

Ecological restoration on former agricultural soils: Feasibility of *in situ* phosphate fixation as an alternative to top soil removal

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ABSTRACT

In Europe, high phosphorus (P) concentrations form the most important constraint on the ecological restoration of biodiverse vegetation on former agricultural soils, because they lead to dominance of highly competitive species like *Juncus effusus* or to algal blooms in flooded situations. Top soil removal is often not sufficient or not possible, so alternative methods have to be found. We therefore investigated whether modified bentonite clay to which 5% lanthanum had been added (LMC) and lime could effectively decrease bioavailable P and phosphate mobilization to the water layer in different soil types.

A container experiment was performed using peaty and sandy soils with different Olsen-P concentrations, mixed with different doses of LMC and lime. The soils were exposed to two different common water regimes (moist and flooded). *J. effusus* seedlings were used as phytometers.

Addition of LMC and lime lowered extractable P concentrations in some of the P-rich sandy soils. Only the highest LMC dose was able to decrease phosphate mobilization to the water layer in the sandy soils. However, neither LMC nor lime was sufficiently effective in reducing Olsen-P concentrations and *J. effusus* growth. Lime addition eventually even led to additional nutrient mobilization by alkalization and increased mineralization of the soil.

Our experiments therefore show that LMC and lime are not feasible alternatives to top soil removal, because they are inefficient in preventing dominance of highly competitive species under moist or shallowly flooded conditions. LMC may only be used to prevent phosphate mobilization to the water layer in deeply flooded situations, which may allow for a more biodiverse vegetation development.

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1. Introduction

Several national and European legislations that aim to connect existing nature conservation areas have strongly increased the area of former agricultural land that is available for ecological restoration (Natura 2000; Smolders et al., 2008). However, a major problem that has to be overcome in these areas is that they have been heavily fertilized in recent decades, resulting in the accumulation of huge amounts of phosphorus (P) and nitrogen (N) in the soil (Barberis et al., 1995). It is especially P, being less mobile, which accumulates in the top layer of the soil (Schärer et al., 2007), whereas N can easily leach out to deeper layers (Johnston, 1994).

These exceptionally high nutrient concentrations form the most important constraint on the development and maintenance of biodiverse plant communities at these sites.

An additional problem may arise if former agricultural lands are converted into wetlands, which often also serve as water storage areas. This can cause serious phosphate mobilization to the water layer, depending on the biogeochemical properties of the soil and the quality of the water that is used for rewetting (Lamers et al., 2005; Pant and Reddy, 2003). Flooding of a site results in anaerobic soil conditions and the reduction of nitrate, manganese and iron. Iron reduction decreases the binding capacity of iron for P and may subsequently lead to the release of iron-bound P (Patrick and Khalid, 1974; Ponnampereuma, 1984). This P release during flooding has been shown to depend on the availability of organic matter and the P saturation of iron binding sites (Loeb et al., 2008).

Juncus effusus (soft rush or common rush) is one of the notorious eutrophic species that tends to dominate strongly on soils with a high P availability (Smolders et al., 2008). This easily dispersing species can germinate and grow very fast and outcompete

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other plant species by shading them (Ervin and Wetzel, 2001, 2002). In densely populated regions it is not expected that N will be the growth limiting factor for *J. effusus*, because N depositions are generally high (Bobbink et al., 1998). This is supported by studies showing that the most biodiverse nature areas are found on P-limited soils (Janssens et al., 1998; Wassen et al., 2005). A good indication of the bioavailable P fraction in the soil is given by the Olsen-P concentration (Olsen et al., 1954), which should not exceed 200–300 $\mu\text{mol kg}^{-1}$ soil if domination of *J. effusus* is to be prevented (Smolders et al., 2008).

In the past, different methods have been used to reduce P concentrations in surface water, pore water, sediments and soils. In terrestrial systems, top soil removal seems to be the most efficient measure to create a nutrient-poor situation within a relatively limited time span (Lamers et al., 2005; Smolders et al., 2008; Wetzel and Howe, 1999), although it is an expensive measure in many cases. Sometimes it is not even possible to remove enough soil to create P limitation, because deeper soil layers still contain high P concentrations, and additional measures have to be taken. Removing the top soil also results in the disappearance of the diaspore bank, although in agricultural soils this mainly contains seeds from eutrophic species and not from rare target species (Smolders et al., 2008). In addition to or instead of top soil removal, different compounds have been used to immobilize P *in situ*. Water treatment residuals (WTRs) are widely used in agricultural soils to increase the P sorption capacity of the soil and reduce off-site P leaching (Agyin-Birikorang et al., 2009; Novak and Watts, 2004). WTRs contain either iron, aluminum or calcium as potential P immobilizers (Elliott et al., 2002), with aluminum-WTRs having the highest ability to immobilize P.

Calcium has been used in different forms to control eutrophication in both lakes and terrestrial systems (Anderson, 2004; Beltman et al., 2001; Brouwer et al., 2002; Varjo et al., 2003). Liming with CaCO_3 has turned out to be an effective additional measure to reduce P availability after top soil removal (Smolders et al., 2008). However, liming leads to an increase in pH and alkalinity and could therefore increase decomposition rates in organic soils, which will also result in additional nutrient mobilization (Smolders et al., 2006). Iron compounds have also been used to bind P both in terrestrial systems (Schärer et al., 2007) and, especially, in aquatic systems (Boers et al., 1994; Hansen et al., 2003; Smolders et al., 2001), but iron has the disadvantage of being redox-sensitive (Ann et al., 2000). Aluminum addition is a widely used method to immobilize P in lakes (Reitzel et al., 2003; Rydin and Welch, 1998). Although aluminum is not redox-sensitive, it is sensitive to pH changes (Cooke et al., 1993; Driscoll and Schecher, 1990). This method is most effective between pH 6 and 8 and can cause serious toxicity problems at lower pH. Moreover, aluminum addition itself can lead to decreased pH values in soil and surface water (Malecki-Brown et al., 2007). Studies have also shown that the aluminum will crystallize over time and form gibbsite, leading to a lower binding capacity for P (Berkowitz et al., 2005).

Because all the above-mentioned compounds may show some serious drawbacks, we investigated whether the addition of the lanthanum-modified clay (LMC) Phoslock®, a bentonite clay to which 5% lanthanum has been added (Douglas, 2002; Phoslock Water Solutions Ltd, Sydney, Australia), would be an effective alternative method to immobilize P in former agricultural soils. To our knowledge, LMC addition is a novel method in terrestrial systems. The results were compared with those of lime addition. The advantage of LMC is that it forms highly stable minerals in the presence of phosphates (Douglas et al., 2000), which are relatively insensitive to changes in pH, redox potential and oxygen concentrations (Ross et al., 2008). LMC proved to be successful in immobilizing P and reducing algal blooms in some lakes and

rivers (experiments by Akhurst et al., 2004; Robb et al., 2003; Yang et al., 2004) by trapping P in the water layer and by forming an active layer on top of the sediment, which may immobilize P at the sediment–water interface. If LMC should prove to be effective in decreasing Olsen-P concentrations in former agricultural soils to below the threshold of 200–300 $\mu\text{mol kg}^{-1}$ soil (Smolders et al., 2008), domination of fast growing species like *J. effusus* could be prevented, and chances for the development of a more biodiverse vegetation could be improved. LMC addition is expected to be especially effective in flooded soils, because it can form an active layer on top of the soil that reduces phosphate mobilization to the water layer. This might avert blooms of algae and cyanobacteria in the overlying water layer.

To test the effectiveness of LMC and lime in decreasing bioavailable P and phosphate mobilization to the water layer in different types of former agricultural soils, a container experiment was performed using three sandy soils with different Olsen-P concentrations and one peaty soil, which are commonly encountered in restoration sites. We used these very different soil types because of the differential impacts LMC and lime may have upon soils with variable organic matter contents and buffering capacities. The soils were mixed with two different doses of LMC and one lime dose, and exposed to two different water regimes (moist and flooded) that frequently occur after restoration measures have been taken. To investigate whether the possible decrease in P concentrations would indeed decrease the growth of eutrophic plant species, *J. effusus* seedlings were planted in the containers and monitored as phytometers (Clements and Goldsmith, 1924; Wheeler et al., 1992) for three months. This enabled us to investigate whether LMC and lime could be used as effective alternatives to the expensive removal of nutrient-rich top soil or as an additional measure after top soil removal.

2. Methods

2.1. Experimental set-up

In January 2008, three soil types from different locations in the Netherlands were collected in 100 L containers. Nutrient-rich sandy soil was collected from a fertilized meadow on De Kieftskamp estate near Vorden (52°05'N, 6°20'E). Nutrient-poor sandy soil was collected from a fallow site on Staverden estate near Uddel (52°16'N, 5°44'E). Peaty soil was collected from a former pasture in the Vossenbroek nature reserve near Epe (52°19'N, 6°00'E).

After the soils had been homogenized in a concrete mixer, samples of each soil type were dried for 24 h at 70 °C and Olsen-P concentrations were determined by extraction according to Olsen et al. (1954). Subsequently, the nutrient-rich sandy soil was mixed with the nutrient-poor sandy soil in such proportions that soils were created with Olsen-P concentrations of about 500, 1000 and 2000 $\mu\text{mol kg}^{-1}$ soil (Table 1). We will refer to these soils as S500, S1000 and S2000.

In a greenhouse at the Botanical Gardens of the Radboud University Nijmegen, 128 plastic containers with a diameter of 15 cm were filled with 10 cm (1.77 L) of soil. Four different treatments were applied to both the sandy soil types and the peaty soil type ($n=8$):

- control treatment
- single lanthanum-modified clay dose using 100 g Phoslock® per g Olsen-P (LMC1x; Table 1; based on the 1:1 molar ratio of La and P)
- 5-fold lanthanum-modified clay dose using 500 g Phoslock® per g Olsen-P (LMC5x; Table 1)

Table 1
Initial Olsen-P and total-P concentrations, bulk density and organic matter content of the peat soil and the three sandy soil types (\pm SD; $n=8$) and the derived concentrations of lanthanum-modified clay (LMC) and lime that were added in the different treatments.

Soil	Olsen-P ($\mu\text{mol kg}^{-1}$)	Total-P (mmol kg^{-1})	Bulk density (kg L^{-1})	Organic matter %	LMC1x (g L^{-1} soil)	LMC5x (g L^{-1} soil)	Lime (g L^{-1} soil)
Peat	3991 (213)	53.1 (4.3)	0.35 (0.01)	38 (0.4)	4.4	21.8	3.5
S500	451 (68)	6.8 (1.3)	1.30 (0.04)	0.8 (0.1)	1.8	9.1	13.3
S1000	804 (158)	8.3 (1.8)	1.28 (0.05)	1.1 (0.1)	3.2	15.9	13.1
S2000	1918 (294)	11.1 (2.6)	1.22 (0.03)	1.8 (0.1)	7.2	36.2	12.5

- a lime dose of 10 g kg^{-1} fresh soil using Dolokal (75% CaCO_3 , 10% MgCO_3 and 5% MgO ; Smolders et al., 2008; Table 1);

LMC and lime were thoroughly mixed with the different soil types. Half of the containers were placed on a dish filled with demineralized water to keep the soils moist. The other half of the containers were flooded with 5 cm of demineralized water on top of the soil. To compensate for evaporation, demineralized water was added to maintain constant water levels. All containers were covered with two layers of white, nontransparent plastic to prevent plant growth and high soil temperatures. Etiolated seedlings were carefully removed.

A permanent soil moisture sampler of 10 cm with a pore diameter of $0.15 \mu\text{m}$ (Rhizon SMS, Eijkelkamp Agrisearch Equipment, Giesbeek, the Netherlands) was inserted diagonally into the soil of each container. Vacuum glass infusion bottles (30 mL) were attached to the soil moisture samplers to anaerobically sample soil pore water on days 1, 8, 47, 104, and 176. Water samples (30 mL) were taken from the water layer of the flooded soils on days 19, 40, 103, and 145. Small samples of the vertical profile of the soils (10 g) were taken on days 1, 8, 41, 110, and 176. These soil samples were dried for 24 h at 70°C and used for Olsen-P extractions (Olsen et al., 1954; Smolders et al., 2008).

After 145 days, the plastic cover was removed and three tufts of *J. effusus* seedlings with a shoot length of 5 mm and a dry weight of 7 mg were planted in each container. The water on the flooded soils was temporarily removed until plants were tall enough to reach the water surface. Plant growth was monitored by measuring the length of the longest shoots each month. After 243 days, three months after planting, the aboveground plant biomass was harvested, dried for 48 h at 70°C and weighed.

2.2. Chemical analysis

The pH of the pore water samples and unfiltered surface water samples was measured using a combined pH electrode with an Ag/AgCl internal reference (Orion Research, Beverly, CA, USA), and a TIM800 pH meter. Alkalinity was determined by titration to pH 4.2 with 0.01 M HCl using an ABU901 Autoburette (Radiometer, Copenhagen, Denmark). The samples were stored in iodated polyethylene bottles at -20°C until further analysis.

The concentrations of PO_4 , NO_3 , and NH_4 were measured colorimetrically with an Auto Analyzer 3 system (Bran+Luebbe, Norderstedt, Germany) according to Geurts et al. (2008). The concentrations of Ca, Fe, La, P, and Olsen-P were measured using an ICP Spectrometer (IRIS Intrepid II, Thermo Electron Corporation, Franklin, USA).

Homogenized portions of 200 mg dry plant material were digested with 4 mL HNO_3 (65%) and 1 mL H_2O_2 (30%), using an Ethos D microwave labstation (Milestone srl, Sorisole, Italy). Digestates were diluted and concentrations of P were determined by ICP as described above. Digestion analyses on 200 mg LMC showed that it contained 4.9% La, 1.0% Ca, 0.8% Al, 0.5% Fe, 0.3% Na, 0.3% Mg, and 0.1% K. Homogenized portions of 3 mg dry plant material were used to determine carbon and nitrogen content, using a Carlo Erba

NA1500 elemental analyzer (Thermo Fisher Scientific, Waltham, MA, USA). Weighted average N:P ratios in plant tissue were then calculated on a total dry weight basis.

2.3. Statistical analysis

All statistical analyses were carried out using SPSS for Windows (version 15.0, 2006, SPSS, Chicago, IL, USA). All data were $\log(+1)$ transformed to make variances less dependent on sample means, and to obtain a normal distribution. Levene's test indicated that equal variances are not assumed. Time effects and time \times treatment effects for chemical variables and *J. effusus* shoot length in the different moist and flooded soil types were tested separately with GLM repeated measures combined with a Tukey post hoc test. Differences between treatments in terms of chemical variables, plant biomass, shoot length and plant element ratios at the end of the experiment were determined by a univariate ANOVA combined with a Tukey post hoc test. Correlations between chemical variables, plant biomass and shoot length were determined by regression analyses. For clarity of presentation, the means and SEM presented in the figures and tables represent the non-transformed data.

3. Results

3.1. Effects on biogeochemistry

Pore water phosphate concentrations during the experiment were highest in the peaty soil and S2000 (Fig. 1). Only in the peaty soil were phosphate concentrations higher in the flooded soils than in the moist soils. In almost all cases, phosphate concentrations were highest in the control treatments, and lower in all other treatments. Phosphate concentrations in the peaty soil were 2–3 times lower in the lime treatments, whereas phosphate concentrations in the sandy soils were 2–10 times lower in the treatment with a 5-fold dose of lanthanum-modified clay (LMC5x). In the long term, pore water phosphate was significantly lower in all treatments of S2000 compared to the control (Tables 2 and 3). Phosphate concentrations in the surface water of the flooded peaty soils were only significantly lower over time in the lime treatment (Fig. 1c). Surface water phosphate concentrations on the sandy soils were also significantly lower in the LMC5x treatment. In the long term, however, surface water phosphate concentrations were only significantly lower than the control treatment in the LMC5x and lime treatments of S500 and in the LMC5x treatment of S2000 (Table 4), whereas phosphate concentrations in the LMC1x treatment of the peaty soils even became significantly higher than in the control situation.

Olsen-P concentrations in the soil remained quite stable over time in all control treatments, but were significantly lower in the LMC5x and lime treatment of the moist S1000, in the LMC5x treatment of the moist S2000 and in all treatments of the flooded S2000 ($P < 0.05$; Fig. 2). In the long term, Olsen-P concentrations were only significantly lower in the LMC5x treatment of the moist S2000, whereas concentrations were actually higher in the lime treatment

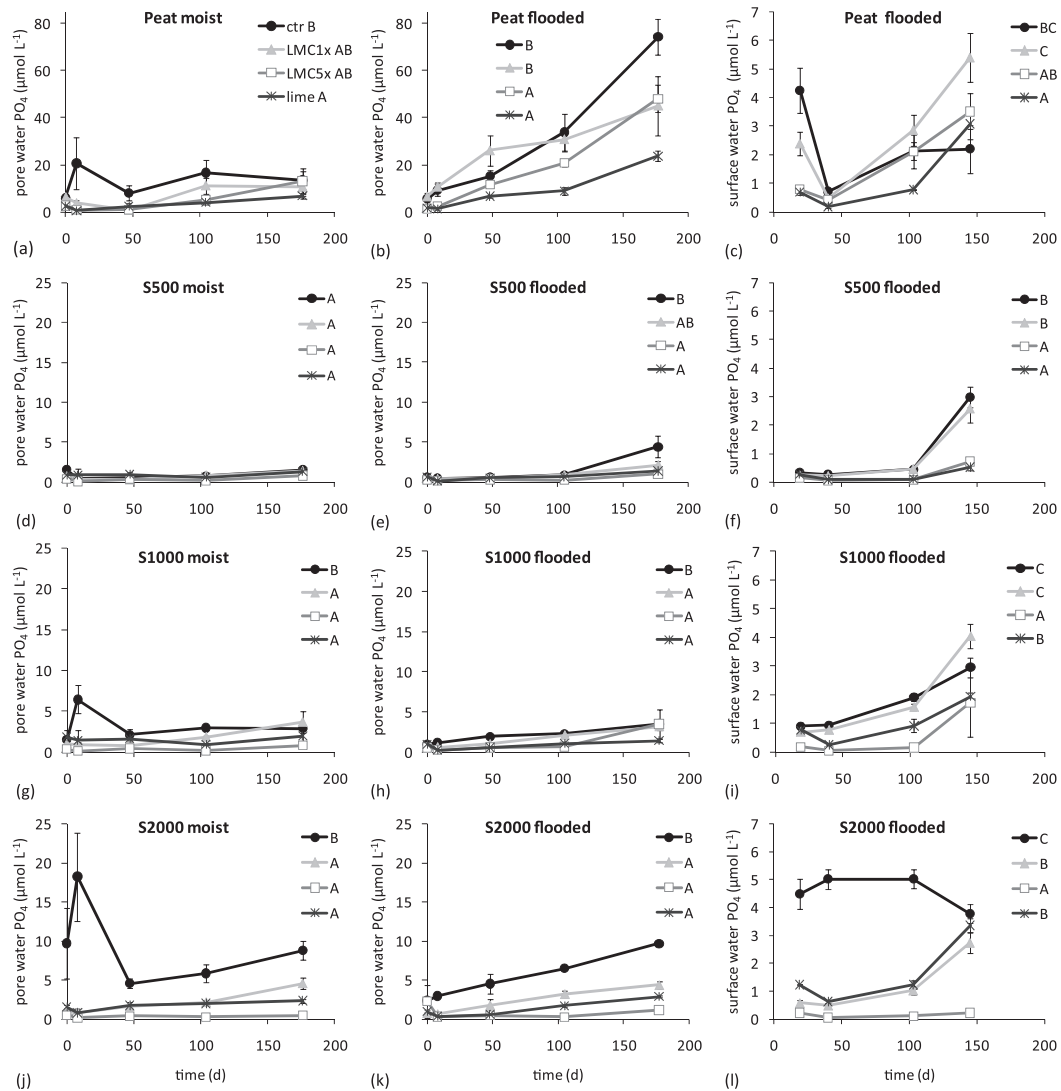


Fig. 1. Phosphate concentrations in pore water and surface water for the peat soil and the three sandy soil types under moist and flooded conditions (\pm SEM). Significant differences between treatments, as tested by GLM repeated measures ($P \leq 0.05$), are indicated by different letters. Note that different scales are used for the peaty soil and surface water graphs. ctr = control; LMC = lanthanum-modified clay; S = sandy soils with three different Olsen-P concentrations.

of this soil (Table 2). In the flooded soils, Olsen-P concentrations were significantly lower compared to the control in all treatments of S1000 and S2000, with the lowest concentrations in the LMC5x treatments (Table 3). After 176 days, Olsen-P concentrations in the LMC5x treatments were even lower than those in the control treatment of S500.

Especially in the short term, pore water ammonium concentrations in the sandy soils increased in all treatments compared to the controls (Fig. 3), with by far the largest increase in the LMC5x treatment (6–90 times higher). Water extractions of LMC revealed that LMC itself released large quantities of ammonium ($8.3 \pm 1.7 \text{ SD } \mu\text{mol g}^{-1} \text{ LMC}$; $n=5$), which could result in ammonium concentrations of 75–300 $\mu\text{mol L}^{-1}$ soil in this experiment. In the peaty soils, ammonium concentrations increased in the short term and remained highest in the LMC5x and lime treatments. In the sandy soils, however, ammonium concentrations did not increase in the long term. Long-term results showed the highest ammonium concentrations in the lime treatments of all soils, where they were twice as high as in the controls (Tables 2 and 3). The LMC treatments had no significant long-term effect on pore water ammonium concentrations, except for the flooded peaty soil,

in which ammonium concentrations were 1.5 times higher than in the control (Table 3). Only transient increases in ammonium concentrations were measured in the surface water, especially in the LMC5x treatments (Fig. 3). Nitrate concentrations in the pore water and the surface water of the LMC treatments increased to as much as 600 $\mu\text{mol L}^{-1}$ in the first weeks (data not shown), after which nitrate concentrations generally decreased again (Table 4). It was only in the moist sandy soils that pore water N concentrations correlated well with Olsen-P concentrations ($r^2 = 0.64$).

Alkalinity was higher in the peaty soils than in the sandy soils (Tables 2 and 3). In the lime treatments, alkalinity and pH in the pore water and the surface water strongly increased in all soils (Tables 2–4). Alkalinity in these soils increased 6–9 times in the pore water and 17–23 times in the surface water, whereas pH values increased by 1–1.5 units. Alkalinity and pH also increased significantly in the LMC treatments of most soils, but this increase was much smaller than in the lime treatments.

Lanthanum concentrations in the pore water and surface water increased in all LMC treatments and were highest in the LMC5x treatments (Tables 2–4). However, lanthanum concentrations only rose to 4–9 $\mu\text{mol L}^{-1}$ in the pore water and 2–5 $\mu\text{mol L}^{-1}$ in the

Table 2

Mean values (\pm SD) of chemical variables in pore water and soil for the different treatments of the moist peat soil and the three sandy soil types after 176 days. Significant differences between treatments are indicated by different letters (univariate ANOVA, $P \leq 0.05$). LMC = lanthanum-modified clay; S = sandy soils with three different Olsen-P concentrations.

	Fe ($\mu\text{mol L}^{-1}$)	NH ₄ ($\mu\text{mol L}^{-1}$)	NO ₃ ($\mu\text{mol L}^{-1}$)	PO ₄ ($\mu\text{mol L}^{-1}$)	Olsen-P ($\mu\text{mol kg}^{-1}$ DW)	pH	Alkalinity (meq L ⁻¹)	La ($\mu\text{mol L}^{-1}$)
Peat								
Control	151 (98)	167 ^{ab} (82)	36.1 (40.0)	13.5 (7.6)	6714 (518)	6.1 ^{ab} (0.0)	0.9 ^a (0.1)	0.0 ^a (0.0)
LMC1x	285 (223)	120 ^a (49)	35.0 (17.6)	10.6 (7.4)	6867 (598)	6.0 ^a (0.0)	1.0 ^a (0.4)	0.8 ^a (0.6)
LMC5x	435 (383)	340 ^{ab} (193)	13.4 (10.7)	13.1 (10.4)	6022 (436)	6.2 ^b (0.1)	1.6 ^a (0.9)	3.4 ^b (2.9)
Lime	273 (133)	406 ^b (172)	34.3 (18.3)	6.7 (1.9)	6275 (836)	6.9 ^c (0.1)	3.7 ^b (0.2)	0.4 ^a (0.3)
S500								
Control	108 ^b (20)	36 ^a (8)	4.4 (2.6)	1.5 (0.3)	454 (154)	5.9 ^a (0.1)	0.5 ^a (0.1)	0.0 ^a (0.0)
LMC1x	110 ^b (12)	43 ^a (2)	2.4 (1.1)	1.4 (0.3)	488 (78)	6.0 ^{ab} (0.1)	0.5 ^a (0.0)	4.0 ^c (0.5)
LMC5x	97 ^{ab} (14)	37 ^a (15)	6.2 (4.2)	0.8 (0.8)	371 (77)	6.2 ^b (0.2)	0.6 ^a (0.1)	7.5 ^d (1.5)
Lime	76 ^a (11)	80 ^b (15)	4.0 (3.4)	1.3 (0.9)	404 (87)	7.0 ^c (0.0)	4.0 ^b (0.3)	0.3 ^b (0.0)
S1000								
Control	89 (22)	63 ^a (19)	3.0 ^{ab} (2.2)	2.9 (0.4)	660 (145)	5.9 ^a (0.1)	0.6 ^a (0.0)	0.0 ^a (0.0)
LMC1x	132 (25)	93 ^{ab} (36)	15.6 ^b (19.7)	3.7 (2.6)	594 (76)	6.1 ^a (0.3)	0.9 ^a (0.3)	4.9 ^b (1.3)
LMC5x	116 (41)	73 ^{ab} (24)	1.7 ^a (0.2)	0.8 (0.2)	809 (252)	6.2 ^a (0.1)	0.8 ^a (0.2)	7.0 ^c (0.6)
Lime	76 (16)	144 ^b (33)	3.5 ^{ab} (3.6)	2.0 (2.0)	772 (113)	7.0 ^b (0.1)	3.8 ^b (0.4)	0.1 ^a (0.0)
S2000								
Control	166 (63)	170 (98)	1.6 (0.7)	8.8 ^c (2.3)	2151 ^b (463)	5.7 ^a (0.1)	0.5 ^a (0.2)	0.0 ^a (0.0)
LMC1x	153 (59)	182 (73)	1.2 (0.5)	4.6 ^b (1.5)	2149 ^b (268)	5.8 ^a (0.1)	0.7 ^a (0.1)	5.4 ^b (0.9)
LMC5x	156 (51)	175 (72)	1.9 (0.4)	0.4 ^a (0.6)	1193 ^a (226)	5.9 ^b (0.0)	0.7 ^a (0.1)	4.6 ^b (1.3)
Lime	97 (21)	284 (64)	1.3 (0.3)	2.3 ^b (0.5)	3775 ^c (170)	6.9 ^c (0.1)	4.0 ^b (0.4)	0.1 ^a (0.0)

shallow surface water. Iron concentrations were generally highest in the peaty soils, although they increased in all sandy soils during the experiment. Iron concentrations in the moist sandy soils became higher than in the flooded sandy soils, whereas the opposite was observed in the peaty soils (Tables 2 and 3). In the long term, iron concentrations were lowest in the lime treatments of the sandy soils (Tables 2 and 3).

3.2. Effects on plant growth

Shoot length and biomass of *J. effusus* were highest on the peaty soil, followed by S2000, S1000 and S500 (Tables 5 and 6). In the long term, shoot length and biomass were significantly higher on the flooded S500 and peaty soils than on the corresponding moist soils. Shoot length increased over time on all soil types, and this increase differed between treatments on S1000 and S2000 ($P < 0.05$). In the long term, however, treatments did not significantly decrease shoot

length and biomass compared to the controls in most cases, under both moist and flooded conditions (Tables 5 and 6). Shoot lengths on the flooded S500 were significantly lower in the lime treatment, while biomass was significantly lower in the LMC5x and lime treatment. On the moist S1000, biomass was significantly lower in both LMC treatments.

Shoot length and plant biomass at the end of the experiment correlated positively with Olsen-P concentrations ($r^2 = 0.4$ – 0.5) and pore water concentrations of phosphate ($r^2 = 0.5$ – 0.6) and nitrogen ($r^2 = 0.5$ – 0.8), as measured in the middle of the growth period (Tables 2 and 3). The correlation with Olsen-P concentrations was highest in the moist sandy soils ($r^2 = 0.7$ – 0.8). There was no correlation between the high pore water N concentrations and the high Olsen-P concentrations ($r^2 = 0.1$) in the peaty soils, although there was still a positive relationship between pore water N concentrations and plant biomass ($r^2 = 0.5$). N:P ratios in plant biomass were generally low (2 – 4 g g^{-1}), with N concentrations ranging between

Table 3

Mean values (\pm SD) of chemical variables in pore water and soil for the different treatments of the flooded peat soil and the three sandy soil types after 176 days. Significant differences between treatments are indicated by different letters (univariate ANOVA, $P \leq 0.05$). LMC = lanthanum-modified clay; S = sandy soils with three different Olsen-P concentrations.

	Fe ($\mu\text{mol L}^{-1}$)	NH ₄ ($\mu\text{mol L}^{-1}$)	NO ₃ ($\mu\text{mol L}^{-1}$)	PO ₄ ($\mu\text{mol L}^{-1}$)	Olsen-P ($\mu\text{mol kg}^{-1}$ DW)	pH	Alkalinity (meq L ⁻¹)	La ($\mu\text{mol L}^{-1}$)
Peat								
Control	454 (213)	571 ^a (57)	2.8 (2.6)	74.3 ^b (15.2)	9110 ^{ab} (1730)	5.7 ^a (0.4)	1.7 ^a (0.1)	0.1 ^a (0.0)
LMC1x	398 (267)	704 ^{ab} (52)	2.2 (1.2)	45.0 ^{ab} (25.3)	7285 ^a (912)	6.1 ^{ab} (0.3)	2.0 ^b (0.1)	1.0 ^b (0.6)
LMC5x	472 (141)	870 ^b (158)	1.5 (0.1)	48.1 ^{ab} (11.7)	6812 ^a (573)	6.1 ^{ab} (0.1)	2.2 ^b (0.2)	5.5 ^c (1.4)
Lime	570 (12)	817 ^b (100)	1.2 (0.2)	23.8 ^a (4.3)	16,865 ^b (9599)	6.3 ^b (0.1)	3.6 ^c (0.3)	0.1 ^a (0.0)
S500								
Control	69 (12)	36 (4.6)	1.0 ^a (0.5)	4.4 ^b (2.7)	400 (155)	5.9 ^a (0.2)	0.4 ^a (0.1)	0.0 ^a (0.0)
LMC1x	68 (10)	37 (6.2)	2.2 ^a (2.1)	2.1 ^{ab} (1.1)	329 (71)	6.1 ^a (0.1)	0.4 ^a (0.1)	4.6 ^b (0.4)
LMC5x	26 (19)	28 (15)	12.9 ^b (6.5)	1.0 ^a (0.1)	427 (121)	6.4 ^b (0.1)	0.5 ^a (0.1)	9.2 ^b (6.9)
Lime	30 (23)	45 (19)	2.6 ^a (1.5)	1.4 ^{ab} (1.0)	434 (129)	7.2 ^c (0.1)	3.5 ^b (0.3)	0.0 ^a (0.0)
S1000								
Control	72 (40)	55 ^a (25)	0.9 (0.5)	3.5 (0.8)	873 ^c (199)	5.9 ^a (0.1)	0.4 ^a (0.0)	0.0 ^a (0.0)
LMC1x	81 (21)	84 ^{ab} (4.2)	1.2 (1.6)	3.2 (0.8)	379 ^{ab} (34)	6.4 ^b (0.1)	0.9 ^b (0.0)	4.2 ^b (0.5)
LMC5x	78 (23)	102 ^{ab} (24)	3.2 (4.3)	3.5 (3.7)	260 ^a (106)	6.4 ^b (0.1)	0.8 ^b (0.0)	8.4 ^c (3.1)
Lime	60 (18)	139 ^b (33)	1.0 (1.0)	1.4 (0.5)	411 ^b (56)	7.1 ^c (0.1)	3.7 ^c (0.3)	0.1 ^a (0.0)
S2000								
Control	115 (10)	71 (34)	1.1 (1.0)	9.6 ^c (0.3)	1012 ^c (63)	5.8 ^a (0.1)	0.4 ^a (0.0)	0.0 ^a (0.0)
LMC1x	92 (68)	89 (52)	1.9 (0.8)	4.4 ^b (1.0)	637 ^b (96)	6.2 ^b (0.2)	0.8 ^{ab} (0.3)	4.4 ^b (1.5)
LMC5x	44 (42)	60 (92)	9.8 (12.2)	1.1 ^a (0.6)	383 ^a (18)	6.4 ^b (0.3)	0.9 ^b (0.3)	5.3 ^b (4.3)
Lime	44 (26)	135 (27)	7.7 (13.4)	2.9 ^b (0.3)	590 ^b (147)	6.9 ^c (0.1)	3.7 ^c (0.4)	0.1 ^a (0.0)

Table 4

Mean values (\pm SD) of chemical variables in the *surface water* for the different treatments of the *flooded* peat soil and the three sandy soil types after 145 days. Significant differences between treatments are indicated by different letters (univariate ANOVA, $P \leq 0.05$). LMC = lanthanum-modified clay; S = sandy soils with three different Olsen-P concentrations.

	Fe ($\mu\text{mol L}^{-1}$)	NH ₄ ($\mu\text{mol L}^{-1}$)	NO ₃ ($\mu\text{mol L}^{-1}$)	PO ₄ ($\mu\text{mol L}^{-1}$)	pH	Alkalinity (meq L ⁻¹)	La ($\mu\text{mol L}^{-1}$)
Peat							
Control	68.7 (36.7)	14.8 (6.2)	7.4 ^a (8.4)	2.2 ^a (1.7)	5.7 ^a (0.1)	0.5 ^a (0.1)	0.1 ^a (0.0)
LMC1x	111.2 (26.8)	14.5 (4.6)	36.2 ^b (12.9)	5.4 ^b (1.7)	5.7 ^a (0.1)	0.6 ^a (0.2)	1.1 ^b (0.3)
LMC5x	70.5 (20.7)	9.5 (1.8)	27.4 ^{ab} (18.2)	3.5 ^{ab} (1.2)	5.9 ^a (0.1)	0.5 ^a (0.1)	3.8 ^c (0.8)
Lime	58.4 (28.2)	8.7 (0.7)	45.0 ^b (15.9)	3.1 ^{ab} (1.1)	6.7 ^b (0.1)	2.2 ^b (0.4)	0.2 ^a (0.1)
S500							
Control	8.9 ^c (2.4)	6.6 (1.8)	22.0 ^{ab} (5.6)	3.0 ^b (0.7)	5.8 ^a (0.0)	0.2 ^a (0.0)	0.0 ^a (0.0)
LMC1x	8.6 ^c (4.0)	5.2 (2.1)	12.3 ^a (12.1)	2.6 ^b (1.0)	5.9 ^a (0.1)	0.2 ^a (0.0)	2.7 ^b (0.5)
LMC5x	2.9 ^b (1.1)	7.1 (2.2)	76.5 ^b (13.7)	0.7 ^a (0.2)	6.4 ^b (0.1)	0.3 ^b (0.1)	4.5 ^c (1.2)
Lime	0.2 ^a (0.2)	4.6 (2.0)	21.9 ^{ab} (7.6)	0.5 ^a (0.2)	7.4 ^c (0.3)	2.8 ^c (0.6)	0.1 ^a (0.1)
S1000							
Control	5.8 ^b (1.9)	6.1 (1.7)	35.7 (11.3)	3.0 ^{ab} (0.7)	5.2 ^a (0.2)	0.1 ^a (0.0)	0.0 ^a (0.0)
LMC1x	8.4 ^b (1.6)	6.3 (2.4)	34.3 (10.4)	4.0 ^b (0.9)	6.1 ^b (0.1)	0.4 ^b (0.1)	2.4 ^b (0.2)
LMC5x	3.9 ^{ab} (5.4)	9.7 (2.0)	39.4 (9.4)	1.7 ^a (2.4)	6.2 ^b (0.1)	0.4 ^b (0.1)	5.2 ^b (4.0)
Lime	0.7 ^a (0.3)	6.4 (2.2)	29.2 (11.7)	1.9 ^{ab} (0.3)	7.5 ^c (0.1)	3.3 ^c (0.2)	0.1 ^a (0.1)
S2000							
Control	4.3 ^b (1.3)	7.0 (2.6)	31.3 (12.6)	3.8 ^b (0.7)	5.1 ^a (0.2)	0.1 ^a (0.0)	0.0 ^a (0.0)
LMC1x	5.3 ^b (0.9)	6.8 (1.8)	25.1 (9.4)	2.7 ^b (0.8)	6.2 ^b (0.1)	0.4 ^b (0.1)	3.1 ^b (0.3)
LMC5x	0.6 ^a (0.8)	6.9 (1.3)	56.7 (27.0)	0.2 ^a (0.2)	6.5 ^b (0.1)	0.3 ^b (0.0)	2.3 ^b (1.9)
Lime	0.7 ^a (0.3)	10.8 (2.0)	30.5 (14.0)	3.4 ^b (0.5)	7.7 ^c (0.1)	3.0 ^c (0.3)	0.1 ^a (0.1)

12 and 27 mg g⁻¹ and P concentrations ranging between 4 and 8 mg g⁻¹ (Tables 5 and 6). N:P ratios on the flooded sandy soils decreased with increasing Olsen-P concentrations. On the moist sandy soils, N:P ratios in the lime treatments were significantly higher than in the controls. Compared to the control treatments, N concentrations in plant tissue increased in all addition treatments on the sandy soils, whereas they decreased on the peaty soil. Lanthanum concentrations in plant tissue only significantly increased in the LMC5x treatments (Tables 5 and 6).

4. Discussion

Although additions of lanthanum-modified clay (LMC) and lime resulted in lower pore water phosphate concentrations over time in the P-rich sandy soils, this could not prevent luxurious growth of the highly competitive species *J. effusus*. Under flooded conditions, only the highest LMC dose was able to decrease phosphate

mobilization to the water layer in the sandy soils. Lower phosphate concentrations in the water layer may prevent blooms of algae and cyanobacteria and create better light conditions for the growth of various aquatic plant species (Jeppesen et al., 2005; Roelofs, 1991). Because LMC was originally developed for application in surface waters, it has only been used in aquatic systems till now (Akhurst et al., 2004; Robb et al., 2003; Yang et al., 2004). In the peaty soils, however, phosphate mobilization to the water layer was highest in the LMC treatments, which makes LMC application not suitable for peaty soils.

LMC was not able to lower Olsen-P concentrations in the peaty soils and P-poor sandy soils. Although the highest LMC dose could reduce Olsen-P concentrations more than 50% in the flooded P-rich sandy soils, they still did not fall below the threshold of 200–300 $\mu\text{mol kg}^{-1}$ soil (260–390 $\mu\text{mol L}^{-1}$ fresh soil; Smolders et al., 2008). Apparently the heterogeneity of the soil was too high to allow for efficient P-binding to the LMC, in contrast to LMC application in the water layer. This means that *J. effusus* would still be

Table 5

Mean values (\pm SD) of shoot length, aboveground biomass, N:P ratios in plant tissue and concentrations of N, P, and La in plant tissue of *Juncus effusus* plants on the peat soil and the three sandy soil types under *moist* conditions 98 days after planting. Significant differences between treatments are indicated by different letters (univariate ANOVA, $P \leq 0.05$). LMC = lanthanum-modified clay; S = sandy soils with three different Olsen-P concentrations.

	Shoot length (cm)	Shoot dry weight (g)	N:P ratio (g g ⁻¹)	N in tissue (mg g ⁻¹ DW)	P in tissue (mg g ⁻¹ DW)	La in tissue (mg g ⁻¹ DW)
Peat						
Control	16.7 (2.5)	1.22 (0.33) ^{ab}	2.6 (0.7) ^b	21.6 (2.5) ^{ab}	8.1 (0.5) ^b	0.00 (0.00) ^a
LMC1x	12.9 (2.9)	0.53 (0.21) ^a	3.6 (1.1) ^a	27.2 (3.9) ^a	7.3 (0.3) ^{ab}	0.01 (0.00) ^a
LMC5x	17.2 (2.2)	1.42 (0.60) ^b	2.9 (0.9) ^{ab}	20.7 (3.0) ^b	6.8 (0.3) ^a	0.03 (0.00) ^b
Lime	18.1 (4.7)	1.67 (0.74) ^b	2.8 (1.4) ^b	19.2 (2.1) ^b	6.9 (0.9) ^{ab}	0.00 (0.00) ^a
S500						
Control	5.6 (0.6) ^{ab}	0.04 (0.02) ^{ab}	2.4 (0.7) ^b	13.3 (1.7)	5.6 (1.8)	0.01 (0.01)
LMC1x	6.3 (0.7) ^b	0.05 (0.01) ^{ab}	2.2 (0.3) ^b	14.7 (0.4)	6.7 (0.3)	0.19 (0.06)
LMC5x	4.2 (1.3) ^a	0.02 (0.01) ^a	2.4 (1.4) ^b	14.5 (1.5)	7.1 (2.9)	0.37 (0.37)
Lime	6.9 (1.4) ^b	0.09 (0.05) ^b	3.5 (1.6) ^a	16.0 (1.8)	4.5 (0.8)	0.01 (0.00)
S1000						
Control	10.4 (1.5)	0.22 (0.01) ^b	2.2 (0.2) ^c	13.0 (0.8) ^b	5.8 (0.3) ^{bc}	0.00 (0.00) ^a
LMC1x	8.7 (1.4)	0.14 (0.02) ^a	2.3 (0.4) ^{bc}	14.7 (1.5) ^{ab}	6.3 (0.7) ^c	0.11 (0.03) ^{ab}
LMC5x	8.5 (1.7)	0.10 (0.06) ^a	2.8 (1.7) ^{ab}	13.8 (0.5) ^{ab}	4.9 (0.2) ^a	0.33 (0.25) ^b
Lime	8.7 (0.9)	0.16 (0.02) ^{ab}	3.1 (0.4) ^a	16.1 (1.8) ^a	5.3 (0.4) ^{ab}	0.00 (0.00) ^a
S2000						
Control	13.0 (0.9)	0.47 (0.06)	2.1 (0.3) ^b	13.4 (1.2)	6.4 (0.8) ^b	0.01 (0.02)
LMC1x	12.3 (0.9)	0.43 (0.03)	2.2 (0.1) ^b	13.4 (1.5)	6.2 (0.5) ^b	0.05 (0.04)
LMC5x	11.9 (1.9)	0.40 (0.11)	3.1 (0.5) ^a	16.8 (4.0)	5.1 (0.3) ^a	0.06 (0.03)
Lime	10.8 (1.0)	0.34 (0.06)	2.8 (0.4) ^a	16.3 (1.1)	5.9 (0.2) ^{ab}	0.00 (0.00)

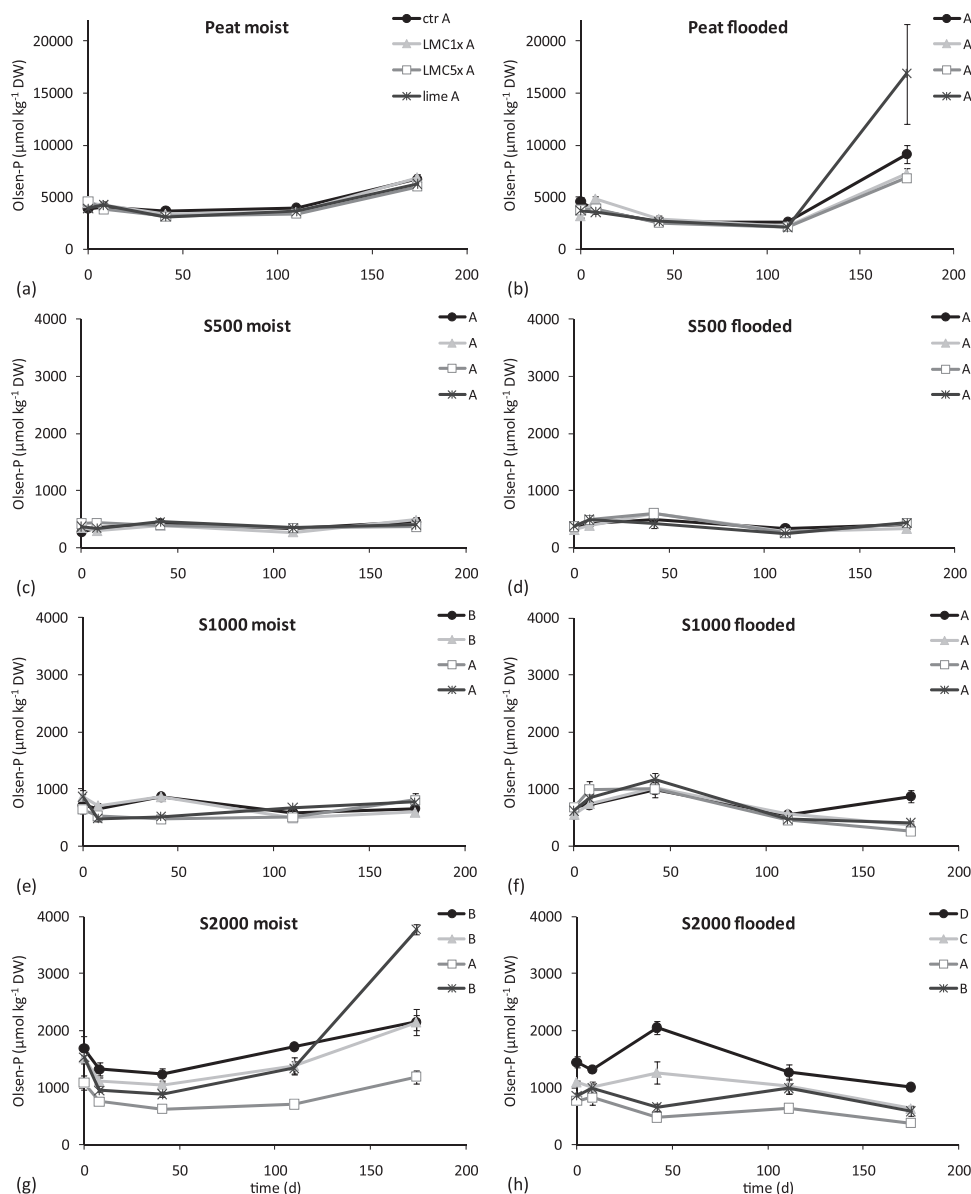


Fig. 2. Olsen-P concentrations in the peat soil and the three sandy soil types under moist and flooded conditions (\pm SEM). Significant differences between treatments, as tested by GLM repeated measures ($P \leq 0.05$), are indicated by different letters. Note that different scales are used for the peaty soil. ctr = control; LMC = lanthanum-modified clay; S = sandy soils with three different Olsen-P concentrations.

able to dominate on these soils during moist or waterlogged conditions or when the water layer covering the soil is thin enough to allow its growth, without opportunities for a biodiverse vegetation to develop (Janssens et al., 1998). LMC and lime were indeed not effective enough in reducing *J. effusus* growth, although shoot length and biomass were decreased on some of the sandy soils. This implies that *J. effusus* could have mobilized organically bound P or P bound by Fe, Al or Ca in these soils (Dinkelaker et al., 1989; Shen et al., 2002; Tweel and Bohlen, 2008). It is less likely that *J. effusus* can mobilize P that is bound by LMC, because lanthanum has strong ionic binding characteristics (Stumm and Morgan, 1996) and forms highly stable minerals with a low solubility in the presence of phosphates (Douglas et al., 2000; Firsching, 1992). Although other anions such as sulphate and nitrate could compete for binding sites, their adsorption is much lower than that of phosphate. The soils were relatively rich in Fe and thus presumably also rich in Fe-bound P, suggesting that the effects of LMC and lime may have been lower than they would have been in Fe-poor soils.

The low N:P ratios in plant tissue that we found on all soil types may indicate that *J. effusus* was limited in its growth by N and not by P (N:P ratio $\ll 14$; Koerselman and Meuleman, 1996), although N:P ratios of individual plant species cannot be used to predict their growth-limiting nutrient (Bedford et al., 1999; Güsewell and Koerselman, 2002). In addition, plant tissue concentrations of N and P were higher than the threshold concentrations for nutrient limitation ($N < 9.5 \text{ mg g}^{-1}$; $P < 1 \text{ mg g}^{-1}$; Güsewell and Koerselman, 2002). It seems more likely, however, that there was at least co-limitation of N and P, because plants grew better with increasing Olsen-P concentrations in the soil. Unlike Smolders et al. (2008), we did find a positive relationship between N concentrations in pore water and plant biomass, which can probably be explained by the positive correlation between the pore water N concentrations and the Olsen-P concentrations. Moreover, LMC and lime decreased pore water phosphate concentrations in the some of the sandy soils and sometimes increased nitrate and ammonium concentrations, whereas they decreased plant biomass and shoot length. This

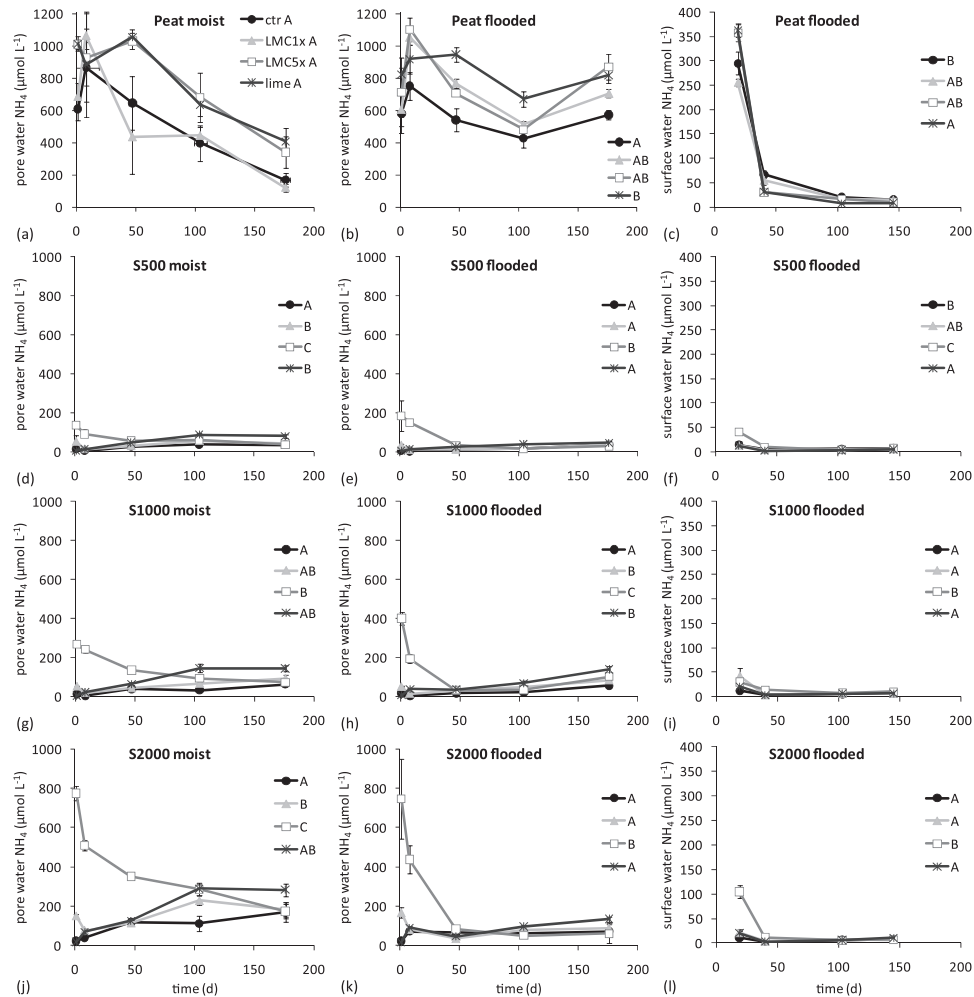


Fig. 3. Ammonium concentrations in pore water and surface water for the peat soil and the three sandy soil types under moist and flooded conditions (\pm SEM). Significant differences between treatments, as tested by GLM repeated measures ($P \leq 0.05$), are indicated by different letters. Note that different scales are used for the peaty soil and surface water graphs. ctr = control; LMC = lanthanum-modified clay; S = sandy soils with three different Olsen-P concentrations.

Table 6

Mean values (\pm SD) of shoot length, aboveground biomass, N:P ratios in plant tissue and concentrations of N, P, and La in plant tissue of *Juncus effusus* plants on the peat soil and the three sandy soil types under flooded conditions 98 days after planting. Significant differences between treatments are indicated by different letters (univariate ANOVA, $P \leq 0.05$). LMC = lanthanum-modified clay; S = sandy soils with three different Olsen-P concentrations.

	Shoot length (cm)	Shoot dry weight (g)	N:P ratio (g g^{-1})	N in tissue (mg g^{-1} DW)	P in tissue (mg g^{-1} DW)	La in tissue (mg g^{-1} DW)
Peat						
Control	21.7 (3.0)	1.53 (0.30)	3.9 (1.0)	24.4 (0.5)	6.3 (0.5)	0.01 (0.00)
LMC1x	18.8 (1.3)	1.91 (0.29)	3.5 (0.3)	22.5 (1.5)	6.4 (0.6)	0.05 (0.01)
LMC5x	20.0 (2.6)	1.98 (1.00)	4.1 (2.0)	24.4 (4.1)	5.8 (0.7)	0.24 (0.25)
Lime	19.9 (2.9)	1.95 (0.51)	3.5 (0.4)	21.3 (3.7)	6.0 (0.7)	0.02 (0.01)
S500						
Control	9.0 (0.4) ^b	0.14 (0.03) ^b	3.2 (0.7)	12.0 (0.3) ^b	3.7 (0.3)	0.01 (0.00) ^a
LMC1x	8.6 (0.3) ^b	0.11 (0.03) ^{ab}	3.0 (1.1)	14.3 (1.4) ^b	5.0 (1.3)	0.36 (0.16) ^a
LMC5x	9.0 (0.6) ^b	0.06 (0.02) ^a	3.7 (1.4)	18.3 (1.6) ^a	5.0 (0.5)	1.30 (0.40) ^b
Lime	5.8 (1.1) ^a	0.06 (0.03) ^a	4.6 (1.0)	20.9 (2.8) ^a	4.4 (2.3)	0.03 (0.04) ^a
S1000						
Control	13.3 (0.7)	0.24 (0.05)	3.0 (0.9)	15.7 (1.2) ^b	5.4 (0.8) ^{ab}	0.00 (0.00) ^a
LMC1x	10.3 (5.4)	0.19 (0.13)	3.4 (2.2)	19.0 (2.1) ^{ab}	5.4 (0.4) ^{ab}	0.22 (0.05) ^a
LMC5x	12.1 (4.0)	0.20 (0.10)	3.6 (2.0)	17.9 (3.1) ^{ab}	5.0 (0.9) ^a	1.59 (0.81) ^b
Lime	7.9 (5.2)	0.12 (0.11)	3.0 (2.6)	23.0 (6.4) ^a	7.3 (1.4) ^b	0.01 (0.00) ^a
S2000						
Control	16.9 (2.9)	0.53 (0.12)	2.4 (0.6)	13.5 (3.3)	5.7 (1.5)	0.00 (0.00) ^a
LMC1x	14.1 (1.8)	0.38 (0.11)	2.8 (0.4)	17.0 (2.6)	6.0 (0.2)	0.26 (0.15) ^a
LMC5x	12.4 (2.7)	0.35 (0.23)	2.7 (1.1)	17.2 (4.6)	5.6 (0.3)	2.13 (1.17) ^b
Lime	13.4 (2.7)	0.38 (0.17)	2.4 (0.9)	16.9 (2.8)	6.8 (1.8)	0.01 (0.00) ^a

implies that phosphate was the main nutrient limiting the growth of *J. effusus* on the sandy soils, which is why mobilization of nitrate and ammonium in some of the treatments did not increase *J. effusus* growth. On the peaty soils, plants only grew better with increasing pore water N concentrations, which could indicate N limitation for *J. effusus* on the peaty soils.

This mobilization of nitrate and ammonium did not last very long in the LMC treatments, because it was primarily released from the LMC itself, whereas it lasted much longer in the lime treatments. This was caused by the extreme increase in pH and alkalinity we observed after lime addition, which may have led to an increase in decomposition rates in the peaty soils and P-rich sandy soils and subsequently to additional nutrient mobilization (Smolders et al., 2006). In the other sandy soils, ammonium was probably dispelled from the soil absorption complex by calcium. The lanthanum concentrations released in the LMC treatments were below $6 \mu\text{mol L}^{-1}$, because lanthanum ions are very tightly adsorbed to the bentonite clay and the majority is therefore not considered to be bioavailable (Ross et al., 2008). The measured concentrations are not expected to be toxic to plants, animals or micro-organisms (Greenop and Robb, 2001; Persy et al., 2006), although plants may accumulate lanthanum in their tissue (Weltje et al., 2002) and concentrations of $7.5 \mu\text{mol L}^{-1}$ can significantly influence the growth rates of *Daphnia magna* (Lüring and Tolman, 2010).

A problem that may arise if LMC or lime is mixed with the upper 10 cm of the soil in the field is that *J. effusus* can form deeper roots, to reach untreated P-rich soil layers. Therefore, these compounds could only be used if the whole P-saturated soil layer is treated. However, even the highest LMC dose turned out to be not effective enough in reducing phosphate concentrations and plant growth in our experiment. Because of its high price, LMC application will not be cost-effective under all hydrological conditions. Only the highest LMC dose appeared to lower phosphate fluxes to the water layer under flooded conditions, which may reduce algal growth in surface waters (Akhurst et al., 2004; Robb et al., 2003; Yang et al., 2004). In deeply flooded situations, for example in water storage areas, this will create a better initial situation for the development of a more biodiverse aquatic vegetation on former agricultural sandy soils than in moist or shallowly flooded situations, where domination of highly competitive plant species like *J. effusus* leads to very low biodiversity.

5. Conclusions

Our experiments show that LMC and lime are not feasible alternatives to top soil removal, because they are inefficient in preventing dominance of highly competitive species under moist or shallowly flooded conditions. LMC may only be used to prevent phosphate mobilization to the water layer in deeply flooded situations, which may allow for a more biodiverse vegetation development.

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