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#### A mesocosm study

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The combined effects of macrophytes (*Vallisneria denseserrulata*) and a lanthanum-modified bentonite on water quality of shallow eutrophic lakes: a mesocosm study

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Running Title: The Combined Effects of Macrophyte and Phoslock®

# 1 Abstract

2 Establishment of submerged macrophyte beds and application of chemical phosphorus 3 inactivation are common lake restoration methods for reducing internal phosphorus loading. 4 The two methods operate via different mechanisms and may potentially supplement each 5 other, especially when internal phosphorous loading is continuously high. However, their 6 combined effects have so far not been elucidated. Here, we investigated the combined impact 7 of the submerged macrophyte Vallisneria denseserrulata and a lanthanum-modified bentonite 8 (Phoslock®) on water quality in a 12-week mesocosm experiment. The combined treatment 9 led to stronger improvement of water quality and a more pronounced reduction of porewater 10 soluble reactive phosphorus than each of the two measures. In the combined treatment, total 11 porewater soluble reactive phosphorus in the top 10 cm sediment layers decreased by 78% 12 compared with the control group without Phoslock<sup>®</sup> and submerged macrophytes. Besides, in 13 the upper 0-1 cm sediment layer, mobile phosphorus was transformed into recalcitrant forms 14 (e.g. the proportion of HCl-P increased to 64%), while in the deeper layers, 15 (hydr)oxides-bound phosphorus species increased 17-28%. Phoslock<sup>®</sup>, however, reduced the 16 clonal growth of V. denseserrulata by 35% of biomass (dry weight) and 27% of plant density. 17 Our study indicated that Phoslock<sup>®</sup> and submerged macrophytes may complement each other 18 in the early stage of lake restoration following external nutrient loading reduction in 19 eutrophic lakes, potentially accelerating the restoration process, especially in those lakes 20 where the internal phosphorus loading is high.

21

22 *Keywords:* Restoration, Phosphorus, Eutrophication, Sediments

23

# 24 **1. INTRODUCTION**

25 Eutrophication, occurring mainly as a result of excessive loading of nutrients such as 26 nitrogen and phosphorus, is a severe problem worldwide (Downing, 2014; Smith and 27 Schindler, 2009). Due to the controllability and effectiveness, reducing phosphorus inputs has 28 generally been accepted as a key method to mitigate lake eutrophication (Schindler et al., 29 2008, 2016). However, numerous studies have affirmed that lake recovery after control of the 30 external phosphorus loading is often delayed (Cooke et al., 2005; Jeppesen et al., 1991), 31 which is in part due to phosphorus release from the sediment (internal loading) maintaining 32 high water phosphorus concentrations and supporting continued algal growth (Søndergaard et 33 al., 2003; Spears et al., 2012). Accordingly, reduction of the internal loading has become a 34 key challenge in lake eutrophication reversal. In the past decades, various methods to reduce 35 internal loading have been tested in laboratory experiments, followed by full-scale field 36 applications. These include biomanipulation ("top-down" control) (Jeppesen et al., 1997, 37 2012), aquatic plant community restoration (Liu et al., 2018; Søndergaard et al., 2000), 38 sediment dredging (Van der Does et al., 1992), sediment oxidation (Ripl, 1976), and chemical 39 phosphorus inactivation (Hansen et al., 2003; Huser et al., 2016; Zamparas and Zacharias, 40 2014).

41 Submerged macrophytes play an important role in the phosphorus cycling process in 42 lakes. They can enhance the phosphorus cycling by mobilizing phosphorus from the sediment 43 through rhizosphere acidification (Long et al., 2008), but they can also reduce the phosphorus 44 release by oxidizing the rhizosphere and metals such as iron (Fe) and manganese (Mn) 45 (Christensen et al., 1997). (Hydr)oxides of these metals are coprecipitated with phosphorus 46 and deposited on the root surface in the form of plaques with high specific surfaces and 47 affinity for adsorbing phosphorus (St-Cyr et al., 1993; Wang et al., 2013). In addition, 48 submerged macrophytes can enhance sedimentation (Barko and James, 1998), deplete labile

49 phosphorus pools in the sediment (Barko and James, 1998), and take up nutrients from the 50 water column (Bole and Allan, 1978; Carignan and Kalff, 1980), thereby transferring the 51 bioavailable phosphorus from the environment into plant tissue. Phosphorus in healthy tissues 52 is rarely released until plant decomposition (Barko et al., 1991; Barko and Smart, 1980).

53 Given the overall favorable effects of submerged macrophytes on reducing the internal 54 loading and their structural role in shallow lake ecosystems, transplantation of submerged 55 macrophyte stands has been recognized as a possibly effective management tool (Barko and 56 James, 1998; Liu et al., 2018), but the effect may be hampered by continuous high internal 57 loading. Chemical intervention might, therefore, be considered as a supplementary tool. 58 Phoslock<sup>®</sup>, a lanthanum-modified bentonite (LMB), developed by the Australian government 59 agency Commonwealth Scientific and Industrial Research Organisation (CSIRO) in the 60 1990s, is one of the most used techniques for chemical phosphorus immobilization (Bishop et 61 al., 2014; Douglas et al., 1999; Lürling and van Oosterhout, 2013; Meis et al., 2013). Previous studies showed Phoslock<sup>®</sup> to be highly efficient at reducing phosphorus 62 63 concentrations in the water column (Crosa et al., 2013; Marquez-Pacheco et al., 2013) and 64 inactivating phosphorus in sediments (Bishop et al., 2014; Meis et al., 2012) over a wide 65 range of physico-chemical conditions in both lab experiments and natural water bodies. 66 However, confounding factors reducing the effect of phosphorus binding have also been 67 reported (e.g. dissolved organic carbon (DOC), high pH (>9)) (Lürling et al., 2014; Reitzel et al., 2013a; Ross et al., 2008). Furthermore, several studies have combined Phoslock® with 68 69 other chemical capping agents (e.g. polyaluminiumchloride (PAC), iron (III) chloride) (Lürling et al., 2016; Waajen et al., 2016b). However, studies combining Phoslock<sup>®</sup> with 70 71 submerged macrophytes are scarce (Waajen et al., 2016a).

72 The mechanisms of submerged macrophytes and Phoslock<sup>®</sup> in reducing internal

73 phosphorus loading differ, and their combined effects on water quality improvement are not 74 clear. Since submerged macrophytes (e.g. Myriophyllum spicatum, Hydrilla verticillata, 75 *Vallisneria spiralis*) take up phosphorus from both the sediment and the water column (Bole 76 and Allan, 1978; Gentner, 1977), mainly from the sediment (Christiansen et al., 2016), the 77 decreased phosphorus availability caused by Phoslock<sup>®</sup> may have negative effects on the 78 growth of these plants. Conversely, the reduced algal biomass in the overlying water after 79 Phoslock<sup>®</sup> addition may provide a better light environment for macrophyte growth (Gunn et al., 2013). To test the combined effects of submerged macrophytes and Phoslock<sup>®</sup> on the 80 81 water quality and the influence of Phoslock<sup>®</sup> on submerged macrophyte growth, we 82 conducted a 12-week mesocosm experiment. Vallisneria denseserrulata, a common perennial 83 meadow-forming species in shallow lakes in China and often used in lake restoration (Liu et 84 al., 2018; Zhou et al., 2016), was chosen for the experiment. We hypothesized that Phoslock<sup>®</sup> 85 treatment in combination with transplantation of submerged macrophytes would complement 86 each other via different mechanisms in reducing the internal phosphorus loading in the early 87 stage of lake restoration.

88

89 2. MATERIALS AND METHODS

### 90 2.1. Experimental set-up

The mesocosm experiment was conducted from August to November 2018 at Dongshan station located at Taihu Lake ecosystem research station near Taihu Lake, Suzhou City (China), and the set-up involved four treatments: (1) control group without Phoslock<sup>®</sup> and macrophyte, (2) Phoslock<sup>®</sup> added; (3) *V. d. (V. denseserrulata)* planted, (4) Phoslock<sup>®</sup> added and *V. d.* planted. *V. denseserrulata* were procured from ponds of Belsun Aquatic Ecology Science and Technology Ltd. Each treatment consisted of four replicate barrels (top diameter

97 84 cm, bottom diameter 66 cm, height 83 cm). All barrels were filled with a 20-cm mixed 98 sediment layer collected from the pond at Dongshan Town near Taihu Lake (Table 1) and 99 50-cm overlying water (water volume 227 L) pumped from the pond nearby. All the barrels 100 were situated in a pond statically. The treatments were randomly assigned to barrels. A week 101 after the initiation of the experiment, sediment cores and overlying water were sampled for 102 analysis of phosphorus speciation and water chemistry. Then, four V. denseserrulata shoots 103 with a wet weight of  $6.40\pm0.46$  g and a length of  $40.4\pm2.9$  cm were transplanted into each 104 barrel of the V. d. treatment and the Phoslock<sup>®</sup>+V. d. treatment. Three hundred and ninety 105 grams Phoslock<sup>®</sup> was mixed into slurry with 2 L overlying water and then added to the water 106 surface in each barrel of the Phoslock<sup>®</sup> treatment and the Phoslock<sup>®</sup> + V. d. treatment, 107 corresponding to a Phoslock<sup>®</sup>: P<sub>mob</sub> (mobile phosphorus) mass ratio of 100:1. Subsequently, 108 the mesocosms were incubated for 12 weeks. The P<sub>mob</sub> pool was calculated as the sum of 109 potentially mobile phosphorus consisting of porewater phosphorus, phosphorus bound to 110 reducible Fe/Mn, and labile organic phosphorus (i.e. H<sub>2</sub>O-P, BD-P, NaOH-OP). NaOH-OP is 111 organic phosphorus in the extractant of sediment treated with NaOH (see 2.2.4).

112 2.2. Sampling and measurements

#### 113 2.2.1 Water samples

The pH and temperature of the water column were measured by portable multiparameter water monitoring probes (Aquread AP-2000, UK) (Fig. S1, S2) every two weeks. Water samples were collected every two weeks and analyzed for total phosphorus (TP), total nitrogen (TN), and chlorophyll a (Chl.a). TP rather soluble reactive phosphorus (SRP) was taken as a key parameter that reflecting the effects resulted from Phoslock<sup>®</sup> and *V*. *denseserrulata* in reducing phosphorus concentrations. The changes in SRP concentrations in the water column are the result of the dual effects of the uptake by algae and submerged 121 macrophytes and sediment release. The part absorbed by algae will occur as particulate 122 phosphorus. In addition, sediments may also release dissolved organic phosphorus forms that 123 are only measured after wet oxidation (Jensen et al., 2017). These forms also show up in TP 124 analyses but not in SRP analysis. However, we put SRP figure in the supplementary material 125 file to give a more complete understanding (Fig. S3). TP and TN concentrations were 126 spectrophotometrically determined after digestion with K<sub>2</sub>S<sub>2</sub>O<sub>8</sub> and H<sub>2</sub>SO<sub>4</sub> at 120 °C for 30 127 min, as described in Jin and Tu (1990). Chl.a was measured spectrophotometrically from the 128 matter retained on a GF/C filter after extraction in a 90% (v/v) ethanol/water solution (Chen 129 and Gao, 2000).

130

# 131 2.2.2 Light attenuation

132 Light intensity (in  $\mu$ mol quanta m<sup>-2</sup> s<sup>-1</sup>) was measured using an underwater 133 photosynthetically active radiation meter (Apogee MQ-510, USA) at a depth of 0.3 m (near 134 the top of the plant shoots) every two weeks, and the vertical light attenuation coefficient (K<sub>d</sub>) 135 (in m<sup>-1</sup>) was calculated by the equation (1) (McPherson and Miller, 1987):

136

$$K_d = \ln(I_0/I_z)/z \tag{1}$$

137 where  $I_0$  is light intensity at the water surface,  $I_z$  is light intensity at depth z, and z is the depth 138 where measurements were made (in m).

139

# 140 2.2.3 Porewater soluble reactive phosphorus

Porewater samples were gathered every four weeks with HR-Peeper probes (vertical resolution of 5.0 mm, www.easysensor.net). The probes were randomly inserted into the barrels and left for 48 h to equilibrate. After retrieval, the sediment solids adhering to the surfaces of the probes was wiped off and the probes were rinsed with deionized water. 145 Samples were then immediately analyzed for SRP according to a miniaturized photometrical 146 method described in Laskov et al. (2007). Besides from presenting porewater SRP files the 147 total SRP content (mg m<sup>-2</sup>) in the surface 10 cm sediment layers was calculated by the 148 equation (2):

149

$$SRP_{total} = \sum_{1}^{20} C_i \cdot \left(\frac{M_i}{D}\right) / S$$
<sup>(2)</sup>

where i is the number of the layer and there are 20 layers in the 10 cm sediment;  $C_i$  is the concentration of SRP in each layer (in mg mL<sup>-1</sup>);  $M_i$  is the mass of porewater in each layer of the sediment core and equals the wet weight of sediment minus the dry weight of sediment (in g); D is the density of porewater, i.e. 1 g ml<sup>-1</sup>; and S is the area of the cross section of the sediment core (in m<sup>2</sup>).

155

### 156 2.2.4 Sediment characteristics

157 One sediment core from each barrel (16 cores in all) was sampled by a lucite tube 158 (internal diameter 36 mm) at both the beginning and at the end of the experiment. The initial 159 sediment cores were collected and the 0-5 cm sediment in each core was mixed to analyze 160 phosphorus fractions and calculate P<sub>mob</sub> (Rydin, 2000), while the upper 8 cm of the terminal 161 cores were sliced at 1 cm intervals to investigate the changes of phosphorus forms with depth. 162 TP in the sediments (0.5 g DW) was determined following ignition of the sediment at 550 °C 163 and subsequent digestion in 1 M HCl (50 ml) (Aspila et al., 1976). Identification of major 164 pools of phosphorus in the sediments was made following the sequential extraction scheme 165 modified by Paludan and Jensen (1995). Labile phosphorus was extracted from 1 g wet 166 sediment by H<sub>2</sub>O; reducible Fe and Mn hydroxide-bound phosphorus were extracted with BD 167 reagent (bicarbonate-dithionite); metal oxide-bound phosphorus (NaOH-IP) and labile 168 organic phosphorus (NaOH-OP) were extracted with 0.1 M NaOH; and inorganic phosphorus pools, e.g., CaCO<sub>3</sub>-bound phosphorus were extracted with 0.5 M HCl. Residual phosphorus
was calculated as TP minus the sum of the extracted phosphorus pools. The concentration of
each phosphorus fraction was converted to dry matter by the equation (3):

172 
$$C_{P(DW)} = \frac{C \cdot V}{m_w \cdot (1 - water \ content)}$$
(3)

173 where  $C_{P(DW)}$  is the concentration of phosphorus fractions in dry matter (in mg g DW<sup>-1</sup>); C is 174 the concentration of phosphorus in the extractant (in mg L<sup>-1</sup>); V is the volume of extractant 175 (in L); m<sub>w</sub> is the wet weight of sediment (in g); water content = (wet weight – dry weight)/ 176 wet weight. Dry weight was measured after drying the sediment at 105 °C for 24 h.

177

# 178 2.2.5 Macrophyte traits

179 Macrophyte (V. denseserrulata) traits (i.e. biomass, length, shoot number) were 180 determined at the start and at the end of the experiment. Also, at the start, an additional 10 181 shoots were chosen to measure the water content, which was used to calculate the initial dry 182 weight. At the end of the experiment, all plants were uprooted by hand and rinsed carefully to 183 remove attached material on leaves and roots. Dry weight (biomass dried at 45 °C) and 184 physical dimensions were estimated using an electronic balance (to the nearest 0.01 g) and 185 ruler (to the nearest 1 mm), respectively. The relative growth rate (RGR) of the plant in each 186 barrel was calculated using the equation (4) (Hunt, 1982):

187

RGR (d<sup>-1</sup>) = ln 
$$\left(\frac{W_{\rm f}}{W_{\rm i}}\right)$$
/days (4)

188 where  $W_f$  (g) and  $W_i$ (g) are the final and initial total biomass (DW) in each barrel, 189 respectively.

190

#### 191 2.3. Statistical analysis

192 The effects of Phoslock<sup>®</sup> and V. denseserrulata on the water chemistry and light

193 attenuation coefficient versus time were analyzed by repeated measures analysis of variance 194 (rm-ANOVA) in SPSS 20.0. If the assumption of sphericity was violated, we used the 195 Greenhouse-Geissler correction of the degrees of freedom when the epsilon was <0.75 and 196 the Huynh-Feldt correction of the degrees of freedom when the epsilon was >0.75 (Lürling 197 and Faassen, 2012). Two-way ANOVA was used to identify the effects of Phoslock<sup>®</sup> and V. 198 denseserrulata on SRP concentrations, and depth was set as a random factor. If a significant 199 interaction was observed, a simple effects test (Bonferroni method) was conducted to identify 200 where the difference occurred. One-way ANOVA followed by post hoc test (Tukey method) 201 was conducted to analyze the difference in SRP<sub>total</sub> in the surface 10 cm sediment between each of the two treatments. t-test was applied to elucidate the effects of  $Phoslock^{(0)}$  on V. 202 203 *denseserrulata* traits. The level of significance was set to p < 0.05 for all tests.

204

#### **205 3. RESULTS**

#### 206 3.1. Water quality and light condition

Both Phoslock<sup>®</sup> and V. denseserrulata significantly reduced TP concentrations in the 207 208 water column (Table S1, p<0.001 for both), and the most obvious reduction of TP was observed in the Phoslock<sup>®</sup> + V. d. treatment (Fig. 1). However, Phoslock<sup>®</sup> significantly 209 210 increased TN concentrations while V. denseserrulata significantly reduced TN concentrations 211 relative to the control (Fig. 1; Table S1, p=0.002 and p=0.009, respectively). Demonstrating a 212 similar pattern as that of TP, the effects of Phoslock<sup>®</sup> and V. denseserrulata on the reduction 213 of Chl.a were statistically significant (Table S1, p=0.008 and 0.012, respectively), and the 214 most rapid and obvious reduction of Chl.a was observed in the Phoslock<sup>®</sup> + V. d. treatment 215 (Fig. 1). V. denseserrulata significantly reduced K<sub>d</sub> while Phoslock<sup>®</sup> had no effect (Fig. 1; 216 Table S1, p<0.001 and p=0.076, respectively). Using mean TP and Chl.a concentrations

during the experiment period, reduction rates were calculated by comparing the different treatments with the controls (Fig. S4). The reduction rates of TP and Chl.a concentrations in the combined treatment was higher than that in the single treatments, however, the combined effects were not additive (less than the sum of the two single effects).

- 221
- 222

# 3.2. Phosphorus in the sediment

During the experiment, both Phoslock<sup>®</sup> and V. denseserrulata significantly reduced the 223 224 porewater SRP concentrations (Fig. 2; Table S2, p<0.001 at three time points), interaction 225 being observed only in week 4 (Table S2, p<0.001). In the two treatments with Phoslock<sup>®</sup>, 226 SRP concentrations in the mesocosms with V. denseserrulata were significantly lower than in 227 those without V. denseserrulata (Bonferroni test, p<0.001). In the two treatments without Phoslock<sup>®</sup>, SRP concentrations did not differ significantly in either the with- or the without- V. 228 229 denseserrulata mesocosms (p=0.825). At the end of the experiment, total SRP in the surface 230 10 cm sediment layers had decreased by 78% in the Phoslock<sup>®</sup> + V. d. treatment compared 231 with the control group without Phoslock<sup>®</sup> and macrophytes (Tukey test, p<0.001), while in 232 the Phoslock<sup>®</sup> treatment and the V. d. treatment it had decreased by 35% and 33%, 233 respectively (p=0.033, 0.046, respectively). No significant difference appeared between the 234 two single treatments (p=0.996) (Fig. 3).

235

In the Phoslock<sup>®</sup> treatment, the most obvious changes in phosphorus fractions were observed in the 0-1 cm layer where HCl-P increased to 0.51 mg gDW<sup>-1</sup> and became the major pool (accounting for 68% of TP), while other potentially mobile phosphorus fractions (H<sub>2</sub>O-P, BD-P, NaOH-P) decreased compared with the control group without Phoslock<sup>®</sup> and macrophyte (Fig. 4). In contrast to the Phoslock<sup>®</sup> treatment, metal (hydr)oxides-bound phosphorus (i.e. BD-P and NaOH-IP) in the surface sediment layer in the *V. d.* treatment increased by 50% and HCI-P decreased by 20% compared with the control group. BD-P and
NaOH-IP also increased in the deeper sediments compared with the control group. In the
treatment with both Phoslock<sup>®</sup> and *V. denseserrulata*, HCI-P increased to 0.53 mg gDW<sup>-1</sup> and
constituted 64% of TP in the 0-1 cm layer. However, BD-P in the sediments below 3 cm
exhibited an increase within the range of 17% to 28% compared with the control group (Fig.
4).

248

249

### 49 3.3. Submerged macrophyte traits

Compared with the *V. d.* treatment, the biomass and density of plants significantly decreased by 35% and 27% (p=0.002, 0.009, respectively) in the Phoslock<sup>®</sup> + *V. d.* treatment, respectively, whereas no significant changes occurred in individual weight (p=0.098) (Fig. 5a-c). RGR decreased markedly by 17% (p=0.002) (Fig. 5d). The biomass and total length of stolons decreased significantly by 24% and 30% (p=0.010, 0.019, respectively) (Fig. 5e, 5f).

255

#### 256 4. **DISCUSSION**

# 257 4.1. Effects of Phoslock<sup>®</sup> and submerged macrophytes on phosphorus and nitrogen 258 concentrations

The Phoslock<sup>®</sup>+*V. d.* treatment led to a much stronger improvement of water quality than if the two measures were used alone, since the combined treatment had stronger effects on phosphorus in both the water column and the sediment. However, water TP decreased and clarity increased over time in all treatments and in the control, which can be explained by both clam water conditions (no stirring) and decreasing water temperature over the course of the experiment (Fig. S2).

265 In the two Phoslock<sup>®</sup> treatments, Phoslock<sup>®</sup> not only led to fast removal of phosphorus

266 from the water column during the addition, it also capped phosphorus on the surface of the 267 sediment, retarding the internal phosphorus loading. The capping layer depleted the SRP 268 diffused from deep sediment. At the end of the experiment, however, total SRP in the top 10 cm sediment of Phoslock<sup>®</sup> treatment had decreased only by 35% compared with the control 269 270 group without Phoslock<sup>®</sup> and macrophytes, while total SRP in the Phoslock<sup>®</sup>+V. d. treatment 271 had decreased by 78%, indicating that the combination of Phoslock<sup>®</sup> and macrophytes had a stronger efficiency than if Phoslock<sup>®</sup> was used alone. This likely reflects that V. 272 273 denseserrulata enhanced the P-binding capacity by oxidizing metals in the deep sediment, 274 adsorbing more porewater SRP and thus increasing the content of metal (hydr)oxides-bound 275 phosphorus species as detected. Moreover, submerged macrophytes can also take up 276 porewater SRP by root for growth (Christiansen et al., 2016).

277 In the two treatments with Phoslock<sup>®</sup>, the strong transformation of phosphorus forms in 278 the top layers was in line with previous studies on Phoslock<sup>®</sup> application (Bishop et al., 2014; 279 Meis et al., 2012; Reitzel et al., 2013b). However, BD-P in the deep sediment layers in the combined treatment increased relative to the treatment with Phoslock<sup>®</sup> implemented alone. In 280 281 the upper sediment layer, the significant decrease of BD-P and NaOH-IP and the increase of 282 the HCl-P pool not only indicate a stronger binding capacity of Lanthanum (La) with 283 phosphorus compared with metal (hydr)oxides, but also phosphorus re-adsorption onto 284 available La during the sequential phosphorus extraction by the BD and NaOH solution 285 (Reitzel et al., 2013b). Furthermore, since BD-P is sensitive to redox and can be released under anoxia or low redox conditions (Boström et al., 1988), the surface Phoslock<sup>®</sup> layer will 286 287 be capable of re-adsorbing the phosphorus released from BD-P in the deeper sediments when 288 reductive conditions occur (Reitzel et al., 2013b). Submerged macrophytes release oxygen 289 produced during photosynthesis into sediments through their roots (Santner et al., 2015),

which leads to oxidation of the metals and thus increase the phosphorus binding capacity.
Hence, influenced by both Phoslock<sup>®</sup> and macrophytes, the Phoslock<sup>®</sup>+*V*. *d*. treatment had
the lowest phosphorus concentration in the water column.

However, a side effect of Phoslock<sup>®</sup> in the form of increased nitrogen efflux appeared, possibly reflecting the addition of ammonium with Phoslock<sup>®</sup> (Reitzel et al., 2013b; van Oosterhout and Lürling, 2013). Moreover, the clonal growth of *V. denseserrulata* led to a remarkable reduction in TN relative to the control treatment. This may result from absorption of nitrogen by the plants or enhanced nitrification and denitrification (Barko and James, 1998; Reddy et al., 1989). Nevertheless, the changes in nitrogen species are a topic warranting further studies as we did not study the nitrogen-cycling in detail in present study.

Thus, compared with the single treatment, the combined treatment had two ways of binding phosphorus and retarding the phosphorus release into the water column, which is more conducive to reducing internal phosphorus loading. In addition, *V. denseserrulata* can offset the effect of Phoslock<sup>®</sup> on the nitrogen increase, as shown in the Phoslock<sup>®</sup> + *V. d.* treatment.

305

# 306 4.2. Effects of Phoslock<sup>®</sup> on submerged macrophyte growth

Being one of the fundamental factors for photosynthesis, light plays an important role for plant growth. In this study, however, the application of Phoslock<sup>®</sup> improved light conditions only insignificantly, indicating that light was not the major influencing factor for macrophyte growth. Therefore, the recorded negative effect on submerged macrophyte clonal growth might be related to the reduction of bioavailable phosphorus in the surface sediment. According to our observations, the clonal growth of *V. denseserrulata* was through elongation of the stolon from the leaf sheath of the mother plant near the sediment-water interface, 314 followed by growth of leaves and roots from the apex of the stolon and formation of a new 315 ramet. Then the roots kept growing and penetrated into the deep sediment. In this study, 316 phosphorus fractions transformed mainly in the top 1 cm, and P<sub>mob</sub> consisting of H<sub>2</sub>O-P, BD-P 317 and NaOH-OP declined to 0.08 mg gDW<sup>-1</sup>, accounting for only 10% of TP compared with 318 0.20 mg gDW<sup>-1</sup> in the control group without Phoslock<sup>®</sup> and macrophytes. Since submerged 319 macrophytes can absorb phosphorus by roots and shoots (Gentner, 1977), and mainly through 320 roots (Christiansen et al., 2016), the low content of P<sub>mob</sub> in the top sediment seems to be 321 unfavorable to the new ramets in their early life stage. With the slower growth of new ramets, 322 clonal growth was overall inhibited and, eventually, the shoot density and total biomass of V. denseserrulata decreased. However, for the individual plant in the Phoslock<sup>®</sup> + V. d. 323 324 treatment,  $P_{mob}$  was not significantly different from the V. d. treatment when its roots 325 elongated into the sediments below 1 cm, and in its later life stages it can obtain a similar 326 level of phosphorus as in the V. d. treatment. However, the long-term (say >1 year) effects of 327 Phoslock<sup>®</sup> on submerged macrophytes require further studies.

328

# 329 **5.** Conclusion

In this study, the largest improvement in water quality was observed in the Phoslock<sup>®</sup> + V. 330 331 d. treatment; thus, using the methods in combination had a stronger effect than using them 332 individually. The combined treatment led to the most significant and dramatic decrease in 333 porewater SRP, and total SRP in the top 10 cm sediment layers decreased by 78% compared with the control group without Phoslock<sup>®</sup> and macrophytes. In the 0-1 cm sediment layer, 334 335 HCl-P increased to 0.53 mg gDW<sup>-1</sup> and constituted 64% of TP, and BD-P in the sediment 336 below 3 cm increased 17-28%. The phosphorus inactivation by  $La^{3+}$  in the surface layer as 337 well as the oxidization of metals by roots likely increased the P-binding capacity in the

338 sediment. Additionally, Phoslock<sup>®</sup> had a negative effect on *V. denseserrulata* growth, mainly 339 clonal growth with a decrease by 35% in biomass (dry weight) and 27% in plant density, 340 whereas the impact on individual weight was negligible, which likely can be ascribed to 341 phosphorus inactivation in the surface sediment. Hence, Phoslock<sup>®</sup> and submerged 342 macrophytes may complement each other in the early stage of lake restoration following 343 external nutrient loading reduction, potentially accelerating the restoration process in 344 eutrophic lakes, especially those where the internal phosphorus loading is high.

345

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# 356 References

Aspila, K.I., Agemian, H., Chau, A.S., 1976. A semi-automated method for the determination
of inorganic, organic and total phosphate in sediments. Analyst 101, 187-197.
https://doi.org/10.1039/an9760100187

Barko, J.W., Gunnison, D., Carpenter, S.R., 1991. Sediment interactions with submersed
macrophyte growth and community dynamics. Aquat. Bot. 41, 41-65.
https://doi.org/10.1016/0304-3770(91)90038-7

363 Barko, J.W., James, W.F., 1998. Effects of submerged aquatic macrophytes on nutrient

- 364 dynamics, sedimentation, and resuspension, in: Jeppesen, E., Søndergaard, M., 365 Søndergaard, M., Christoffersen, K. (Eds.), The Structuring Role of Submerged 366 Lakes. 197-214. Macrophytes in Springer, New York, NY, pp. 367 https://doi.org/10.1007/978-1-4612-0695-8\_10
- Barko, J.W., Smart, R.M., 1980. Mobilization of sediment phosphorus by submersed
  freshwater macrophytes. Freshwat. Biol. 10, 229-238.
  https://doi.org/10.1111/j.1365-2427.1980.tb01198.x
- Bishop, W.M., McNabb, T., Cormican, I., Willis, B.E., Hyde, S., 2014. Operational
  evaluation of phoslock phosphorus locking technology in Laguna Niguel Lake,
  California. Water Air Soil Pollut. 225. https://doi.org/10.1007/s11270-014-2018-6
- Bole, J., Allan, J., 1978. Uptake of phosphorus from sediment by aquatic plants, *Myriophyllum spicatum* and *Hydrilla verticillata*. Water Res. 12, 353-358.
  https://doi.org/10.1016/0043-1354(78)90123-9
- Boström, B., Andersen, J.M., Fleischer, S., Jansson, M., 1988. Exchange of phosphorus
  across the sediment-water interface, in: Persson, G., Jansson, M. (Eds.), Phosphorus in
  Freshwater Ecosystems. Springer, Dordrecht, pp. 229-244.
  https://doi.org/10.1007/978-94-009-3109-1\_14
- Carignan, R., Kalff, J., 1980. Phosphorus sources for aquatic weeds: water or sediments?
  Science 207, 987-989. https://doi.org/10.1126/science.207.4434.987
- Chen, Y., Gao, X., 2000. Comparison of two methods for phytoplankton chlorophyll-a
  concentration measurement. J. Lake Sci. 12, 185-188. (in Chinese)
  https://doi.org/10.18307/2000.0215
- Christensen, K.K., Andersen, F.O., Jensen, H.S., 1997. Comparison of iron, manganese, and
  phosphorus retention in freshwater littoral sediment with growth of *Littorella uniflora*and benthic microalgae. Biogeochemistry 38, 149-171.
  https://doi.org/10.1023/A:1005736930062
- Christiansen, N.H., Andersen, F.Ø., Jensen, H.S., 2016. Phosphate uptake kinetics for four
   species of submerged freshwater macrophytes measured by a <sup>33</sup>P phosphate radioisotope
   technique. Aquat. Bot. 128, 58-67. https://doi.org/10.1016/j.aquabot.2015.10.002
- Cooke, G.D., Welch, E.B., Peterson, S., Nichols, S.A., 2005. Restoration and management of
  lakes and reservoirs, 3th ed. CRC press, Boca Raton, Florida.

- Crosa, G., Yasseri, S., Nowak, K.-E., Canziani, A., Roella, V., Zaccara, S., 2013. Recovery of
  Lake Varese: reducing trophic status through internal P load capping. Fundam. Appl.
  Limnol. 183, 49-61. https://doi.org/10.1127/1863-9135/2013/0427
- Douglas, G., Adeney, J., Robb, M., 1999. A novel technique for reducing bioavailable
   phosphorus in water and sediments, International Association Water Quality Conference
   on Diffuse Pollution.
- 401 Downing, J.A., 2014. Limnology and oceanography: two estranged twins reuniting by global
  402 change. Inl. Waters 4, 215-232. https://doi.org/10.5268/Iw-4.2.753
- 403 Gentner, S.R., 1977. Uptake and transport of iron and phosphate by *Vallisneria spiralis* L.
  404 Aquat. Bot. 3, 267-272. https://doi.org/10.1016/0304-3770(77)90028-6
- Gunn, I.D.M., Meis, S., Maberly, S.C., Spears, B.M., 2013. Assessing the responses of
  aquatic macrophytes to the application of a lanthanum modified bentonite clay, at Loch
  Flemington, Scotland, UK. Hydrobiologia 737, 309-320.
  https://doi.org/10.1007/s10750-013-1765-5
- Hansen, J., Reitzel, K., Jensen, H.S., Andersen, F.Ø., 2003. Effects of aluminum, iron,
  oxygen and nitrate additions on phosphorus release from the sediment of a Danish
  softwater lake. Hydrobiologia 492, 139-149. https://doi.org/10.1023/a:1024826131327
- 412 Hunt, R., 1982. Plant growth curves. The functional approach to plant growth analysis.
  413 Edward Arnold Ltd., London.
- Huser, B.J., Egemose, S., Harper, H., Hupfer, M., Jensen, H., Pilgrim, K.M., Reitzel, K.,
  Rydin, E., Futter, M., 2016. Longevity and effectiveness of aluminum addition to reduce
  sediment phosphorus release and restore lake water quality. Water Res. 97, 122-132.
  https://doi.org/10.1016/j.watres.2015.06.051
- 418 Jensen, M., Liu, Z., Zhang, X., Reitzel, K. and Jensen, H.S. 2017. The effect of 419 biomanipulation on phosphorus exchange between sediment and water in shallow, 420 tropical Huizhou West Lake, China. Limnologica 63. 65-73. 421 http://dx.doi.org/10.1016/j.limno.2017.01.001
- Jeppesen, E., Kristensen, P., Jensen, J.P., Søndergaard, M., Mortensen, E., Lauridsen, T., 1991.
  Recovery resilience following a reduction in external phosphorus loading of shallow,
  eutrophic Danish lakes: duration, regulating factors and methods for overcoming
  resilience. Mem. Ist. Ital. idrobiol. 48, 127-148.

- Jeppesen, E., Peder Jensen, J., Søndergaard, M., Lauridsen, T., Junge Pedersen, L., Jensen, L.,
  1997. Top-down control in freshwater lakes: the role of nutrient state, submerged
  macrophytes and water depth. Hydrobiologia 342/343, 151-164.
  https://doi.org/10.1007/978-94-011-5648-6\_17
- 430 Jeppesen, E., Søndergaard, M., Lauridsen, T.L., Davidson, T.A., Liu, Z., Mazzeo, N., 431 Trochine, C., Özkan, K., Jensen, H.S., Trolle, D., Starling, F., Lazzaro, X., Johansson, 432 L.S., Bjerring, R., Liboriussen, L., Larsen, S.E., Landkildehus, F., Meerhoff, M., 2012. 433 Biomanipulation as a restoration tool to combat eutrophication: recent advances and 434 Res. future challenges. Adv. Ecol. 47, 411-488. 435 https://doi.org/10.1016/B978-0-12-398315-2.00006-5
- Jin, X., Tu, Q., 1990. The standard methods for observation and analysis in lake
  eutrophication, 2nd ed. Environmental Science Press, Beijing. (in Chinese)
- 438 Laskov, C., Herzog, C., Lewandowski, J., Hupfer, M., 2007. Miniaturized photometrical 439 methods for the rapid analysis of phosphate, ammonium, ferrous iron, and sulfate in pore 440 of freshwater sediments. Limnol. Oceanogr. 5. 63-71. water Methods 441 https://doi.org/10.4319/lom.2007.5.63
- Liu, Z., Hu, J., Zhong, P., Zhang, X., Ning, J., Larsen, S.E., Chen, D., Gao, Y., He, H.,
  Jeppesen, E., 2018. Successful restoration of a tropical shallow eutrophic lake: Strong
  bottom-up but weak top-down effects recorded. Water Res. 146, 88-97.
  https://doi.org/10.1016/j.watres.2018.09.007
- Long, M.H., McGlathery, K.J., Zieman, J.C., Berg, P., 2008. The role of organic acid
  exudates in liberating phosphorus from seagrass-vegetated carbonate sediments. Limnol.
  Oceanogr. 53, 2616-2626. https://doi.org/10.4319/lo.2008.53.6.2616
- Lürling, M., Faassen, E.J., 2012. Controlling toxic cyanobacteria: Effects of dredging and
  phosphorus-binding clay on cyanobacteria and microcystins. Water Res. 46, 1447-1459.
  https://doi.org/10.1016/j.watres.2011.11.008
- 452 Lürling, M., Mackay, E., Reitzel, K., Spears, B.M., 2016. Editorial A critical perspective on
  453 geo-engineering for eutrophication management in lakes. Water Res. 97, 1-10.
  454 https://doi.org/10.1016/j.watres.2016.03.035
- 455 Lürling, M., van Oosterhout, F., 2013. Controlling eutrophication by combined bloom
  456 precipitation and sediment phosphorus inactivation. Water Res. 47, 6527-6537.
  457 https://doi.org/10.1016/j.watres.2013.08.019

- 458 Lürling, M., Waajen, G., van Oosterhout, F., 2014. Humic substances interfere with
  459 phosphate removal by Lanthanum modified clay in controlling eutrophication. Water
  460 Res. 54, 78-88. https://doi.org/10.1016/j.watres.2014.01.059
- 461 Marquez-Pacheco, H., Hansen, A.M., Falcon-Rojas, A., 2013. Phosphorous control in a
  462 eutrophied reservoir. Environ Sci Pollut Res. 20, 8446-8456.
  463 https://doi.org/10.1007/s11356-013-1701-2
- McPherson, B.F., Miller, R.L., 1987. The vertical attenuation of light in Charlotte Harbor, a
  shallow, subtropical estuary, south-western Florida. Estuar. Coast. Shelf Sci. 25, 721-737.
  https://doi.org/10.1016/0272-7714(87)90018-7
- Meis, S., Spears, B.M., Maberly, S.C., O'Malley, M.B., Perkins, R.G., 2012. Sediment amendment with Phoslock<sup>®</sup> in Clatto Reservoir (Dundee, UK): Investigating changes in sediment elemental composition and phosphorus fractionation. J. Environ. Manage. 93, 185-193. https://doi.org/10.1016/j.jenvman.2011.09.015
- 471 Meis, S., Spears, B.M., Maberly, S.C., Perkins, R.G., 2013. Assessing the mode of action of
  472 Phoslock<sup>®</sup> in the control of phosphorus release from the bed sediments in a shallow lake
  473 (Loch Flemington, UK). Water Res. 47, 4460-4473.
  474 https://doi.org/10.1016/j.watres.2013.05.017
- Paludan, C., Jensen, H.S., 1995. Sequential extraction of phosphorus in freshwater wetland
  and lake sediment: Significance of humic acids. Wetlands 15, 365-373.
  https://doi.org/10.1007/Bf03160891
- Reddy, K. R., Patrick, W. H., Lindau, C. W., 1989. Nitrification-denitrification at the plant
  root-sediment interface in wetlands. Limnol. Oceanogr. 34, 1004-1013.
  https://doi.org/10.4319/lo.1989.34.6.1004
- 481 Reitzel, K., Andersen, F.O., Egemose, S., Jensen, H.S., 2013a. Phosphate adsorption by
  482 lanthanum modified bentonite clay in fresh and brackish water. Water Res. 47,
  483 2787-2796. https://doi.org/10.1016/j.watres.2013.02.051
- 484 Reitzel, K., Lotter, S., Dubke, M., Egemose, S., Jensen, H.S., Andersen, F.Ø., 2013b. Effects
  485 of Phoslock<sup>®</sup> treatment and chironomids on the exchange of nutrients between sediment
  486 and water. Hydrobiologia 703, 189-202. https://doi.org/10.1007/s10750-012-1358-8
- 487 Ripl, W., 1976. Biochemical oxidation of polluted lake sediment with nitrate: a new lake
  488 restoration method. Ambio 5, 132-135.

- 489 Ross, G., Haghseresht, F., Cloete, T.E., 2008. The effect of pH and anoxia on the performance
  490 of Phoslock<sup>®</sup>, a phosphorus binding clay. Harmf. Alg. 7, 545-550.
  491 https://doi.org/10.1016/j.hal.2007.12.007
- 492 Rydin, E., 2000. Potentially mobile phosphorus in Lake Erken sediment. Water Res. 34,
  493 2037-2042. https://doi.org/10.1016/S0043-1354(99)00375-9
- 494 Santner, J., Larsen, M., Kreuzeder, A., Guld, R. N., 2015. Two decades of chemical imaging
  495 of solutes in sediments and soil a review. Anal Chim Acta. 878, 9-42.
  496 https://doi.org/10.1016/j.aca.2015.02.006
- Schindler, D.W., Carpenter, S.R., Chapra, S.C., Hecky, R.E., Orihel, D.M., 2016. Reducing
  phosphorus to curb lake eutrophication is a success. Environ. Sci. Technol. 50,
  8923-8929. https://doi.org/10.1021/acs.est.6b02204
- Schindler, D.W., Hecky, R.E., Findlay, D.L., Stainton, M.P., Parker, B.R., Paterson, M.J.,
  Beaty, K.G., Lyng, M., Kasian, S.E., 2008. Eutrophication of lakes cannot be controlled
  by reducing nitrogen input: Results of a 37-year whole-ecosystem experiment.
  Proceedings from the National Academy of Science USA 105, 11254-11258.
  https://doi.org/10.1073/pnas.0805108105
- 505 Smith, V.H., Schindler, D.W., 2009. Eutrophication science: where do we go from here?
  506 Trends Ecol. Evol. 24, 201-207. https://doi.org/10.1016/j.tree.2008.11.009
- 507 Søndergaard, M., Jensen, J.P., Jeppesen, E., 2003. Role of sediment and internal loading of
  508 phosphorus in shallow lakes. Hydrobiologia 506, 135-145.
  509 https://doi.org/10.1023/B:HYDR.0000008611.12704.dd
- 510 Søndergaard, M., Jeppesen, E., Jensen, J.P., Lauridsen, T., 2000. Lake restoration in Denmark.
  511 Lakes and Reservoirs: Research and Management 5, 151-159.
  512 https://doi.org/10.1046/j.1440-1770.2000.00110.x
- 513 Spears, B.M., Carvalho, L., Perkins, R., Kirika, A., Paterson, D.M., 2012. Long-term
  514 variation and regulation of internal phosphorus loading in Loch Leven. Hydrobiologia
  515 681, 23-33. https://doi.org/10.1007/s10750-011-0921-z
- 516 St-Cyr, L., Fortin, D., Campbell, P.G.C., 1993. Microscopic observations of the iron plaque of
  517 a submerged aquatic plant (*Vallisneria americana* Michx). Aquat. Bot. 46, 155-167.
  518 https://doi.org/10.1016/0304-3770(93)90043-v
- 519 Van der Does, J., Verstraelen, P., Boers, P., Van Roestel, J., Roijackers, R., Moser, G., 1992.

- Lake restoration with and without dredging of phosphorus-enriched upper sediment
  layers. Hydrobiologia 233, 197-210. https://doi.org/10.1007/BF00016108
- van Oosterhout, F., Lürling, M., 2013. The effect of phosphorus binding clay (Phoslock<sup>®</sup>) in
  mitigating cyanobacterial nuisance: a laboratory study on the effects on water quality
  variables and plankton. Hydrobiologia 710, 265-277.
  https://doi.org/10.1007/s10750-012-1206-x
- Waajen, G., van Oosterhout, F., Douglas, G., Lürling, M., 2016a. Geo-engineering
  experiments in two urban ponds to control eutrophication. Water Res. 97, 69-82.
  https://doi.org/10.1016/j.watres.2015.11.070
- Waajen, G., van Oosterhout, F., Douglas, G., Lürling, M., 2016b. Management of
  eutrophication in Lake De Kuil (The Netherlands) using combined flocculant Lanthanum modified bentonite treatment. Water Res. 97, 83-95.
  https://doi.org/10.1016/j.watres.2015.11.034
- Wang, X., Liu, F., Tan, W., Li, W., Feng, X., Sparks, D.L., 2013. Characteristics of phosphate
  adsorption-desorption onto ferrihydrite: comparison with well-crystalline Fe
  (hydr)oxides. Soil Sci. 178, 1-11. https://doi.org/10.1097/SS.0b013e31828683f8
- Zamparas, M., Zacharias, I., 2014. Restoration of eutrophic freshwater by managing internal
  nutrient loads. A review. Sci. Total Environ. 496, 551-562.
  https://doi.org/10.1016/j.scitotenv.2014.07.076
- Zhou, Y., Li, X., Zhao, Y., Zhou, W., Li, L., Wang, B., Cui, X., Chen, J., Song, Z., 2016.
  Divergences in reproductive strategy explain the distribution ranges of *Vallisneria* species in China. Aquat. Bot. 132, 41-48. https://doi.org/10.1016/j.aquabot.2016.04.005

 Properties
 Mean  $\pm$  SD

 Loss on ignition (LOI) (%)
  $4.31\pm0.87$  

 Water content (%)
  $49.33\pm4.81$  

 Dry bulk density (g cm<sup>-3</sup>)
  $0.74\pm0.10$  

 TP (mg gDW<sup>-1</sup>)
  $0.63\pm0.03$  

 P<sub>mob</sub> (mg gDW<sup>-1</sup>)
  $0.31\pm0.03$ 

**Table 1** Initial sediment properties (n=16).



- 546 Fig. 1. Water chemistry and light attenuation coefficient (K<sub>d</sub>) in the four treatments during the
- 547 experiment. Vertical bars indicate standard deviation.





Fig. 2. Mean values (n=3) of SRP concentrations in depth profiles in near-bottom water and
porewater in the four different treatments at different times. Three replicates for each

551 treatment. Horizontal bars indicate standard deviation.







**Fig. 3.** Total SRP in the surface 10 cm sediment at the end of the experiment. (Different letters indicate a significant difference among treatments, p<0.05). Vertical bars indicate standard deviation.



Fig. 4. Vertical distribution of different phosphorus fractions in the sediments of the different
treatments at the end of the experiment. Four replicates for each treatment. Horizontal bars
indicate standard deviation.



Fig. 5. Macrophyte traits at the end of the experiment. Significance results of t-test relative to
the Phoslock<sup>®</sup> treatments and controls indicated by N (p>0.05); \* (p<0.05); \*\* (p<0.01).</li>
Vertical bars indicate standard deviation.





Vallisneria denseserrulata (V. d.) 1 Internal phosphorus loading

*V. d*.

Phoslock<sup>®</sup> + V. d.

