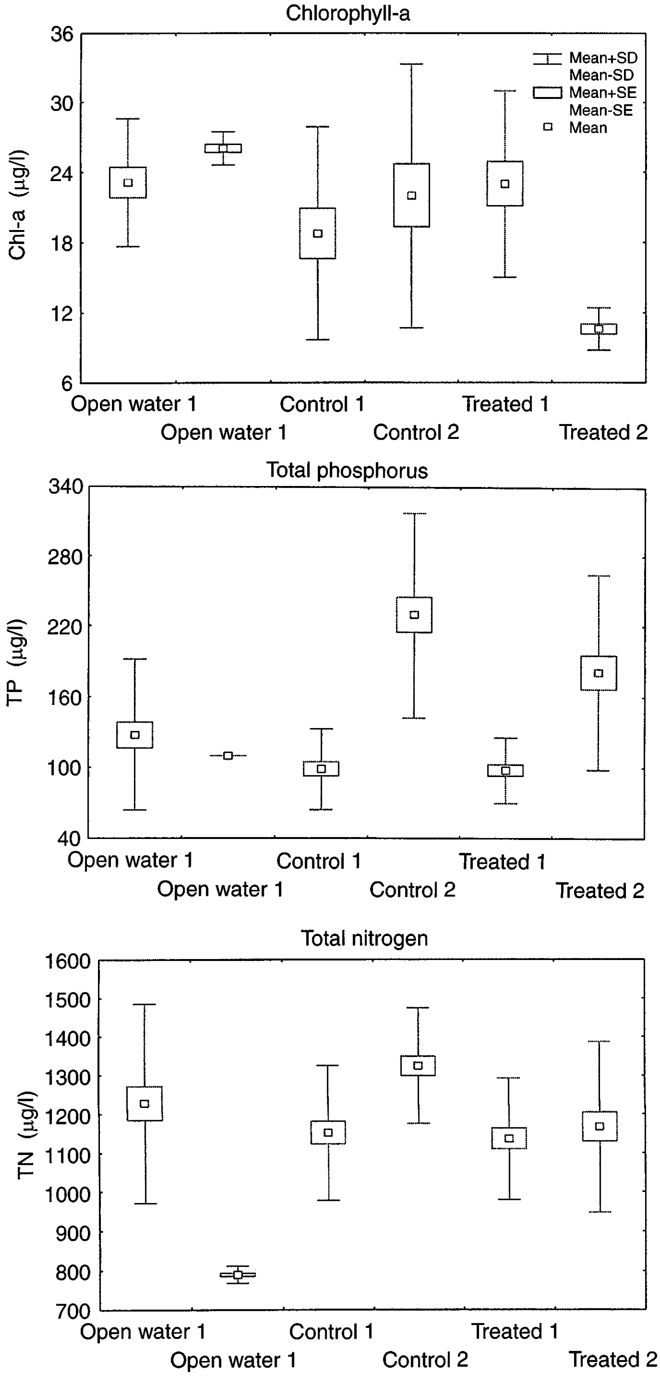

Fig. 5
Chlorophyll-a, total phosphorus and total nitrogen concentrations in studied waters Number *1* figures denote samples taken during oxic conditions and number *2* anoxia

sediment (Lovley and Klug 1986). In the presence of excess sulphate, methanogenesis is limited by the availability of acetate and hydrogen because sulphate reducers are able to outcompete methanogens for these substrates (Kuivila and Murray 1984; Lovley and Klug 1986). Sulphate provides an electron acceptor for sulphur bacteria, which is largely responsible for the composition of the

Table 3
Phosphorus fractions (mg g^{-1} dry weight) in bottom sediments at the end of the experiment

Sample and sediment depth		Labile P (pore-water P)	Fe-, Al-compound-bound P	Apatite P	Organic compound-bound P
5.5 m treated 1	0–2 cm	0.027	0.527	0.119	7.036
	2–4 cm	0.071	0.593	0.145	2.283
	4–6 cm	0.015	0.278	0.074	1.293
5.5 m treated 2	0–2 cm	0.148	0.349	0.051	2.482
	2–4 cm	0.495	0.000	0.327	1.881
	4–6 cm	0.000	0.201	0.113	0.424
5.5 m control basin	0–2 cm	0.583	0.280	0.089	0.058
	2–4 cm	0.043	0.446	0.132	1.477
	4–6 cm	0.033	0.302	0.126	0.613
5.5 m open water	0–2 cm	0.043	0.373	0.096	0.800
	2–4 cm	0.043	0.141	0.091	0.302
	4–6 cm	0.031	0.141	0.088	0.203
7.5 m treated 1	0–2 cm	0.044	0.702	0.073	0.992
	2–4 cm	0.027	0.609	0.066	1.226
	4–6 cm	0.015	0.518	0.062	1.133
7.5 m treated 2	0–2 cm	0.035	0.817	0.110	1.415
	2–4 cm	0.018	0.633	0.078	1.307
	4–6 cm	0.003	0.614	0.063	1.066
7.5 m control basin	0–2 cm	0.179	0.569	0.053	0.802
	2–4 cm	0.096	0.542	0.045	0.964
	4–6 cm	0.091	0.617	0.071	0.917
7.5 m open water	0–2 cm	0.262	1.230	0.080	1.544
	2–4 cm	0.110	0.637	0.235	1.247
	4–6 cm	0.095	0.660	0.280	1.584

organic material, therefore preventing ebullition caused by methanobacteria. Sulphate addition therefore suppresses the most detrimental methane fermentation (Urban and others 1994). Anaerobic methane oxidation occurs in sulphate reduction zone, which limits the methane flux into the water column (Devol and Ahmed 1981; Devol 1983; Kuivila and others 1989). Sulphate-reducing bacteria compete also with bacteria that utilise oxygen, nitrate, ferric iron and manganese to oxidize organic material (Komor 1992). Studies on the effects of gypsum treatment on the formation of gas are still under way and results yet to come.

Under anoxic condition, the total phosphorus and nitrogen concentrations in the water mass of the test basins increased notably less than in the control basins. There was clearly a decreasing trend in the basic productivity within the test basins following the addition of gypsum, while in control basin, basic productivity increased. The sharp difference in productivity can be explained by the difference in the amount of labile phosphorus in the sediments. The labile fraction is most important in evaluating trophic state of lakes; since this fraction is readily available to algae and since 90–100% of interstitial phosphorus is in leachable form, it plays a significant role in internal load (Boström and others 1988). As shown by the increase in chlorophyll-a concentration in control basin, a higher proportion of total phosphorus remained labile and available for algae compared with the test basins (Table 3). Under open water conditions, wind-induced mixing of the water mass and lack of anoxia precluded the development of changes in productivity due to phosphorus loading.

The results from the control basins show that when the oxygen concentrations in the hypolimnion approach zero, phosphorus is released, which leads to an increased basic productivity. This is in agreement with Mortimer's (1941, 1942) theory on internal load. Because this was not observed in the basins treated by gypsum, it can be concluded that the addition of gypsum effectively reduces internal loading.

In addition to sulfate, gypsum also brings calcium cations into the sediment column and thus offers more sites for phosphorus to bind, as hydroapatite is very stable under constant, non-acidic pH conditions. Since there was no measurable change in pH following the gypsum treatment, it is not expected that re-dissolution of apatite will occur.

Gypsum is produced as an industrial by-product, at an annual rate of nearly 1.5 Mt in Finland. Although there are numerous useful applications for gypsum (Häkkinen and Kanerva 1981; Häkkinen and others 1985; Sarvaranta and Kaasinen 1994), gypsum piles are growing and the use of gypsum in new applications would not likely meet with shortages. As a lake sediment restoration technique, gypsum may achieve comparable accomplishments as some other chemicals already in use such as Fe-sulfate, Al-chloride etc. (Sommarlund 1996), but as an industrial by-product, its use might be much more cost-effective.

Conclusion

Gypsum appears to have promising possibilities in lake restoration strategies. It appears well suited for lakes with high methanogenesis and with bottom areas where internal loading by phosphorus accelerates eutrophication. Gypsum accumulates as a thin layer near the sediment/water interface and produces the three following effects:

1. It stabilises the surface sediment where it forms a natural geomembrane preventing resuspension and gas ebullition.
2. It improves the sediment's capacity to bind phosphorus as a result of the addition of Ca and formation of hydroapatite.
3. It provides electron acceptors for sulfate bacteria and thus increases the proportion of degradation process effectuated by them at the expense of the methanobacteriae. This limits the methane formation and the convective effect of ebullition which transports sediment particles and dissolved substances into the water column.

These effects are comparable in magnitude and essence as those resultings from the addition of other chemicals (Fe-nitrate, Fe-chloride, Fe-sulphate, Al-chloride, lime etc.) in lakes. However, gypsum is a much more cost-effective material, as it is an industrial by-product and also a common industrial mineral of which there are large known deposits. This gives it further potential in lake sediment restoration applications.

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