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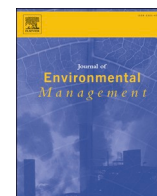
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Research article

Synthetic and biopolymers for lake restoration – An evaluation of flocculation mechanism and dewatering performance

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ABSTRACT

In the frame of the global phosphorus (P) crisis and ongoing eutrophication issues in the environmental sector, lake sediment can be considered as an alternative P source after its removal from eutrophic lakes. However, high water contents make sediment dewatering a crucial step towards the efficient reusability of remaining solids. The application of polymeric substances facilitates solid-liquid separation by flocculation of suspended particles. To lower the environmental risk of contamination with toxic, non-biodegradable monomeric residues during and after the application of synthetic polyacrylamide(PAM)-based polymers, switching to natural polymeric substances (biopolymers), e.g., starch- or chitosan-based, is increasingly emphasized. The dewatering performance of four conventional PAM-based polymers was compared to two starch- and one chitosan-based biopolymer. Laboratory experiments were conducted to determine the dewatering rate, floc size and strength, and reject water quality. Biopolymers generally caused the formation of smaller but less shear-sensitive flocs, and lower P levels in the reject water compared to synthetic polymers. Dewatering performance was correlated to the most important functioning influencing polymer-specific properties intrinsic viscosity (polymer extension) and surface charge density (CD). Due to the high CD and low intrinsic viscosity of the biopolymers, electrostatic patch flocculation seems to be the favored flocculation mechanism, while for synthetic polymers bridging seems to be dominating. Solid-liquid separation technologies should be adjusted to the resulting floc size and structure, while surface CD and intrinsic viscosity are important properties for the choice of biopolymer. Overall, biopolymers can function as a more environmentally friendly alternative to synthetic products for lake sediment dewatering accompanied by the potential for P recovery.

1. Introduction

The essential nutrient phosphorous (P) is currently facing both unsustainable use and resource (P rock) depletion (Boer et al., 2019; Cordell et al., 2009; Jupp et al., 2021). The main P resources are estimated to be exhausted within the next 100 years (Cooper et al., 2011), while an unequal global distribution might cause geopolitical challenges. For instance, in early 2022, China banned P rock exports for the rest of the year. Further, the price for triple superphosphate (conventional mineral P fertilizer) is now peaking and will be prone to continuous rise and fluctuations (Cordell et al., 2009). Meanwhile, ongoing excessive anthropogenic P input to the environment causes severe and costly eutrophication issues globally (Azam et al., 2019; Conley et al.,

2009; Jupp et al., 2021). Eutrophication is the result of excessive (anthropogenic) nutrient inputs, mainly nitrogen and P (Conley et al., 2009), which causes a cascade of nuisances in freshwater systems, such as harmful algae blooms, hypoxia, and a general loss of biodiversity (Battisti et al., 2016; Smith et al., 1999). The widespread pollution of the aquatic environment caused by P leakage from land to water has an estimated cost of £39.5 billion in the UK alone (Re-Focusing Phosphorus Use in the UK Food System, n.d.).

In ecological terms, lake sediment is often understood as a terminal sink for P. Depending on local conditions and sedimentation rates, lake sediment can contain a large legacy pool of potentially releasable P (Sondergaard et al., 2001). The resulting internal P loading from the sediment can be the major contributor to current eutrophication issues.

Abbreviations: PAM, polyacrylamide-based polymer; CD, charge density; MW, molecular weight; CS, chitosan; TDP, total dissolved phosphorus; SRP, soluble reactive phosphorus.

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Dredging, i.e., the removal of upper P-rich sediment, is a common internal restoration tool that has proven to effectively lower the extent of internal P loading (Cooke et al., 1972; Van Wichelen et al., 2007).

Until today, dredged sediment is mostly treated as contaminated waste material in Europe (Renella, 2021), which after removal is often deposited as a presumptive ‘waste product’ without further use. However, depending on local sediment compositions, there is a good opportunity to link lake restoration with resource recovery. Accordingly, few studies are currently investigating the potential for lake sediment as an alternative nutrient source, amongst others within the agricultural sector as P fertilizer (e.g., Kiani et al., 2021). Additionally, in 2020, the EU adopted a Circular economy action plan (2020) which enables a nutrient circular economy in Europe by providing the regulatory framework to facilitate the development and commercialization of recycled nutrient products.

Since surface sediments can contain a small dry matter fraction (DM %) (<10%) but a large amount of water (up to > 90%), sediment removal often involves a dewatering step before transport and disposal for volume and cost reduction (Song et al., 2020; Wei et al., 2018). Sediment dewatering consists of two basic steps: 1) coagulation and flocculation of suspended particles followed by 2) solid-liquid separation. A key mechanism for successful solid-liquid separation is the initial coagulation/flocculation step (Bolto, 1995; Wei et al., 2018). In the wastewater treatment sector, coagulation and flocculation processes are often promoted by the addition of organic polymeric substances, which are traditionally polyacrylamide (PAM) based chain molecules (Wei et al., 2018). Since a major part of suspended particles in sediment (mainly clay, silt, and organic compounds) can carry a negative surface charge (Bolto, 1995), the most effective flocculants for lake sediment dewatering are often cationic PAM-based polymers. The added flocculant readily adsorbs to the oppositely charged surface of suspended particles, which causes particle destabilization, and thus, coagulation due to surface charge neutralization followed by the formation of larger flocculates due to the bridging between smaller coagulates by free, excessive polymer chains (Bolto, 1995; Hogg, 2000; Yang et al., 2016). After the flocculation, several solid-liquid separation technologies can be applied, i.e., drainage by gravity, or the application of mechanical forces, such as filtration, vacuum pumps, belt filter press, or decanter centrifugation (e.g., Hjorth et al., 2011).

However, potentially toxic residual monomers released from the PAM-based polymer products (e.g., acrylamide or sodium acrylate) during dewatering can increase the risk of environmental trace pollution in the reject water (Bolto and Gregory, 2007; Duggan et al., 2019). In addition, the biodegradation of synthetic polymeric substances is extremely slow (Bolto, 1995). The increasing focus on micro- and nano plastics in the environment may hinder the recycling process of the sediment if synthetic polymers are used. Hence, a switch from synthetic PAM-based to products with a natural origin (hereafter biopolymers) has been emphasized recently to minimize the risk of environmental pollution (Bhatia, 2017; Bratskaya et al., 2005; Khachan et al., 2014). Biopolymers are defined as naturally occurring polymeric chain molecules that can be transformed into their basic forms (carbon dioxide, water, methane, and low molecular weight organic compounds) mediated by several abiotic or biotic degradation processes. Biopolymers often consist of polysaccharides such as starch, cellulose, and chitosan, as well as lignin or collagen (Bolto, 1995; Encalada, 2018). Known advantages of biopolymers are better biodegradability, non-toxicity, and cost-effectiveness (e.g., Bolto, 1995; Encalada, 2018; Yang et al., 2016).

An effective and environmentally friendly solution for sediment dewatering is an important step towards more sustainable lake restoration methods. From a circular economy point of view, the use of biodegradable products during sediment dewatering will be crucial for the reusability of the end product. For instance, if considering the potential uptake of micro- and nano plastics by plants (Mateos-Cárdenas et al., 2021; Wang et al., 2022). Several comparative performance studies on cationic PAM-polymers and biopolymers have been

performed (e.g., Bratskaya et al., 2005; Khachan et al., 2014), however, to the author's knowledge, literature is lacking direct polymer performance studies with a focus on dredged lake sediment for lake restoration in combination with its recycling potential in the agricultural sector. Meanwhile, the agronomic value of sediment has been confirmed and increasingly emphasized recently (e.g., Braga et al., 2019; Kiani et al., 2021; Renella, 2021).

To evaluate the possibility of using biopolymers for lake restoration with the final purpose of reusing P in the dredged sediment, e.g., as fertilizer in the agricultural sector, a dewatering performance study was conducted aiming at the development of an application protocol. The tests were also part of the preparation for a planned large-scale sediment removal in the period 2023–2024 from the shallow Danish Lake Ormstrup. The dewatering performance of three different biopolymer products, two based on starch (a polysaccharide originating from grain, roots, etc. composed of α -D-glucose units (Bolto, 1995; Encalada, 2018)), plus one based on chitosan (natural occurring polysaccharide, poly-b-(1/4)-2-amino-2-deoxy-D-glucose; derived from deacetylation of chitin (Bolto, 1995; Yang et al., 2016)) was compared to four commercially available organic cationic PAM-polymers recommended for lake sediment dewatering. The efficiency of the flocculation is dependent on mixing procedures, and the flocculant's specific chemical and structural properties, of which the most important ones have been defined as surface charge density (CD), ionic properties, special functional groups, and molecular weight (MW) (Bolto, 1995; Wei et al., 2018; Yang et al., 2016). The performance of the different flocculants was evaluated by measuring dewatering rates, floc size and strength, and the reject water quality, i.e., particle and nutrient removal. These results were compared to and correlated with the specific performance influencing properties of the respective polymeric compound, i.e., CD and polymer extension (intrinsic viscosity, proxy for the MW and chain flexibility) using simple linear regression.

2. Materials and methods

2.1. Lake sediment sampling and characterization

The shallow hypertrophic Lake Ormstrup (Bjerringbro, Denmark, 56°19'34.0"N 9°38'20.9"E) has an open surface area of 11 ha with an average water depth of 3.4 m (maximum water depth 5.5 m) (Søndergaard et al., 2022). Total sedimentary P concentrations in the upper 5 cm can reach 6 mg g DW⁻¹ (Søndergaard et al., 2022). In 2020–2022 planktivorous fish was removed as part of a large-scale restoration project involving biomanipulation efforts. Surface sediment (approx. depth 10–20 cm; ca. 33 L) was sampled in bulk from the deepest station (5.5 m) in the eastern part of Lake Ormstrup, using a sediment grab sampler. The sediment was stored at 4 °C until further experiments. Homogeneous sediment samples for the various experiments were ensured by thorough mixing of the sediment prior to subsampling and polymer addition. The dry matter content (DM%) of the sediment was 10.2% (loss of weight at 105 °C), and the TP content 1.8 mg P g DW⁻¹ (wet digestion of ~0.1 g ignited sediment, 8 mL 1 M HCl, 120 °C, 1 h, followed by spectrophotometrically soluble reactive P (SRP) measurement via the molybdenum blue method (Koroleff, 1983)). Detailed background information on sediment parameters were established prior to analysis (see supplementary material S1).

2.2. Polymer products and dosage

A summary of the tested polymer products and applied dosages can be found in Table 1. All polymer products were prepared and applied as a 0.2% (w/w for solids, v/v for liquids) solution in pure water (0.5% acetic acid in case of the chitosan) for optimal dosage according to the product supplier's recommendation (personal communication). The specific optimal dosing ratio between polymer and fresh sediment was assessed by preliminary bench-scale studies (supplementary material

Table 1

Tested polymer product names, active material, structure, molecular weight (MW), and optimal dosage (mL 0.2% polymer solution per 100 g fresh sediment) for lake sediment dewatering. Data was partly obtained from DanKalk, Solenis, and Kemira (product manufacturers).

Abbreviation	Active material (%)	Structure	MW	Dosage (mL 100 g ⁻¹)
PAM-1	cationic PAM (50)	crosslinked emulsion	medium – high	9
PAM-2	cationic PAM (50)	crosslinked emulsion	high	9
PAM-3	cationic PAM (50)	crosslinked emulsion	high	9
PAM-4	cationic PAM (50)	linear granulate	medium	9
Starch-1	heavily cationized starch (100)	granulate	amylose and amylopectin components, hard to	9
Starch-2	heavily cationized starch (starch, 2-hydroxy-3- (trimethylammonio) propyl ether, chloride; 1,2-Benzisothiazolin-3-one) (20)	emulsion	determine, but starch-1 > starch-2	10
CS	cationic chitosan (needs to be dissolved at acidic pH (e.g., 0.5% acetic acid) (100; pH dependent)	flakes	< than normal polyacrylamides, but > compared to other chitosan type products	8

S2).

2.3. Dewatering time

A simple gravity filtration test was used to determine the dewatering time. This method was comparable with the procedure used in other studies for testing flocculation processes (Hjorth et al., 2008; Olivier et al., 2018; Karadoğan et al., 2022). Ten mL 0.2% polymer solution, eventually supplemented with pure water to ensure a similar initial water content in all samples (depending on the respective dosage), was added to 100 g fresh sediment in a 100 mL glass beaker. The control treatment (unfloculated) received 10 mL of pure water only. After flocculation, the sediment was applied on a filter mesh (150 µm) and the filtrate (hereafter reject water) was collected in a volumetric glass cylinder beneath. The drained volume (V in mL) was noted every half minute until two and a half minutes, and then after 5 and 10 min (in replicates of 5 for each polymer product). The dewatering performance experiment was repeated with centrifugation for solid-liquid separation. The DM% of the remaining solid (hereafter cake) was determined as loss of weight at 105 °C.

2.4. Compressibility test

Analytical centrifugation was used to study the compressibility of flocculated lake sediment cakes (Lumiziser 613 Dispersion Analyser from L.U.M. GmbH, Berlin, Germany). The method was developed to have a fast and simple comparison of the compression of the cakes for different flocculants. For each test, two mL of flocculated sediment were added to rectangular cuvettes (10 mm). All polymers were tested as well as unfloculated sediment (control). The experiment was operated at 200 rpm (6×g) for 1000 s, 400 rpm (23×g) for 1000 s, and 600 rpm (52×g) continuing this procedure to 3000 rpm (1308×g) for 1000 s. After 1000 s at 600 rpm an observable cake was obtained. The cake height was measured at 600 rpm (h_{600}) and 3000 rpm (h_{3000}). Cake compressibility was calculated as

$$\text{compressibility} = \frac{h_{600} - h_{3000}}{h_{600}} \quad (1)$$

The maximum speed of the centrifuge was 3000 rpm. At 600 rpm all particles were deposited as a cake, thus, 600 and 3000 rpm were chosen to ensure a large change of compression pressure that gives the best possibility for measuring the compressibility.

2.5. P concentration in the reject water

After flocculation and solid-liquid separation by drainage, the reject water was filtered (1.2 µm GFC syringe filter) and the soluble reactive P (SRP) concentration was determined via the molybdenum blue method (Koroleff, 1983). Total dissolved P (TDP) was measured on the ICP-OES

in the unfiltered reject water.

2.6. Floc size and structure

The particle size distribution in the unfloculated and flocculated sediment was directly determined using a Hydro Mastersizer 3000 (Malvern). Each sample was run in four separate cycles (internal system settings) to check for floc destruction and possible variances during the measurements. Floc size and structure were visualized under a stereo microscope (Leica) equipped with a Leica Application Suite (LAS) EZ camera and using the LAS EZ software version 3.4.0. Freshly flocculated sediment was transferred to a glass Petri dish placed under the stereo microscope. Pictures were taken with 8 times magnification on a plain white background.

2.7. Residual turbidity

Residual turbidity was measured in the reject water after flocculation and centrifugation. Twenty-five grams of fresh lake sediment were flocculated in a 50 mL Falcon tube using the respective amount of 0.2% polymer solution (in replicates of four). Flocs were gently removed from the water phase by centrifugation (2 min, 3000 rpm), and absorption at 650 nm in the supernatant was measured spectrophotometrically (Thermo Scientific, Genesys 10 S UV–Vis) in a 1 cm cuvette to determine the residual turbidity in the reject water. Absorption values were recalculated into the standard unit for turbidity NTU (nephelometric turbidity unit) using a standard NTU curve determined at 650 nm and prepared from a 1000 NTU stabilized Formazin turbidity standard (StablCal® Standard, Hach®, Cat. 2660642).

2.8. Floc strength

Floc strength (shear sensitivity) was determined in a deflocculation experiment modified after Mikkelsen and Keiding (1999, 2002). Fresh lake sediment (350 mL) was flocculated in a 500 mL plastic beaker with the respective amount of 0.2% polymer solution and transferred into a four-baffled incubation reactor ($d = 10.5$ cm; $h = 16$ cm). Pure water (350 mL) was added to reach a final sample volume of 700 mL (34.5 ± 0.2 g SS L⁻¹). The sample was exposed to increasing mixing intensity (i. e., shear stress) using a Heidolph stirrer (RZR 2051 control) equipped with a single-bladed paddle at a set paddle stirring speed adjusted to different turbulent shear rates. The root mean square (RMS) velocity gradient G (s⁻¹) developed by Camp (1943) was used as a measurement for turbulent shear rates. Accordingly, the applied G values were 100, 200, and 800 corresponding to 380, 450, and 900 rpm paddle stirring speed, respectively. The samples were exposed for a total of 120 min of mixing and subsamples of five mL were taken after 0, 5, 10, 20, 40, 80, and 120 min using a ten mL plastic syringe attached to a silicon tube and transferred into clean 15 mL Falcon tubes for centrifugation (3000 rpm,

2 min). Residual turbidity was determined in the supernatant as described earlier (see section 2.7). One replicate per polymeric substance at the respective stirring speed was performed.

2.9. Polymer charge density and intrinsic viscosity

Charge density (CD) was determined using a standard protocol for colloid titration of polyelectrolytes modified after Ueno and Kina (1985). Half a milliliter of the 0.2% polymer solution of unknown CD was added to 100 mL pure water and mixed for 2 min at 150 rpm. One milliliter of cationic dye toluene blue (TB; 0.1 g L^{-1}) was added as the adsorptive indicator. The solution was titrated (200 μL steps) with the anionic polyelectrolyte potassium polyvinyl sulfate (0.2027 g L^{-1} ; PVSK) of known CD ($\sigma = 6.1651 \text{ meq g}^{-1}$). After each titration step absorption at 620 nm was measured spectrophotometrically. At the endpoint PVSK becomes excessive and TB is adsorbed to the anionic PVSK which is indicated by a color change from blue to red-purple

(metachromasy phenomenon). The CD of the polymer (σ_{polymer}) in meq g^{-1} was calculated as follows:

$$\sigma_{\text{polymer}} = \frac{N(V_{\text{PVSK, Blind}} - V_{\text{PVSK, sample}})}{CV} \quad (2)$$

where C is the concentration of the polyelectrolyte of unknown CD, V is the added volume of PVSK, and $N = 1.25 \text{ meq L}^{-1}$. $V_{\text{PVSK, Blind}}$ is the titration volume without polymer (blind), and $V_{\text{PVSK, sample}}$ the titration volume with the polymer just before the absorbance starts to decline.

Viscosity was determined using an ASTM kinematic Cannon-Fenske viscosimeter (size 150) after the Standard Test ASTM D-445, IP-71, ISO-3104 method. Nine mL 0.2% polymer solution in different dilutions was transferred into the viscosimeter and the time (seconds) needed for the liquid to run through was measured and used to calculate the reduced viscosity (η_{red}):

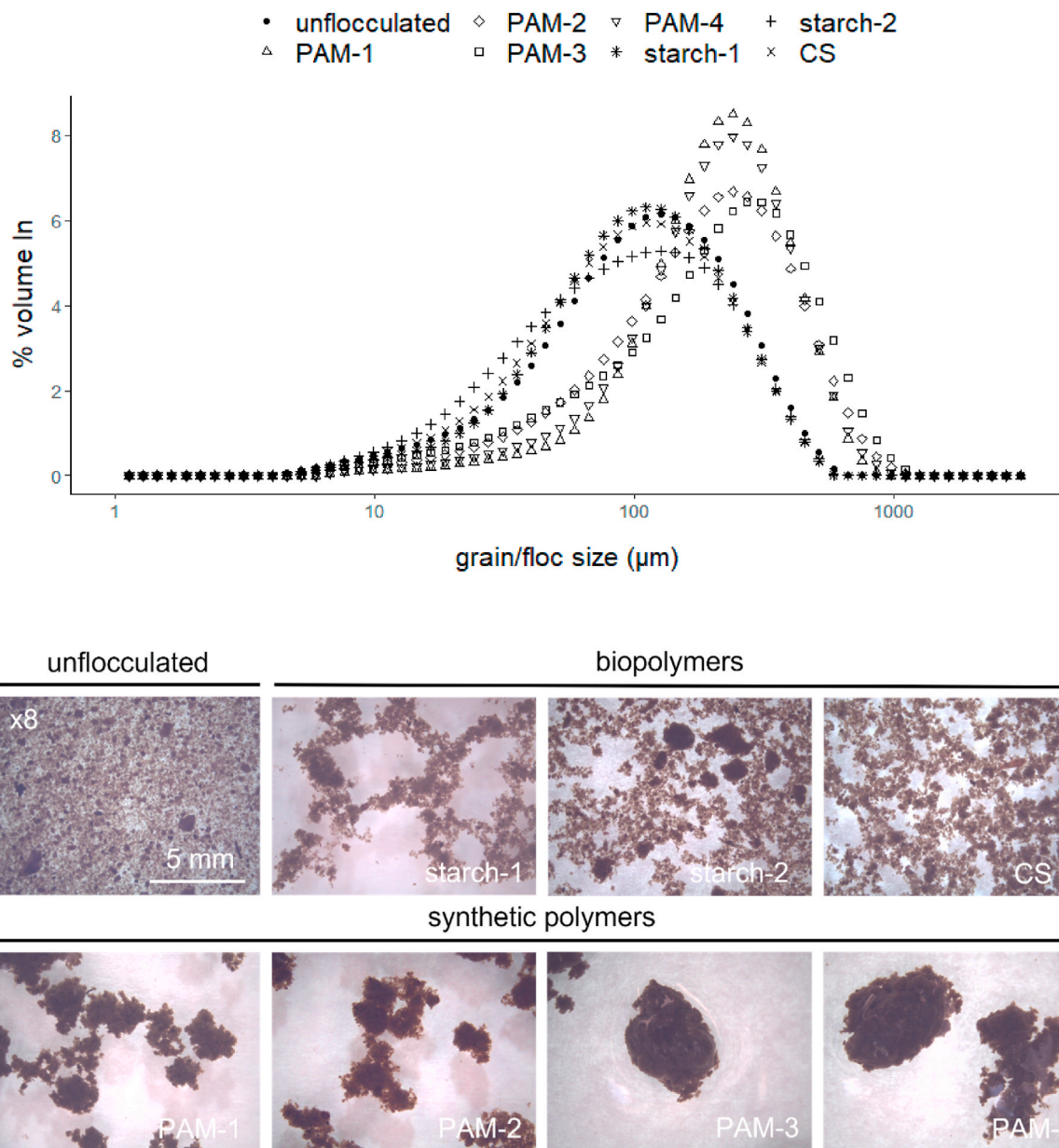


Fig. 1. Top: Grain/floc size distribution in the sediment after flocculation with PAM polymers, biopolymers, and the unflocculated sediment (control). Bottom: Stereo microscope images at 8× magnification, $10 \times 14 \text{ mm}$, on a white background using a Leica stereo microscope equipped with a Leica Application Suite (LAS) EZ camera and the LAS EZ software version 3.4.0.

$$\eta_{red} = \left(\frac{t}{t_0} - 1 \right) \varphi^{-1} \quad (3)$$

Where t is the time for the solution, t_0 is the time for the solvent and φ is the concentration of the polymer (%). The reduced viscosity was measured at different concentrations and plotted as a function of concentration. To get an estimate of the MW and extension of the polymer molecules, linear regression was used, and data was extrapolated to a concentration of zero to determine the intrinsic viscosity $[\eta]$:

$$[\eta] = \lim_{\varphi \rightarrow 0} \eta_{red} \quad (4)$$

3. Results and discussion

3.1. Flocculation efficiency – floc size, structure, and residual turbidity

Flocculation with biopolymers caused the formation of smaller flocs with a wider size distribution compared to synthetic polymers (Fig. 1; Table 2). Flocculation with all four PAM polymers resulted in a median floc size $>200 \mu\text{m}$, whereas flocculation with the three biopolymers produced flocs with a median size of around $100 \mu\text{m}$.

In sludge, Mikkelsen and Keiding (2002) classify all particles sized $0.5\text{--}5 \mu\text{m}$ as primary particles, whereas particles ranging from 25 to $100 \mu\text{m}$ are categorized as flocs. In sediment, most suspended particles are abundant as fine inorganic particles typically sized $<63 \mu\text{m}$ and representing the silt and clay fraction. Fine sand ranges between 63 and $250 \mu\text{m}$, whereas all particles $>2 \text{ mm}$ are termed gravel. Therefore, the silt and clay fraction in sediments can be seen as an indicator for the abundance of suspended primary particles. In the unflocculated sediment, the silt and clay fraction constituted $25 \text{ vol } \%$ (Table 2). After flocculation with the four PAMs, the silt and clay fractions in the solid phase were drastically reduced (5.9 , 12.2 , 13.8 , and 7.8% , respectively), which indicates that most fine inorganic particles sized $<63 \mu\text{m}$ were successfully incorporated in the flocs. Contrastingly, the flocculation by biopolymers resulted in a slight increase in the silt and clay fraction (25.1 , 33.0 , and 28.1% , respectively). However, due to the overall smaller floc size after biopolymer application, it might be difficult to distinguish between suspended particles and small particle aggregates based on particle size only. Therefore, residual turbidity in the reject water was used as a measure for the degree of primary particle incorporation into the flocs (Gregory, 2009). It can be assumed that the residual turbidity increases linearly with the amount of suspended primary particles (Mikkelsen and Keiding, 1999). Residual turbidity after flocculation with biopolymers ($0.97\text{--}2.9 \text{ NTU}$) and synthetic polymers ($1.4\text{--}2.7 \text{ NTU}$) was drastically reduced compared to the control (unflocculated; 225.4 NTU), indicating a high degree of primary particle incorporation into the flocs (Table 2). The lowest residual turbidity was detected after flocculation with starch-1 (0.97 NTU).

The flocs were inspected under a Leica stereo microscope to explore

differences in structure (Fig. 1). For the synthetic polymers, large and compact aggregates were noticed, whereas the biopolymer flocs showed a more open fractal structure. This can be linked to differences in the underlying collision and aggregation kinetics (Fig. 2). Here, two possible models were described by Gregory (1997). If one individual particle binds to larger aggregates (particle-cluster model), a compact structure is often formed. On the contrary, if aggregates bind to other aggregates (cluster-cluster model) more open structures are formed. Thus, flocs formed with biopolymers are most likely a result of cluster-cluster aggregation, whereas flocs with synthetic polymer are a result of the single particle-cluster model.

Additionally, the observed differences in floc size and structure indicated variations in the underlying flocculation mechanisms. Two possible flocculation mechanisms following the initial particle charge neutralization step have been described by Bolto (1995): 1) polymer bridging and 2) electrostatic patch flocculation (Fig. 2). Highly extended polymers (i.e., high intrinsic viscosity and MW) are expected to favor polymer bridging (Khachan et al., 2014; Wei et al., 2018). After a negatively charged particle is adsorbed to the positively charged polymer surface, the formation of larger flocs is caused by the looping and tangling of free leftover polymer chain molecules that exceed the electrical double layer and can interconnect particles in close vicinity (Bolto, 1995; Bolto and Gregory, 2007; Yang et al., 2016). In contrast, polymers with high surface CD will likely adsorb to the particles in a rather flat configuration, creating high-intensity patches or domains with localized excessive charges on the particle's surfaces, causing local charge reversal, and resulting in positive-negative attraction between particles (Gregory and Barany, 2011). Consequently, the direct electrostatic attraction between the particles is encouraged by the attractive forces between the domains of opposite charge resulting in enhanced agglomeration.

Accordingly, intrinsic viscosity was found to be higher for the tested PAM-based polymers, while biopolymers had a higher surface CD, except for starch-2 (Table 3). Intrinsic viscosity is a measure for the degree of the polymer extension. It should be noted that the backbone structure of synthetic polymers is a single carbon strain whereas for the biopolymer it is saccharide units which lowers the polymer chain flexibility, and thus, its intrinsic viscosity (Gregory and Barany, 2011).

The collected data suggest that biopolymers are functioning via electrostatic patch flocculation due to high surface CD, whereas polymer bridging is the favored flocculation mechanism after the application of the PAM-based polymers due to higher intrinsic viscosity, MW, and coiled configuration. This has been confirmed by several earlier studies, e.g., Bolto (1995), Hjorth and Jørgensen (2012), Hu et al. (2021), Song et al. (2020), Wei et al. (2018), and Yang et al. (2016). Additionally, Hjorth and Jørgensen (2012) describe a counteractive effect of very high CD for the bridging effect, since not enough charge on a particle surface was left to bridge the particles. They further state that small polymer size shows a reduced likelihood for polymer-particle collisions, i.e., less effective bridging, but enhances the formation of local charge patches on the particle surfaces. Likewise, in our study floc size was positively correlated with intrinsic viscosity ($R^2 = 0.68$; Fig. 3). Floc size and CD were only weakly correlated ($R^2 = 0.32$) but showed a negative relation. It indicates that the polymeric chain configuration (i.e., intrinsic viscosity) is of greater importance during flocculation than the surface CD.

In summary, biopolymers yielded smaller floc size, but similar residual turbidity compared to synthetic polymers, meaning that the flocculation efficiency of biopolymers in lake sediment is not affected, but filtration methods should be adapted to the final floc size. The observed differences in floc size and structure can be traced back to differences in the underlying flocculation and particle collision/aggregation mechanisms. Biopolymers are well known for their high surface charge but rather low chain length. Based on our data it is recommended to consider high surface CD but also moderate intrinsic viscosity for the choice of biopolymer for efficient lake sediment dewatering. It can also be taken into account to blend synthetic with biopolymers to benefit

Table 2

Median floc size ($n = 1$), silt and clay fraction ($n = 1$), and residual turbidity in the reject water (mean \pm SD, $n = 4$; co-efficient of variation ranging from 12.5 to 54.6% ; low level of values should be noted) after application of the respective polymeric substance and for the unflocculated fresh sediment (control).

	Median floc size (μm)	Silt and clay fraction (%)	Residual turbidity (NTU)
Control	115	25.0	225.4 ± 28.3
PAM-1	229	5.9	1.4 ± 0.7
PAM-2	206	12.2	1.5 ± 0.0
PAM-3	231	13.8	2.7 ± 1.5
PAM-4	224	7.8	1.9 ± 0.4
Starch-1	110	25.1	0.97 ± 0.4
Starch-2	99	33.0	2.9 ± 0.4
CS	106	28.1	1.9 ± 0.4

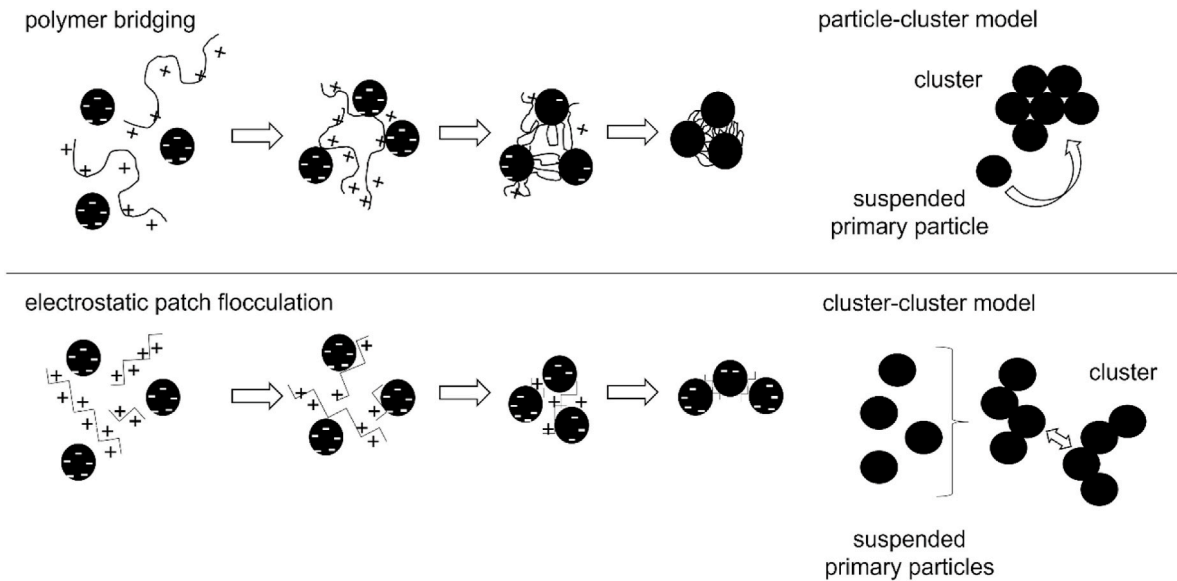


Fig. 2. Schematic illustration of the potential underlying differences in flocculation mechanism, particle collision and aggregation kinetics (redrawn from Bolto, 1995; Gregory, 1997) between biopolymers and synthetic polymers.

Table 3
Polymer surface CD measured by colloid titration (in replicates of 5, 3, and 1), and intrinsic viscosity (n = 1).

Polymer	Charge density (meq g ⁻¹)	Intrinsic viscosity (n.d.)
PAM-1	2.0	30.0
PAM-2	1.1	12.1
PAM-3	1.4	22.5
PAM-4	1.3	14.4
Starch-1	5.0	3.5
Starch-2	0.7	1.6
CS	5.7 (pH ~ 5.1)	10.6

from a combined effect and be able to increase dewatering efficiency and reduce the environmental impact, simultaneously.

3.2. Dewatering performance

Flocculation with biopolymers resulted in lower dewatering rates compared to synthetic polymers (Fig. 4). Almost twice as much reject water was collected after flocculation with the synthetic polymers and solid-liquid separation using drainage by gravity. The final reject water volume was similar for all tested synthetic polymers, while sediment

flocculated with PAM-1 (median floc size 229 μm; CD = 2 meq g⁻¹; intrinsic viscosity = 30) reached the maximum drained volume the fastest (<1.5 min). Differences in dewatering rate and final reject water volume were not observed among the biopolymers, however, the least water could be removed after application of the CS biopolymer (median floc size 106 μm; CD = 5.72 meq g⁻¹; intrinsic viscosity = 10.6).

Floc size seemingly determined the dewatering performance, as also stated by Gregory (1997), Hogg (2000), and Wei et al. (2018). Due to the overall smaller floc size after flocculation with biopolymers, the observed reduced dewatering efficiency could be explained by 1) the slower filtration kinetic because of a higher hydraulic resistance or 2) by the equilibrium dry matter which can be explained by an increased water holding capacity (WHC). Decreased particle size is known to increase the maximum WHC in soils due to the larger surface area (Verheijen et al., 2019). At decreased particle size the relative surface area (surface per area particle) increases dramatically, which in turn increases the hydraulic resistance and thereby dewaterability (Happel, 1958).

Wu et al. (2020) describe the water retained in biological flocs in form of different water fractions reflecting the solid-water binding strength which influences the water removal efficiency. Surface-adhered water refers to the water bound to the surface via hydrogen bonds. Interstitial water is an effect of the open orientation of the flocs and

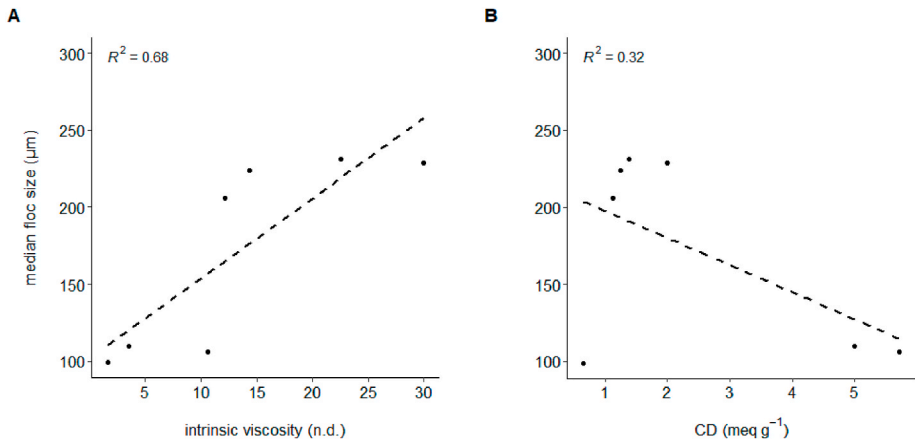


Fig. 3. Correlation between the polymer's (A) intrinsic viscosity (polymer extension; n. d.), (B) CD (meq g⁻¹), and the median floc size (μm).

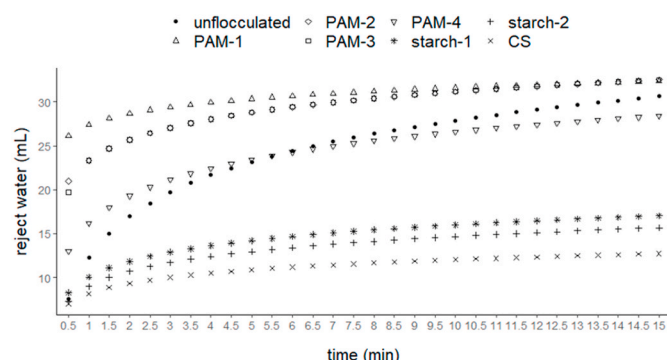


Fig. 4. Dewatering efficiencies plotted as the drained reject water volume (mL) over time (min) for each polymer product ($n = 5$).

small pores formed during filtration, which can create capillary effects. The latter is harder to remove due to the open particle structure, which might explain why the dewatering rate by drainage was reduced after biopolymer application. Concurrently, Wei et al. (2018) stated a good dewatering performance of organic synthetic polymers due to their larger floc size, and thus, more void spaces and less resistance for the water to drain.

The compressibility of all PAM-based flocculated samples was found to be high (Table 4) which might be due to the more open structure of the solid during the centrifugation and a result of the liquid between the flocs, suggesting that water can be removed at relatively low pressure and after that be consolidated to ensure high dry matter content. In contrast, the rather low observed compressibility after biopolymer application (Table 4) demonstrates that more water is retained, and thus, requires mechanical forces for solid-liquid separation, such as a belt filter press, decanter centrifugation, or application of vacuum to ensure a higher dry matter content of the dewatered sediment.

Conclusively, it might be necessary to combine sediment dewatering using biopolymers with a mechanical solid-liquid separation tool after flocculation. In our study, the highest DM% contents in the cake were reached after the application of biopolymers followed by solid-liquid separation via centrifugation (Table 4; 15 and 16% for starch-1 and CS, respectively), whereas sediment flocculated with PAM-based polymers resulted in cake DM% ranging from 12 to 15% after centrifugation. Lower DM% in the cake was reported for all tested polymers when the flocculated sediment was drained only, ranging from 7.1 to 8.9%. Hence, combining biopolymer flocculation with solid-liquid separation using, i.e., centrifugation or vacuum application, can be recommended to increase dewatering efficiency. Likewise, Wu et al. (2020) recommend centrifugation for high floc strength and vacuum for middle floc strength. Incurring energy costs might be overweight by the dewatering efficiency as well as the environmental benefit and reusability of the resulting end product when a biopolymer is used. Although the tested biopolymer products are not yet commercially available, price

estimations assuming large-scale production were made and communicated by the product manufacturer. Biopolymer production costs are expected to be the same as for synthetic ones, even showing a potential for cost reduction.

3.3. Floc strength

A deflocculation experiment was performed to investigate the sensitivity towards shear stress of the different polymer flocs, i.e., floc strength. An increase of residual turbidity with the product of shear force and time ($G \times t$) indicates a breakdown of the flocs. Flocs by synthetic polymers were more shear sensitive over time compared to biopolymer flocs (Fig. 5). Residual turbidity increased drastically with increasing $G \times t$ when the sample was flocculated with synthetic polymers reaching values > 500 NTU, while the maximum detected residual turbidity for biopolymer flocculated samples remained below.

The observed differences in floc size and structure might be a possible explanation for the observed trend in shear sensitivity. Hogg (2000) mentions that large-sized flocs are more fragile and sensitive to breakage during exposure to turbulent shear stress. Generally, the underlying mechanisms of deflocculation are defragmentation (splitting) and erosion (removal of single particles from the surface). The larger size of the synthetic flocs might exceed the Kolmogorov microscale length at the tested average shear rates (Thomas et al., 1999). Typically, flocs are fractal, meaning that density decreases with increasing floc size (Gregory, 1997). This can result in enhanced fragmentation and erosion of larger flocs which causes the release of smaller particles into solution, and thus, increase in residual turbidity. Likewise, a positive relation between median floc size and final residual turbidity was found at all applied shear rates (Fig. 5). Further, the lower shear sensitivity of biopolymer flocs can be explained by the observed differences in particle collision kinetics and flocculation mechanisms. The high surface CD of the biopolymers likely creates stronger electrostatic interparticle bonding via the local patch flocculation mechanism, making them less susceptible to mechanical defragmentation or surface erosion of particles. Additionally, the dominating bridging function of the PAM polymers can be deteriorated due to partly breakdown of the long chain molecules during shear stress.

In conclusion, the observed reduced shear stress sensitivity of flocs after biopolymer application enables the application of a wider range of mechanical solid-liquid separation tools other than only sedimentation or filtration, which is often recommended for the application of synthetic polymeric substances (Bolto, 1995; Bolto and Gregory, 2007).

3.4. Reject water quality – P retention

The removal of inorganic and organic pollutants is of high priority during the wastewater treatment process, since these can cause pollution of the receiving environment and add tax penalties to the treatment plant, as in Denmark. Overall, reject water quality was comparable between the application of biopolymers and PAM-based substances. An overview of the general reject water quality can be found as supplementary material (S3). In our study, P retention in the cake was of particular interest considering the circularity of the approach: reintroduction of the clean, low on nutrients reject water to surface waters, while the dewatered sediment can be reused in the, e.g., agricultural sector as fertilizer or soil amendment.

Soluble reactive P (SRP) concentrations in the reject water ranged from 0.22 to 0.83 mg L⁻¹ (starch-1 and PAM-3, respectively), which is below the maximum permissible limit of reactive P (1 mg L⁻¹) according to the standards for effluent discharge regulations (United Nations, 2002). It is also below peaking water column concentrations of Lake Ormstrup in summer (ca. 1 mg L⁻¹). Biopolymer application resulted in the lowest SRP concentrations (Fig. 6), with CS reaching the lowest level of total dissolved P (TDP; 0.22 mg L⁻¹). After flocculation with CS, the SRP concentration was almost equal to TDP (0.26 and 0.22 mg L⁻¹,

Table 4

Cake DM% after two different solid-liquid separation methods 1) drainage and 2) centrifugation (mean \pm SD, $n = 4$; co-efficient of variation $< 5\%$), and cake compressibility determined by analytical centrifugation.

Polymer	DM%		Compressibility %
	Drainage (150 μ m)	Centrifugation (3000 rcf)	
Control	7.1 \pm 0.1	13.8 \pm 0.1	42
PAM-1	8.9 \pm 0.1	15.1 \pm 0.2	52
PAM-2	8.4 \pm 0.1	14.8 \pm 0.2	57
PAM-3	7.8 \pm 0.2	12.9 \pm 0.2	56
PAM-4	7.9 \pm 0.4	12.3 \pm 0.3	53
Starch-1	8.3 \pm 0.1	15.2 \pm 0.1	49
Starch-2	8.5 \pm 0.1	12.7 \pm 0.2	46
CS	8.4 \pm 0.2	15.7 \pm 0.1	48

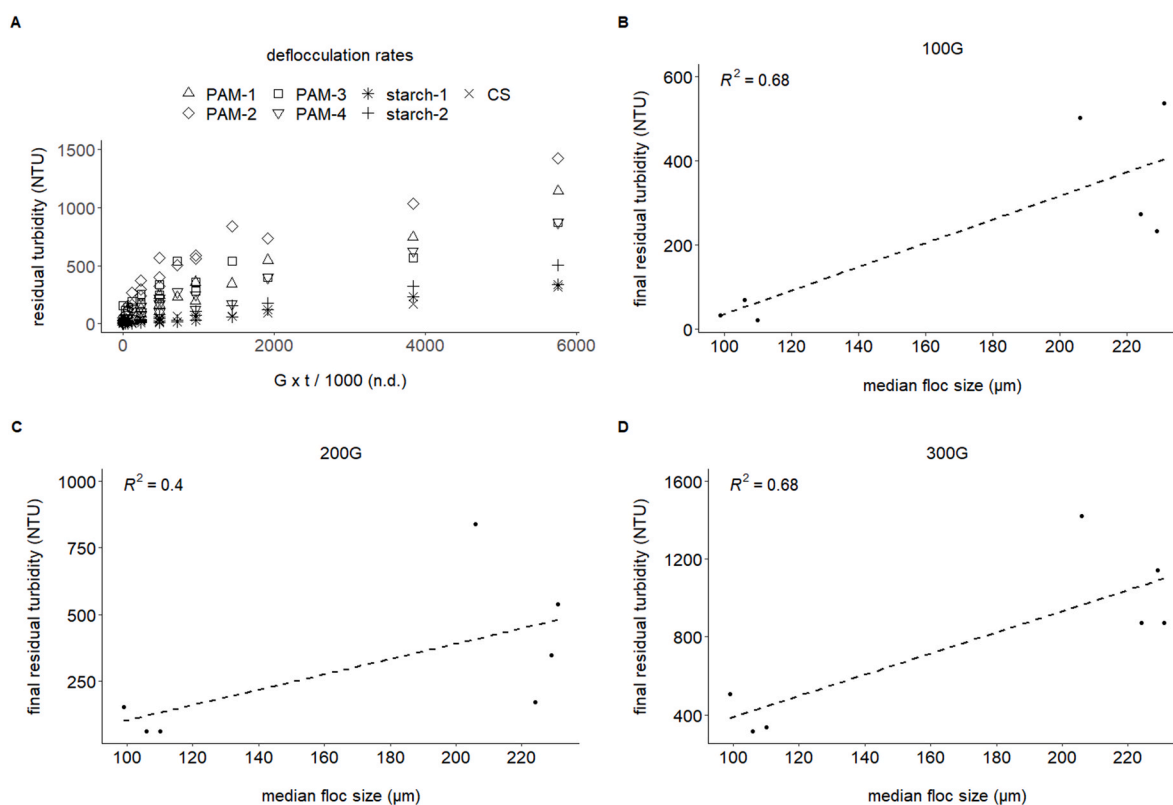


Fig. 5. Results from the deflocculation experiment. A: Residual turbidity (NTU) as a function of the applied RMS velocity gradient G (s^{-1}) multiplied by time (s) and divided by 1000 (for better readability). B–D: Final residual turbidity (NTU) as a function of median floc size (μm) at the three different applied shear rates.

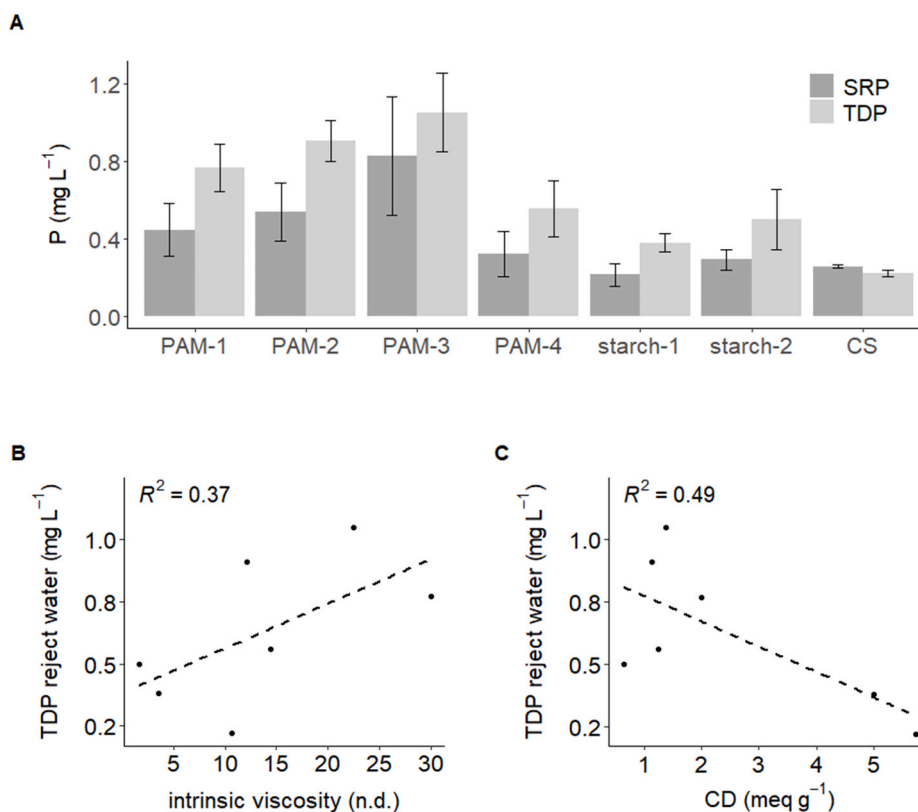


Fig. 6. A: Concentration of SRP and TDP in the reject water (mg L^{-1}) after flocculation and solid-liquid separation via drainage (150 μm). Mean \pm SD, $n = 4$. B and C: Correlation between TDP in the reject water and the polymer properties intrinsic viscosity and surface CD.

respectively), which indicated that all particulate P was successfully retained in the solid. The good performance of the highly positively charged CS was followed by starch-1, starch-2, and finally PAM-4.

Kofinas and Kioussis (2003) describe P removal via sorption processes using a cross-linked polyamine polymeric hydrogel as a sorbent. A positive relation between intrinsic viscosity and TDP, but a negative relation between CD and TDP levels in the reject water, was found. However, correlations were rather weak (Fig. 6). Based on the obtained data it was possible to calculate the overall amount of P being lost in the reject water. Theoretically, <1% of the initial TP in the sediment before dewatering would end up in the reject water in the case of all tested polymeric substances which favors the agronomic and nutritional value of the cake.

3.5. Environmental relevance and future perspectives

This study has proven that a transition from synthetic PAM-based polymers to natural-based substances, such as starch and chitosan, is possible and recommended for lake sediment dewatering. The application of biopolymers in lake sediment dewatering is beneficial for both lowering the environmental risk and broadening recycling options of the reject water and remaining solid phase towards a more circular approach in lake restoration. However, on the transformative pathway from sink to resource several intermediate challenges must be considered. Sediments can often be polluted with heavy metals (e.g., Cu, Zn, and Pb), as well as persistent organic pollutants depending on their origin (Ahlf and Förstner, 2001). Hence, further research on the removal of such pollutants by polymeric substances is necessary. Additionally, biodegradability assessments for biopolymers are recommended and the bioavailability of P from the cake needs to be assessed.

4. Conclusions

- Biopolymers caused the formation of smaller but less shear-sensitive flocs compared to PAM-based polymers, which did not impact flocculation efficiency but lowered the dewatering rates by drainage only;
- Differences in flocculation mechanism, as well as particle collision and aggregation kinetics, might be underlying due to higher surface CD but lower intrinsic viscosity of biopolymers compared to PAM-based polymers;
- Biopolymer application is recommended to be combined with mechanical solid-liquid separation adjusted to the respective floc size, such as the application of vacuum or centrifugation;
- Lower shear stress sensitivity of flocs after biopolymer application widens the options for mechanical solid-liquid separation tools and can increase the efficiency with regards to the final reject water quality and cake DM%;
- Concentrations of TDP and SRP in the reject water were generally lower after biopolymer application compared to PAM-based polymers, which favors P retention in the cake, and thus, increases the nutritional value of the remaining solid;
- It can be recommended to choose a biopolymer of high CD and moderate intrinsic viscosity to achieve high dewatering efficiency and (P) nutrient retention in the solid, alternatively, biopolymers could be blended with synthetic polymers to combine effects;
- Biopolymers represent an efficient alternative to synthetic substances for more sustainable lake sediment dewatering, increasing the potential to reintroduce the reject water, as well as reusability options of the remaining solid, e.g., in the agricultural sector as P fertilizer.

Author contributions

The manuscript was written through equal contributions of all authors. All authors have approved the final version of the manuscript.

Sina Haasler: conceptualization, methodology, visualization, investigation, writing – original draft preparation, editing, and revision. Morten Lykkegaard Christensen: conceptualization, methodology, supervision, writing – reviewing and editing. Kasper Reitzel: conceptualization, supervision, writing – reviewing and editing.

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Notes

No additional notes.

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.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jenvman.2022.117199>.

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