Interactions between calcite precipitation (natural and artificial) and phosphorus cycle in the hardwater lake

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Abstract

The influence of calcite precipitation on the phosphorus cycle in stratified hardwater lake was studied before and during experiments with a new restoration technique. Surveys of the chemical composition of water column and monitoring of settling particles of Lake Luzin (North–East) showed that calcite precipitation occurs each year over 2–3 periods during spring and summer. The change of the phosphorus content influenced the calcite precipitation intensity. The sedimentation fluxes of phosphorus and the calcite precipitation were closely associated. Based on the hypothesis that calcite precipitation acts as an improvement to the trophic state by enhancing the internal phosphorus sink, this new technology for lake restoration was developed. The hypolimnetic $\text{Ca}(\text{OH})_2$ addition during summer stratification in 1996–1997 induced the calcite precipitation in the deep water layer of Basin Carwitz of Lake Schmalzer Luzin. The treatment also supported the natural calcite precipitation in the epilimnion. The annual total phosphorus content decreased from 0.46 tons in 1995 to 0.35 tons in 1997. The annual SRP content decreased from 0.02 tons in 1996 to 0.01 tons in 1997 after beginning the artificial calcite precipitation in 1996. The decrease of the annual Chl-*a* concentration in 1998 on 38% compared with that in 1996 pointed out the lake recovering. According to the one box model, the artificial calcite precipitation affected the P cycle in the lake by suppressing the P release from the sediments.

Introduction

For the restoration of eutrophic lakes, development of new technologies (ecotechnologies) based on natural mechanisms are desired. One such ecotechnology is based upon calcite precipitation, which occurs in most hardwater lakes and is considered to be a self-cleaning process due to coprecipitation of phosphorus and enhanced sedimentation (Rossknecht, 1980; Koschel et al., 1983; Murphy et al., 1983).

Although autochthonous calcite precipitation is a general phenomenon in hardwater lakes (Stabel, 1986; Thompson et al., 1997), it is not clearly understood whether the phosphorus concentration influences the intensity and dynamics of the calcite precipitation and visa versa. From one point of view, the calcite precipitation is induced by the shift of in pH caused

by photosynthesis. The enhancing of the calcite precipitation will follow the increase in photosynthesis. The higher level of photosynthesis or primary production is coupled with higher phosphorus concentration. Therefore, one expects the simultaneous increase of the phosphorus concentration and calcite precipitation. From the other point of view, it was shown in laboratory experiments that dissolved phosphorus is able to inhibit the calcite precipitation (House, 1987). This means that the calcite precipitation will be lower in the case of the higher phosphorus concentration. In this study we analyzed the concentration of phosphorus and calcite during 1991–1997 to find out which kind (acceleration/inhibition) interactions between phosphorus evolution and CaCO_3 precipitation occur in Basin Carwitz.

The natural CaCO_3 precipitation can be supported by injecting CaO or Ca(OH)_2 , increasing pH and Ca concentration and thus favoring calcite precipitation. Recently, lime (CaO , CaCO_3 , Ca(OH)_2) has been added to the surface water of several eutrophic lakes and ponds in Canada (Murphy & Prepas, 1990). The treatment was effective for controlling both phytoplankton and macrophyte biomass. However, the resulting high pH could be toxic for organisms living in the epilimnion. Another disadvantage is the difficulty to mix the lime homogeneously into the water body. Additionally, CaCO_3 precipitated near the surface may re-dissolve in the deeper water layer. Therefore, coprecipitation of P is likely more efficient in a hypolimnion with high P concentration than in a P depleted epilimnion. This is why the hypolimnetic addition of the Ca(OH)_2 has been preferred for the restoration of hardwater stratified lake.

Hypolimnetic calcite precipitation combined with aeration to homogeneously mix the added suspension of Ca(OH)_2 into the hypolimnion has been successfully tested as a restoration technology in the eutrophic hardwater Lake Schmalzer Luzin (Carwitz Basin) (Koschel et al., 1998a). The purpose of this part of the study was also to determine how the induced hypolimnetic calcite precipitation influences the phosphorus balance.

Description of sites studied

The investigations were carried out in one of two basins of Lake Schmalzer Luzin situated in north-eastern Germany, Basin Carwitz (Fig. 1). Lake Schmalzer Luzin was oligotrophic until the 1930s (Thienemann, 1925) but became eutrophic due to agricultural and municipal runoff in its catchment (Koschel et al., 1985). Morphometrical and chemical characteristics of both basins, Carwitz Basin (CB) and Middle Basin (MB), are very similar (Jordan et al., 1986; Table 1). An underwater sill at 8 m separates them. The Basin Carwitz has one outflow, the river Bäk, and its inflow is the Basin Middle. The Lake Schmalzer Luzin is rich in calcium and carbonate (Table 1) and autochthonous calcite precipitation was observed during summer stagnation, producing a high concentration of calcite (Koschel et al., 1998a).

Materials and methods

The soluble reactive phosphorus (SRP) – and calcium (Ca) – cycling of the lake were analyzed with the help of the following data sets:

1. Water column data were obtained from a biweekly or monthly monitoring program sampling at 0 m, 5 m, 10 m, 20 m, 25 m, 31.5 m depths at the deepest site. This program was initiated in 1990. SRP and CaCO_3 -content calculations for each date were based on the concentration-depth profiles and corresponding volume data. Methods for measurements of SRP, total phosphorus (TP) and calcite are described in Koschel et al. (1998a).
2. P-output: The annual P output into the river Bäk was estimated from P/Q relationships of river specific concentration P and water discharge Q and based on the continuous water discharge measurements.
3. Sediment traps were exposed for 14 days at 20 m depth during 1995–1998. The proportion of dry matter was determined after centrifugation (11 000 rpm, 10 min) and subsequent drying. The weight loss after drying at 105 °C revealed the proportion of water (dry weight). Two samples were used for CaCO_3 determination by infrared spectroscopy for CO_2 measurement after acidification. The total phosphorus content was analyzed in replicate as well (Dittrich et al., 2000).
4. Phosphorus diffuse flux from the sediment was estimated from the porewater concentration according to Fick's first law (Dittrich et al., 2000). The average flux from 1996 was used for the model calculation.

Artificial hypolimnetic calcite precipitation

Artificial hypolimnetic calcite precipitation was induced in CB by hypolimnetic injection of Ca(OH)_2 combined with aeration during the summer stratification periods in 1996, 1997 and 1998 (Koschel et al., 1998a). Two cycles of injection were carried out two to three times for approximately 3 weeks each summer (Table 2). The design and performance of the technology are described in Koschel et al. (1998a).

Model for P prediction

Expected effects of artificial calcite precipitation on the P content of Basin Carwitz were predicted with a one box model (Gächter & Wehrli, 1998). According

Table 1. Characteristics of Lake Schmalzer Luzin, the Basin Carwitz and Basin Middle (morphometric data according to Jordan et al., 1986)

Parameter	Lake Schmalzer Luzin	Carwitz Basin, (CB)	Middle Basin, (MB)
Surface (km ²)	1.34	0.58	0.76
Volume (10 ⁶ m ³)	20.6	10.5	10.1
Max. depth (m)	34	33	34
Mean depth (m)	14.7	18.1	13.3
Retention time (a)	4.4	2.2	2.2
CaCO ₃ ^a (mg l ⁻¹)		0.30	0.35
Ca ²⁺ ^a (mg l ⁻¹)		41.1	42.7
O ₂ ^b (mg l ⁻¹)		0	0

^aMean summer 0–10 m 1994.

^bMean summer, below 15 m.

Table 2. Injection of Ca(OH)₂ in Lake Schmalzer Luzin, Basin Carwitz 1996–1998

Year	Air addition		Ca(OH) ₂ addition	
	Time	Amount (m ³)	Time	Amount (t)
1996	26.06–19.07	416 000	26.06–19.07	140
	31.07–08.08		31.07–08.08	
	08.08–15.08			
	05.09–05.10		05.09–05.10	
1997	26.05–25.06	532 000	26.05–12.06	91
	20.08–14.10		20.08–03.10	

to the model, changes in the P content of the lake depend on rates of input (I), release from sediments (R), gross sedimentation (S), and output (O), where V is the lake volume and $[P]$ the average P concentration (Equation (1)):

$$V \frac{d[P]}{dt} = I + R - S - O \text{ [tons yr}^{-1}\text{]}. \quad (1)$$

In a simplified approach, output O and net sedimentation ($S-R$) are modeled as first order processes with ρ and σ as rate constants [yr^{-1}], then the P export via the outflow is defined as:

$$O = \beta \rho [P] V \text{ [tons yr}^{-1}\text{]}, \quad (2)$$

where the flushing rate ρ equals the reciprocal water residence time τ [yr] and β [–] relates the average P concentration in the outflow to the average concentration of the entire lake. P net sedimentation ($S-R$) equals:

$$S - R = \sigma [P] V \text{ [tons yr}^{-1}\text{]}, \quad (3)$$

where σ equals the sedimentation rate [yr^{-1}]. However, if $[P]$ exceeds a critical concentration $[P]^{\text{crit}}$, then net sedimentation or permanent P burial rate ($S-R$) no longer increases linearly with $[P]$ (Gächter et al., 1989). ($S-R$) rather approaches a constant value c , which likely does not depend on redox conditions at the sediment–water interface, but on the sedimentation of minerals that can precipitate or adsorb P in the anoxic conditions after the early diagenesis of the settled material. Therefore, when gross sedimentation S exceeds the maximum P retention capacity c , the excess amount R is released to the water column and P net sedimentation becomes independent of the lakes P concentration:

$$S - R = c \text{ for } [P] > [P]^{\text{crit}}. \quad (4)$$

Results

Seasonal and annual variations of phosphorus and CaCO₃ contents of the surface water (0–5 m) from

Figure 1. Map showing the locations of Lake Schmalzer Luzin.

March 1991 through December 1997 of Carwitz basin are presented in Figure 2 (A, B). The SRP content showed substantial variation during the investigated period, ranging from a maximum of 0.09 tons in 1991 to a minimum of 0.003 tons in 1997. The annual SRP content decreased from about 0.05 tons in 1991 to 0.01 tons in 1997. Although the seasonal evolution of calcite content is distinguished from that of phosphorus, it showed the same characteristic time period. Furthermore, in 1991, the maximum calcite content was estimated at about 1 ton and increased to 3.5 tons in 1994. The maximum value of calcite content (6 tons) was observed in 1995, when the maximum of SRP content was lowest. The annual SRP content decreased from 0.02 tons in 1996 to 0.01 tons in 1997 after beginning the artificial calcite precipitation in 1996. The dynamics of the CaCO_3 content indicated that calcite precipitation occurred each year over 1–3 periods during spring and summer.

The interaction between P sedimentation rate and calcite concentration is presented for 1995 in Figure 3. The typical calcite concentration pattern exhibits the

first concentration maximum in early summer during the peak of the primary production. The calcite precipitated in the surface water reached a maximum of about 2.3 mg l^{-1} at the beginning of June. A second peak (1.7 mg l^{-1}) and a third peak (1.89 mg l^{-1}) were observed in July and September 1995, respectively. Much of the calcite crystals dissolved in the CO_2 -rich hypolimnetic water and did not reach the sediment. As a result, the calcite concentration in the deep water was lower than that in epilimnion. The CaCO_3 concentration at 20 m depth amounted to a maximum of 1.02 mg l^{-1} in July 1995. Simultaneously, the P sedimentation flux exhibited a significant increase ($50 \text{ mg m}^{-2} \text{ d}^{-1}$).

The addition of Ca(OH)_2 into the hypolimnion in summer 1996–1997 increased the pH and lead to calcite precipitation (Koschel et al., 1998a). Furthermore, the treatment influenced the P concentration in basin Carwitz (Fig. 4). SRP and TP concentrations in the hypolimnion during 1995 differed from those during 1996–1997. Maximum SRP- and TP-concentrations at 25 m depth were 0.09 mg l^{-1} and 0.12 mg l^{-1} , respectively, in 1995. The autumn turnover 1995/1996

Figure 2. Soluble reactive phosphorus and calcium carbonate contents in 0–5 m in Basin Carwitz (A). The arrows indicate the $\text{Ca}(\text{OH})_2$ additions. P- and CaCO_3 -contents were calculated from the depth concentration profiles (0 m, 5 m, 10 m, 20 m, 25 m, 31.5 m) and water volume according to Jordan et al. (1986). The annual contents were estimated as average content value for the year (B).

did not occur due to ice cover. The P concentrations at 25 m depth were higher than those at 0 m depth. The warm spring 1996 lead to the thermal stratification immediately after winter stratification and as a result the mixing period did not occur in 1996. Therefore, one may expect high P accumulation in the hypolimnion, however, compared to 1995, both SRP- and TP-concentrations were lower in 1996. This trend continued through 1997, where the maximum SRP was 0.02 mg l^{-1} and TP was 0.04 mg l^{-1} at the end of the summer stagnation period at 25 m depth. As a result, the concentrations of SRP and TP in the entire water column decreased in the euphotic zone after circulation periods.

Discussion

SRP/ CaCO_3 interaction

The strong connection between calcite precipitation and trophic state hardwater lakes was previously ob-

served (Koschel, 1990; Nürnberg, 1998). While the calcite precipitation is often induced by photosynthetic activity of alga, the eutrophication (increasing of the primary production and P content) enhanced the calcite precipitation (Koschel et al. 1983; Groleau et al., 1999). However, the high eutrophic level was often characterized by low calcite precipitation. Stabel (1986) described the absence of the precipitation at high saturation indexes in Lake Constance. Our observations in Basin Carwitz during 1991–1993 support the hypothesis about the interaction of P content and the CaCO_3 precipitation. While the P content during the spring circulation was at the highest level from 1991 to 1993, the CaCO_3 content stayed at a low level. Afterwards, the P content decreased and the precipitation of calcite enhanced. The highest CaCO_3 content occurred in 1995, when the P content was at the lowest level. Therefore, the evolutions of the annual CaCO_3 and SRP content in surface water were opposite during 1991–1996 before the hypolimnetic calcite treatment began in 1996 (Fig. 2B). However, the dynamics of the evolution of calcite and phosphorus has some differences. While the SRP content sank drastically by about 80% permanently from 1991 to 1995, the calcite content changed only slightly, maximum of about 46% (1992–1993). The spring concentration of phosphorus plays a key role in the primary production during summer and consequently for the trophic state. It seems that an SRP-threshold value exists. When the spring SRP concentrations are lower than this value, the CaCO_3 precipitation in summer is enhanced. For basin Carwitz, this value amounts to approximately 0.015 mg l^{-1} . Therefore, we can conclude that the calcite precipitation is sensitive to the trophic state of the lake and to the SRP content in spring.

We did not observe the simultaneous decrease in the SRP content in the surface water and enhancement of CaCO_3 precipitation for different years. This would support the hypothesis of an inhibition effect of dissolved phosphorus on calcite precipitation (House, 1987; Kleiner, 1988). One possibility is that the differences between the concentrations are too small (0.001 mg l^{-1}) and lower than the sensitivity of our measurements. The P decrease directly after the CaCO_3 precipitation was measured in all years (Fig. 2A). Another possible interaction between phosphorus and calcite is the P-coprecipitation with calcite (Kleiner, 1988; House, 1990; Hartley et al., 1995; Danen-Louwerse et al., 1995). This process is considered to be an important ‘self-cleaning mechanism’ for lakes due to phosphorus reduction and increase in the sed-

Figure 3. Phosphorus sedimentation rate in 20 m depth and CaCO_3 concentration in water column in 0 m and 20 m depth in Basin Carwitz during 1995.

Figure 4. (A) Soluble reactive phosphorus (SRP) and (B) total phosphorus (TP) concentrations in surface (0 m) and deep water (25 m) before (1995) and during the artificial calcite precipitation (1996–1998).

imentation rates (Koschel, 1990). The literature data were fitted by Danen-Louwerse et al. (1995) as:

$$Y = 10.9X^{0.5}, \quad (5)$$

where Y is the incorporation of SRP in CaCO_3 in $\text{mg P g}^{-1} \text{Ca}$ and X is SRP concentration in mg l^{-1} . The evolution of the P-incorporation in CaCO_3 is shown in Figure 5. In this estimation, the inhibition effect was not considered. The fraction of the incorporated phosphorus increased from 1991 to 1997 and amounted to about 70% in summer 1997.

Another evidence of the coprecipitation with calcite is the occurrence of the maximum P sedimentation rate during the calcite maximum at 20 m depth. The P incorporation rate on calcite was calculated based on data from sedimentation traps and amounted to 0.72 mgP/gCa (Dittrich et al., 2001). This is in good agreement with the estimation from Equation (5). The simultaneous occurrence of the calcite maximum and phosphorus flux has been observed in the alpine Lake Bourget (Groleau et al., 1999).

Effect of artificial calcite precipitation on P cycling

Due to the hypolimnetic artificial calcite precipitation, the hypolimnion is saturated with respect to calcite and the excess calcite crystals do not dissolve in the deep water. Therefore, the hypolimnetic calcite precipitation supports the phosphorus transport with calcite from the upper water layer through the deep water to the sediment and enhances the P sedimentation rate. The artificial calcite precipitation changed the P binding forms and the proportion of the CaCO_3 -bound P binding form increased in settled material (Dittrich et al., 2001). This binding form is redox insensitive and increased the P-immobilization in the sediment.

Figure 5. Phosphorus incorporation in calcite (mg P/g Ca) calculated according to Danen-Louwerse et al. (1995) and data for SRP- and CaCO_3 -concentrations.

Table 3. Model parameters. The net sedimentation for scenario (1) equals to the gross sedimentation rate $\sigma = S/[P] * Z$, where S is the gross sedimentation, P is the P concentration and Z is the mean depth. The gross sedimentation was estimated from the sedimentation trap measurements and the P concentration was calculated for 1996. The net sedimentation in scenario (2) was assumed at $0.17 \text{ tons yr}^{-1}$ according to Koschel et al. (1998a). The P release from the sediment in scenario (3) was estimated at $2.7 \text{ g m}^{-2} \text{ yr}^{-1}$ (Dittrich et al., 2000) and $\sigma_{\text{release rate}} = R/[P] * Z$ was calculated as 3.4 yr^{-1} , where R is the P release from the sediment, P is the P concentration and Z is the mean depth

Input tons yr^{-1}	ρ yr^{-1}	$\text{pCB}_{\text{conc.}}$ mg m^{-3}	$\text{pOutflow}_{\text{conc.}}$ mg m^{-3}	β $\text{pCB}_{\text{conc.}}/\text{pOutflow}_{\text{conc.}}$	Gross Sed. $\text{g m}^{-2} \text{ yr}^{-1}$	σ yr^{-1}
0.12	0.45	43	30	0.70	3.1	4.0

Figure 6. Observations (dots) and model predictions (lines) of the total P content in the basin Carwitz. The dotted line assumes no P release from sediments, $R=0$ and $S=N$. Prediction presented by solid thick line assumes a constant net sedimentation (0.17 tons yr). The solid thin line assumes that the calcite precipitation affect the P release from sediment $S = \sigma[P]V$ and $R = \sigma_{\text{release rate}}[P]V$ (Table 3).

Figure 7. TP content in the deep layers 20–33 m in the basin Carwitz.

The influence of the treatment on the P budget was analysed by the balance approach. If the artificial calcite precipitation would really suppress the P release from the sediment, then according to Equation (1), the P content of the lake had responded drastically to the treatment. We assume that the average external loading with P was $0.12 \text{ tons yr}^{-1}$ since 1996, and that the yearly gross sedimentation S and export via the outflow can be described by the first order reactions $S = \sigma[P]V$ and $O = \beta\rho[P]V$ (Table 3). With these boundary conditions we can predict the development of the P content for three scenarios since 1996:

1. After the onset of the treatment, the release of P from the sediment was zero (net sedimentation equaled the gross sedimentation).
2. Net sedimentation was unaffected by treatment.
3. Net sedimentation was affected by treatment and the diffuse flux defined the P release from the sediment.

In contrast to the scenarios that assume a constant net sedimentation or zero P release from the sediment, the scenario with an influence of net sedimentation via P release from the sediment agrees well with the observed data (Fig. 6). This conclusion is also supported by the observation of the hypolimnetic P accumulation (Fig. 7). The highest accumulation (0.22 tons) of the TP content was from 0.07 tons at the end of April to 0.296 tons at the beginning of November in 1993. The accumulation of TP was about 0.1 tons in 1995 before the treatment. After the artificial calcite precipitation, there was almost no phosphorus accumulation in the hypolimnion (0.01 tons). This caused decrease of the phosphorus content in Lake Schmalzer Luzin and lead to the change of the phytoplankton quality and quantity. In comparison to 1996, the dominance of the blue

alga shifted to the diatoms (Koschel et al., 1998b). The annual Chl-*a* concentration decreased on 38% from 0.005 mg l^{-1} to 0.003 mg l^{-1} in 1998 pointing out the lake recovering (Koschel et al., 1998b).

Conclusion

Calcite precipitation in Lake Schmalzer Luzin occurred each year with reproducible intensity. The maximum concentration of calcite occurs simultaneously with phosphorus fluxes. In this way, the calcite precipitation forced the transport of phosphorus to the sediment and, therefore, it contributed to the recovery of a mesotrophic status of the lake. However, the majority of the calcite dissolved in the anoxic, CO_2 -rich hypolimnion and not all coprecipitated phosphorus reached the sediment. To support this interaction between calcite and phosphorus, the artificial calcite precipitation was induced in the hypolimnion of Lake Schmalzer Luzin during the stratification period from 1996 to 1997. The artificial hypolimnetic calcite precipitation was combined with aeration to homogeneously mix the added suspension of Ca(OH)_2 . The treatment lead to the decrease of the P content in the whole basin. The annual concentration of total phosphorus in the euphotic zone of the Carwitz Basin decreased from 0.038 mg l^{-1} (1992–1994) to 0.02 mg l^{-1} (1996–1997). Consequently, the annual Chl-*a* in 1998 was about 38% of that at the beginning of the hypolimnetic treatment in 1996. The decrease in the hypolimnetic phosphorus accumulation after the artificial calcite precipitation suggested a decrease in P release from the sediment.

According to the balance model, the artificial calcite precipitation critically influenced the net sedimentation. The scenario that assumes the constant net sedimentation and the scenario that assumes zero P re-

lease from the sediment underestimated the P content values. The best fit of the measurements was found by assuming the dependence of the gross sedimentation and P release on the P content.

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