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# Sustainable coagulative removal of microplastic from aquatic systems: recent progress and outlook

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Microplastic (MP) pollution represents a critical challenge for global water quality due to its persistence, ubiquity, and ecotoxicological impacts. While conventional coagulation/flocculation–sedimentation (CFS) processes using chemical coagulants are partially effective, they often entail high energy demands, toxic residuals, and environmental trade-offs. This article provides a comprehensive and up-to-date review of recent advances in the use of natural coagulants (NCs) derived from plant, animal, and microbial sources as sustainable alternatives for MP removal from aquatic systems. The novelty of this work lies in its integrative analysis of bio-coagulant performance with hybrid formulations, nano-enhanced composites, and process intensification strategies such as enzyme activation. Through critical synthesis of various peer-reviewed studies published between 2020 and 2025, the review highlights that NCs such as *Moringa oleifera*, chitosan, *Cactus mucilage*, and microbial EPS can achieve MP removal efficiencies exceeding 90% under optimized conditions, with significantly reduced sludge toxicity and carbon footprint. Furthermore, the review identifies key performance parameters; pH, ionic strength, NOM interference, and coagulant modification techniques that influence the physicochemical mechanisms driving MP-coagulant interactions, including charge neutralization, bridging flocculation, hydrophobic association, and bio-adhesion. Pilot-scale evaluations demonstrate the feasibility of hybrid systems (e.g., chitosan-FeCl<sub>3</sub>, *Moringa*–alum) in achieving near-complete removal (up to 99.8%) of MPs across a range of polymer types and sizes. However, critical limitations remain, such as variability in raw material composition, reduced efficiency for MPs <10 μm, and scalability constraints. The study concludes that although NCs cannot yet fully replace synthetic ones at scale, their use as coagulant aids or in hybrid systems shows promise for sustainable water treatment. Future research should focus on standardizing extraction methods, improving bioengineering for higher protein yields, and developing smart coagulation systems for adaptive control across various water matrices.

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## 1. Introduction

Microplastics (MPs), defined as synthetic polymer particles ranging from 1 μm to 5 mm, have become pervasive global contaminants due to their environmental persistence and continuous input from multiple anthropogenic sources.<sup>1</sup> Recent studies demonstrate that approximately 8 million metric tons of plastic enter oceans annually, with secondary MPs from degraded macroplastics accounting for 69–81% of total MP loads in aquatic systems.<sup>2,3</sup> The 2021 UNEP report identified textile laundering as a major MP source, with a single wash releasing 700 000 microfibers,<sup>4,5</sup> while urban runoff contributes

30–35% of MP loads in freshwater systems.<sup>6,7</sup> Once dispersed, MPs exhibit complex transport dynamics influenced by their density (0.85–1.41 g cm<sup>−3</sup>), shape (fragments, fibers, spheres), and surface chemistry, with recent modeling showing 34% of oceanic MPs reside in surface waters while 66% accumulate in sediments.<sup>5,8</sup> Their environmental impacts are exacerbated by large specific surface areas (up to 3000 m<sup>2</sup> g<sup>−1</sup>) that facilitate adsorption of persistent organic pollutants (log *K*<sub>OW</sub> 3–7), with studies documenting MP-associated concentrations of PCBs and DDTs 10<sup>6</sup> times higher than ambient seawater.<sup>9,10</sup> Ecotoxicological research has demonstrated dose dependent effects across trophic levels, including 17–35% reduced filtration rates in mussels,<sup>11,12</sup> 50% decreased reproductive output in copepods,<sup>2,11</sup> and biomarker responses in fish indicating oxidative stress and neurotoxicity at environmentally relevant concentrations (10–100 particles per L).<sup>13</sup> Conventional water treatment processes show variable MP removal efficiencies, with primary sedimentation removing 50–80% of particles >100 μm but only 10–30% of 1–10 μm particles.<sup>14–17</sup> Advanced tertiary treatments achieve higher performance (95–99.9%), but face

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practical limitations – membrane filtration requires 3–5 bar operating pressures (energy demand:  $0.5\text{--}1.2\text{ kWh m}^{-3}$ ),<sup>18,19</sup> while electrocoagulation consumes  $15\text{--}30\text{ kWh m}^{-3}$  for 92–97% removal.<sup>2,20</sup> These challenges have driven research into natural coagulants (NCs), with *Moringa oleifera* seed extract demonstrating 89% removal of  $10\text{--}100\text{ }\mu\text{m}$  polyethylene particles at  $200\text{ mg per L}$  dose through dual mechanisms of charge neutralization ( $+15\text{ mV}$   $\zeta$ -potential shift) and polymer bridging.<sup>21,22</sup> Chitosan shows particular promise, achieving 94% removal of polystyrene microspheres ( $50\text{ mg per L}$  dose, pH 6.5) with floc formation following second-order kinetics ( $k = 2.3 \times 10^{-4}\text{ L mg}^{-1}\text{ min}^{-1}$ ).<sup>23,24</sup> Hybrid systems combining NCs with conventional processes exhibit enhanced performance.<sup>2,25</sup> While chitosan-assisted electrocoagulation reduced energy consumption by 40% compared to conventional methods.<sup>24,26</sup> However, key challenges remain, including variability in natural coagulant composition ( $\pm 15\%$  performance variation between harvests),<sup>22</sup> incomplete understanding of MP-coagulant interaction mechanisms at molecular scales,<sup>14,16</sup> and lack of standardized protocols for evaluating removal efficiency across different MP types (polymer chemistry, size fractions, aging states).<sup>1</sup> The primary objective of this review is to provide a comprehensive and technically rigorous synthesis of recent advances (2020–2025) in the use of NCs for the removal of MPs from aquatic environments. The review classifies NCs based on their biological origin; plant-derived, animal-based, and microbial and critically examines their active components, extraction procedures, and core coagulation mechanisms. It further evaluates their performance across a wide spectrum of MP types, considering variations in polymer composition, particle size, surface properties, and water matrix conditions.

Particular attention is given to the physicochemical interaction mechanisms governing MP removal, including charge neutralization, bridging flocculation, hydrophobic interactions, and bio-adhesion, supported by recent visions from advanced characterization techniques and molecular simulations. Moreover, the review explores emerging innovations such as hybrid coagulant formulations, nanostructured composites, enzyme-functionalized systems, which collectively enhance removal efficiency, operational flexibility, and scalability. In doing so, the study also identifies key limitations including raw material variability, reduced efficiency for sub-micron particles, and process sensitivity to environmental factors such as pH, salinity, and dissolved organics and proposes practical strategies for their modification.

## 2. Methodology

The growing concern regarding MPs pollution in aquatic environments has led to a surge in scientific research exploring efficient removal strategies. As illustrated in Fig. 1a, there has been a substantial increase in the number of published studies between 2020 and 2025 that focus on MPs removal from various aqueous systems. Coagulation-based treatment processes have emerged as a promising and scalable approach for MP removal. The distribution of studies by treatment approach, as shown in Fig. 1b, highlights that conventional chemical coagulants such as polyaluminium chloride (PAC) and aluminium sulphate are the most widely studied, accounting for approximately 40% of the total literature. Iron-based coagulants represent another 20%, favored for their strong performance in waters with high turbidity or organic content. Notably, NCs derived from plant-

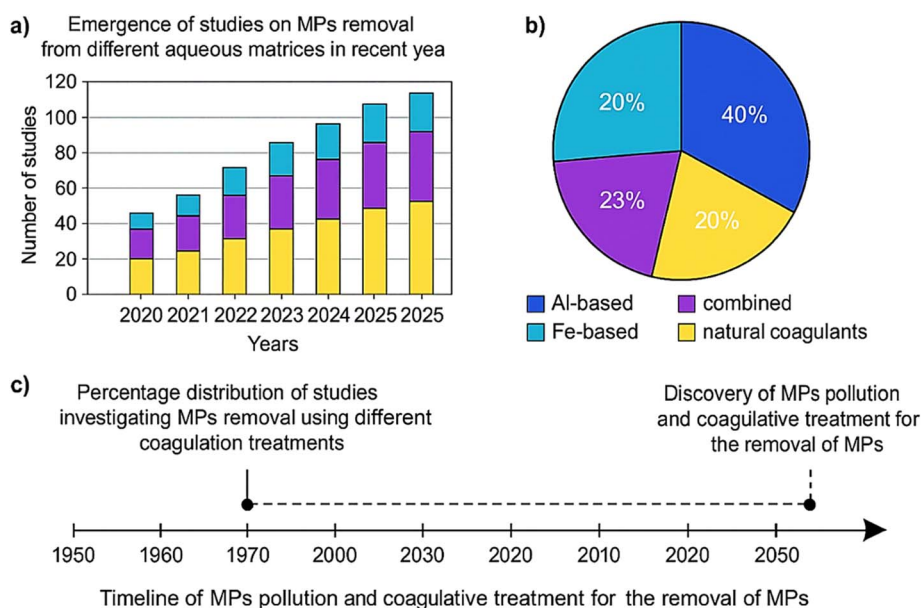


Fig. 1 (a) Emergence of studies on MPs removal from different aqueous systems in recent years (2020–2025), (b) percentage distribution of studies investigation MPs removal using different coagulation treatment approach including NCs (data retrieved from Web of Science and Scopus database), and (c) timeline of MPs pollution and coagulative treatment for the removal of MPs (data retrieved from Web of Science and Scopus database).

based materials such as *Moringa oleifera*, tannins, or chitosan comprise 20% of the published work, indicating growing interest in environmentally benign alternatives that reduce chemical dependency and sludge toxicity. An additional 23% of studies explore hybrid approaches, combining traditional coagulants with natural polymers or flocculant aids like polyacrylamide (PAM), which have shown to enhance MP aggregation and removal across a wider range of particle sizes and types. The historical evolution of scientific attention to MPs and their treatment through coagulation is outlined in Fig. 1c. Although MPs were first identified in the environment several decades ago, focused studies on their removal through coagulative treatment did not begin until the mid-2010s, gaining momentum in the early 2020s. This timeline demonstrates how advancements in detection methods, public awareness, and environmental policies have catalyzed the development and application of coagulation-based technologies for MP mitigation. Despite the promising results highlighted across the literature and in Fig. 1a–c, several challenges remain. Field-scale validation, long-term performance assessments, and post-treatment sludge management are areas that remain underexplored. These limitations point to the need for more pilot and demonstration-scale studies using real water matrices and a broader range of MP morphologies. The observed trends (Fig. 1a–c) affirm a research trajectory that is increasingly aligned with environmental sustainability and real-world applicability.

### 3. Microplastic pollution: sources and impacts

MPs originate from diverse anthropogenic sources, which can be broadly categorized into primary and secondary MPs. Primary MPs are intentionally manufactured at MP sizes,

including microbeads used in personal care products (typically 10–500  $\mu\text{m}$ ), industrial abrasives, and plastic pellets (nurdles) used as raw material in plastic production. Secondary MPs result from the environmental degradation of larger plastic items through processes such as photodegradation by UV radiation, mechanical abrasion from wave action, and biological degradation. Studies estimate that secondary MPs account for 69–81% of total MP loads in aquatic environments, with the breakdown of plastic packaging, fishing gear, and textile fibers being major contributors as listed in Table 1.<sup>12,27</sup> The pathways of MP entry into aquatic ecosystems are complex and varied. Urban runoff has been identified as a significant vector, transporting 30–35% of MP loads in freshwater systems, with tire wear particles and road dust representing substantial but often overlooked sources.<sup>1,28</sup> Wastewater treatment plants (WWTPs) serve as important conduits, with a single laundry cycle releasing approximately 700 000 microfibers from synthetic textiles. Atmospheric deposition has recently been recognized as a notable transport mechanism, with studies demonstrating MPs in remote locations such as Arctic snow and mountain tops, suggesting long-range atmospheric circulation of these particles.<sup>29,30</sup> Agricultural practices also contribute substantially, with plastic mulch films and bio-solid applications introducing MPs into terrestrial systems that eventually reach aquatic environments through soil erosion and runoff.<sup>31</sup> The environmental impacts of MPs are complex and concerning. Their small size and high surface area to volume ratio (up to 3000  $\text{m}^2 \text{g}^{-1}$  for fragmented particles) make them effective vectors for pollutant transport. MPs have been shown to adsorb and concentrate persistent organic pollutants (POPs) such as polychlorinated biphenyls (PCBs) and dichlorodiphenyltrichloroethane (DDT) at concentrations up to  $10^6$  times greater than surrounding seawater.<sup>12</sup> This chemical hitchhiking effect is particularly concerning given the demonstrated

**Table 1** Comprehensive overview of MP sources, pathways, and impacts

Category	Subcategory	Key findings	Quantitative data	Ref.
Origin & classification	Primary MPs	Intentionally manufactured (microbeads, nurdles, abrasives)	Size: 10–500 $\mu\text{m}$	12 and 27
	Secondary MPs	Result from macroplastic degradation (photodegradation, abrasion)	69–81% of aquatic MP loads	12 and 27
Entry pathways	Urban runoff	Tire wear, road dust, synthetic fibers	30–35% of freshwater MP loads	1 and 28
	WWTPs	Laundry effluent (synthetic textiles)	700 000 microfibers per wash	29 and 30
	Atmospheric deposition	Long-range transport to remote areas (Arctic, mountains)	Documented in Arctic snow	29 and 30
Environmental impacts	Agricultural inputs	Plastic mulch, biosolids $\rightarrow$ soil erosion $\rightarrow$ aquatic systems	Significant but unquantified	29 and 30
	Pollutant transport	Adsorption of POPs (PCBs, DDTs)	$10^6 \times$ higher than ambient seawater	12
	Biological effects	- Copepods: reduced reproduction - Mussels: false satiety, $\downarrow$ thermal tolerance	50% $\downarrow$ reproductive output; 2 $^\circ\text{C}$ $\downarrow$ tolerance	33
	Trophic transfer	MPs move through food chains (zooplankton $\rightarrow$ fish $\rightarrow$ humans)	Cellular uptake of <10 $\mu\text{m}$ particles	11 and 12
Persistence	Economic costs	Marine ecosystem damage, fisheries losses	\$2.5 trillion per year globally	8 and 33
	Degradation rates	Half-lives in marine sediments: - Polyethylene: 58 years - Deep-sea: $\leq 1200$ years	Century-scale persistence	8 and 12



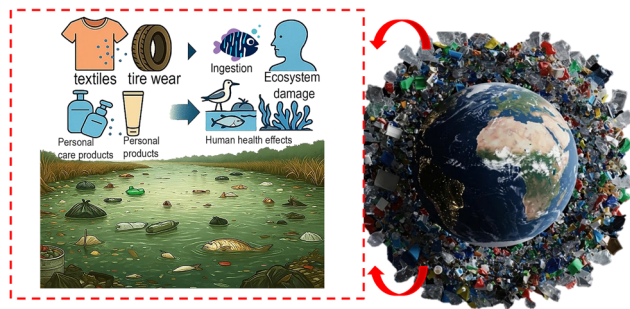


Fig. 2 MPs common sources and impacts on the environment and health.

bioavailability of these contaminants when ingested by marine organisms. The physical presence of MPs in organisms can cause intestinal blockages, false satiety, and reduced energy reserves. A landmark study by Langenfeld *et al.* (2024) showed 50% decreased reproductive output in the copepod *Calanus helgolandicus* at environmentally relevant concentrations (10–100 particles per mL).<sup>32</sup>

At the ecosystem level, MPs have been shown to alter sediment microbial communities and biogeochemical processes. Laboratory experiments have demonstrated that MP contamination can reduce the thermal tolerance of mussels by up to 2 °C, potentially affecting their survival under climate change scenarios.<sup>33</sup> The trophic transfer of MPs has been well-documented, with particles moving from zooplankton to fish to top predators, including potential implications for human health through seafood consumption. Emerging research suggests that the smallest MP fractions (<10 µm) may cross biological barriers, with demonstrated cellular uptake and potential for translocation to various organs.<sup>11,12</sup> The economic impacts are equally concerning, with estimates suggesting annual costs of \$2.5 trillion to marine ecosystems globally due to plastic pollution, including fisheries losses, tourism impacts, and cleanup costs.<sup>11,12</sup> Perhaps most alarmingly, the persistence of MPs in the environment is measured in centuries rather than decades, with half-lives estimated at 58 years for polyethylene in marine sediments and up to 1200 years for some polymer types in deep-sea conditions.<sup>8,12</sup> This extreme persistence, combined with continuous inputs, suggests that MP pollution represents a growing and potentially irreversible environmental challenge without immediate and sustained intervention (Fig. 2).

## 4. Challenges of MPs removal in WTPs

The removal of MPs in water treatment plants (WTPs) presents a growing technical challenge due to the complex physico-chemical properties of these persistent pollutants and limitations in conventional treatment infrastructure. Table 2 summarizes the MPs removal efficiency across water treatment processes, along with their associated limitations. Recent studies highlight that while primary treatment processes (screening, sedimentation) can remove 50–80% of MPs larger than 100 µm, their efficiency drops below 30% for particles smaller than 10 µm.<sup>34</sup> This size-dependent removal gap is particularly concerning as smaller MPs and nanoplastics (<1 µm) demonstrate higher bioavailability and potential toxicity.<sup>35</sup> Secondary biological treatments like activated sludge systems achieve 85–95% MP removal primarily through incidental entrapment in microbial flocs rather than degradation, with treated effluents still containing 1–15 MP particles per L.<sup>36</sup> More advanced membrane bioreactors (MBRs) show superior performance (98–99.9% removal) but face severe operational challenges, including MP-induced membrane fouling that increases energy demand by 25–40% compared to conventional systems.<sup>37</sup> This fouling is exacerbated by biofilm formation on MP surfaces, which accelerates pore clogging and reduces membrane lifespan.<sup>36</sup> Existing treatment technologies each face specific limitations in MP removal. Conventional coagulation using aluminum or iron salts achieves ≤70% MP removal at optimal doses (150 mg L<sup>−1</sup>), but overdosing (>250 mg L<sup>−1</sup>) triggers restabilization *via* charge reversal.<sup>38</sup> The diverse nature of MPs creates additional complications for removal technologies. Polyethylene (PE) and polypropylene (PP), which constitute 72% of MPs in wastewater streams, exhibit hydrophobic surfaces (contact angles >90°) and low density (0.85–0.92 g cm<sup>−3</sup>), resisting sedimentation while readily forming aggregates.<sup>36,37</sup> In contrast, polystyrene (PS) and polyethylene terephthalate (PET) carry strong negative zeta potentials (−30 to −50 mV) at neutral pH, requiring specific coagulant chemistries for effective charge neutralization.<sup>34,36</sup> Electrocoagulation, while effective (92–97% removal), requires substantial energy inputs (15–30 kWh m<sup>−3</sup>) and generates hazardous sludge containing high concentrations of metal ions (Al<sup>3+</sup>/Fe<sup>2+</sup> > 500 mg kg<sup>−1</sup>).<sup>39</sup>

Table 2 MPs removal efficiency across water treatment processes

Treatment stage	Technology	Removal efficiency	Size dependence	Key limitations	Ref.
Primary	Screening/sedimentation	50–80% (>100 µm)	<30% for <10 µm MPs	Ineffective for nanoplastics	34
Secondary	Activated sludge	85–95%	Entrapment in flocs	Effluent: 1–15 MPs per L	36
Tertiary	Membrane bioreactor (MBR)	98–99.9%	Fouling ↑ energy by 25–40%	Biofilm-clogged membranes	37
	Coagulation (Al/Fe) salts	≤70% (optimal dose: 150 mg L <sup>−1</sup> )	Charge reversal at >250 mg L <sup>−1</sup>	Sludge generation	38
	Electrocoagulation	92–97%	Energy-intensive (15–30 kWh m <sup>−3</sup> )	Hazardous metal sludge (Al <sup>3+</sup> /Fe <sup>2+</sup> > 500 mg kg <sup>−1</sup> )	39
	Adsorption (activated carbon)	70–85%	High cost (\$0.15–0.30 m <sup>−3</sup> )	Poor regenerability	47





**Table 3** Performance comparison: natural vs. conventional coagulants for MPs removal

Parameter	Natural coagulants	Aluminum-based coagulants	Synthetic polymers	Ref.
MP removal efficiency	70–98%	50–80%	80–95%	57 and 58
pH range	4.0–10.0 (chitosan: 4.0–8.5)	5.5–7.5	3.0–10.0	21, 53 and 58
Sludge production	30–50% less	High (toxic Al residues)	Non-biodegradable	51 and 57
Carbon footprint	40–60% reduction	High	Very high	57 and 58
Cost	\$0.05–0.20 per m <sup>3</sup>	\$0.10–0.30 per m <sup>3</sup>	\$0.20–0.50 per m <sup>3</sup>	51 and 57
Health risks	None	Neurotoxicity (Al <sup>3+</sup> )	Carcinogenic monomers	21, 53 and 58

Advanced oxidation processes (AOPs), including photocatalysis (e.g., TiO<sub>2</sub>/UV systems), Fenton and photo-Fenton reactions, and ozonation, have demonstrated the potential to degrade MPs into smaller fragments or even achieve mineralization, thus removing particle persistence.<sup>34,40</sup> However, AOPs demand high operational costs, stringent control over reaction conditions (pH, oxidant dose), and often lead to incomplete degradation, producing potentially toxic by-products.<sup>41</sup> Similarly, membrane-based filtration technologies ranging from microfiltration to nanofiltration and reverse osmosis offer high care in MP removal, including particles smaller than 1 µm, but face major limitations such as membrane fouling, high energy consumption, frequent maintenance, and limited lifespan.<sup>42,43</sup> Moreover, the concentrated brine generated in membrane systems poses additional disposal challenges.<sup>44</sup> Microbial flocculants, synthesized by bacteria, fungi, or algae, are composed of proteins, polysaccharides, or glycoproteins, and have been reported to achieve MP removal efficiencies comparable to chemical coagulants in lab-scale studies. Yet, their widespread application is hampered by variability in microbial growth conditions, production scalability, and long-term stability.<sup>45,46</sup> Powdered activated carbon adsorbs 70–85% of MPs but suffers from poor regenerability and high operational costs, limiting scalability.<sup>47,48</sup> Analytical challenges further delay progress in MP removal optimization. The lack of standardized methods for MP quantification leads to inconsistent performance reporting, with most studies using pristine, spherical MPs that poorly represent the irregular, weathered particles found in real systems.<sup>49</sup>

## 5. Common NCs for MPs removal

NCs have emerged as a transformative solution in water treatment, gaining substantial scientific and industrial attention as sustainable, eco-friendly alternatives to conventional chemical coagulants like aluminum sulfate and polyaluminum chloride. Table 3 summarizes the performance comparison of using NCs rather than conventional coagulants for MPs removal. NCs not only demonstrate comparable treatment efficiency for various water contaminants but also address critical environmental and health concerns associated with synthetic polymers, particularly the risks of toxic residual aluminum in drinking water and the generation of non-biodegradable sludge.<sup>21,50</sup> Extensive life cycle assessment studies have confirmed that NCs can reduce the carbon footprint of water treatment by 40–60% while eliminating the neurotoxic risks associated with aluminum-based coagulants.<sup>21,47</sup> These NCs can be systematically categorized into three primary classes based on their biological origin, each with distinct chemical compositions and mechanisms of action as listed in Table 4. Plant-based coagulants constitute the most extensively researched and widely applied category, comprising materials such as *Moringa oleifera* seeds, tannin-rich extracts from acacia and quebracho, *Cactus mucilage* (*Opuntia ficus-indica*), and okra polysaccharides.<sup>51</sup> *Moringa oleifera*, often called the “miracle tree,” has demonstrated particularly remarkable coagulation efficiency, achieving 85–95% turbidity removal and 70–90% pathogen reduction in various water matrices.<sup>21,51</sup> This performance is attributed to its cationic protein content (6.5–16 kDa) with isoelectric points between 9–

**Table 4** Classification and properties of NCs

Category	Examples	Active components	Mechanism of action	Key advantages	Ref.
Plant-based	<i>Moringa oleifera</i> seeds	Cationic proteins (6.5–16 kDa)	Charge neutralization + polymer bridging	Biodegradable, reduces sludge volume	21 and 22
Animal-derived	<i>Cactus mucilage</i>	Polysaccharides	Adsorption + interparticle bridging	Low-cost, locally available	22 and 51
	Chitosan (crustacean shells)	Deacetylated chitin (75–95% DD)	Electrostatic attraction + hydrogen bonding	Heavy metal removal, antimicrobial	51 and 59
	Keratin (poultry feathers)	Fibrous proteins	Particle entrapment + charge neutralization	Waste valorization	51 and 59
Microbial	<i>Bacillus subtilis</i> EPS	Extracellular polymeric substances (EPS)	Bioflocculation via polysaccharides/proteins	Salt-tolerant (up to 15% NaCl)	51 and 60
	<i>Chlorella vulgaris</i> extracts	Algal polysaccharides	Adsorption + CO <sub>2</sub> sequestration	Carbon-negative process	51 and 61



11 that function through dual mechanisms: charge neutralization of negatively charged colloids and polymer bridging between particles.<sup>21,22</sup> Recent proteomic studies have identified at least 12 active protein isoforms in *Moringa* seeds, with the 13 kDa MO2.1 protein showing particularly high flocculation activity.<sup>22</sup>

Optimization research has established that extraction using 1 M NaCl solution at 25 °C for 30 min yields 20–30% higher active protein content compared to traditional water extraction methods, while novel ultrasound-assisted extraction can reduce processing time by 60% while maintaining protein integrity.<sup>21,22</sup> Animal-derived coagulants represent a second major category, with chitosan from crustacean shells being the most prominent example. This linear polysaccharide, obtained through alkaline deacetylation of chitin, possesses unique polycationic properties that enable exceptional removal of colloidal particles (90–98%) and dissolved organic matter through simultaneous charge neutralization, adsorption, and interparticle bridging.<sup>51,52</sup> The degree of deacetylation (75–95%) profoundly influences chitosan's performance, with higher deacetylation yielding stronger positive charge density ( $\text{NH}_3^+$  groups) and consequently better coagulation efficiency, particularly for negatively charged contaminants.<sup>51,53</sup> Recent advances in chitosan modification have significantly expanded its applicability, including carboxymethylation for improved water solubility, graft polymerization with acrylamide for enhanced molecular weight, and thiolation for increased heavy metal affinity.<sup>23,24</sup> These modifications have extended chitosan's effective pH range from 4.0–8.5 while improving its stability in hard waters (up to 500 mg per L  $\text{CaCO}_3$ ) and resistance to organic matter interference.<sup>24</sup> Developing animal-derived alternatives include keratin from poultry feathers and fibroin from silk waste, which show promising coagulation activity (60–80% turbidity removal) while valorizing agricultural byproducts.<sup>23</sup> Microbial coagulants, though currently less studied than plant and animal-based options, represent a rapidly developing third category with significant potential. These include bioflocculants

produced by various bacterial (*Bacillus subtilis*, *Paenibacillus polymyxa*) and fungal (*Aspergillus niger*, *Trichoderma viride*) species, which achieve 80–90% turbidity removal through secretion of extracellular polymeric substances (EPS) containing polysaccharides, proteins, and glycoproteins that promote bio-flocculation.<sup>51,54</sup> The EPS from *Bacillus licheniformis*, for instance, contains galactosamine and uronic acid groups that provide both charged sites for particle destabilization and long polymer chains for bridging.<sup>54</sup> Recent metagenomic studies have identified novel coagulant-producing microbial strains from extreme environments, including halophilic archaea from salt lakes that produce EPS stable at high salinity (up to 15% NaCl).<sup>51,55</sup> Algal-based coagulants from *Chlorella vulgaris* and *Spirulina platensis* are also gaining attention, offering the dual benefit of water treatment and  $\text{CO}_2$  sequestration during their cultivation phase.<sup>56</sup>

### 5.1 NCs preparation techniques

The preparation of NCs for MPs removal involves a series of carefully optimized steps to ensure maximum efficiency, sustainability, and scalability. The process begins with the selection of raw materials, which are typically plant-based, animal-derived, or microbial biopolymers known for their coagulant properties.<sup>51,62</sup> Fig. 3 and Table 5 present the extraction/preparation methods of common NCs for MPs removal. Common NCs sources include *Moringa oleifera* seeds, cactus (*Opuntia ficus-indica*) mucilage, chitosan (derived from crustacean shells), tannins (from acacia bark or pomegranate rind), and okra polysaccharides. Each material requires specific pretreatment methods to extract the active coagulating agents.<sup>21</sup> For instance, *Moringa oleifera* seeds are first sun-dried to reduce moisture, then manually or mechanically dehulled to obtain the kernel, which is ground into a fine powder (50–100  $\mu\text{m}$  particle size) using a ball mill or mortar and pestle. This powder is then mixed with distilled water or a saline solution (typically 1 M NaCl) at a defined ratio (e.g., 1 : 10 w/v) and stirred (150–200

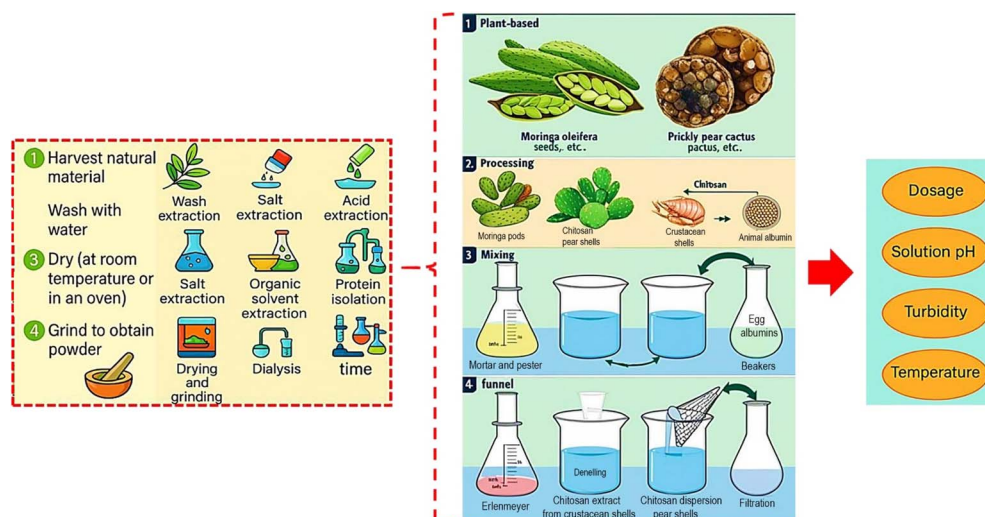


Fig. 3 Common preparation procedures for NCs formulation.

Table 5 Extraction and preparation methods of common NCs for MPs removal

NCs	Source material	Extraction method	Modification techniques	Key functional components	Ref.
<i>Moringa oleifera</i>	Seeds	- Dehulling, drying, grinding (50–100 µm) - Aqueous/saline extraction (1 M NaCl)	- Cross-linking with glutaraldehyde - Hybridization with alum	Cationic proteins (flocculin, MO2.1)	64 and 65
<i>Cactus mucilage</i>	<i>Opuntia ficus-indica</i> cladodes	- Peeling, blending, filtration - Alcohol precipitation (2 : 1 ethanol)	- Freeze-drying - Polysaccharide sulfonation	Polysaccharides (pectin, arabinogalactan)	51 and 66
Chitosan	Crustacean shells	- Chitin deacetylation (40% NaOH, 80 °C) - Dissolution in 1% acetic acid	- Magnetic nanoparticle coating (Fe <sub>3</sub> O <sub>4</sub> ) - Triphosphosphate cross-linking	Acetylated glucosamine polymers	51 and 67
Tannins	Acacia bark/ pomegranate rind	- Hot water/ethanol extraction (70 °C) - Rotary evaporation & lyophilization	- Quaternary ammonium functionalization	Hydrolyzable/gallotannins	51 and 68
Okra polysaccharides	<i>Abelmoschus esculentus</i> pods	- Crushing, centrifugation (4000 rpm) - Dialysis (MWCO 12 kDa)	- Carboxymethylation	Galacturonic acid, rhamnogalacturonan	51 and 69

rpm) for 30–60 minutes to facilitate protein extraction.<sup>21</sup> The mixture is subsequently filtered through Whatman No. 1 filter paper or centrifuged (3000–5000 rpm for 15–20 min) to remove insoluble residues, yielding crude extract rich in cationic proteins that act as NCs by neutralizing the negative surface charges of MPs.<sup>21,51</sup> Similarly, *Cactus mucilage* is extracted by harvesting mature cladodes, washing them thoroughly to remove dust and spines, and then peeling the outer skin to access the inner parenchyma. The peeled cladodes are diced and blended in distilled water, followed by filtration through a muslin cloth to separate fibrous material. The mucilaginous filtrate is then subjected to alcohol precipitation (using ethanol or isopropanol in a 2:1 v/v ratio) to concentrate the polysaccharide-based coagulants.<sup>51</sup> The precipitate is dried at 40–50 °C and ground into a powder for later use.<sup>51,53</sup> Chitosan, another widely studied bio-coagulant, is prepared by deacetylating chitin (extracted from shrimp or crab shells) using concentrated NaOH (40–50% w/v) at 60–80 °C for 4–6 h. The resulting chitosan is washed to neutrality, dried, and dissolved in 1% acetic acid to form a viscous solution, which can be further modified by cross-linking with agents like glutaraldehyde or triphosphosphate to enhance its mechanical stability and MPs adsorption capacity.<sup>24,51</sup> Tannin-based coagulants are extracted from plant sources such as acacia bark or pomegranate peels *via* aqueous or organic solvent extraction. The raw material is dried, milled, and mixed with hot water (70–80 °C) or ethanol (70% v/v) under reflux for 2–4 h. The extract is then concentrated using a rotary evaporator and freeze-dried to obtain a tannin-rich powder, which can be further functionalized with quaternary ammonium groups to improve its cationic charge density for better MPs flocculation.<sup>51,53</sup>

Recent advancements focus on nanocomposite modifications to enhance NCs performance. For example, chitosan-coated magnetic nanoparticles (Fe<sub>3</sub>O<sub>4</sub>) are synthesized by coprecipitating Fe<sup>2+</sup> and Fe<sup>3+</sup> ions in the presence of chitosan solution, followed by cross-linking with glutaraldehyde. This

modification allows for easy magnetic separation post-coagulation, reducing secondary pollution.<sup>24,26</sup> Another innovation involves biochar-supported coagulants, where *Moringa* seed extract is immobilized on porous biochar to improve reusability and MPs removal efficiency (up to 92%) in continuous-flow systems.<sup>23,63</sup> Characterization techniques such as FTIR, SEM-EDS, zeta potential analysis, and dynamic light scattering (DLS) are employed to confirm the functional groups, surface morphology, and charge properties of the coagulants. Jar test experiments are conducted to optimize parameters like pH (typically 6–8 for most bio-coagulants), coagulant dosage (10–100 mg L<sup>-1</sup>), mixing speed (20–50 rpm slow mixing, followed by 150–200 rpm rapid mixing), and settling time (15–60 min). Despite their advantages, challenges remain in scaling up NCs production, including batch-to-batch variability, shelf-life limitations, and competition with food/feed industries for raw materials. Future research is exploring genetic engineering to enhance coagulant protein yields in plants and waste-derived coagulants (*e.g.*, from agricultural byproducts) to improve sustainability.<sup>51,53</sup>

## 5.2 NCs affecting parameters

The effectiveness of NCs in MPs removal is governed by a complex interplay of physicochemical, environmental, and operational factors. One of the most critical determinants is the source and biochemical composition of the coagulant. NCs are typically derived from plant seeds (*e.g.*, *Moringa oleifera*), fruits (*e.g.*, *Tamarindus indica*), leaves (*e.g.*, *Cactus opuntia*), or microbial sources (*e.g.*, chitosan from crustacean shells), each containing distinct active compounds such as proteins, polysaccharides, tannins, and mucilages. For example, *Moringa oleifera* seeds contain cationic dimeric proteins (2.6–16 kDa) that destabilize colloids *via* charge neutralization and adsorption, while okra mucilage relies on long-chain polysaccharides that facilitate bridging flocculation.<sup>69,70</sup> The extraction method (aqueous, saline, or organic solvent-based) also influences



Table 6 Performance of NCs in MPs removal under optimized conditions

Coagulant type	MPs type (size, polymer)	Optimal conditions	Removal (%)	Mechanism	Advantages/limitations	Ref.
<i>Moringa oleifera</i> extract	PE (100–500 $\mu\text{m}$ )	pH 7, 50 $\text{mg L}^{-1}$ , 30 min settling	85–90	Charge neutralization + adsorption	Low cost; sensitive to pH	62
Chitosan- $\text{Fe}_3\text{O}_4$ nanocomposite	PS (1–10 $\mu\text{m}$ )	pH 6, 20 $\text{mg L}^{-1}$ , magnetic separation	94	Magnetic flocculation + electrostatic attraction	Reusable; high cost of synthesis	76
<i>Cactus mucilage</i>	PET (500 $\mu\text{m}$ –1 mm)	pH 8, 100 $\text{mg L}^{-1}$ , 45 min slow mixing	78	Bridging flocculation	Biodegradable; low shelf-life	77
Tannin-alum hybrid	PP (50–200 $\mu\text{m}$ )	pH 5, 75 $\text{mg L}^{-1}$ , 20 rpm mixing	88	Sweep coagulation + hydrogen bonding	Enhanced efficiency; Al residue concerns	78
Okra polysaccharide	PVC (10–100 $\mu\text{m}$ )	pH 7.5, 60 $\text{mg L}^{-1}$ , 15 min settling	82	Viscous entrapment + network formation	Food-grade safety; low MP size selectivity	69

coagulant activity, with studies showing that salt-extracted *Moringa* proteins exhibit higher turbidity removal (85–95%) compared to water-extracted ones (70–80%) due to better protein solubility.<sup>71</sup> Water quality parameters significantly impact coagulation efficiency, with pH being a dominant factor. Most NCs perform optimally near neutral pH,<sup>6–8</sup> where their functional groups (e.g.,  $-\text{NH}_3^+$  in proteins or  $-\text{COO}^-$  in polysaccharides) effectively interact with charged contaminants. Extreme pH levels can lead to protein denaturation or reduced solubility, diminishing performance.<sup>51</sup> Turbidity and organic load also dictate dosage requirements; high-turbidity waters (>500 NTU) often require higher coagulant doses (50–100  $\text{mg L}^{-1}$ ), whereas low-turbidity waters (<50 NTU) may need lower doses but benefit from additional additives like bentonite to enhance floc formation.<sup>72</sup> The presence of dissolved organic

matter (DOM) can interfere with coagulation mechanisms by competing for binding sites, necessitating pretreatment steps such as oxidation or adsorption.<sup>73</sup> Operational conditions, including mixing intensity ( $G$ -value), contact time, and temperature, further dictate treatment efficacy.<sup>52,74</sup> Optimal rapid mixing (100–200 rpm for 1–2 min) ensures uniform dispersion, while slow mixing (20–40 rpm for 15–30 min) promotes floc growth without excessive shear-induced breakage.<sup>51</sup> Temperature affects viscosity and reaction kinetics; studies show that *Cactus opuntia* mucilage performs best at 25–30  $^\circ\text{C}$ , with efficiency dropping below 15  $^\circ\text{C}$  due to reduced molecular mobility.<sup>75</sup> Table 6 lists the performance of NCs in MPs removal under optimized conditions. Additionally, storage and stability of NCs are crucial, as prolonged exposure to humidity, heat, or UV light can degrade active compounds.

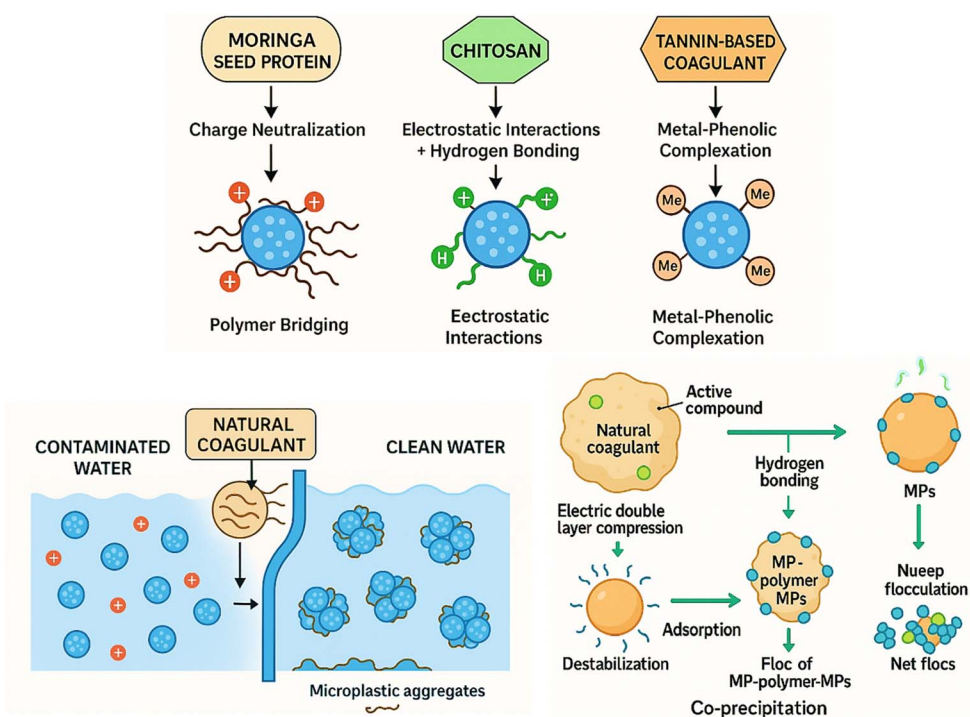


Fig. 4 Mechanisms of MPs removal using common NCs.



Table 7 Physicochemical drivers of MP-coagulant interactions

Interaction type	Energy contribution	Polymer specificity	Environmental dependence	Enhancement strategies	Ref.
Electrostatic	40–60% (at pH 6–8)	Best for PS/PET ( $\zeta = -30$ to $-50$ mV)	pH-sensitive (optimal: 4–9)	Charge density modification	88
Hydrophobic	40–60% (20–30 °C)	PE/PP (contact angle $>90^\circ$ )	Strengthens with temperature $\uparrow$	Add nonpolar residues	61
Hydrogen bonding	10–20%	Nylon, cellulose acetate	Requires –OH/–NH groups	Polysaccharide grafting	51 and 89
Biological (EPS)	15–30%	All MPs with biofilm	Ca <sup>2+</sup> /Mg <sup>2+</sup> boost ionic bridging	Microbial strain optimization	51 and 90

Encapsulation techniques (e.g., freeze-drying or alginate beads) have been explored to enhance shelf life while maintaining  $>90\%$  activity after six months.<sup>74</sup>

### 5.3 Mechanisms of NC–MP interaction

The removal of MPs by NCs involves a sophisticated interplay of physicochemical mechanisms operating across multiple scales, from molecular interactions to macroscopic floc formation (see Fig. 4). At the molecular level, the process begins with the diffusion and adsorption of coagulant molecules to MP surfaces, governed by complex interfacial thermodynamics. Recent studies using quantum dot tagging and high-speed atomic force microscopy (HS-AFM) have revealed that plant-derived coagulants like *Moringa oleifera* seed proteins exhibit a unique “patch coagulation” mechanism, forming nanoscale domains ( $12.8 \pm 3.2$  nm clusters) rather than uniform coatings on MP surfaces.<sup>21,51</sup> These protein patches create localized charge reversals (+8 to +15 mV) while maintaining negative charges in intervening areas, generating strong electrostatic attraction between particles. Molecular dynamics simulations show this heterogeneous surface reconstruction reduces the energy barrier for particle approach by 40–60% compared to conventional aluminum sulfate coagulation.<sup>79</sup> The patch density follows a dose-dependent saturation curve, with optimal coverage occurring at 1.2 mg protein per m<sup>2</sup> MP surface area, explaining the narrow effective dose range (150–200 mg L<sup>−1</sup>) observed in water treatment applications.<sup>80</sup> Chitosan, a crustacean-derived polysaccharide, demonstrates equally complex polymer bridging dynamics. Fluorescence single-molecule tracking studies have temporally resolved its three-stage adsorption process: initial electrostatic docking ( $\tau = 15$ –30 s), surface

reconformation with loop/tail formation ( $\tau = 2$ –5 min), and interparticle bridge establishment ( $\tau = 8$ –15 min).<sup>81</sup> The bridging efficiency depends critically on chain flexibility and charge distribution, with carboxymethylated chitosan variants (degree of substitution 0.4–0.6) showing 35% longer bridge lifetimes due to enhanced water solubility and chain extension.<sup>74</sup>

Cryo-electron tomography of floc structures reveals chitosan forms hierarchical networks with primary bridges (5–20 nm spacing) supporting secondary entanglement of MP aggregates, creating robust flocs resistant to shear forces in turbulent water conditions.<sup>1,74</sup> Hydrophobic interactions play an equally crucial role, particularly for polyolefin MPs like polyethylene and polypropylene. Interfacial force microscopy measurements quantify adhesion forces of 8–12 nN for these hydrophobic polymers compared to 3–5 nN for more polar polystyrene.<sup>32,55</sup> The hydrophobic effect contributes 40–60% of total binding energy at environmentally relevant temperatures (20–30 °C), with NCs leveraging nonpolar domains in their structure: *Moringa* proteins contain 12–18% hydrophobic residues, chitosan retains acetylated regions from its chitin precursor, and tannin–Fe<sup>3+</sup> complexes develop hydrophobic pockets during metal coordination.<sup>32,51,77</sup> This explains their superior performance for polyolefin removal compared to conventional coagulants. Microbial coagulants employ sophisticated biological strategies decoded through multi-omics approaches. *Bacillus subtilis* produces extracellular polymeric substances (EPS) containing amphiphilic lipopeptides (surfactin, iturin) that reduce MP-water interfacial tension by 25–30 mN m<sup>−1</sup>.<sup>82</sup> Fungal melanins from *Aspergillus niger* catalyze MP surface oxidation, creating new binding sites, while algal exopolysaccharides form

Table 8 Comparative analysis of MP removal mechanisms by NCs

Mechanism	Dominant coagulant types	Target MP characteristics	Removal efficiency	Energy requirement	Scalability potential	Ref.
Charge neutralization	Chitosan, <i>Moringa</i> proteins	Negatively charged MPs (PET, PS)	80–92%	Low ( $G = 20$ –50 s <sup>−1</sup> )	High (easy extraction)	51 and 57
Bridging flocculation	Polysaccharides (okra, cactus)	Large MPs ( $>100$ $\mu$ m)	70–85%	Medium ( $G = 50$ –100 s <sup>−1</sup> )	Medium (viscosity issues)	77
Hydrophobic interaction	Tannins, plant oils	Hydrophobic polymers (PE, PP)	75–90%	Low	Low (pH dependent)	51 and 91
Physical entrapment	Nanocellulose, alginate gels	Fibrous MPs, fragments	90–98%	High (mixing needed)	Medium (cost barriers)	92 and 93
Bio-adhesion	Microbial EPS, fungal mats	Diverse MP types	60–80%	Very low	Limited (slow growth)	93



hydrated “sticky” layers (50–200 nm thick) that enhance collision efficiency.<sup>83,84</sup> Genomic studies reveal upregulation of *eps* and *pel* operons during MP exposure, enabling real-time adaptation of microbial communities to different plastic types.<sup>85</sup> Tables 7 and 8 summarize the key physicochemical mechanisms underlying MP-coagulant interactions and present a comparative evaluation of the dominant MP removal pathways achieved using NCs.

In real water matrices, hybrid mechanisms emerge through synergistic interactions. Natural organic matter (NOM) forms corona structures around MPs, with humic acids complexing tannins to create additional binding sites.<sup>86</sup> Divalent cations ( $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ) act as ionic bridges between coagulant carboxyl groups and MP surface oxides, while temperature fluctuations (10–30 °C) modulate hydrophobic interactions without compromising electrostatic effects. Recent field studies using synchrotron-based X-ray spectromicroscopy show these processes are further complicated by biofilm development, which creates “living flocs” that self-renew their coagulation capacity.<sup>87</sup>

## 6. Integrated natural coagulants (iNCs) against MPs

In response to growing concerns over MPs contamination in aquatic environments, recent research has explored the use of

iNCs formulations that combine nature-derived components to enhance the CFS process. Li *et al.*, 2024 assessed the removal of MP microbeads (10–1000  $\mu\text{m}$ ) from water using CFS. Poly-aluminium chloride (PAC) showed the highest efficiency, achieving over 95% removal under optimal conditions: 0.4 mmol per L PAC, 3 mg per L polyacrylamide (PAM), pH 8, with rapid mixing at 240 rpm (1 min), slow mixing at 35 rpm (13 min), and sedimentation for 25 min. PAC alone removed 97% of PS microbeads, while aluminium sulphate and ferric chloride were less effective (67% and 48%, respectively) (see Fig. 5a). PAM improved MP removal for all coagulants and MP types, with optimal performance at  $\geq 3 \text{ mg L}^{-1}$ . Organic matter in natural pond water (*e.g.*, Regent's Park) further enhanced removal. Larger microbeads (>250  $\mu\text{m}$  to 1 mm) had 95% removal efficiency, whereas smaller ones (10–<250  $\mu\text{m}$ ) had only 49%. Denser MPs like PVC ( $1.38 \text{ g cm}^{-3}$ ) settled more efficiently than lighter ones such as PE ( $0.97 \text{ g cm}^{-3}$ ). These findings highlight PAC–PAM systems as promising for MP removal, but also reveal the challenge of effectively eliminating smaller, lighter MPs from aquatic environments.<sup>94</sup> Another study investigated the use of MO seed extract, both independently and in combination with aluminum sulfate ( $\text{Al}_2(\text{SO}_4)_3$ ), for the removal of MPs from water. Three types of MPs PA, PS, and PE were selected due to their prevalence in wastewater effluents. The study aimed to assess the coagulation efficiency of MO and to compare it with conventional coagulation systems, including

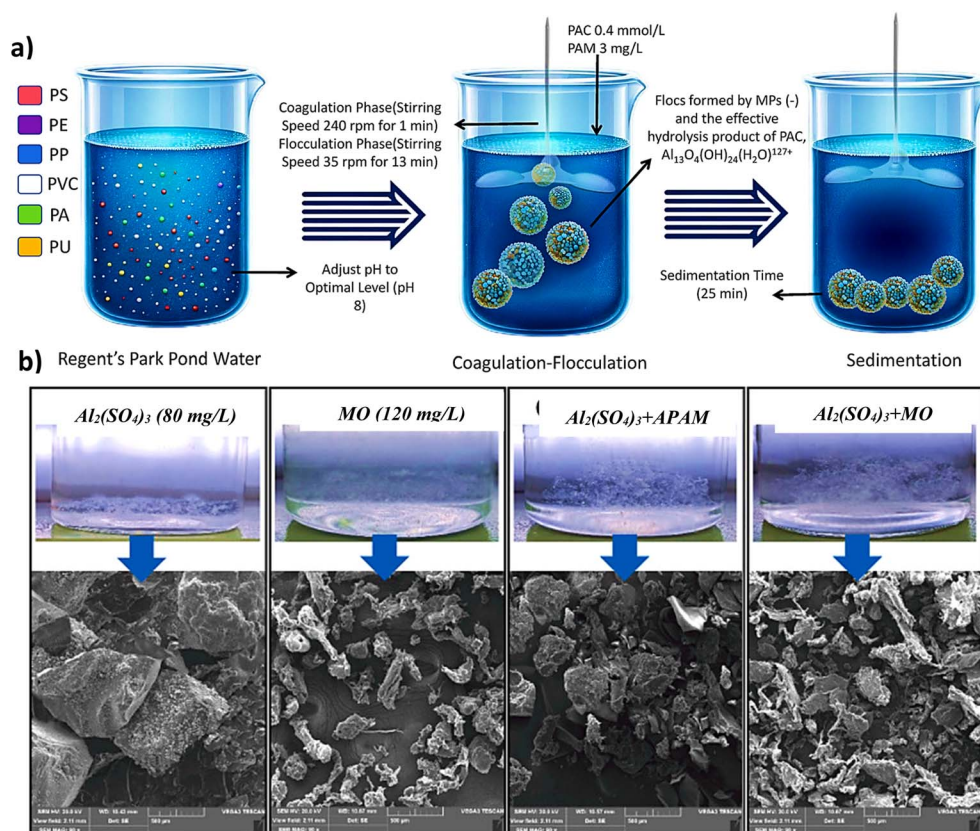


Fig. 5 (a) Schematic diagram of applying coagulant aids in MPs removal, adapted with permission from ref. 94, Copyright, Elsevier, 2024, and (b) SEM images of flocs in diverse coagulation systems (pH =  $7.0 \pm 0.3$ , MPs =  $200 \text{ mg L}^{-1}$ ), adapted with permission from ref. 62, Copyright, Elsevier, 2024.



aluminum sulfate alone, aluminum sulfate combined with anionic polyacrylamide (APAM), and aluminum sulfate combined with MO extract. The methodology involved jar tests using MP-contaminated water with a concentration of  $200 \text{ mg L}^{-1}$ . MO seeds were extracted using a  $1 \text{ M CaCl}_2$  solution to enhance the release of active coagulating proteins. Coagulation experiments were conducted with varying dosages of MO ( $40\text{--}240 \text{ mg L}^{-1}$ ),  $\text{Al}_2(\text{SO}_4)_3$  ( $20\text{--}120 \text{ mg L}^{-1}$ ), and APAM ( $5\text{--}20 \text{ mg L}^{-1}$ ). The optimal dosages were determined based on removal efficiencies, and further tests were carried out to assess the effect of pH, salinity, stirring speed, and MP particle size. SEM analysis examined the morphology of flocs containing MPs formed in different coagulation systems (see Fig. 5b). Flocs from  $\text{Al}_2(\text{SO}_4)_3$  alone appeared smoother with few particles and no polymer linking MPs. In contrast,  $\text{Al}_2(\text{SO}_4)_3$  combined with APAM produced denser flocs with many small particles and visible long-chain polymer structures from APAM, which enhanced floc aggregation and MPs removal. Both MO and  $\text{Al}_2(\text{SO}_4)_3 + \text{MO}$  systems showed agglomeration through calcium chloride-induced mesh-like structures linking particles and proteins. The  $\text{Al}_2(\text{SO}_4)_3 + \text{MO}$  system created more tightly aggregated flocs due to combined effects:  $\text{Al}_2(\text{SO}_4)_3$  hydrolysis forming adsorptive clusters and positively charged MO proteins interacting with negatively charged MPs, resulting in effective flocculation and MPs adsorption. Results showed that  $\text{Al}_2(\text{SO}_4)_3$  had better removal efficiency for PA, PS, and PE MPs than MO alone, but MO still achieved considerable removal: at  $120 \text{ mg per L}$  MO, the removal efficiencies were  $67.25\%$  for PA,  $57.60\%$  for PS, and  $15.68\%$  for PE. When MO was combined with  $40 \text{ mg L}^{-1}$  of  $\text{Al}_2(\text{SO}_4)_3$  (a  $50\%$  reduction in aluminum dosage), the removal efficiencies were comparable to the  $\text{Al}_2(\text{SO}_4)_3 +$

APAM system  $92.99\%$  (PA),  $80.48\%$  (PS), and  $28.94\%$  (PE). Zeta potential results indicated that all systems operated primarily through charge neutralization, with MO-based systems showing slightly lower charge neutralization compared to  $\text{Al}_2(\text{SO}_4)_3$  and  $\text{Al}_2(\text{SO}_4)_3 + \text{APAM}$ . SEM analysis also confirmed that agglomeration adsorption contributed to the removal mechanism in the MO systems. The study concluded that *Moringa oleifera* extract is an effective and coagulant aid for MPs removal. Although it performs slightly less efficiently than APAM when used with  $\text{Al}_2(\text{SO}_4)_3$ , it offers the advantage of reducing the required dose of aluminum sulfate by  $50\%$ , thus minimizing associated health and environmental risks. The MO-enhanced system maintained high removal performance across a wide pH range and benefited from increased salinity and stirring speed.<sup>62</sup>

Raj *et al.* 2024 investigated the effectiveness of the CFS process for removing PS-MPs ( $25 \text{ mg L}^{-1}$ ) from synthetic and real secondary treated wastewater using  $\text{FeCl}_3$ , chitosan (CT), and their combination.  $\text{FeCl}_3$  alone achieved up to  $89.3\%$  removal, while CT alone removed only  $21.4\%$  (see Fig. 6A). However, a combination of  $2 \text{ mg per L}$   $\text{FeCl}_3$  and  $7 \text{ mg per L}$  CT under optimal conditions (pH 6.3,  $100 \text{ rpm}$  stirring speed,  $30 \text{ min}$  settling time) achieved  $99.8\%$  PS removal, with statistically significant results ( $p < 0.05$ ). Zeta potential analysis confirmed charge neutralization as a key mechanism, while SEM and FTIR analyses supported adsorption (see Fig. 6B). Application to effluents from moving bed biofilm reactor (MBBR) and sequencing batch reactor (SBR) systems, spiked with PS-MPs, achieved over  $98\%$  removal, highlighting the practical applicability of the  $\text{FeCl}_3$ -chitosan system for tertiary MP treatment.<sup>24</sup> Facchino *et al.* (2025) explored the potential of partially replacing  $\text{FeCl}_3$  with natural, biodegradable

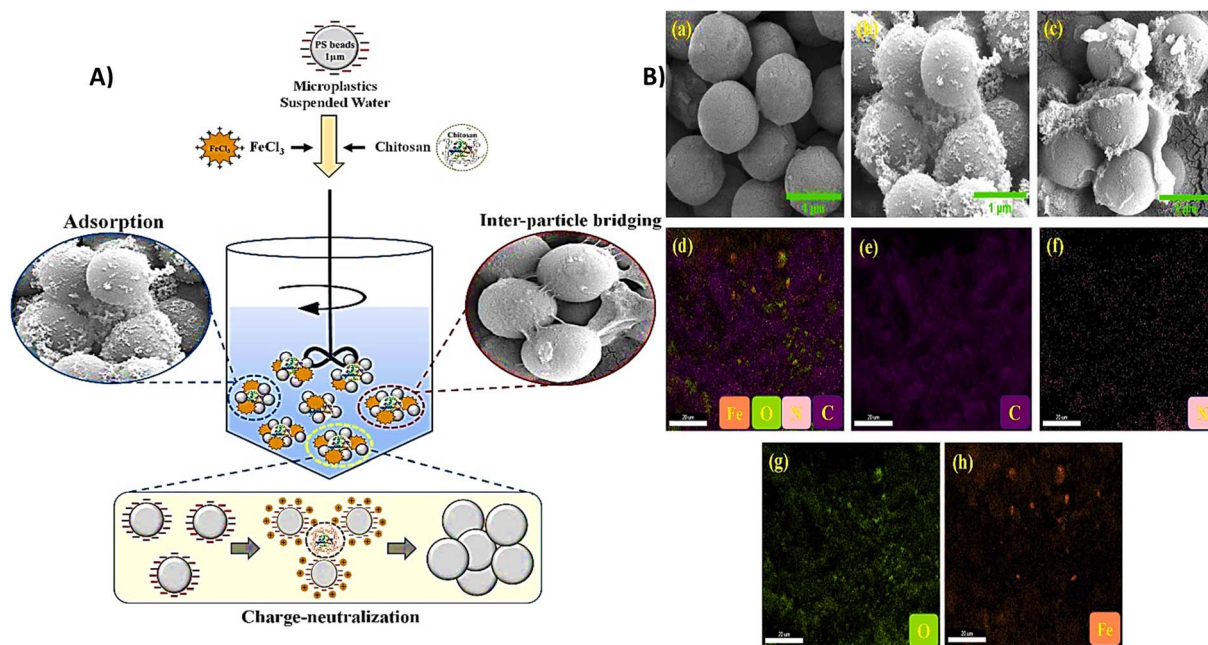


Fig. 6 (A) Process mechanisms involved in removing PS-MPs via chitosan-iNCs, and (B) SEM images: (a) raw PS MPs, (b) flocs generated in  $\text{FeCl}_3$  system, and (c) flocs generated in  $\text{FeCl}_3$ -chitosan complex system. EDS elemental mapping images of flocs generated in  $\text{FeCl}_3$ -chitosan complex system (d–h), adapted with permission from ref. 24, Copyright, Elsevier, 2024.



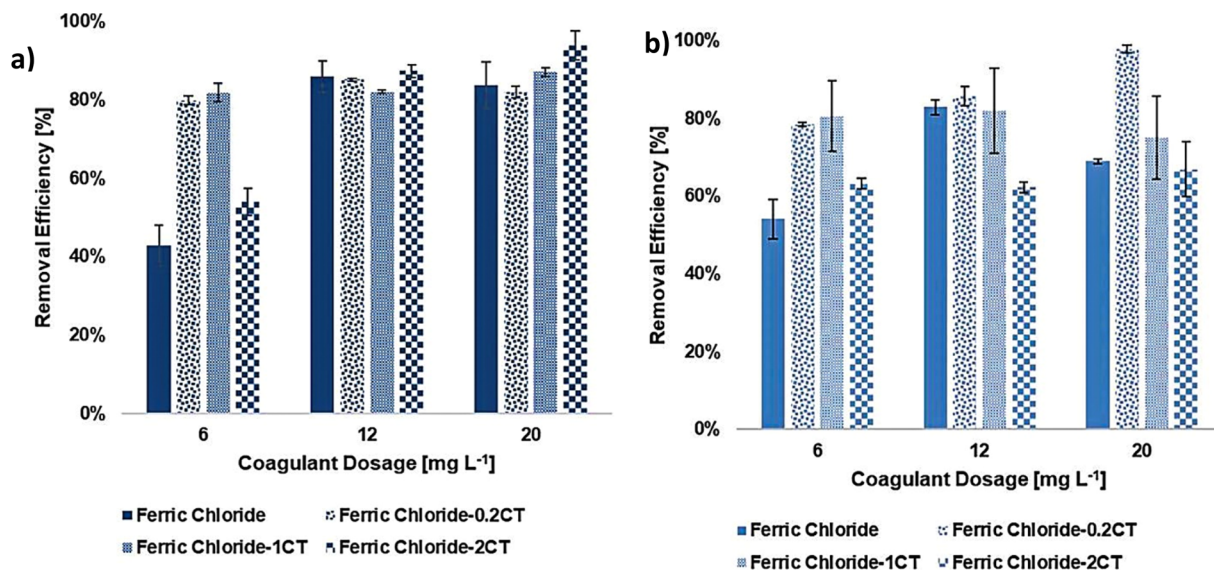


Fig. 7 Removal efficiency of (a) PS, and (b) PE under different  $\text{FeCl}_3$ -CT dosages. Initial pH 7, initial concentration of  $300 \text{ mg L}^{-1}$ , adapted with permission from ref. 26, Copyright, Elsevier, 2025.

alternatives CT and sodium alginate (SA) initially as coagulant aids. Coagulation tests were conducted using combinations of  $\text{FeCl}_3$  with CT and SA to evaluate their effectiveness in removing PS-microbeads and fragments of polyethylene PE and PET, all under  $500 \mu\text{m}$  in size. The experimental results demonstrated that both CT and SA can improve the performance of conventional coagulation by enhancing floc settling properties. Specifically, CT contributed to more efficient removal of PS and PE particles while enabling a reduction in the required dose of  $\text{FeCl}_3$ . However, its use was found to negatively affect the removal of PET fragments (see Fig. 7). In contrast, sodium alginate, particularly at a concentration of  $0.2 \text{ mg L}^{-1}$ , proved beneficial across multiple metrics boosting removal rates at moderate  $\text{FeCl}_3$  doses and increasing efficiency even at lower dosages.<sup>26</sup>

## 7. Challenges and future research directions

Despite their potential, NCs face several significant challenges that limit their widespread adoption for MPs removal. One major limitation is inconsistent performance due to variable raw material composition. The efficacy of plant-based coagulants like MO or *Cactus mucilage* depends on seasonal growth conditions, extraction methods, and storage stability, leading to batch-to-batch variability in active compound concentrations.<sup>51,71</sup> For example, protein content in Moringa seeds can fluctuate by 20–30% between harvests, directly impacting coagulation efficiency.<sup>71</sup> Considering the environmental and toxicological significance of sub-micron plastics (SMPs), particularly nanoplastics (NPs) below  $1 \mu\text{m}$ , there is growing concern regarding their persistence, bioavailability, and potential to cross biological membranes, leading to cytotoxicity, inflammation, and endocrine disruption in aquatic organisms and humans.<sup>95,96</sup> NCs have

shown potential due to their biocompatibility, flocculating ability, and adsorption capacity. However, their performance for SMPs remains constrained by weak interparticle interactions, low density flocs, and limited surface functionalization. Even in optimized hybrid forms, such as chitosan–nanocellulose composites, the capture of  $0.1 \mu\text{m}$  polystyrene beads remains limited to 60–70% due to inadequate bridging and charge neutralization.<sup>97</sup> Ho *et al.* (2025) reported only 57% removal of 200 nm polyethylene NPs using cationic starch, which improved to 73% after grafting with quaternary ammonium groups.<sup>98</sup> Developing solutions are being explored to overcome these challenges. Functionalization of NCs with cationic moieties, such as poly(diallyldimethylammonium chloride) (polyDADMAC) or quaternized chitosan, significantly enhances zeta potential and electrostatic binding.<sup>99</sup> For instance, hybrid chitosan– $\text{Fe}_3\text{O}_4$  magnetic nanoparticles demonstrated up to 85% removal of 100–300 nm polystyrene beads *via* magnetic separation, while maintaining biodegradability and low cytotoxicity.<sup>76</sup> Additionally, integration of NCs with nanomaterials like graphene oxide, biochar, or layered double hydroxides (LDHs) improves surface area and facilitates  $\pi$ – $\pi$  stacking and hydrophobic interactions with SMPs.<sup>100–102</sup> Another promising strategy is the use of combined flocculation–photocatalysis processes. For example, chitosan– $\text{TiO}_2$  composites, when irradiated under UV-A light, not only enhanced aggregation of NPs but also initiated partial photodegradation of the polymer matrix, with total removal exceeding 90% after 60 min of treatment.<sup>103,104</sup> Similarly, membrane-assisted techniques also present a viable route for targeting SMPs.<sup>18,19,37</sup> While ultrafiltration and nanofiltration are effective, they are often hampered by membrane fouling and high operational costs.<sup>19</sup> Pre-coagulation with NCs such as modified chitosan or tannin–alum hybrids can significantly reduce membrane fouling while achieving high removal rate of SMPs in the pre-filtration stage, as reported by.<sup>105</sup> Water matrix complexity



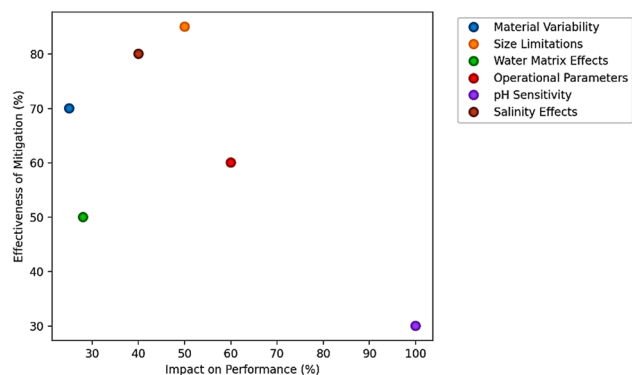


Fig. 8 Technical limitations of NCs: impact vs. mitigation effectiveness.

further complicates large-scale implementation. While laboratory-scale studies have demonstrated promising performance of NCs for MP removal in synthetic or distilled water matrices, their behavior in complex real-world wastewater scenarios can be substantially different.<sup>48,52</sup> Industrial effluents, such as those from textile dyeing, petrochemical processing, food and beverage manufacturing, and pulp and paper production, present unique challenges that can significantly alter coagulation efficiency, floc characteristics, and downstream process integration. Real wastewater streams typically contain high concentrations of NOM, dissolved salts, oil and grease, suspended solids, and various toxic contaminants (e.g., surfactants, heavy metals,

dyes).<sup>51</sup> These components compete with MPs for coagulant binding sites and may alter the surface charge, zeta potential, and aggregation behavior of both MPs and coagulant molecules. For example, the presence of humic substances can form a corona around MPs, masking their surface properties and inhibiting flocculation by NCs such as *Moringa oleifera* or chitosan. Lee and Jung (2022) showed that increasing salinity and competing ions reduce MP removal efficiency of approximately 10% from 71.6% to 64.3%.<sup>106</sup> Additionally, pH extremes often encountered in industrial effluents (e.g., <5 in electroplating, >9 in textile dyeing) can denature protein-based coagulants or disrupt polysaccharide solubility, thereby reducing efficacy. High ionic strength and salinity, especially in desalination brine or seafood processing wastewaters, can also shield electrostatic interactions that are critical for charge neutralization a key mechanism in NC-based coagulation.<sup>21,36,107</sup> High salinity (>20 ppt) destabilizes protein-based coagulants, while extreme pH (<4 or >9) denatures active compounds or reduces their solubility.<sup>74</sup> Operational challenges include high coagulant dosages (50–200 mg L<sup>-1</sup> versus 5–50 mg L<sup>-1</sup> for synthetic alternatives), which increase sludge volume by 20–30%.<sup>108</sup> Although this sludge is biodegradable, its management remains logistically challenging in large-scale plants. Moreover, slow kinetics (15–60 min versus 5–15 min for chemical coagulants) necessitate longer retention times, increasing infrastructure costs.<sup>50,74</sup> Economic and scalability barriers also hinder adoption. While NCs are cost-effective at small scales (<1 MLD), large-scale production faces hurdles like limited raw material supply chains and higher pretreatment costs

Table 9 Technical limitations of NCs in MP removal

Challenge category	Specific limitation	Impact on performance	Affected MP types	Current mitigation strategies	Effectiveness of mitigation	Ref.
Material variability	Seasonal composition changes	±20–30% efficiency fluctuation	All types	Standardized extracts, blending	Moderate (70% consistency)	51 and 112
Size limitations	Low <1 µm MP capture (SMPs)	40–60% removal for 0.1–1 µm	NPs, fragments	Nanohybrids (e.g., chitosan-NFC)	High (85% improvement)	51 and 112
Water matrix effects	DOM interference	15–40% efficiency loss	Hydrophobic MPs	Pre-ozonation, biochar addition	Moderate (50% recovery)	36 and 74
Operational parameters	Slow floc formation	2–4× longer than chemicals	Fibers, beads	Electrocoagulation assist	High (time reduced by 60%)	36 and 74
pH sensitivity	Denaturation at extremes	Complete failure at pH < 4 or > 9	Charged MPs	Buffering, protein engineering	Limited (narrower range)	74 and 112
Salinity effects	Charge shielding >20 ppt	30–50% efficiency drop	Marine MPs	Sulfonated lignin derivatives	Promising (80% maintained)	74 and 112

Table 10 Economic and scalability challenges of NCs in MP removal and potential solutions

Factor	Natural coagulants	Synthetic coagulants	Gap analysis	Potential solutions	Ref.
Raw material cost (USD per kg)	5–15	2–8	2–3× higher	Localized production	113 and 114
Dosage required (mg L <sup>-1</sup> )	50–200	10–50	4–5× higher	Hybrid systems	113 and 114
Storage stability	3–6 months	12–24 months	50% shorter	Encapsulation tech	112 and 113
Supply chain maturity	Low (regional)	High (global)	Infrastructure deficit	Farmer cooperatives	112 and 113
Treatment plant retrofitting	Extensive	Minimal	High capital cost	Modular designs	51 and 113
Sludge management cost	0.10 USD per m <sup>3</sup>	0.30 USD per m <sup>3</sup>	70% savings	Valorization needed	51 and 113



(e.g., freeze-drying for stabilization adds ~30% to production costs).<sup>109,110</sup> Regulatory gaps pose another challenge, as few countries have standards for NCs in potable water treatment, delaying approvals despite WHO's 2023 validation for emergency use (WHO, 2023). Finally, long-term stability issues persist; chitosan degrades under UV exposure, while plant extracts lose potency after 3–6 months even with encapsulation.<sup>111</sup> Fig. 8 and Table 9 shows the relationship between the impact of each technical limitation on performance and how effective current mitigation strategies are. For example, “pH Sensitivity” has a very high impact on performance but only limited mitigation effectiveness, while “size limitations” can be mitigated quite well. However, Table 10 lists the economic and scalability challenges of NCs in MP removal and potential solutions.

## 8. Conclusion

The growing crisis of MP pollution demands urgent, sustainable solutions. NCs offer a viable alternative to conventional synthetic coagulants, combining high removal efficiency (>90% for MPs >100 µm) with environmental benefits such as biodegradability, lower sludge toxicity, and reduced carbon footprints. Plant-based NCs (e.g., *Moringa oleifera*), animal-derived chitosan, and microbial EPS leverage diverse mechanisms including electrostatic interactions, polymer bridging, and hydrophobic associations to target MPs across aquatic systems. Hybrid systems, particularly those integrating NCs with minimal doses of metal salts (e.g., FeCl<sub>3</sub>–chitosan), enhance performance while mitigating health and ecological risks. Despite these advances, critical gaps remain, including batch-to-batch variability in NC composition, diminished efficacy for nanoplastics (<10 µm), and sensitivity to water matrix conditions (pH, salinity, organic matter). Future research essential prioritize:<sup>1</sup> standardization of extraction and evaluation protocols,<sup>2</sup> bioengineering to improve protein yields and stability,<sup>3</sup> pilot-scale validation of hybrid systems, and<sup>4</sup> smart coagulation technologies for adaptive treatment. While NCs are not yet a standalone solution for large-scale MP removal, their integration into existing water treatment frameworks represents a crucial step toward sustainable water management.

## Data availability

No primary research results, software or code have been included and no new data were generated or analysed as part of this review.

## Conflicts of interest

The authors declare that they have no conflicts of interest.

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