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Phosphorus control and dredging decrease methane emissions from shallow lakes

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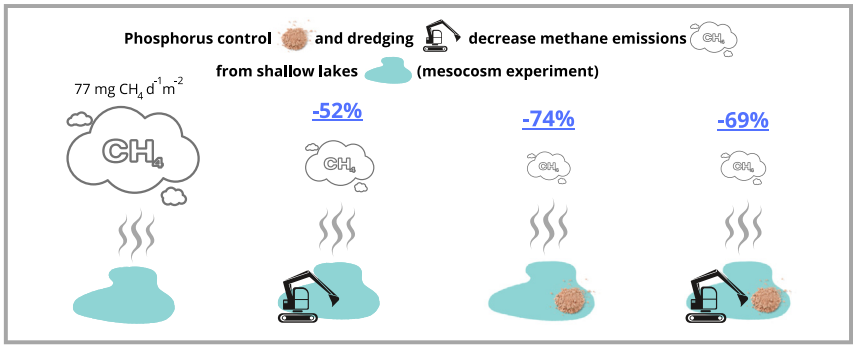
HIGHLIGHTS

- Mesocosms were used to test if phosphorus control and dredging decrease CH_4 emissions.
- CH_4 emissions were lower in mesocosms with phosphorus control and dredging.
- Phosphorus control and dredging can be used to mitigate CH_4 emissions from lakes.

GRAPHICAL ABSTRACT



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ABSTRACT

Freshwater ecosystems are an important source of the greenhouse gas methane (CH_4), and their emissions are expected to increase due to eutrophication. Two commonly applied management techniques to reduce eutrophication are the addition of phosphate-binding lanthanum modified bentonite (LMB, trademark Phoslock©) and dredging, but their effect on CH_4 emissions is still poorly understood. Here, this study researched how LMB and dredging affected CH_4 emissions using a full-factorial mesocosm design monitored for 18 months. The effect was tested by measuring diffusive and ebullitive CH_4 fluxes, plant community composition, methanogen and methanotroph activity and community composition, and a range of physicochemical water and sediment variables. LMB addition decreased total CH_4 emissions, while dredging showed a trend towards decreasing CH_4 emissions. Total CH_4 emissions in all mesocosms were much higher in the summer of the second year, likely because of higher algal decomposition and organic matter availability. First, LMB addition lowered CH_4 emissions by decreasing P-availability, which reduced coverage of the floating fern *Azolla filiculoides*, and thereby prevented anoxia and decreased surface water NH_4^+ concentrations, lowering CH_4 production rates. Second, dredging decreased CH_4 emissions in the first summer, possibly it removed the methanogenic community, and in the second year by preventing autumn and winter die-off of the rooted macrophyte *Potamogeton cripsus*. Finally, methanogen community composition was related to surface water NH_4^+ and O_2, and porewater total phosphorus, while methanotroph community composition was related to organic matter content. To conclude, LMB addition and dredging not only improve water quality, but also decrease CH_4 emissions, mitigating climate change.

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1. Introduction

Methane (CH_4) contributes 13–20 % to the total greenhouse effect, making it the second most important greenhouse gas (IPCC et al., 2021). Lakes contribute around 151 Tg CH_4 per year of these emissions (Rosentreter et al., 2021), making it essential to find ways to decrease their emissions. Lakes emit CH_4 mostly through diffusive flux, ebullitive flux (bubbles), and through plant structures (plant-mediated flux, Bastviken et al., 2004). While diffusive emissions have to pass through the top layer of the sediment, where methanotrophs can oxidize up to 90 % of the CH_4 produced (Oremland and Culbertson, 1992). CH_4 that passes the sediment as bubbles or through plant structures can reach the surface without being oxidized (Bastviken et al., 2004; Oliveira Junior et al., 2021). Eutrophic systems tend to emit more CH_4 than oligotrophic systems (Beaulieu et al., 2019; Davidson et al., 2018) due to accumulation of organic matter on the sediment. Such sediment accumulation is also a source of nutrients – referred to as internal loading – that keeps freshwater ecosystems in a eutrophic state even when external nutrient inputs have been controlled (Søndergaard et al., 2013). Consequently, in managing eutrophication in-lake interventions are often needed, and those measures potentially also decrease CH_4 emissions from shallow lakes.

Two commonly applied management techniques that target internal loading are adding P-binding agents such as lanthanum-modified bentonite (LMB, trademark Phoslock© (Douglas et al., 1999; Robb et al., 2003)), and dredging (Bray et al., 1996). LMB lowers water column P by immobilizing phosphate in the water column and by hampering phosphate release from the sediment through formation of an insoluble lanthanum-phosphate mineral (Copetti et al., 2016). Dredging removes the top layer of the sediment, which is rich in nutrients and organic matter, and thus reduces internal nutrient loading and mitigates eutrophication (Wan et al., 2020; Zhi-ying et al., 2008). LMB addition may be more effective in reducing sediment P fluxes than dredging (Yin et al., 2021), and LMB addition increased bacterial diversity, while dredging temporarily reduced it (Yin et al., 2021). Furthermore, dredging can also lower N-loading (Zhong et al., 2021), potentially also decreasing nitrous oxide emissions (Tian et al., 2020), which are predicted to rise with warming and eutrophication of freshwater lakes (Velthuis and Veraart, 2022). In two experiments (Lürling and Faassen, 2012; Yin et al., 2021), the combination of dredging and LMB decreased eutrophication even more, showing that the combination of these techniques is an effective way to reduce eutrophication.

However, the impact of these measures on CH_4 emissions has rarely been studied and findings are contrasting. For LMB, the effect on CH_4 emissions is largely unknown but a study in a laboratory experiment showed that LMB did not lower the CH_4 concentration in the water (Zhan et al., 2021). For dredging, one study found that dredging increased CH_4 concentrations in the Black Sea (Gar'kusha et al., 2019) but another study found that dredging decreased CH_4 emissions from aquaculture ponds (Zhao et al., 2021). Both of these studies do not represent natural shallow lakes, because saltwater ecosystems emit much less CH_4 than freshwater ecosystems (Saunois et al., 2020), and fish ponds have a much higher fish density, changing their dynamics (Colina et al., 2021; Kosten et al., 2020; Oliveira Junior et al., 2019). Finally, no studies so far investigated the combined effect of LMB and dredging on CH_4 emissions. Therefore, the effects of LMB and dredging on CH_4 emissions from shallow lakes are still poorly understood.

LMB and dredging likely lower CH_4 emissions, because they reduce P loading. This can lower chlorophyll *a* concentrations, decreasing CH_4 emissions (Beaulieu et al., 2019). Also, reduced P-loading can shift shallow lakes from an algae-dominated state to a macrophyte-dominated state (Scheffer and Van Nes, 2007), further decreasing CH_4 emissions (Davidson et al., 2015). However, dredging could also increase CH_4 emissions in the short-term because dredging causes a brief increase in sediment resuspension, and consequently a reduction in surface water O_2 (Suedel et al., 2008), decreasing CH_4 oxidation. Importantly, this resuspension is temporary (Manap and Voulvoulis, 2016). Moreover, because dredging can prevent cyanobacterial blooms (Wan et al., 2020), the turbidity caused by these blooms can be prevented in the long-term.

Here, mesocosms were used to study how LMB and dredging affect CH_4 emissions from shallow lakes and unravelled the underlying processes affecting CH_4 production and oxidation. The expectation was that both techniques would reduce CH_4 emissions by decreasing P loading. Furthermore, dredging was expected to increase short-term CH_4 emissions by reducing CH_4 oxidation.

2. Methods

This study used a full-factorial mesocosm design with LMB addition and dredging to test how CH_4 emissions were affected by LMB addition, dredging, and their combination. First, mesocosms (ø: 1.15 m, h: 0.75 m) were filled with dredged sediment from the eutrophic lake Wylerbergmeer (Latitude, longitude: 51°49′35″N 5°56′39″E). This sediment was gathered in July 2019, using freshly dredged sediment from the upper one meter of the organic sediment layer (see table A.1 for sediment characteristics). Sediment was mixed using a concrete mixer, and 200 l were added to every mesocosm, after which the rest of the mesocosm was filled with tap water, resulting in a sediment layer of ~20 cm with 50 cm overlying water (519 l). The mesocosms were left to stabilize until June 2020, after which the treatments were applied. Then, CH_4 emissions, and the development of the macrophyte community, sediment microbiome, and water quality variables of the mesocosms were monitored for 18 months, to explore the effects of LMB addition and dredging.

2.1. LMB addition and dredging

For the LMB treatment, lanthanum-modified bentonite clay (trademark Phoslock©, Douglas et al., 1999) was added based on the amount of available P in the mesocosm sediments, in a 100:1 LMB:P ratio as recommended by the manufacturer (Haghseresht et al., 2009). This ratio is based on 1:1 M La:P ratio (mass ratio 4.48:1) and 5 % La in LMB (Copetti et al., 2016). Available P was determined with a sequential P-fractionation method performed on the top 10 cm of all mesocosm sediments (Golterman, 1996), adding up the loosely available P and the Fe-bounded P. The available P in the whole sediment was targeted, based on the assumption that P in the deeper layers of the sediment would diffuse upwards after P in the upper layers was bound by LMB. The highest [P] in the mesocosm sediments was 27.8 mg/L sediment, which resulted in ~5.55 g P per mesocosm that could be immobilized using 555 g LMB. Each LMB mesocosm was treated by slowly adding LMB as small particles while continuously mixing the water in the mesocosm with a shovel. In the dredged mesocosms, the top five centimetre of the sediment was removed. First, the water was temporarily pumped out to an empty tank. Then, using a bucket (30 cm length x 40 cm width x 35 cm height) attached to a horizontal wooden pole to ensure that all mesocosms were dredged at exactly the same depth, the top five cm of the sediment was removed (this way of removing sediment can also be called excavation). As a consequence, all plants and filamentous algae were also removed similar to a real dredging scenario (Bray et al., 1996). Last, the water from the respective mesocosm was pumped back, taking care not to further disturb the sediment by pumping via the side of the tank.

2.2. CH_4 emissions

2.2.1. Diffusion

Both diffusive and ebullitive CH_4 emissions were measured. Diffusive emissions were measured biweekly with a transparent acrylic glass floating chamber (ø 30 cm) attached to either an LI-7810

Greenhouse Gas Analyser (LI-COR Biosciences, Lincoln, United States) or an Ultraportable Greenhouse Gas Analyser (Los Gatos Research, San Jose, United States, comparison between gas analysers in Fig. A.1). After a stable slope was established, diffusive flux was measured for 4 min, while monitoring for ebullition, in which case the measurement was restarted. The linear change in gas concentration over time was used to calculate the diffusive flux of CH_4 in mg CH_4 m^{-2} day^{-1}, as described in Almeida et al. (2016).

2.2.2. Ebullition

Ebullition was measured with bubble traps, using a funnel with a 20 cm diameter attached to a 280 ml glass bottle. The bubble traps were checked biweekly in summer and monthly in the other seasons. If they contained at least 20 ml of bubbles, the amount of gas was noted down and a headspace sample was taken to analyse the CH_4 concentration. The ebullitive flux (φ_{eb}, mg CH_4 m^{-2} day^{-1}) was then estimated as follows:

$$\varphi_{eb} = \frac{pCH_{4gas}\ x\ V_{ebb}}{t * A}$$

where pCH_{4gas} is the CH_4 concentration in the headspace of the bubble trap, V_{ebb} is the volume of the total volume of the gas in the bubble traps, t is the time since the last sampling of the bubble trap and A is the area of the bubble trap (i.e., 0.0314 m^2).

During four sampling intervals, storms caused bubble traps to flip and release the gas that was trapped (one in November 2020, two in December 2020, two in April 2021, one in July 2021). Additionally, the bubble traps were removed between 5 February and 4 April 2021 to prevent damage by ice formation. For these times, ebullitive flux was estimated by extrapolating ebullition rates before and after the time without bubble traps.

The analysis focused on summer emissions (June–September) when investigating how LMB and dredging affected CH_4 emissions per year, because these tend to have the vast majority of the annual CH_4 emissions (Schrier-Uijl et al., 2011), which was also confirmed by the last 12 months of measurement in this experiment (Summer 2021 represented 92 % of annual diffusive and 85 % of annual ebullitive emissions, Fig. A.2–3). Focusing on the summer emissions also allowed us to make comparisons between the summer of 2020 and 2021 (as the experiment in was started the summer of 2020, and in 2021 winter emissions were not measured).

2.3. Physicochemical variables and plant and algal community

2.3.1. Physicochemical variables

Directly after measuring diffusive CH_4 flux, O_2 concentration and temperature were measured at the top and bottom of the water column of the respective mesocosm, using a HQ40D Portable Multi Meter (Hach, Loveland, United States) with an Intellical LDO101 oxygen sensor (Hach, Loveland, United States).

Furthermore, surface water samples were taken from the centre of the mesocosms at 10 cm depth, while porewater samples were taken at 10 cm depth in the sediment at 20 cm from the mesocosm wall using Rhizon samplers (Rhizosphere Research Products, Wageningen, The Netherlands). An additional porewater sample was taken to determine the porewater CH_4 concentration, as described in Nijman et al. (2021). Water samples were transported immediately to the laboratory where the chlorophyll *a* concentration was measured using a PHYTO-PAM (Heinz Walz GmbH, Effeltrich, Germany), and pH and alkalinity were measured using an Ag/AgCl electrode (Orion Research, Beverly, MA, USA) and TIM 840 Titration Manager (Radiometer Analytical SAS, Villeurbanne, France).

Surface water and porewater samples were analysed for orthophosphate (o-PO_4^{3-}), nitrate (NO_3^-), and ammonium (NH_4^+) concentrations using colorimetric methods (Auto Analyser III, Bran and Luebbe GmbH, Norderstedt, Germany). Total phosphorus (TP) and elemental concentrations of Al, Ca, Fe, K, Mg, Mn, Na, S, Si, and Zn were measured using inductively coupled plasma optical emission spectrometry (ICP-OES, Thermo Fischer Scientific, Bremen, Germany). To stop microbial activity, samples that were not measured within three days were either frozen until their analysis for Auto Analyser samples or acidified with 0.1 ml 65 % HNO_3 for ICP-OES samples. Due to technical issues, data of o-PO_4^{3-} and NO_3^- concentrations from frozen porewater samples were excluded (Appendix A.1.1, Fig. A.4).

CH_4 in the headspace of the porewater samples was measured using an HP 5890 GC equipped with a flame ionization detector and a Porapak Q column (80/200 mesh).

2.3.2. Plant and algal community

The percentage cover of the aquatic plants, macroalgae, and filamentous algae was determined by visual estimation (their development is shown in Fig. A.5). The percentage was always evaluated by the same person (Thomas P.A. Nijman) to reduce sampling bias. Plant and macroalgal samples were taken to the lab to identify the species.

2.4. Sediment microcosm incubations

2.4.1. Sediment sampling and slicing

Potential rates of CH_4 oxidation and CH_4 production in the sediment were measured using microcosm incubations. CH_4 oxidation was determined biweekly in the summer of 2020 and once in June 2021, while CH_4 production was only measured in June 2021. Since there were no treatment effects on CH_4 oxidation in 2020, their setup is further discussed in the Appendix A.1.2.

For CH_4 oxidation and production in June 2021, the sediment manually was sampled manually, using 2.2 cm ø cores. Separate cores were taken to determine potential CH_4 oxidation and CH_4 production. Five sediment cores were taken per mesocosm for CH_4 oxidation and one sediment core for CH_4 production. The top 1 cm was used for CH_4 oxidation and the layer of 1–11 cm was used for CH_4 production, because O_2 was depleted in the top cm of the sediment in the mesocosms with highest O_2 concentration (Appendix A.1.3). For CH_4 oxidation samples, cores were sliced and mixed immediately after collection, while cores for potential CH_4 production were sliced in a glove bag (Aldrich® AtmosBag, Sigma-Aldrich) flushed with N_2-gas to prevent O_2 exposure.

Water content and organic matter content of the layers that were used for potential CH_4 oxidation and production were also determined by first drying fresh sediment samples for 72 h at 70 °C to obtain water content, and then muffling the dried sediment samples for 4 h at 550 °C to obtain organic matter content.

2.4.2. CH_4 oxidation incubations

One potential CH_4 oxidation incubation was set up per mesocosm. Five grams of wet sediment and ten ml of surface water from the respective mesocosm were added to 115 ml serum bottles, after which they were closed with airtight butyl stoppers, capped, and CH_4 (purity grade 4.5) was added to reach a 1 % headspace CH_4 concentration. The bottles were incubated at 20 °C for 48 h on a gyratory shaker at 150 rpm. CH_4 concentration in the headspace was measured in triplicate immediately ($t = 0$) and after 18, 24, and 42 h, using an HP 5890 GC equipped with a flame ionization detector and a Porapak Q column (80/200 mesh). The concentration was determined by comparing to a standard curve and rates were calculated based on the slope of decrease in CH_4 concentration.

2.4.3. CH_4 production incubations

For the potential CH_4 production incubations, extra care was taken not to expose the methanogenic community to O_2. Therefore, incubations were prepared in an anoxic chamber. Furthermore, the surface

water from the corresponding mesocosm used in the incubations was bubbled with N_2-gas for 30 min. Five g of sediment and ten ml of anoxic water were added to 115 ml serum bottles, after which they were closed with airtight butyl stoppers and capped. To remove any residual O_2, five cycles of vacuum-gassing with N_2-gas were performed. Finally, overpressure of an additional 0.5 bar N_2-gas was added to the bottles to prevent any O_2 intrusion during GC measurements. The CH_4 concentration in the headspace was measured after 0, 26, 51 and 75 h, using the same GC and standards as for CH_4 oxidation, while correcting for the overpressure by measuring with a GMSD 2B pressure sensor connected to a GMH 3111 pressure gauge (Greisinger, Regenstauf, Germany). The rates were calculated based on the slope of increase in CH_4 concentration.

2.5. Microbial community composition

Methanotroph and methanogen community composition were assessed in June 2021 using amplicon sequencing. First, DNA was extracted from the same sediment as the incubations (0–1 cm and 1–4 cm for methanotrophs, 1–11 cm for methanogens) using the DNeasy PowerSoil kit (Qiagen), with some small modifications to account for the higher water content of the samples as described in Nijman et al. (2021). Then, amplicon sequencing was performed by Novogene, on the Novaseq platform. The bacterial V4 region was sequenced using the 515F (GTGCCAGCMGCCGCGGTAA) and 806R (GGACTACHVG GGTWTCTAAT) primers (Caporaso et al., 2011), and the archaeal V4-V5 region was sequenced using the Arch519F (CAGCCGCCGCGGTAA) and Arch915R (GTGCTCCCCCGCCAATTCCT) primers (Coolen et al., 2004). Data was processed using the *dada2* pipeline (Callahan et al., 2016), as described in the supplementary methods (A.1.3). The microbial data were analysed using the *phyloseq* (McMurdie and Holmes, 2013) and *microbiome* (Leo and Shetty, 2017) packages.

2.6. Statistical analysis of LMB and dredging effects on mesocosm CH_4 emissions and the variables driving these emissions

2.6.1. Summer CH_4 emissions in relation to sampling year, LMB addition, and dredging

First, the effects of sampling year, LMB addition, and dredging on summer CH_4 emissions were tested. The CH_4 emissions were log-transformed to achieve normality and a three-way repeated measures ANOVA using the *rstatix* package (Kassambara, 2020) was used to test if they were significantly affected by LMB addition, dredging, and sampling year. Diffusive and ebullitive emissions were also tested separately using three-way repeated measures ANOVAs.

Principal component analysis (PCA) using the *vegan* package (Oksanen et al., 2013) was used to test how diffusive and ebullitive summer CH_4 emissions were related to the treatments and the other lake variables, including the lake community (plants, algae, macroalgae), chlorophyll *a* concentration, o-PO_4^{3-}, NO_3^-, NH_4^+, and TP concentrations in porewater and surface water, and physicochemical variables (O_2, pH, temperature).

2.6.2. Effect of LMB addition one year after applying treatments

Second, to study the effect of LMB addition on CH_4 emissions, the analysis then zoomed in on the state of the mesocosms one year after adding LMB (June 2021), focusing on the concentrations of o-PO_4^{3-} and TP in the surface water and porewater, the cover of *Azolla filiculoides*, and the community composition and potential activity of methanogens and methanotrophs. For methanotrophs, the focus was on the layer of 0–1 cm depth, as that was the layer that still had O_2 and therefore likely contained most methanotrophs. Variables were tested for normal distribution, after which surface water o-PO_4^{3-} was square-root transformed. Then, the effect of LMB addition and dredging on the variables was tested using two-way ANOVA, except for cover of *A. filiculoides*, which could not be converted to a normal distribution. Instead, percentage cover of *A. filiculoides* was converted to seven vegetation classes (1: 0–0.1 %, 2: 0.1–1 %, 3: 1–5 %, 4: 5–25 %, 5: 25–50 %, 6: 50–75 %, 7: 75–100 %, according to Braun-Blanquet, 1964), using the Wilcoxon rank sum test on these classes to test if LMB significantly affected cover of *A. filiculoides*.

To better understand the effects of the presence of the floating fern *A. filiculoides*, the relationship between *A. filiculoides* cover in summer and O_2 concentration at the bottom of the mesocosms, and the relationship between *A. filiculoides* cover in the summer of 2021 and NH_4^+ concentration in the surface water two weeks later were also explored.

For the microbial community, the drivers of community composition were investigated through redundancy analysis (RDA) using the *vegan* package (Oksanen et al., 2013). To find the drivers significantly correlating to community composition, a forward-backward selection model was run, including only the significant variables in the final figure. Last, the community members were plotted using the *envfit* function, grouping at the order level for methanogens and at the genus level for methanotrophs.

2.6.3. Macrophyte dynamics and CH_4 emissions in response to dredging

Finally, to better understand the mechanisms behind the effect of dredging on CH_4 emissions, the abundance of dominant macrophytes was examined. The decrease in coverage of *Potamogeton crispus* from 13 August 2021 until 26 February 2022 (Fig. A.5A) was compared between dredged and non-dredged mesocosms, as was also done for the ebullitive emissions in early summer (2 June – 30 July 2022, including the first peak in ebullitive emissions in 2021 during a heatwave in early July). Because it was not possible to convert the decrease in coverage of *P. crispus* autumn and winter to a normal distribution, the effect of dredging was statistically analysed using a Wilcoxon rank sum test. The ebullitive emissions in early summer were square-root transformed to achieve normality and a two-way ANOVA was used to test the effect of dredging and LMB addition. All analyses were performed in R version 3.6.3 (R Core Team, 2019).

3. Results

3.1. CH_4 emissions in response to LMB addition and dredging

This study investigated how LMB addition and dredging affected CH_4 emissions, in a full-factorial mesocosm design, monitored for 18 months. LMB significantly decreased total CH_4 emissions ($F_{1,12} = 5.51$, $p = 0.037$, average 61.5 % lower than without LMB, Fig. 1, table A.2A) while dredging showed a trend towards decreasing total CH_4 emissions ($F_{1,12} = 4.31$, $p = 0.060$, average 38.0 % lower than without dredging). Furthermore, there was a strong increase in total CH_4 emissions in the summer of 2021 relative to the summer of 2020 ($F_{1,12} = 41{,}34$, $p < 0.001$, average 19 times higher), likely because of overall increased algal growth and organic matter availability (Fig. A.6). Interestingly, only dredging seemed to influence CH_4 emissions in the first summer, although there was no interaction between sampling year and LMB or dredging ($F_{1,12} = 1.47$, $p = 0.249$ and $F_{1,12} = 0.10$, $p = 0.758$). The majority of CH_4 emissions originated from diffusion (86.3 %) with ebullition accounting for 13.7 %. LMB addition significantly affected diffusion ($F_{1,12} = 6.65$, $p = 0.024$, table A.2B) but not ebullition ($F_{1,12} = 1.57$, $p = 0.234$, table A.2C). Dredging showed a slightly different pattern, significantly affecting diffusion ($F_{1,12} = 4.72$, $p = 0.050$) but also showing a trend towards decreasing ebullition ($F_{1,12} = 3.62$, $p = 0.081$). Both diffusion and ebullition were much higher in 2021 than in 2020 ($F_{1,12} = 39.96$, $p < 0.001$ and $F_{1,12} = 25.59$, $p < 0.001$).

A principal component analysis to better understand the mechanisms driving these changes in CH_4 emissions showed that LMB was negatively correlated to variables related to eutrophication, including o-PO_4^{3-} and TP in surface water and porewater, chlorophyll *a* concentration and NH_4^+

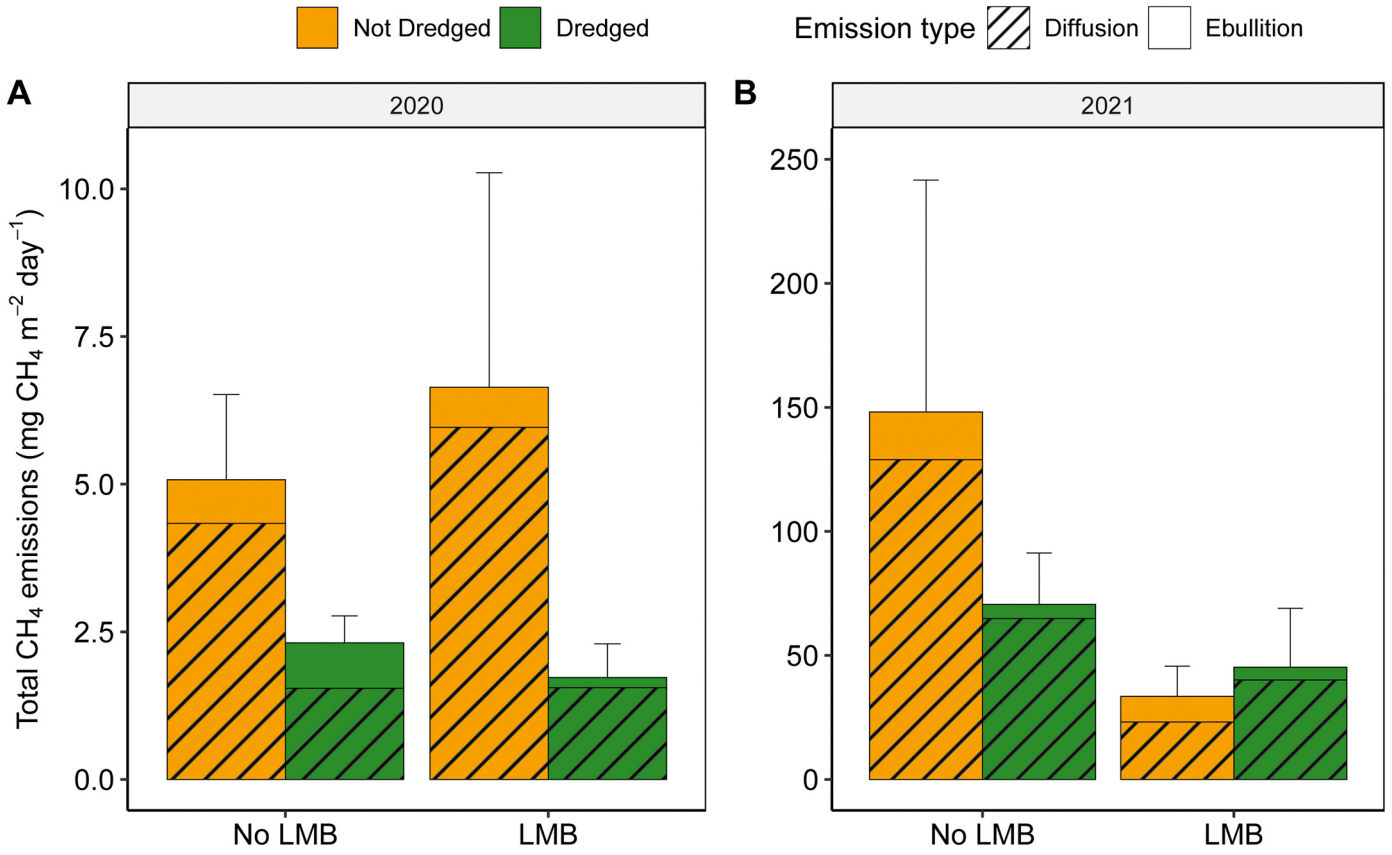

Fig. 1. CH_4 emissions in mg CH_4 m^{-2} day^{-1} in summers of (A) 2020 and (B) 2021 in relation to LMB and dredging. The left and right panel have different y-axes because of the large differences in total emissions between 2020 and 2021. Bars show means, whiskers indicate standard deviation, patterns with and without stripes indicate diffusive and ebullitive emissions respectively. Colour represent dredging treatments, $n = 32$. Note the difference in scale between the two panels.

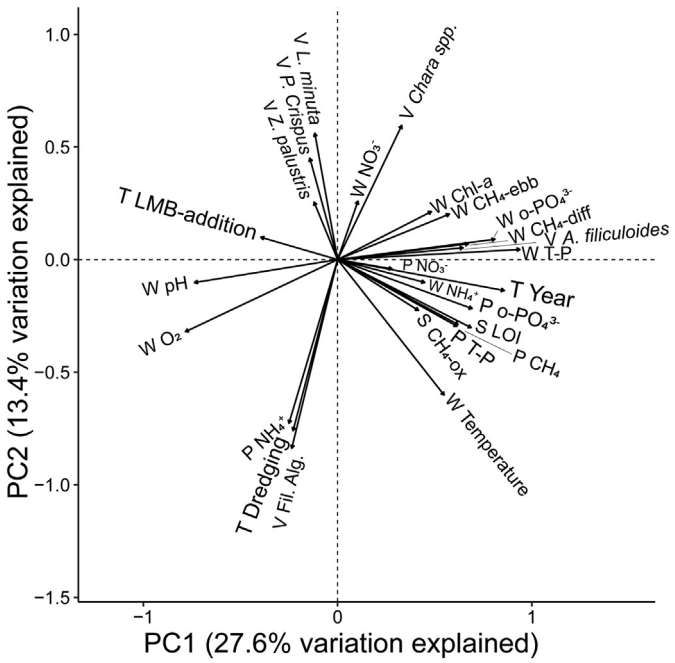

Fig. 2. PCA of averages of all environmental variables in the summer of 2020 and 2021, including diffusive and ebullitive CH_4 emissions. W and P indicate surface water and porewater variables, T indicate treatments, V indicates vegetation variables, and S indicates sediment variables.

concentration in the surface water (Fig. 2). Likely, LMB indeed bound P, especially in the sediment (Fig. A.7), lowering P-availability and subsequently changing the trophic state of the mesocosms. Dredging showed a different pattern and was positively correlated to the presence of filamentous algae, but negatively to *P. crispus* cover. Dredging was also positively correlated to porewater NH_4^+ concentration, however this may be a sampling effect rather than a mechanistic effect: after dredging, porewater samplers were reinserted at 10 cm depth from the new sediment surface, bringing them deeper into the anoxic zone where ammonification takes place.

When zooming in on CH_4 emissions, both CH_4 ebullition and diffusion were negatively correlated to LMB addition and dredging, with ebullition being more strongly correlated to dredging while diffusion was more strongly correlated to LMB addition. Both also showed a strong positive correlation to *A. filiculoides* cover, surface water NH_4^+ concentration, and chlorophyll *a* concentration, while being negatively correlated to dissolved O_2 concentration in the water.

3.2. Effect of LMB addition on water and sediment quality

LMB likely indirectly affected CH_4 emissions by lowering P availability. One year after LMB addition, o-PO_4^{3-} and TP in the porewater were significantly lower in mesocosms with LMB addition ($F_{1,12} = 17.40$, $p = 0.001$ and $F_{1,12} = 13.57$, $p = 0.003$, Fig. 3A/B). TP and o-PO_4^{3-} in the surface water were also lower, although there was only a trend for TP ($F_{1,12} = 3.45$, $p = 0.088$) and no significant difference for o-PO_4^{3-} ($F_{1,12} = 1.54$, $p = 0.238$, Fig. 3C/D). The changes in P-availability in response to LMB addition also affected the macrophyte community dynamics, which impacted the mesocosm

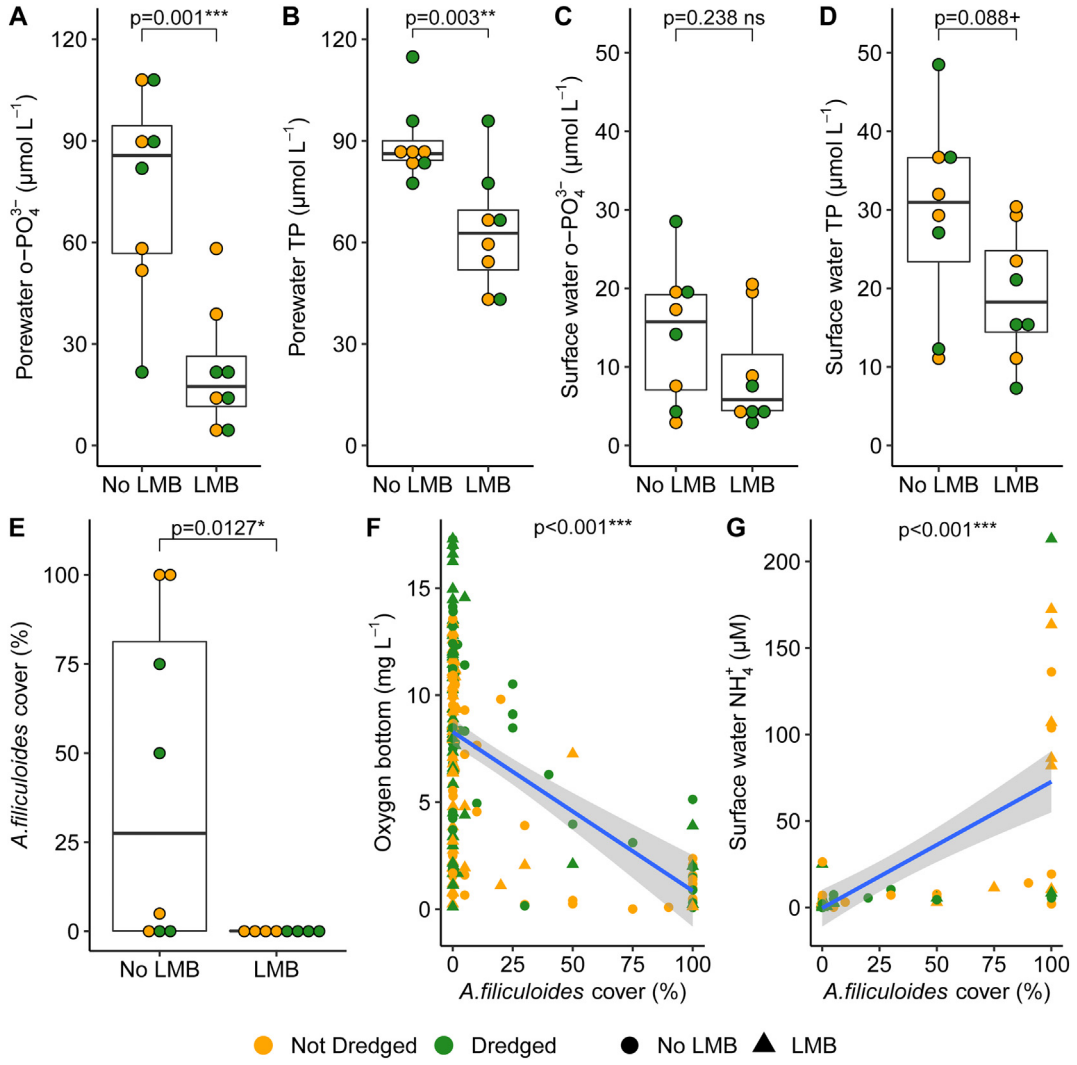

Fig. 3. Mesocosm characteristics one year after adding LMB and dredging. The figures show porewater (A) o-PO_4^{3-} in week 50, and (B) TP, surface water (C) o-PO_4^{3-} and (D) TP, and (E) *A. filiculoides* cover in week 52 after LMB addition. Furthermore, the relationship between (F) O_2 concentration at the sediment surface of the mesocosm in response to *A. filiculoides* cover during the summers of 2020 and 2021 is shown, and (G) the relationship between *A. filiculoides* cover and surface water NH_4^+ concentration two weeks later for weeks 52–60. Colours represent dredging treatments. For figures A-E, boxes indicate the first and third quartiles, lines indicate the median, whiskers indicate outer data points if <1.5*interquartile range from quartiles. For fig. F and G, lines represent linear regressions between *A filiculoides* cover and O_2 concentration at the sediment surface and surface water NH_4^+ respectively, while shaded area indicates 95 % confidence interval.

biogeochemistry. One year after LMB addition, *A. filiculoides* was nearly absent (0–0.1 % cover) in all mesocosms with LMB addition, while half of the mesocosms without LMB showed at least 50 % *A. filiculoides* cover ($Z = 52$, $p = 0.013$, Fig. 3E). Later in the season, some of the mesocosms with LMB addition also had high *A. filiculoides* cover (Fig. A.5B), but this occurred about six weeks later than in the mesocosms without LMB. Meanwhile, *A. filiculoides* likely affected biogeochemistry, as observed from a significant negative correlation between *A. filiculoides* cover and dissolved O_2 concentration (Fig. 3F, $r^2 = 0.2495$, $F_{1,190} = 64.48$, $p < 0.001$) and a significant positive correlation between *A. filiculoides* cover and NH_4^+ concentrations in the surface water (Fig. 3G, $r^2 = 0.4177$, $F_{1,62} = 46.18$, $p < 0.001$), thus creating a more eutrophic state of the mesocosms. This could explain why CH_4 emissions were higher in mesocosms without LMB addition.

The changes in O_2 concentration and NH_4^+ in the surface water coincided with changes in the microbial community one year after applying the treatments. Potential CH_4 production rates were higher in mesocosms without LMB addition ($F_{1,12} = 4.96$, $p = 0.046$, Fig. 4A), while potential CH_4 oxidation rates were not affected by LMB ($F_{1,12} = 0.15$, $p = 0.710$, Fig. 4C), and neither was affected by dredging ($F_{1,12} = 2.715$, $p = 0.125$ for CH_4 production and $F_{1,12} = 1.201$, $p = 0.295$ for CH_4 oxidation). Potential CH_4 oxidation rates were also not affected in the summer of 2020 ($F_{1,12} = 0.09$, $p = 0.780$, Fig. A.8). In terms of community composition, redundancy analysis showed that methanogen community composition was significantly related to NH_4^+ in the surface water ($F_{1,11} = 4.19$, $p = 0.008$) and TP in the porewater ($F_{1,11} = 5.22$, $p = 0.004$), while also showing a small significant correlation with *Zannichellia palustris* cover ($F_{1,11} = 4.28$, $p = 0.007$, Fig. 4B), and a trend towards O_2 concentration ($F_{1,11} = 2.19$, $p = 0.084$, Fig. 4B). On the other hand, methanotroph community composition was only significantly associated to organic matter content ($F_{1,14} = 2.08$, $p = 0.028$, Fig. 4D). However, there were no strong treatment effects on methanogen or methanotroph community composition (Fig. A.9).

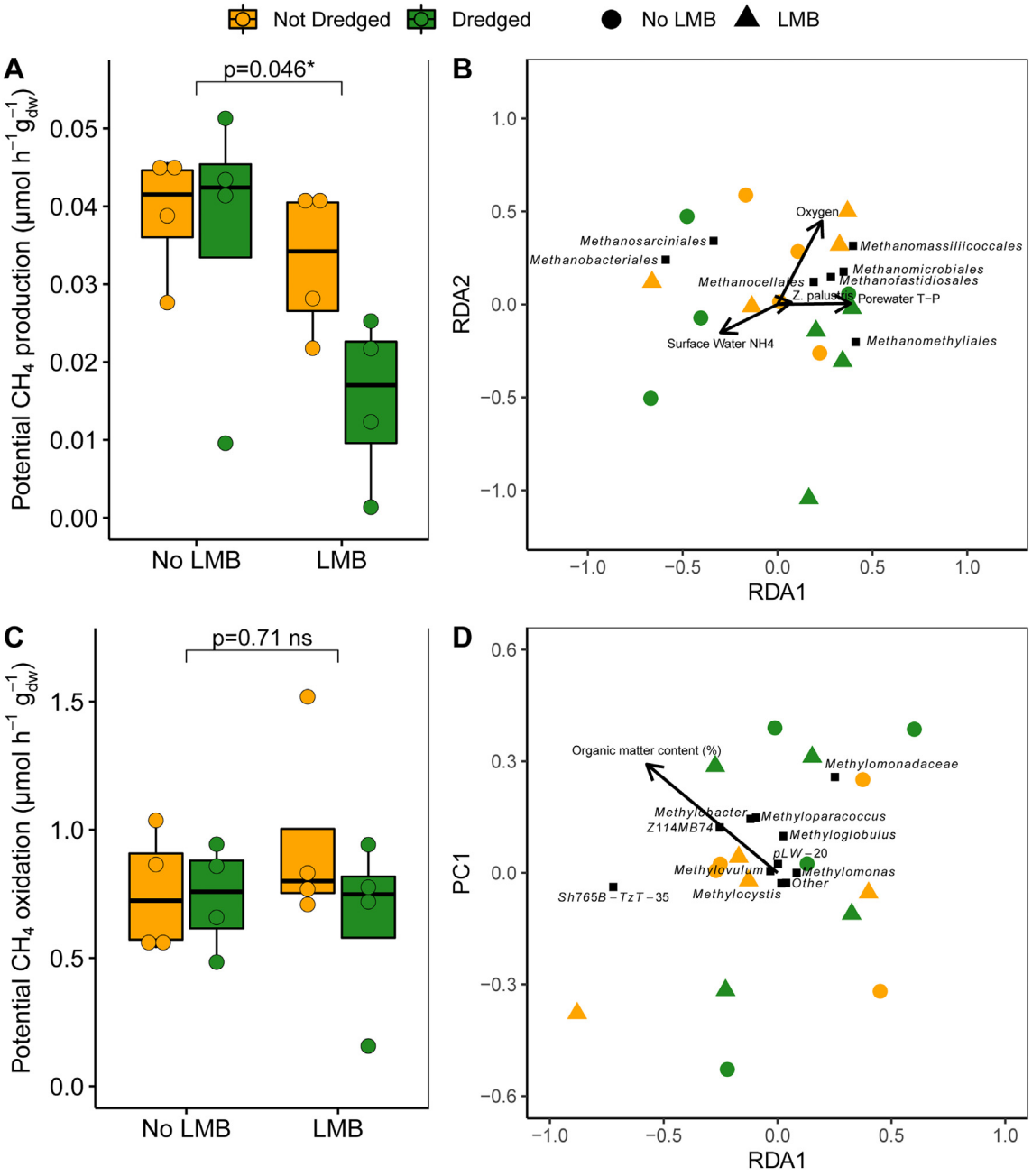

Fig. 4. Potential methane (A) production and (C) oxidation in response to LMB addition and dredging one year after applying the treatments, and a redundancy analysis showing the drivers significantly associated to (B) methanogenic and (D) methanotrophic communities (chord-transformed). The graph showing methanotroph community composition only has one RDA axis, because only one environmental variable was included in the redundancy analysis. Colours represent dredging treatments. For figs. A and C, boxes indicate the first and third quartiles, lines indicate the median, whiskers indicate outer data points if <1.5*interquartile range from quartiles. For fig. B and D, dots represent mesocosms without LMB, triangles mesocosms with LMB, and squares the methanogen orders and methanotroph genera. Arrows were plotted using the *envfit* function.

3.3. Relationship between macrophyte dynamics and ebullitive emissions in early and late summer

To better understand the drivers of ebullitive CH_4 emissions, the development of dominant macrophytes was monitored during the experiment. There was a large die-off of *P. crispus* in non-dredged mesocosms ($Z = 53$, $p = 0.018$, Fig. 5A), while *P. crispus* was removed from the dredged mesocosms during dredging (Fig. A.5A) and therefore did not show a decrease in coverage during autumn and winter. Furthermore, dredged mesocosms had a strong trend towards having higher ebullitive emissions in early summer ($F_{1,12} = 4.55$, $p = 0.054$, on average ~ 6 times higher).

4. Discussion

In this mesocosm study, LMB addition decreased total CH_4 emissions, while dredging showed a trend towards decreasing total CH_4 emissions, with both treatments significantly decreasing diffusive CH_4 emissions. First, LMB addition decreased phosphorus concentrations, especially in the porewater, which likely changed the timing at which *A. filiculoides* became dominant, decreased potential CH_4 production rates, and impacted methanogen community composition (Fig. 6). Second, dredging changed submerged macrophyte development in the mesocosms. Dredged mesocosms had overall lower standing stocks of

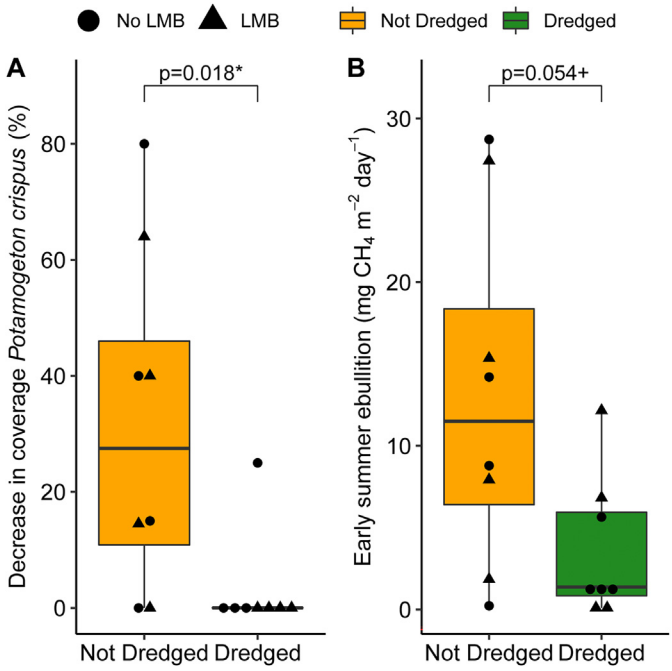

Fig. 5. (A) Decrease in coverage of *P. crispus* during autumn and winter and (B) early summer ebullitive CH_4 emissions. Colours represent dredging treatments. Dots indicate mesocosms without LMB, triangles mesocosms with LMB. Boxes indicate the first and third quartiles, lines indicate the median, whiskers indicate outer data points if <1.5*interquartile range from quartiles.

P. crispus, resulting in much smaller autumn and winter die-off events than non-dredged mesocosms. Dredging thereby decreased diffusive emissions while showing a strong trend towards decreasing early summer ebullitive emissions in the summer of 2021. Dredged mesocosms also had lower CH_4 emissions in the summer of 2020, possibly because of the removal of the methanogenic community. Last, CH_4 emissions were much higher in the second year, likely because of higher algal growth and decomposition, as observed from increased surface water chlorophyll-*a* concentrations and sediment organic matter content.

4.1. LMB effects on CH_4 emissions

LMB decreased CH_4 emissions, likely by decreasing phosphorus availability, in line with the hypothesis. Mesocosms with added LMB had significantly lower total phosphorus and orthophosphate concentrations in the porewater. The decrease in phosphorus coincided with decreased potential CH_4 production rates in LMB-amended mesocosms, which can explain the lowered CH_4 emissions. A previous study showed no direct effect of P on methanogenesis (Kim et al., 2015), and in this experiment, the effect of phosphorus also seemed to be indirect. Mesocosms without LMB addition had higher abundance of *A. filiculoides*, a floating fern that has a symbiosis with N_2-fixing cyanobacteria (Lumpkin and Plucknett, 1980), which makes it more competitive in P-polluted waters where it can outcompete N-limited species. Depending on the system, floating plants can lower CH_4 emissions by reducing the water-atmosphere exchange and facilitating CH_4 oxidation in the rhizosphere, or increase CH_4 emissions by increasing organic matter availability and anoxia (Kosten et al., 2016). In this experiment, increased *A. filiculoides* abundance changed the mesocosm biogeochemistry by lowering O_2 concentrations and increasing surface water NH_4^+ concentrations. Because lack of O_2 stimulates methanogenesis (Bastviken et al., 2004), and increased N causes eutrophication, which has been shown to increase methane emissions (Davidson et al., 2018; DelSontro et al., 2016), the increased abundance of *A. filiculoides* in mesocosms without LMB addition likely explained why they had higher potential CH_4 production rates.

The results are in contrast to a previous study which showed that LMB did not decrease CH_4 concentration of the surface water of LMB-amended sediment cores (Zhan et al., 2021). This may be due to the longer duration of our experiment (18 months vs 21 days), as well as the larger and more

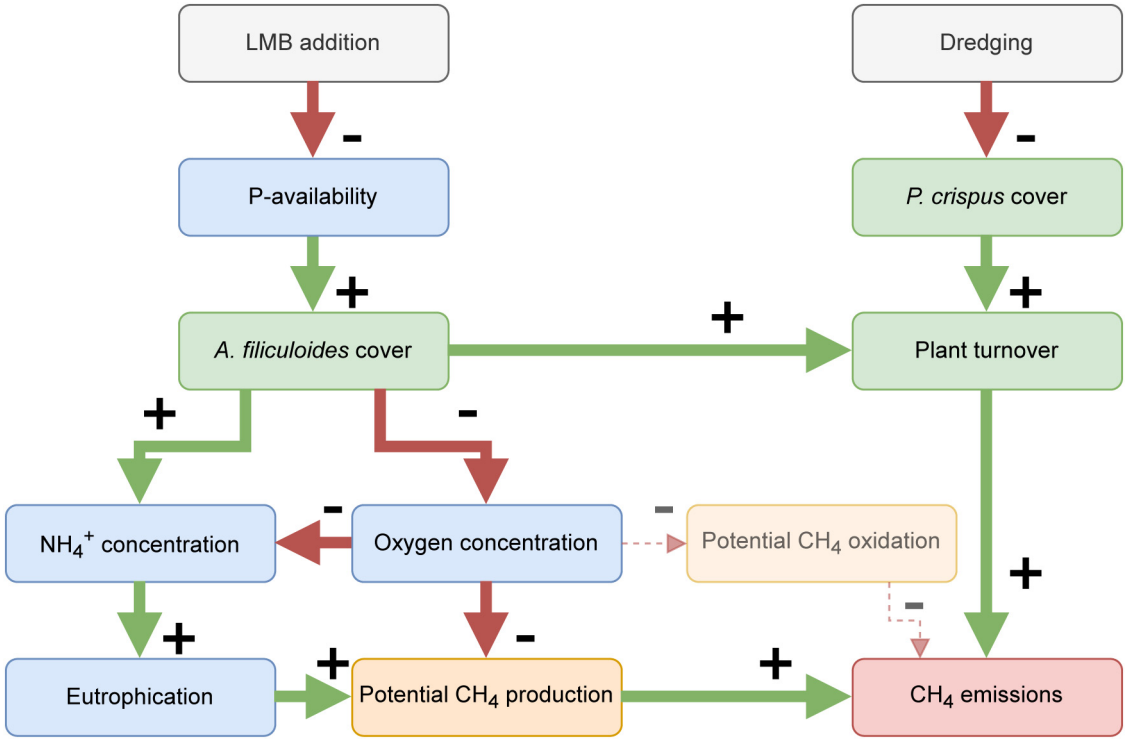

Fig. 6. Proposed mechanisms through which LMB addition and dredging influenced diffusive and ebullitive CH_4 emissions in this mesocosm experiment. Blue boxes indicate water-related variables, green boxes macrophyte-related variables, brown boxes sediment-related variables, and red boxes CH_4 emissions. The dashed arrows and partially transparent box of potential CH_4 oxidation indicate that this effect was not observed in this study.

dynamic mesocosm set up, which also captured indirect effects of macrophyte development. Our assumption is therefore that our experiment better approximates the potential effects in real lakes. Interestingly, when focussing on the first year of summer emissions from the mesocosms, the results are similar to the study by Zhan et al. (2021), as there were no clear differences between mesocosms with and without LMB addition. Therefore, this result shows that LMB affects lakes especially in longer time frames.

A limitation of this study is that the P-fractionation method used to estimate the amount of LMB to be added did not include organic P (P in microorganisms, including poly-P, organic P in detritus, and P bound in humic compounds, Paludan and Jensen, 1995). Thus, potential releasable P might have been underestimated, and LMB effects might have been stronger if more LMB had been added.

4.2. Dredging effects on CH_4 emissions

Dredging decreased diffusive CH_4 emissions, while also showing a strong trend towards decreasing ebullitive CH_4 emissions. However, although not significant, the total amount of ebullitive emissions in non-dredged mesocosms was 200 % higher than in dredged mesocosms, while diffusive emissions were only 63 % higher in non-dredged mesocosms. The fact that ebullitive emissions tend to be highly variable (DelSontro et al., 2016; Linkhorst et al., 2020) might explain why the result was not significant.

The reason that dredging lowered CH_4 emissions in the summer of 2021 was likely through changes in the macrophyte community. In particular, *P. crispus* was removed in the dredged mesocosms, while most non-dredged mesocosms were dominated by *P. crispus* by the end of the first year. During autumn and winter, *P. crispus* died, providing easily degradable carbon for later CH_4 emissions (Grasset et al., 2019). Dredged mesocosms did not have this source of carbon and could therefore produce less CH_4. In line with these results, Zhang et al. (2018) showed that in constructed wetlands, those dominated by *P. crispus* had increased methanogen abundance and CH_4 emissions. This decrease in *P. crispus* likely drove the increase in diffusive CH_4 emissions, as well as showing a trend towards increasing ebullitive CH_4 emissions in early summer.

Contrary to the hypothesis, dredging did not cause a short-term increase in CH_4 emissions in the first summer but instead decreased these emissions, which could have several reasons. First, the way in which the mesocosms were dredged aimed to minimize resuspension, and therefore there was no strong increase in turbidity and decrease in O_2 after dredging. This can also explain why there was no significant change in potential CH_4 oxidation after dredging (Fig. A.8). Second, the short-term effects might occur on a shorter time scale, e.g., in the first weeks rather than the first months. When looking at the diffusive CH_4 emissions in more detail (Fig. A.2), it is clear that the increase in emissions in non-dredged mesocosms in the first summer occurred in the final months of the summer, while there were no differences in the first half of the summer. Third, by removing the top layer of the sediment, it is likely that the most active part of the methanogenic community was removed (On et al., 2005), which could explain why emissions were decreased in the first summer.

In this study, the effect of dredging on CH_4 emissions was lower than effects found for aquaculture fishponds (Zhao et al., 2021), in which dredging decreased CH_4 emissions by over 90 %. However, because only the top five centimetres of the organic sediment was removed while leaving the remainder of the organic sediment intact, there was still a high potential to produce CH_4, leading to CH_4 emissions. This indicates that the newly exposed sediment bed after dredging is important for CH_4 emissions. For example, if this exposed layer would be composed of nutrient-poor sandy soil (Kankaala and Bergstrom, 2004), CH_4 emissions would likely be much more reduced than in this experiment. Furthermore, previous studies found that dredging can lower P release from sediments (Shigaki et al., 2008), decrease sediment organic matter, N and P content (Wan et al., 2020), and reduce N cycling rates (Zhong et al., 2021). However, in our experiment there were no effects of dredging on pore- and surface water N and P concentrations or on organic matter content. This could also be because of the limited dredging depth and the lack of a depth gradient in sediment quality, and could explain the modest decrease in CH_4 emissions.

Our argument is therefore that the observed dredging-induced decrease in CH_4 emissions is likely a lower range estimate of the effect common dredging practices. It is likely that removal of a thicker sediment layer, as is common in dredging practices (Bray et al., 1996), has a stronger impact on CH_4 emissions as sediment CH_4 emission tends to be higher when the sediment layer is thicker (Maeck et al., 2013). Future studies quantifying the effect of dredging on CH_4 emissions under realistic field situations – e.g., when dredging is applied as a management technique – will help to further elucidate large scale dredging effects on CH_4.

How the dredged sediment is managed likely affects the amount of CH_4 that is mitigated by this measure. First, dredged material can produce CH_4 which is then emitted from the location where it is deposited (Gebert et al., 2019). Importantly, once the removed sediment dries, these emissions are much lower than if the dredged sediment was still part of their originating shallow lakes (Paranaíba et al., 2021). Consequently, a decrease in CH_4 emissions from shallow lakes through dredging will likely be partly offset by emissions from the dredged sediment. Furthermore, it is important to prevent the sediment from resuspending while dredging, and to deposit the dredged sediment in such a way that the sediment does not flow back into the lake, e.g. during rain (Bray et al., 1996). Therefore, the handling of the dredged sediment would likely impact whether or not dredging will effectively mitigate CH_4 emissions.

4.3. Increased CH_4 emissions in 2nd year

In the second year, both diffusive and ebullitive CH_4 emissions were much higher, which showed that the mesocosms needed time to develop. In the second year, there was a large increase in the chlorophyll *a* concentration of the surface water and there was higher organic matter availability in the sediment. Earlier studies also showed a positive relationship between chlorophyll *a* and CH_4 emissions (Davidson et al., 2018; DelSontro et al., 2016), which is likely a consequence of associated high sedimentation and decomposition of dead algae (West et al., 2012). After one year, nutrient availability in the mesocosms had increased as they went through a full cycle of plant growth and decay, in addition to nutrient release into the water from the sediment (Smolders et al., 2006), which likely explained the increased algal growth. Furthermore, rooted macrophytes could have harvested nutrients from the deeper layers of the sediment, in particular below the targeted LMB sediment layer, leading to higher nutrient concentrations. The results therefore demonstrate the importance of running mesocosm experiments for at least one year.

4.4. Microbial community

While mesocosms with LMB addition had lower potential CH_4 production rates, there was no LMB or dredging effect on CH_4 oxidation rates and methanogen or methanotroph community composition. Possibly, changes in the methanotrophic community require repeated or more drastic disturbances before effects on the CH_4 oxidation rate can be observed (Ho et al., 2016; van Kruistum et al., 2018). Furthermore, the changes within the community might also be at the species level rather than at the genus level, since small changes within genera can modulate CH_4 oxidation (Bodelier et al., 2013). Although some methanotrophs require lanthanum to grow (Pol et al., 2014), the results did not indicate that LMB-associated lanthanum addition positively affected the methane oxidation rates, possibly because of a lack of bioavailability of lanthanum.

For methanogen community composition, while there were no clear treatment effects, correlations were observed to porewater TP, O_2 concentration and NH_4^+ concentration of the surface water. All of these variables were related to LMB addition, as LMB directly decreased P availability, and also decreased *A. filiculoides* abundance, which influenced O_2 and NH_4^+ concentration of the surface water. Likely, the duration of the experiment was not long enough to find treatment effects of LMB on methanogen

community composition, as these changes occur in more extended time frames, and their analysis is further complicated by the fact that DNA from dead cells can remain in sediments for a long time (Carini et al., 2016).

Based on this study, there were no direct effects of dredging on methanotroph and methanogen activity and community composition. Furthermore, while other studies found decreased bacterial abundance (Wan et al., 2020) and diversity (Yin et al., 2021) after dredging, here dredging did not show such effects, possibly because of the limited amount of sediment removed. However, it is likely that the most active methanotroph and methanogen communities were removed during the dredging process, since these generally reside in the top part of the sediment (Conrad, 1996; On et al., 2005). Possibly, the removal of the most active methanogenic community could explain why CH_4 emissions decreased in the summer directly after dredging.

4.5. Application of LMB and dredging in management context

While it is clear that LMB and dredging can decrease CH_4 emissions, it is also important to consider limiting P input to lakes when aiming to decrease CH_4 emissions by reducing eutrophication. Many lakes suffer from severe external P pollution, e.g., from nearby agricultural fields (Schoumans et al., 2014), and such P input can potentially quickly offset benefits gained by applying LMB or dredging. Furthermore, especially the application of LMB is expensive, costing up to €200 kg P^{-1} bound compared to €15–35 kg P^{-1} reduced input for catchment measures (Mackay et al., 2014). Therefore, in cases where reducing P input can decrease eutrophication sufficiently, it is advisable to focus on P prevention rather than P remediation. However, in some countries, including the Netherlands, lakes also suffer from severe internal eutrophication due to P mineralization in the sediment (Smolders et al., 2006). In these circumstances, limiting P input is not sufficient and (repeated) in lake-measures are needed to control lake trophic status (Lürling and Mucci, 2020). In these situations, P prevention can be supplemented by P remediation through LMB addition and dredging to lower internal eutrophication and decrease CH_4 emissions.

5. Conclusion

To conclude, this study shows that LMB decreased CH_4 emissions, while dredging showed a trend towards decreasing total CH_4 emissions and significantly decreased diffusive CH_4 emissions. LMB decreased P availability, and decreased potential CH_4 production rates, likely by limiting the growth of *A. filiculoides*, an important driver of CH_4 emissions. The reduction in CH_4 emissions after LMB addition was higher than previously found (Zhan et al., 2021), likely because LMB affects lakes especially in longer time frames. Dredging prevented autumn and winter die-off of *P. crispus*, thereby likely decreasing carbon availability for CH_4 production. However, dredging decreased CH_4 emissions less than in a previous study (Zhao et al., 2021), which could be because of the limited dredging depth of the sediment in this study. Neither LMB nor dredging significantly affected microbial community composition, possibly because these changes require stronger disturbances and/or longer time frames. These results show that management of water quality targeting internal eutrophication has the potential to lower CH_4 emissions, which provides another incentive to improve water quality using in-lake measures in cases where P prevention cannot sufficiently reduce eutrophication.

CRediT authorship contribution statement

Thomas P.A. Nijman: Conceptualization, Methodology, Investigation, Formal analysis, Visualization, Writing – original draft. **Maxime Lemmens:** Investigation, Writing – review & editing. **Miquel Lurling:** Methodology, Resources, Writing – review & editing. **Sarian Kosten:** Resources, Writing – review & editing. **Cornelia Welte:** Conceptualization, Methodology, Supervision, Writing – review & editing. **Annelies J. Veraart:** Conceptualization, Methodology, Supervision, Writing – review & editing.

Data availability

Experimental data are available at the Data Archiving and Networked Services (DANS) EASY (https://doi.org/10.17026/dans-xkm-6k6k). Sequencing data are deposited at the European Nucleotide Archive under accession number PRJEB54968.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2022.157584.

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