

## CRITICAL REVIEW

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Cite this: DOI: 10.1039/d5su00634a

# Microplastic removal from wastewater through biopolymer and nanocellulose-based green technologies

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Microplastics (MPs) in wastewater are a growing environmental issue that needs effective solutions. This review examines the use of nanocellulose and biopolymers as sustainable options for removing these pollutants from water. Nanocellulose (NC) is efficient due to its large surface area and biodegradable nature, achieving up to 98% removal of microplastics through various processes, including adsorption and filtration. Similarly, biopolymers like polysaccharides, lignin, and pectin can remove up to 99% of particles by clumping and settling them out. However, some microplastics are not easily removed by these materials on their own. Combining different materials, such as cellulose and chitosan, can enhance removal efficiency to about 75%. Integrating these solutions into existing wastewater treatment plants could help reduce microplastics and save costs; however, it is essential to ensure compatibility with current systems and establish appropriate regulations. The review also highlights the need for future research to support the widespread use of these methods in water treatment.

Received 30th July 2025  
Accepted 27th October 2025

DOI: 10.1039/d5su00634a

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## Sustainability spotlight

This review addresses the sustainability challenges associated with microplastic contamination in wastewater and highlights green technologies for their removal. It focuses on nanocellulose and biopolymer-based materials derived from renewable resources, emphasizing their biodegradability, low toxicity, and potential to replace synthetic treatment agents. The paper evaluates the environmental benefits of adsorption, coagulation, and filtration mechanisms compared to conventional chemical methods, outlining strategies to reduce secondary pollution and energy demand. By exploring scalable integration into existing wastewater treatment systems, this review offers a pathway toward sustainable water management practices aligned with circular economy principles.

## 1. Introduction

Plastic materials first appeared in the late 19th century, with commercial use beginning in 1870.<sup>1</sup> Due to their light weight, durability, and corrosion resistance, plastics have rapidly gained popularity across various industries. As a result, plastic production increased nearly 180 times between 1950 and 2018, reaching over 400.3 million tons globally by 2022.<sup>2</sup> These synthetic macromolecular polymers, such as polyethylene terephthalate, polyethylene, polypropylene, polyvinyl chloride, polyamides, polystyrene, and polyurethanes, have become indispensable in daily life,<sup>3–5</sup> especially in sectors like textiles, construction, motor vehicles, consumer goods, medical, and

food packaging.<sup>6,7</sup> A detailed overview of plastic evolution and its key historical milestones is presented in Fig. 1.<sup>8</sup>

Among these sectors, the textile industry is a leading consumer of synthetic polymers. Since 1995, synthetic fibers have surpassed cotton as the most widely used textile material, accounting for approximately 65% of global fiber production by 2020.<sup>9</sup> China and India dominate this sector, accounting for 66% and 8% of global output, respectively, followed by Taiwan and the United States, each at approximately 4%.<sup>10,11</sup> However, the widespread use of synthetic fibers has led to increasing environmental concerns, particularly regarding MP pollution. Synthetic fibers shed MPs during washing and wear, contributing significantly to the presence of MPs in water bodies. These particles often end up in wastewater treatment systems, where they persist due to their resistance to degradation.

The rising accumulation of plastic waste, primarily from single-use products such as shopping bags, bottles, and lids, has exacerbated global pollution issues.<sup>7,12</sup> While some plastics, such as bottles, are recyclable, most single-use items still accumulate in landfills or are incinerated, adding to environmental degradation.<sup>13,14</sup> Plastic pollution has now been reported in a variety of ecosystems, including coastlines,<sup>15</sup> oceans,<sup>16</sup>

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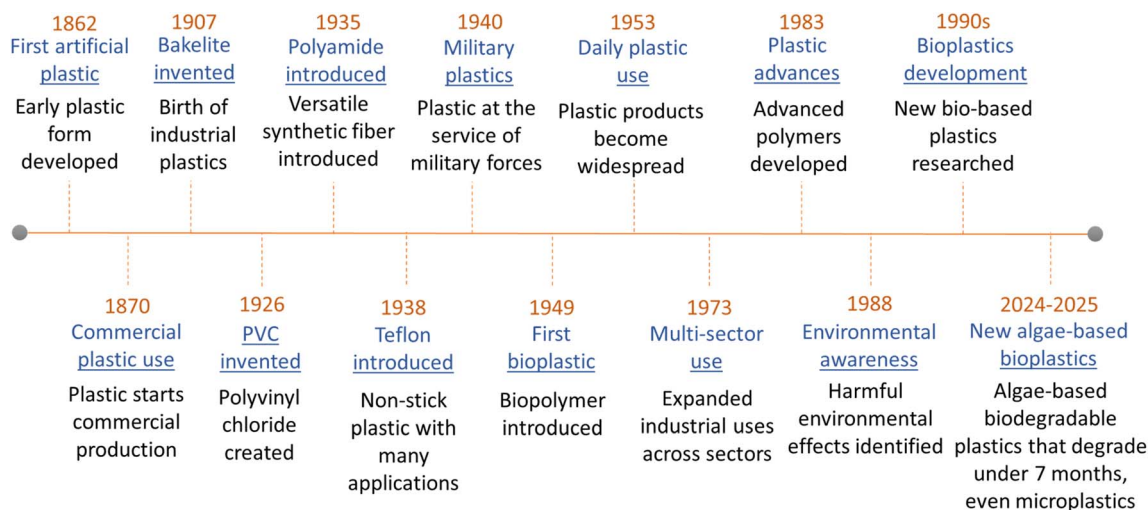


Fig. 1 Historical timeline depicting the development and use of plastics (created with MS PowerPoint).

deep-sea areas,<sup>17</sup> and even remote islands.<sup>18</sup> Plastics degrade into MPs, which are categorized into primary and secondary types.<sup>19–21</sup> Primary MPs are intentionally produced in small sizes (2 to 5 mm), often found in products such as microbeads in cosmetics,<sup>21–23</sup> whereas secondary MPs result from the fragmentation of larger plastics through physical abrasion or UV radiation.<sup>22–24</sup> These degradation processes can produce particles as small as 1.6  $\mu\text{m}$ , which have been detected in marine environments.<sup>25,26</sup> Interestingly, the concentration of MPs in surface ocean waters is lower than expected due to their tendency to aggregate with marine particles and settle into sediments, often facilitated by biofilm formation.<sup>27</sup>

Wastewater treatment plants (WWTPs) are significant entry points for MPs into the environment. According to Sun *et al.*,<sup>5</sup> MP concentrations in WWTP effluents can reach up to 447 particles per liter, with polystyrene (PS) being among the most detected polymers. Even with tertiary treatment processes (TTPs), effluents may still contain up to 51 particles per liter, and only 24% of WWTPs globally currently implement TTPs. Consequently, daily median MP discharges can reach approximately 2 million particles.

Traditional methods of removing MPs, such as filtration, coagulation, and sedimentation, face limitations in terms of efficiency and sustainability, especially for nanoscale particles.<sup>28,29</sup> Filtration becomes energy-intensive when extremely fine pores are required and often suffers from clogging.<sup>29,30</sup> Similarly, Zhang *et al.*<sup>31</sup> demonstrated that coagulation and sedimentation techniques are usually insufficient for the complete removal of both micro- and nanoplastics. Moreover, many conventional filtration materials are non-renewable, non-biodegradable, and relatively expensive, raising concerns over their long-term environmental impact.<sup>28,32</sup> As a result, biopolymers and NC have emerged as promising, eco-friendly alternatives due to their renewability, biodegradability, high surface area, and ease of functionalization.<sup>33–35</sup> These materials, sourced from plant cellulose and crustacean chitosan, can be tailored for effective MP adsorption while minimizing their

ecological footprint.<sup>33,34,36–40</sup> Furthermore, the biodegradability of NC is critical for its application in removing MPs, as it ensures that the material itself will not persist in the environment or generate secondary pollution after use. This attribute strengthens its role as a genuinely sustainable, high-performing option for long-term water purification technologies.

Given the urgent need for clean water and a sustainable environment, there is increasing research interest in developing renewable membrane materials for pollutant removal. However, despite numerous studies on MP removal, comprehensive evaluations explicitly linking NC and biopolymer-based adsorbents to practical wastewater treatment performance remain limited. Existing reviews often focus narrowly on material synthesis or laboratory-scale performance without addressing integration challenges, cost-effectiveness, or regulatory considerations. Moreover, recent trends—including the push for sustainable textile processing, tightening environmental regulations, and advances in bio-based nanomaterials—underscore the urgent need for a critical synthesis of current knowledge. This review consolidates and critically evaluates recent advances in NC- and biopolymer-based systems for MP removal from wastewater, focusing on adsorption, filtration, and flocculation mechanisms. It links material performance to practical considerations for integrating wastewater treatment and achieving cost-effectiveness. It situates these findings within emerging regulatory and policy contexts to provide insights that guide future research and sustainable implementation strategies.

### 1.1 Source of the MPs

According to Babaei *et al.*,<sup>41</sup> MPs in water come from numerous sources, which can be classified into primary and secondary categories. Understanding these sources is essential for formulating effective mitigation strategies. To visualize these origins, the primary and secondary MP pathways are depicted in Fig. 2.<sup>42</sup> This study also illustrated that various sources voluntarily produce primary MPs, which typically measure less than





Fig. 2 Sources and pathways of MP formation. Primary MPs come directly from products like personal care items, textiles, tires, paints, laundry, and plastic manufacturing. Secondary MPs result from fragmentation of larger plastics such as bottles, bags, fishing nets, and shipping waste. Published under the CC-BY License.<sup>42</sup> Copyright 2023, The authors. Published by Springer-Verlag GmbH Germany.

5 mm in size. Familiar primary sources include microbeads found in personal care products such as toothpaste, exfoliating scrubs, and several cosmetics. During use, these microbeads are rinsed down the drain and eventually end up in the water. Plastic pellets, commonly referred to as nurdles, serve as essential raw materials in plastics manufacturing. However, nurdles unintentionally leave the environment during handling processes or transportation.<sup>43–45</sup> Another major contributor is the use of synthetic fibers and textiles, as their production, usage, and disposal result in considerable microfiber emissions into the environment.<sup>46</sup> Microfibers are notably released from garments during washing due to wear and tear, as shown in a study by Parbhakaran *et al.*,<sup>47</sup> which tested polyester and nylon fabrics using brush-washing methods. In contrast, secondary MPs are formed when larger plastic materials degrade into smaller pieces due to environmental factors, including weathering, UV radiation, and physical abrasion.<sup>41,47</sup> The degradation of plastic waste, such as bottles, bags, and packaging materials, occurs through exposure to wind, sunlight, and wave action.<sup>48,49</sup>

In the field of agriculture, plastic mulches degrade over time, releasing MPs into the soil, and animals may ingest these particles when they graze on crop residues mixed with plastic mulch remnants.<sup>48,50</sup> WWTPs can eliminate a considerable amount of MPs, but they remain a significant source of discharge. MPs originating from sources like domestic laundry and industrial wastewater can both enter and exit these facilities, ultimately reaching receiving waters.<sup>46,51,52</sup> Urban runoff, *via* stormwater, transports MPs from polluted soil into aquatic environments,<sup>53</sup> while atmospheric deposition also contaminates urban water systems.<sup>54</sup> Moreover, abrasion of road markings and tire wear generate plastic particles, making them contributors to MP pollution.<sup>46,55</sup>

Table 1 summarizes the composition, physical form, and size ranges of MPs from various domestic, industrial, and environmental sources, providing context to their diversity and scale. Particular examples and geographical factors further emphasize the matter: in the Han River, China, the presence of MPs varied from  $2315 \pm 603$  to  $8406 \pm 2055$  n m<sup>-3</sup>, indicating a rising trend throughout the river.<sup>56</sup> Research conducted in textile industrial regions has revealed elevated levels of MPs,

predominantly consisting of polyester as the primary polymer type.<sup>57</sup> Tsang *et al.*<sup>58</sup> illustrated that in Hong Kong, the levels of particles found in marine waters ranged from 51 to 27 909 particles per m<sup>3</sup>, while in sediments, they varied from 49 to 46 143 particles per kg. A recent investigation conducted in Indiana rivers has highlighted notable MP contamination, emphasizing the extent of this problem in the Midwest's flowing waters.<sup>59</sup>

## 1.2 Impact of MPs on the ecosystem

The ecological impacts of MP pollution are varied and influence organisms at various trophic levels. A wide range of aquatic organisms, extending from plankton to fish, can ingest MPs, resulting in both chemical and physical toxicity.<sup>84</sup> Fig. 3 shows the transfer of MPs through different trophic levels.<sup>85</sup> In addition to the absorption of harmful substances that build up on the surface of MPs, ingestion of MPs may end in physical injury, such as blockages in the digestive tract.<sup>86</sup> It can also disrupt food chains by accumulating within organisms, which leads to higher levels that can affect how ecosystems work and are structured.<sup>87,88</sup> A study by Moto *et al.*<sup>89</sup> illustrated that MPs have the potential to change habitats, disturb ecological equilibrium, and pollute water and soil. Sing *et al.*<sup>90</sup> indicate that MPs have negative impacts on plant development, soil pollution, and the contamination of subsurface aquifers. The ability of MPs to absorb and interact with organic pollutants can change their toxic effects and complicate treatment efforts.<sup>91</sup> The appearance of MPs may facilitate the movement and colonization of particular microbes, resulting in disruptions within the affected ecosystems.<sup>92</sup>

## 1.3 MPs in drinking water

MPs have been found in both tap and bottled water, emphasizing widespread contamination.<sup>93,94</sup> The source of MPs in drinking water can vary, including unfiltered water sources, packaging materials, treatment processes, and distribution systems, as depicted in Fig. 4.<sup>94,95</sup> A study by Semmouri *et al.*<sup>96</sup> found the presence of MPs in surface water, ground water, and treated sewage water that serve as sources of drinking water.



**Table 1** Comprehensive overview of MP sources, their polymer compositions, physical morphologies, and size distributions

Source	Composition	Physical form	Size range	Ref.
Facial cleaners	Polyethylene	Spherical	Higher than 0.5 mm	60
Beverage products	Polyamide, acrylonitrile–butadiene–styrene, poly(ester-amide), poly(ethylene terephthalate)	Fibres, fragments	0.1–3 mm	61
Textile factory	Polyester	Fibres	0.1–1 mm	62
Plastic mulch	Polyester, polypropylene	Fibres, fragments, foam, films	Higher than 500 µm	63
Mariculture activities	Polyester, polypropylene, polyethylene, polyamide (nylon), polystyrene, polyetherurethane, polybutylene terephthalate	Fragments, flakes, fibres, foam	Less than 0.25 mm	64
Anthropogenic activity	Polystyrene, polyethylene, polypropylene	Fibres, styrofoam, fragments, films, pellets	Less than 0.5 mm	65
Urban sewage	Polyethylene, polystyrene, polypropylene	Fragments, lines, foam, films	1–4.75 mm	66
Construction, fishery activities, and human domestic sewage	Polyvinylchloride, polyethylene, polyamide	Fibres, pellets, films, fragments	Less than 0.5 mm	67
Industrial area	Polyethylene, polypropylene, nylon	Fibres, fragments	0.1–5 mm	68
Tertiary industry	Polyethylene, polypropylene, polyacrylonitrile, polyethylene terephthalate	Fragments, fibers, films	500 µm to 5 mm	69
Artificial ecosystems	Polyethylene, rayon, polypropylene	Fibres, flakes, films, granules	Less than 1 mm	70
Plastic industries	Polypropylene, polyester, nylon, polystyrene	Fibres, lines, spherules, fragments/granules, films	Less than 0.5 mm	71
Shower gels	Polyethylene	Irregular shapes	422 ± 185 µm	72
Car tires	Polypropylene, acrylic, nylon, rubber	Fragments, fibres	Higher than 500 µm	73
Facial scrubs	Polyethylene, polyvinyl chloride	Spherical, irregular, granular	85 to 186 µm	74
Cosmetic products	Polyethylene	Irregular, granular, spherical	54–115 µm	75
Industrial sources	Polyethylene, nylon, polypropylene	Films, fragments, lines, granules, sheets, lines	0.5–1.0 mm	76
Fishing and shipping activities	Ionomer surlyn, acrylic (acryl fibre), polyetherimide, polyphenylene sulphide, ethylene vinyl alcohol, acrylonitrile, nylon, polyisoprene, polyvinyl chloride, ethylene–vinyl acetate, polyurethane	Fibres, pellets, fragments	1489 ± 1017 µm	77
Personal care products, facial cleansers, sewage sludge	Polystyrene, polyester, amino thermoset plastic, polyallyl diglycol carbonate	Fragments, pellets, foam, films, lines	0.355–0.999 mm	78
Urbanization	Polyethylene, polypropylene	Pellets, fragments, films, lines, foam	0.3–4.75 mm	79
Sludge and WWTPs	Polyamide, polyethylene, polypropylene	Fragments, fibres, films, granules	0.003–0.05 mm	80
Local inputs, ocean transport	Polypropylene, polyester, polyester, polyethylene	Fibres, flakes, films, granules	2.0–2.5 mm	81
Domestic, agriculture effluent, industry, upstream inflow, and airborne settlement	Polyethylene terephthalate, polyethylene, polypropylene, polystyrene, polycarbonate, polyvinyl chloride, cellulose propionate, polyamide, ethylene–vinyl acetate copolymer	Pellets, fragments	0.05–5 mm	82
Commercial fish species	Polyethylene terephthalate, polyethylene, polypropylene, polyamide, phthalocyanine	Fibres, fragments	Higher than 215 µm	83





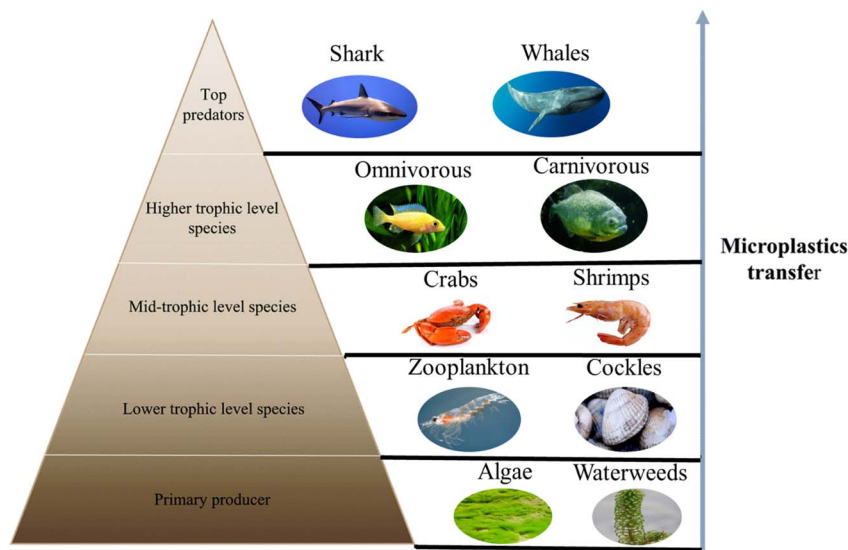


Fig. 3 MP transfer across trophic levels in the aquatic food chain (created with MS PowerPoint).



Fig. 4 Schematic representation of human MP exposure through the use of disposable plastic drinking cups. Published under the CC-BY License.<sup>95</sup> Copyright 2024, The authors. Published by MDPI.

Table 2 Summary of MP particle size, affected organisms, and reported biological effects

Origin	MP exposure/size	Effects	Ref.
<i>Emys orbicularis</i>	500–1000 mg kg <sup>−1</sup>	Induced pathological changes in liver and kidney tissues	104
<i>Ascidian ciona intestinalis</i>	1 µm	Impaired food uptake efficiency and reduced growth rate	105
Nematode	1 µm	Triggered oxidative stress and induced intestinal injury	106
<i>Crepidula onyx</i>	2 µm	Growth reduction	107
<i>Sardinella gibbosa</i>	1 µm	Decreased body weight and altered feeding behavior	108
<i>Dania rerio</i>	70 µm	Disrupted normal gut structure	109

Another study conducted by Zhou *et al.*<sup>97</sup> indicated that the amount of MPs released from plastic cups was around  $556.80 \pm 31.39$  particles/L. In village areas, MPs can contaminate drinking water systems from purification plants and reservoirs. According to Meshram and Mhatre,<sup>98</sup> these particles can pass through the intestinal membrane and aggregate in tissues. They have the potential to promote inflammatory responses and oxidative stress within the body.<sup>99,100</sup> Sharma *et al.*<sup>100</sup> discovered that MPs can interfere with typical composition and

functioning of the intestinal microbiota, which plays a crucial role in immune and digestion response.

To further illustrate these impacts, Table 2 summarizes recent findings on MP sizes, affected organisms, and their observed toxicological effects. Also, they can be found in sources of drinking water, presenting a significant risk to aquatic ecosystems.<sup>84</sup> Aquatic organisms have the ability to consume MPs, which can result in stunted growth, reduced feeding efficiency, and reproductive toxicity.<sup>101,102</sup> MPs may also function as carriers for harmful chemicals, leading to biomagnification and



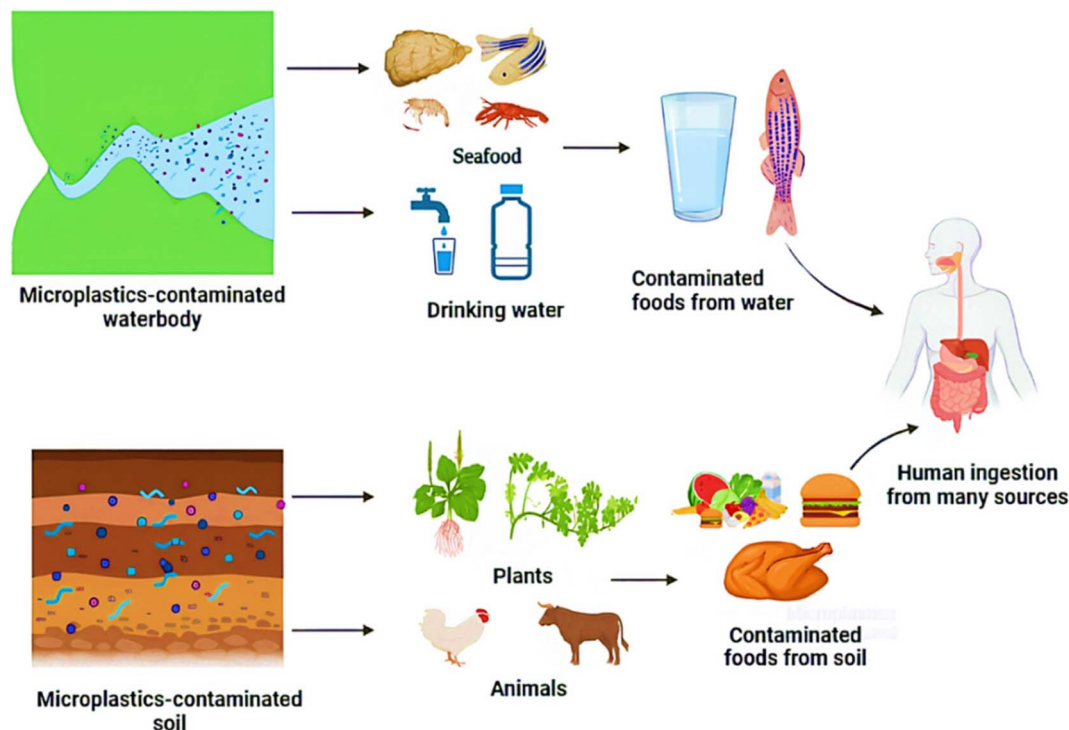


Fig. 5 Transfer routes of MPs from water, soil, and biota to the human body. Published under the CC-BY License.<sup>110</sup> Copyright 2024, The authors. Published by Springer Nature Switzerland AG.

bioaccumulation within the food chain<sup>101</sup>. This trophic transfer and its cascading ecological consequences are illustrated in Fig. 5, which outlines the movement of MPs through various food web levels.<sup>103</sup>

In addition, nanoplastics, due to their small size ( $<1\ \mu\text{m}$ ), show unique toxicological characteristics and pose risks distinct from those of MPs. They can translocate across biological barriers and accumulate in vital organs, potentially leading to inflammatory responses, oxidative stress, and genotoxicity.<sup>111</sup> Moreover, they can interact with cells at the molecular level, affecting gene expression, signaling pathways, and cellular functions.<sup>112</sup> A research conducted by Christopher *et al.*<sup>113</sup> found that nanoplastics have been detected in human placentas, indicating maternal exposure and negative effects on early-life development. Exposure routes include inhalation, dermal contact, and ingestion.<sup>114</sup> They can also accumulate biota, leading to histological damage, neurotoxicity, and metabolic disruption in fish. Additionally, they can impact soil function and food-chain transfer.<sup>115</sup>

## 2. MP detection

The identification of MPs in wastewater involves a series of steps that include sampling, pre-treatment, and separation, followed by identification and quantification. Many methods, such as Fourier-transform infrared spectroscopy (FTIR), pyrolysis-gas chromatography/mass spectroscopy (Py-GC/MS),<sup>†</sup>

and Raman spectroscopy, are used to find and measure MPs in the environment.<sup>116–118</sup> A comprehensive overview of these MP detection techniques, including their analytical capabilities, advantages and limitations, as well as the types of polymers identified in biotic and aquatic environments, is presented in Table 3.

The initial phase involves sampling, an essential procedure focused on gathering samples from WWTPs. The accuracy of this phase depends on the type of water, including freshwater, seawater, or wastewater, since timing is crucial to account for temporal and spatial fluctuations in MP levels.<sup>149</sup> Sampling techniques can be customized according to the type of water-be it freshwater, seawater, or wastewater-because of the variations in particulate load and composition.<sup>150</sup>

After sampling, separation and pretreatment steps are carried out to separate the sample from the wastewater matrices. Filtration stands out as the most prevalent method for separation, generally utilizing filters with stainless steel basket filters that employ smaller and smaller mesh sizes (such as  $10\ \mu\text{m}$ ,  $50\ \mu\text{m}$ , and  $100\ \mu\text{m}$ ), which are effective at capturing different particle sizes.<sup>151</sup> The process becomes better through density separation, which requires the differences in density between MPs and other particulate matter. Researchers used high-density salt solutions to enable the plastics to float, while the heavier materials settle.<sup>152</sup> Digestion steps eliminate organic matter that could obstruct spectroscopic identification. Agents for chemical digestion, such as HCl,  $\text{H}_2\text{O}_2$ , and various enzymes, are applied to break down organic components while preserving the structural integrity of plastic particles.<sup>149,152</sup> The identification of MPs is primarily carried out using

<sup>†</sup> Py-GC/MS: a thermoanalytical method that thermally decomposes polymers into fragments for identification *via* gas chromatography and mass spectrometry.



**Table 3** Comparative evaluation of MP detection techniques, highlighting their analytical strengths and limitations, sample origins and types, measurable size ranges, and polymers identified in environmental and biological matrices<sup>a</sup>

Detection methods	Advantages	Limitations	Source and sample type	Size ranges	Identified polymers	Ref.
Optical microscopy (MO) and Fourier transform infrared spectroscopy (FTIR)	<ul style="list-style-type: none"> <li>Optical microscopy assists in preliminary sorting and size classification, while FTIR provides chemical identification<sup>119,120</sup></li> <li>Combining methods reduces error rates compared to optical microscopy alone<sup>119</sup></li> </ul>	<ul style="list-style-type: none"> <li>Still labor-intensive and time-consuming due to manual sorting and FTIR analysis steps<sup>120</sup></li> <li>FTIR spatial resolution limits detection of smaller particles, and visual sorting may miss small or transparent MPs<sup>119</sup></li> <li>Requires expertise and is prone to subjective bias during visual pre-sorting<sup>120</sup></li> </ul>	Indian white shrimps	0.157–2.785 mm	PA, PES, PE, PP	121
			Deep-sea fish	<1 mm	CPH, PA, PET	122
			Bivalve (oyster, mussel, manila clam and scallop)	0.1–0.2 mm	PE, PP, PS, PES, PEVA, PET, PU	123
			Different fish species	0.2–5 mm	PE, PP	124
			Thamnaconus septentrionalis	0.04–5 mm	CPH, PET, PES	125
			Mussels ( <i>Mytilus edulis</i> )	0.033–4.7 mm	CPH, PET, PES	126
			Benthic organisms	0.05–5 mm	PP, PE, PS, PET, NY	127
			Deep benthic invertebrates	0.023–6.25 mm	ALK, PES	128
			Different fish species	0.656 mm	PS, PE, PP	129
			Benthic and pelagic fish	0.217–4.81 mm	PP, PE, ALK, RY, PES, NY	130
			Pelagic and demersal fish	0.13–14.3 mm	PA, cellulose, RY	131
			Whales	0.3–7 mm	RY, PES, acrylic, PP, PE	132
			Atlantic herring, sprat, common dab, and whiting	0.300–0.400 mm	PMMA	133
			Plastic bottled water	6.5–>100 µm	PET, PP	80
			Glass bottled water	>100 µm	PA, PE, PP	80
			Cardboard bottled water	>100 µm	Cellulose, PE, PP	80
			Ground drinking water	50–150 µm	PE, PA, PES, PVC	135
Fourier transform infrared spectroscopy (FTIR)	<ul style="list-style-type: none"> <li>Widely used and well-established technique with extensive reference databases<sup>134</sup></li> </ul>	<ul style="list-style-type: none"> <li>Cannot identify MPs smaller than about 20 µm (ref. 119)</li> <li>Difficult to analyze opaque and black MPs<sup>134</sup></li> </ul>	Tap water	100–5000 µm	—	136
			Plastic bottled water	6.5–>100 µm	PP	138
			Glass bottled water	6.5–>100 µm	—	138
			European anchovies	0.124–0.438 mm	PE	139
			Bivalves ( <i>Mytilus edulis</i> and <i>Crassostrea gigas</i> )	>0.005 mm	—	140
Optical microscopy (MO) and Raman microspectroscopy (RMS)	<ul style="list-style-type: none"> <li>MO provides rapid visual identification and preliminary sorting based on size, shape, and color; RMS offers detailed chemical characterization and polymer identification</li> </ul>	<ul style="list-style-type: none"> <li>Both methods can be affected by MP heterogeneity and environmental aging</li> </ul>	Mussels ( <i>Mytilus edulis</i> ) and lugworms ( <i>Arenicola marina</i> )	0.015–1 mm	—	141
			Glass bottled water	<5 µm	PE, SBR	142
			Plastic bottled water	<5 µm	PE	142





Table 3 (Contd.)

Detection methods	Advantages	Limitations	Source and sample type	Size ranges	Identified polymers	Ref.
Optical microscopy (MO)	<ul style="list-style-type: none"><li>• Convenient and economical method</li><li>• Simple operation and low cost</li><li>• Can identify MPs with particle sizes of hundreds of microns and above<sup>119</sup></li><li>• Capable of analyzing non-volatile macromolecules by breaking them into volatile fragments for identification</li><li>• Provides detailed chemical characterization of polymers and additives in MPs</li><li>• Useful for complex samples where other methods may fail<sup>146</sup></li><li>• FTIR for larger particles and Raman for smaller particles and detailed chemical imaging.<sup>134</sup></li><li>• Enhances detection accuracy and chemical identification of MPs across a wide size range<sup>119,134</sup></li><li>• Automated or semi-automated systems can reduce analysis time and human bias<sup>148</sup></li></ul>	<ul style="list-style-type: none"><li>• Accuracy is relatively low; error rates of 20–70% for transparent MPs</li><li>• Time consuming and laborious</li><li>• Cannot provide chemical composition information<sup>119</sup></li><li>• Sample is decomposed during analysis</li><li>• Requires specialized instrumentation and expertise</li><li>• Does not provide physical information such as particle size, shape, or morphology directly<sup>146</sup></li><li>• High cost and complexity due to combining two sophisticated instruments<sup>134</sup></li></ul>	Dogfish, hake, red mullet	0.38–3.1 mm	—	143
Pyrolysis-gas chromatography-mass spectrometry (pyrolysis-GC-MS)			Semi-pelagic fish	0.5 mm	—	144
			Ground drinking water	—	PE	145
Fourier transform infrared spectroscopy (FTIR) & micro-Raman imaging microscopy			Treated water from water treatment plants	1–10 µm	PET, PP, PE	147

<sup>a</sup> Abbreviations: ABS: acrylonitrile butadiene styrene; ALK: alkyl resin; CPH: cellphone; EPM: ethylene-propylene rubber; EP: epoxy resin; EVOH: ethylene vinyl alcohol copolymer; LLDPE/Oct: linear low-density polyethylene-octene copolymer; NY: nylon; PA: polyamide; PAN: polyacrylonitrile; PAS: polyacrylate-styrene; PCL: polycaprolactone; PE: polyethylene; PEA: polyethylacrylate; PET: polyethylene terephthalate; PEVA: polyethylene-vinyl acetate; PMMA: polymethyl methacrylate; PP: polypropylene; PR: phenoxy resin; PS: polystyrene; PES: polyester; PTFE: polytetrafluoroethylene; PU: polyurethane; PVC: polyvinyl chloride; PVA: polyvinyl alcohol; PVAc: polyvinyl acetate; PVS: polyvinyl sulfonate; RY: rayon; SAN: styrene-acrylonitrile; SBR: styrene-butadiene rubber; SR: synthetic rubber.



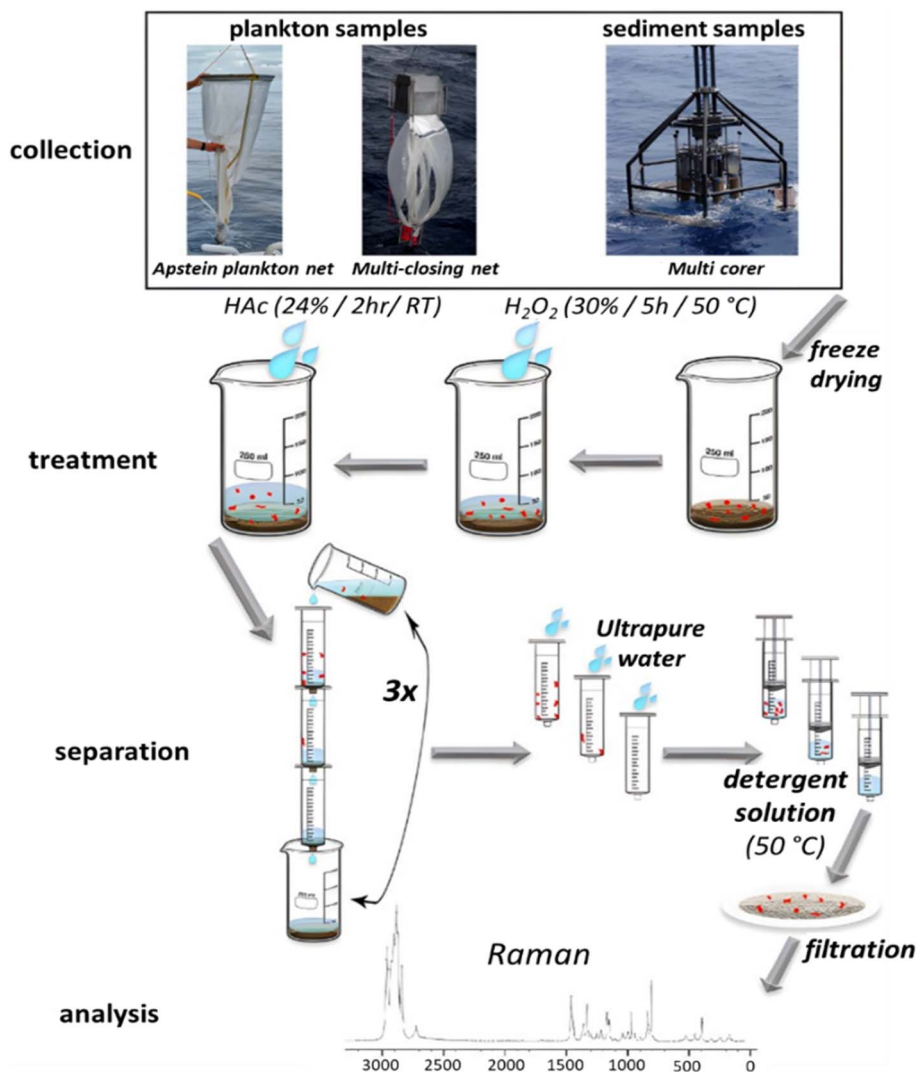


Fig. 6 Workflow for MP detection: (1) sampling via multi-corers or plankton nets; (2) freeze-drying and digestion with H<sub>2</sub>O<sub>2</sub> and acetic acid; (3) separation using syringe cascade and detergent desorption; (4) analysis by Raman spectroscopy. Published under the CC-BY License.<sup>120</sup> Copyright 2021, The authors. Published by Elsevier B.V.

spectroscopic and thermal analytical techniques. FTIR spectroscopy determines the chemical composition of MPs through the analysis of infrared absorption patterns. Using micro-FTIR along with imaging allows for a close-up and detailed study of individual particles.<sup>116,150,153</sup> Raman spectroscopy, a widely used technique, provides information at the molecular level by analyzing rotational and vibrational modes. More advanced methods, such as Surface Enhanced Raman Spectroscopy (SERS), drastically improve detection sensitivity.<sup>153–155</sup> Pyrolysis-GC/MS, functioning as a thermoanalytical method, thermally breaks down MPs into smaller molecular fragments for subsequent identification and separation, which is particularly useful for complex or mixed samples where direct spectroscopic identification is difficult.<sup>109,155</sup>

Methods of quantification involve both chemical and visual tactics. Optical microscopy is often employed for quantifying microplastic particles, whereas scanning electron microscopy offers a thorough morphological understanding.<sup>149,152,153</sup>

Research into the topic of spectroscopic quantification continues, utilizing FTIR and Raman spectral data to determine the mass of MPs, although this area is still under development.<sup>156</sup> Pyrolysis-GC/MS provides a more detailed characterization of specific polymers by determining the prevalence of their distinct degradation products.<sup>157</sup> Innovative detection methods are consistently improving the precision and effectiveness of MP detection. Micro-flow imaging (MFI)<sup>‡</sup> assists in the real-time determination of MPs *in situ*.<sup>158</sup> Hyperspectral imaging uses near-infrared technology and FTIR to provide very detailed images, and when combined with chemometric modeling, it helps quickly and accurately identify different types of plastics.<sup>152,159</sup> Xue *et al.*<sup>154</sup> demonstrated that new methods, such as the extreme learning machine paired with differential Raman spectroscopy, combine advanced computer

<sup>‡</sup> MFI: an optical technique for real-time, *in situ* analysis of suspended microplastic particles in liquids.





**Table 4** Overview of various cellulose-based nanomaterials used for MP removal, detailing their source, size, surface characteristics, synthesis methods, mechanical properties, removal efficiency, recyclability, and application performance

Type	Source	Size (nm)	Surface area (m <sup>2</sup> kg <sup>-1</sup> )	Synthesis method	Surface chemistry/modification	Mechanical properties	Removal efficiency (%)	Recyclability/stability	Application	Ref.
NCC	Wood pulp, cotton, BC	Width: 2–5; length: 100–300	150–250	Acid hydrolysis (typically H <sub>2</sub> SO <sub>4</sub> ), enzymatic pretreatment	Sulfate groups from acid hydrolysis, PEI coating, carboxylation	Rigid rod-like crystals	>98% removal of MPs within 20 min of adsorption	—	Used in membranes, composites for rapid adsorption	144 and 146
CNCs	Wood pulp, cotton, BC	Width: 3–10; length: 100–300	Up to 150	Acid hydrolysis (typically H <sub>2</sub> SO <sub>4</sub> ), mechanical pretreatment	Sulfated groups, PEI, or polymer coatings	Rigid crystalline rods	>98% removal of MPs; rapid adsorption kinetics	—	Incorporated in filtration membranes, aerogels	144 and 146
CNFs	Wood, cotton, BC, <i>Agave americana</i>	Diameter: 10–30	High	Mechanical fibrillation (homogenization), enzymatic/chemical pretreatment (carboxymethylation)	Sulfonation (–SO <sub>3</sub> H), carboxylation (–COOH), PEI coating	Flexible, forms entangled 3D porous aerogels	88.8% removal of PS-NH <sub>2</sub> MPs; adsorption capacity 586.95 mg g <sup>-1</sup>	>78% efficiency after 10 cycles	Used in aerogels, filters	29, 144 and 145
NFC	Wood pulp, agricultural residues	Diameter: 10–30	High	Mechanical fibrillation, chemical pretreatment (TEMPO oxidation)	TEMPO-oxidized carboxyl groups	Flexible, entangled fibrils	Comparable to CNFs; good adsorption	Good recyclability	Used in filtration membranes, aerogels	144 and 145
Micro fibrillated cellulose (MFC)	Bleached wood pulp	Width: 5–30	Moderate to high	Enzymatic or chemical pretreatment + high-pressure homogenization	—	Moderate mechanical strength	Up to 59% removal efficiency	Low recyclability	Used in preliminary MP removal	144 and 148
CMF	Various cellulose sources	Diameter: 200–300	Moderate	Microwave-assisted acid hydrolysis + g C <sub>3</sub> N <sub>4</sub> synergy (10 min)	—	—	99% removal with an attapulgite (APT) composite	Moderate	Composite filters for MP removal	148
Bacterial NC	BC ( <i>Acetobacter xylinum</i> )	Diameter: 20–100	High	Bacterial biosynthesis	Typically unmodified, high purity	Uniform size	—	—	—	144 and 148
Cellulose benzoate (modified CNCs)	Cellulose esterified in an ionic liquid (AmimCl)	—	—	Esterification in an ionic liquid + CNT/MCNT incorporation	Modified with carbon nanotubes (CNTs), magnetic carbon nanotubes (MCNTs)	—	>97% removal of MPS with CNT/MCNT modification	—	Enhanced $\pi$ - $\pi$ interactions and zeta potential for adsorption	147
CNF-coated delignified wood (CNF-CDW)	Balsa wood	CNF diameter up to 10 to 20; wood pores micron scale	—	Delignification (NaClO <sub>2</sub> , Na <sub>2</sub> SO <sub>3</sub> /NaOH, DES) + CNF coating + CaCl <sub>2</sub> crosslinking	CNF-film coating on wood; cross-linked	Flux 1146 L m <sup>-2</sup> h <sup>-1</sup>	95.97% removal of MPs	Sustainable, reusable filter	Combined mechanical filtration and adsorption	28

learning with light detection, leading to enhanced sensitivity for biological samples. Furthermore, nanoparticle tracking analysis has been used to measure the number of nanoplastics present and their size, in response to growing concerns about plastic particles that are smaller than one micron.<sup>119</sup> A study by Rein-eccius *et al.*<sup>120</sup> illustrated the MP detection workflow shown in Fig. 6, highlighting key steps such as sampling using multicorers or plankton nets, freeze-drying and digestion with hydrogen peroxide and acetic acid, separation through syringe cascades and detergent desorption, followed by detailed analysis *via* Raman spectroscopy.

### 3. NC

NC, the most abundant biopolymer on the planet, is an excellent substrate for the massive production of affordable, ecologically sound water treatment media.<sup>121</sup> Based on the origin of raw materials, the synthesis method, and the morphological structure, there are multiple kinds of NC materials. A variety of names have been given to these NC materials, such as nanocrystalline cellulose (NCC), cellulose nanocrystals

(CNCs), cellulose nanofibrils (CNFs), cellulose microfibrils (CMFs), cellulose nanofibers, nanofibrillated cellulose (NFC), microfibrillated cellulose, and various combinations of the aforementioned types.<sup>122</sup> Each type possesses an identical chemical composition, but due to disparities in sources and extraction techniques, they differ in structure, particle size, crystallinity, and other characteristics.<sup>123</sup> Table 4 provides a comparative overview of these NC types, highlighting their synthesis methods, surface modifications, mechanical properties, recyclability, and MP removal efficiencies.

NCC—also referred to as CNCs, nanocrystals of cellulose, or cellulose nanowhiskers—is an exceptionally robust form of NC usually retrieved from cellulose fibrils by acid hydrolysis.<sup>123</sup> The extended, malleable, and interwoven NC that can be mechanically separated from cellulose fibrils is called NFC, often referred to as cellulose nanofibrils (CNFs) or nanofibrillar cellulose.<sup>124,125</sup> Another sort of NC is bacterial NC, which is primarily manufactured by *G. xylinus* and can be recovered from the ingestion of tiny molecular carbohydrates by bacteria.<sup>126</sup> Hydrolysis with extremely potent acids yields the nanocrystalline particles by breaking local crystalline bonds between

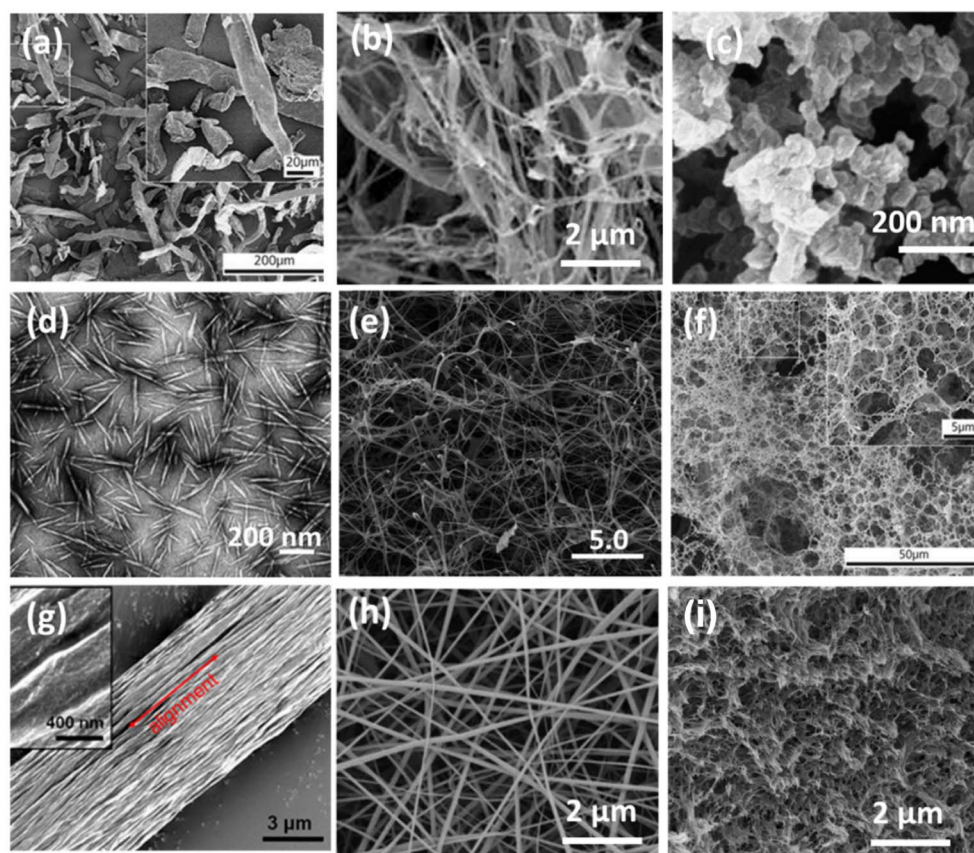


Fig. 7 Microscopy image of (a) CMFs. Reproduced with permission from ref. 129. Copyright 2021, Springer Nature; (b) cellulose fibers (CFs). Published under the CC-BY License.<sup>130</sup> Copyright 2017, The authors. Hindawi; (c) cellulose nanoparticles (CNPs). Reproduced with permission from ref. 131. Copyright 2007, Elsevier Ltd.; (d) CNCs. Published under the CC-BY License.<sup>132</sup> Copyright 2018, The authors. Published by MDPI; (e) cellulose nanofibers. Reproduced with permission from ref. 133. Copyright 2009, Royal Society of Chemistry; (f) BC and (g) cellulose nanofibers assembled into a material stronger than spider silk. Published under the CC-BY License.<sup>134</sup> Copyright 2018, The authors. Published by American Chemical Society; (h) electrospun cellulose nanofibers. Reproduced with permission from ref. 135. Copyright 2012, Elsevier Ltd.; (i) cellulose aerogels. Published under the CC-BY License.<sup>136</sup> Copyright 2018, The authors. Published by MDPI.





nanofibrils and rupturing amorphous domains.<sup>127</sup> Cellulose fibers undergo mechanical breakdown into CNFs most frequently by refining, high-pressure homogenization, and grinding.<sup>125</sup> Bacterial cellulose (BC) can be produced by *Acetobacter aceti*, and it can be optimized by the fermentation process.<sup>128</sup> Fig. 7 shows different types of NC materials.<sup>129–137</sup>

However, the synthesis of various NC materials faces several challenges primarily related to processing conditions and material properties. Traditional methods for NC preparation, such as mechanical, biological, and chemical treatments, have limitations including long treatment times, high energy consumption, and environmental concerns.<sup>138</sup> For instance, the production of CNCs often involves acid hydrolysis, which requires careful control to avoid degrading the cellulose while removing amorphous regions.<sup>139,140</sup> CNFs, produced *via* mechanical methods, can suffer from high energy consumption during fibrillation.<sup>141</sup> BNC, although pure, requires specific culture conditions and can be costly to produce at scale.<sup>142</sup> Modified CNCs, like cellulose benzoate, and composites like CNF-CDW require additional chemical modification steps that can introduce complexities in maintaining structural integrity and desired properties.<sup>143</sup>

### 3.1 NC characteristics relevant to MP removal

**3.1.1 High surface area and porosity.** The high specific surface area of NC is one of its outstanding characteristics, and it contributes to wastewater treatment. The adsorption capacity of materials based on NC for capturing pollutants and the size exclusion capability of eliminating undesired elements may be improved by increasing the specific surface area of NC and its interactions with the surrounding matrix.<sup>144</sup> The adsorption of MPs is greatly dependent on the high specific surface area of the NC and the microporous structure of the NC aerogel.<sup>160</sup> Among many other qualities, excellent mechanical properties of NC, ease of processing into porous 2D or 3D structures, and exceptionally reactive surface with high surface area (resulting

in high density) make it a great building block for designing functional nanomaterials by mineralization.<sup>161</sup> Pickering foam constructed from CNCs has a low density, which makes it possible to dry it to create porous materials with numerous pore structures and a large specific surface area. Reducing inner density is an achievable approach to improve the specific surface area of objects with known shapes and volumes.<sup>162</sup>

**3.1.2 Biodegradability.** NC is one of the most intriguing plant-based biodegradable biopolymers.<sup>163</sup> Nanomaterials derived from biological sources have particular characteristics of nanomaterials along with the further benefits of sustainability, abundance, and biodegradability.<sup>33</sup> This biodegradability arises from its composition of glucose units linked by  $\beta$ -1,4-glycosidic bonds, which are susceptible to enzymatic breakdown by cellulose enzymes produced by different microorganisms.<sup>164</sup> However, the biodegradation of NC can be affected by several factors, including its source, surface modification, production process, and environmental conditions to which it is exposed.<sup>165</sup> Environmental factors such as pH, temperature, moisture levels, and the presence of certain organisms play an important role in influencing the degradation of NC.<sup>166</sup> When conditions are ideal, the performance of cellulose enzymes is enhanced, leading to a higher decomposition rate of NC into basic substances. Hossain *et al.*<sup>164</sup> found that the degree of crosslinking in NC-based superabsorbent polymers can influence their biodegradability. Higher crosslinking density may impede enzyme accessibility, thereby delaying the degradation process. Another study conducted by Frank *et al.*<sup>165</sup> demonstrated that dispersibility within polymer matrices can also impact its biodegradation. For example, research has illustrated that hydrophobic modifications *via* silanization can reduce the biodegradability of NC.

### 3.2 Mechanisms of MP removal using NC

The application of NC in MP remediation operates through several key mechanisms. Fig. 8 provides a schematic overview of

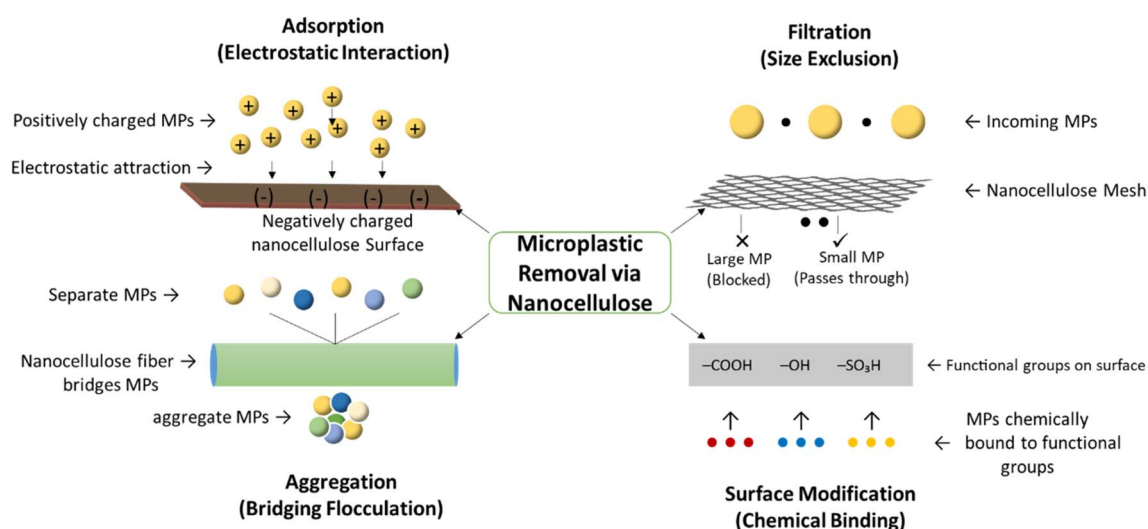


Fig. 8 Schematic representation of four NC-based mechanisms for MP removal (created with MS PowerPoint).



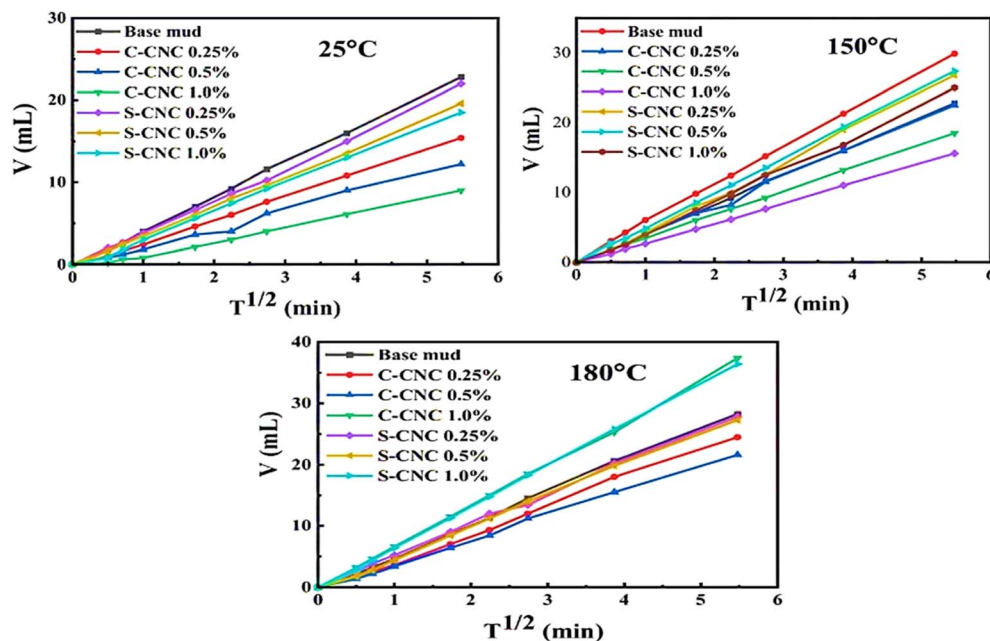


Fig. 9 Filtration volume ( $V$ ) versus square root of time ( $T^{1/2}$ ) for base mud and CNC-modified muds (C-CNC and S-CNC) at 25 °C, 150 °C, and 180 °C, demonstrating the influence of CNC concentration and temperature on fluid loss behavior. Published under the CC-BY License.<sup>170</sup> Copyright 2024, The authors. Published by Springer Nature B.V.

four mechanisms such as adsorption, filtration, aggregation, and surface modification, each of which is elaborated below.

**3.2.1 Adsorption.** TEMPO-mediated oxidation has been identified as an exceptionally effective technique for generating cellulose nanofibers due to its selectivity and low energy consumption.<sup>167</sup> Due to the high attraction between surfaces with opposite charges, the positively charged MPs (polystyrene) instantly bind to the anionic TEMPO-CNF surface during adsorption. Additionally, anionic MPs can more readily enter the NC network in the anionic system because the attraction energy between negatively charged surfaces is half that of surfaces with opposite charges. To boost the effectiveness of removing anionic and nonionic contaminants from NC, the hydroxyl groups can also be chemically modified.<sup>144</sup> These adsorption studies emphasize that surface chemistry and functionalization strongly influence MP capture, providing a conceptual bridge to the membrane selectivity principles discussed in the following filtration section.

**3.2.2 Filtration.** NC can be used to create membrane filters that physically trap MPs.<sup>40,168</sup> These membranes are designed with pore sizes smaller than the MPs, ensuring their retention while allowing water to pass through.<sup>168</sup> NC-based membranes offer advantages such as low energy consumption, high separation efficiency, and eco-friendliness.<sup>40</sup> The effectiveness of these membranes can be improved by modifying the NC to improve its stability and mechanical strength.<sup>169</sup> For instance, the addition of sulfonated or cationic CNCs (S-CNCs and C-CNCs) to water-based drilling fluids reduces filtration volume at different temperatures, illustrating their capability in filtration applications. Fig. 9 demonstrates the filtration volume against the square root of time at various temperatures with

varying concentrations of C-CNC and S-CNC.<sup>170</sup> These findings suggest that coupling adsorption pretreatments with filtration could mitigate fouling and enhance overall MP removal, underscoring the value of integrated treatment designs.

**3.2.3 Aggregation.** NC can induce the aggregation of MPs, leading to the formation of larger particles that are easier to remove.<sup>171,172</sup> This process can be improved through the addition of coagulants, such as aluminum and ferrous sulfate, which destabilize the MPs and promote their clumping together. For example, coagulation with aluminum and ferrous sulfates has proven effective in removing polystyrene and polyvinyl chloride MPs from natural surface water. The efficiency of MP removal can be further enhanced by optimizing the dosage of coagulants and by using NC to create a matrix that entraps the aggregated MPs.<sup>173</sup>

**3.2.4 Surface modification.** Pristine NC contains numerous hydroxyl groups that can be chemically modified to introduce functional groups with a high affinity for MPs.<sup>174,175</sup> Several methods can be employed, such as chemical modification, graft copolymerization, adsorption and bioconjugation. In chemical modification, oxidation, esterification, and etherification can alter the surface properties of NC, making more effective for adsorption.<sup>176</sup> A study by Kopač *et al.*<sup>177</sup> introduced cationic groups that can increase the electrostatic attraction between NC and negatively charged MPs. Fig. 10 shows SEM micrographs of different types of cellulose-based nanofibers, including cationic CNFs, cationic microfibrillated cellulose, freeze dried cellulose nanofibers, and quaternized CNFs, illustrating their individual morphologies.<sup>177</sup> Another technique, graft copolymerization, involves grafting polymers onto the NC surface to fine-tune its properties. By grafting specific polymers, NC can be engineered to





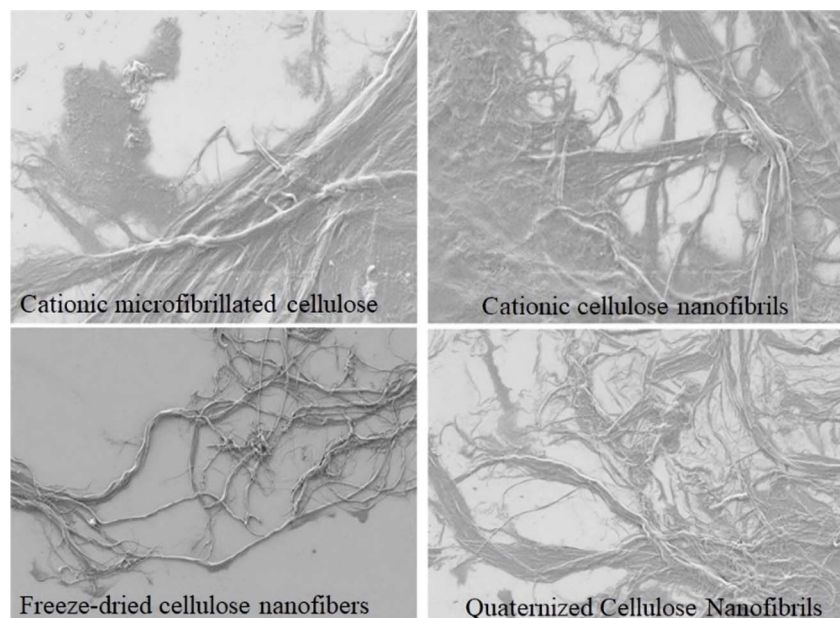


Fig. 10 Micrographs of different types of cellulose-based nanofibers. Published under the CC-BY License.<sup>177</sup> Copyright 2022, The authors. Springer Nature B.V.

selectively bind to different types of MPs.<sup>178</sup> Furthermore, modifying the surface of NC through adsorption and bioconjugation can improve its functionality and biocompatibility. This can lead to better cell adhesion and controlled degradation, making it suitable for various biomedical applications.<sup>179</sup>

Several techniques are available for surface modification, each offering unique advantages depending on the application. For instance, TEMPO (2,2,6,6-tetramethylpiperidine-1-oxyl) is a stable nitroxyl radical widely used in surface modification.<sup>180</sup> It is frequently employed to modify CNFs by introducing carboxy groups.<sup>181,182</sup> These carboxy groups can then be further

functionalized; for instance, primary-amine-terminated polyalkylene glycol (PAG) can react with carboxy groups in TEMPO-CNFs to form amide bonds, improving thermal and dimensional stability.<sup>181</sup> Another surface modification technique is amine functionalization, which involves introducing amine groups ( $-NH_2$ ) onto the surface of a material.<sup>183</sup> A common method for amine functionalization is silanization using 3-aminopropyltriethoxysilane (APTES).<sup>184,185</sup> The ethoxy groups in APTES react with hydroxyl groups on the surface, forming stable siloxane ( $Si-O-Si$ ) covalent bonds.<sup>184</sup> Other surface modification techniques such as cationic surface functionalization using cetrimonium bromide can improve interfacial interactions between CNCs and polymeric matrices in nanocomposites.<sup>186</sup>

## 4. Biopolymers for MP removal

Biopolymers are gaining increasing attention as sustainable materials for several applications, including MP removal from wastewater. These polymers offer eco-friendly alternatives to traditional synthetic polymers due to their renewability and biodegradability. Various types of biopolymers have shown promise in MP removal, as shown in Fig. 11 and detailed in Table 5.

### 4.1 Chitin and chitosan

Chitosan, a naturally derived biopolymer, is a modified form of chitin.<sup>210</sup> It is the most abundant natural polysaccharide found in the exoskeletons of crustaceans (crab, shrimp, and lobster), insects, and fungal cell walls.<sup>211,212</sup> The deacetylation of chitin yields chitosan.<sup>211,213,214</sup> The structure of chitosan features amino and hydroxyl groups, as depicted in Fig. 12, which play an important role in its ability to interact with pollutants.<sup>214,215</sup>

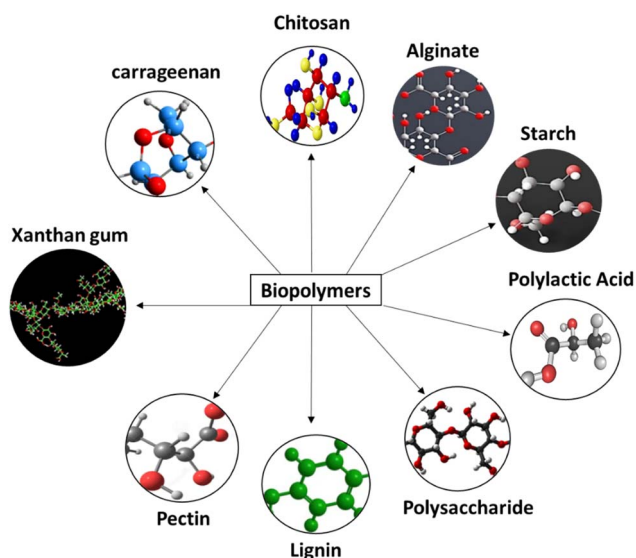


Fig. 11 Different types of biopolymers for MP removal (created with MS PowerPoint).





Table 5 Comparative evaluation of biopolymers based on source, functional groups, MP removal efficiency, and underlying mechanisms

Biopolymer	Source	Functional groups	MP removal efficiency	Mechanism	Ref.
Chitosan	Derived from chitin in crustaceans (shrimp, crab), fungi, insects	-NH <sub>2</sub> , -OH (primary hydroxyl groups at the C-6 position and secondary hydroxyl groups at the C-3 position)	>90% (depends on pH, dosage, type of MP)	Electrostatic attraction (between cationic amino groups and anionic MPs), H-bonding, van der Waals; enhanced by grafting/cross-linking	187–189
Alginate	Brown seaweed (phaeophyceae), bacteria	-COOH, -OH	62–99.79%	Flocculation, electrostatic interaction, hetero-aggregation, physical encapsulation in a Ca-alginate matrix	190–193
Starch	Corn, potato, wheat, cassava	-OH, N <sup>+</sup> (CH <sub>3</sub> ) <sub>3</sub> (cationic modified)	Up to 90% with additives	Charge neutralization, bridging, flocculation, physical entrapment; performance influenced by particle density & age	194–197
Polysaccharides	Plants, algae, animals (e.g., laminarin, starch, alginate, chitosan)	-COOH, -OH, -NH <sub>2</sub>	93–99% (e.g., laminarin gels)	H-bonding, sweeping/entrapment, bridging, van der Waals, electrostatic interactions	195, 197 and 198
Lignin	Wood, pine bark, <i>Miscanthus</i> , paper industry byproduct	-OH (phenolic/aliphatic), -COOH, OCH <sub>3</sub>	Raw: up to 26%; modified: up to 99%	$\pi$ - $\pi$ stacking, hydrophobic interaction, H-bonding, electrostatic, flocculation (hydrogels, Fe-biochar), magnetic separation	199–202
Pectin	Citrus fruits, apples	-COOH, -OH	93–99% (gel systems with coagulants)	H-bonding, sweeping/entrapment, bridging, van der Waals, coagulation & flocculation	198 and 203
Xanthan gum	Fermentation by <i>Xanthomonas campestris</i>	-OH, -COOH, acetyl groups	>90% (coagulant blends)	H-bonding, electrostatic interactions, bridging, gel flocc entrapment; can be enhanced <i>via</i> chemical modification	204, 205 and 197
Carrageenan	Red algae (e.g., <i>Kappaphycus</i> , <i>Gigartina</i> )	-OSO <sub>3</sub> <sup>-</sup> (sulfate), -OH, -OCH <sub>3</sub>	>90% (gels and hydrogels)	Physical entrapment in the gel matrix, electrostatic attraction (sulfated groups), H-bonding	206 and 207
Cyclodextrins	Enzymatic conversion of starch (corn, potato)	-OH (primary and secondary), hydrophobic cavity	>80–90% (for organics)	Host-guest inclusion, H-bonding, van der Waals; enhanced when in composites for aggregation/flocculation	208 and 209

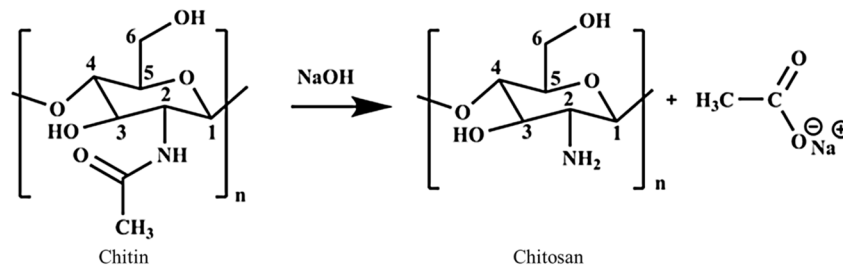


Fig. 12 Derivation of chitosan via chitin deacetylation (created with MS PowerPoint).

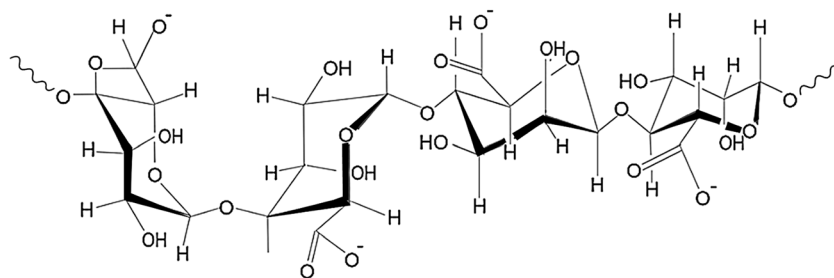


Fig. 13 Structural composition of alginate (created with MS PowerPoint).

The amino groups on the structure of chitosan become positively charged under acidic conditions, facilitating electrostatic interactions with negatively charged MPs.<sup>213</sup> This leads to flocculation and coagulation, where small MPs aggregate into larger flocs that can be easily separated from the water.<sup>216</sup> The porous structure and reactive functional groups of chitosan enable it to adsorb MPs, effectively trapping them within its matrix.<sup>217</sup> Its effectiveness in MP removal stems from its ability to act as an adsorbent and bioflocculant.<sup>34,216</sup> As a bioflocculant, chitosan promotes the aggregation of MPs, leading to their sedimentation or easier removal through filtration.<sup>216,218</sup> Additionally, shaping chitosan into beds increases its surface area and makes it easier to handle and regenerate.<sup>212</sup> Nano-sized chitosan exhibits a high surface area and improved adsorption capacity, and higher MP removal efficiency.<sup>219</sup>

## 4.2 Alginate

Alginate, a naturally occurring biopolymer extracted from brown algae,<sup>220</sup> is a developing sustainable and efficient

solution for MP removal from wastewater.<sup>220,221</sup> Its biocompatible, cost-effective, and nontoxic features make it suitable for various applications.<sup>222</sup> It is a linear polysaccharide composed of two uronic acid monomers, guluronic acid and mannuronic acid.<sup>223</sup> The ability of alginate to form gels in the presence of divalent cations, such as calcium ions, is a key property utilized in MP removal.<sup>224</sup> Zhang *et al.*<sup>221</sup> showed that calcium alginate hydrogel can be used as a flocculant to remove MPs from water, with a removal efficiency of up to 99.5% for certain MPs. Although the effectiveness of this method can vary depending on the composition and size of the MPs. For more effective removal, Wang *et al.*<sup>222</sup> demonstrated that alginate can be combined with other materials to form composites with enhanced adsorption capabilities. Additionally, alginate can be used to entrap other materials, such as nano-zerovalent iron, to degrade organic pollutants through advanced oxidation processes.<sup>225</sup> The functional groups present in alginate can also interact with the surface of MPs, enhancing their removal.<sup>152</sup> It can be processed into various useable forms, such as beads or

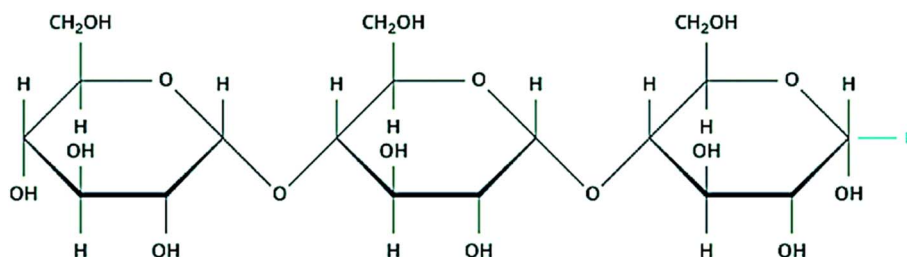


Fig. 14 Fundamental molecular structure of starch polymer (created with MS PowerPoint).





demonstrated biofloculant capacity when exposed to micro- and nano-plastics.<sup>243</sup> Jagadeesh and Sundaram<sup>244</sup> found that modified polysaccharides can absorb MPs onto their surface, facilitating their removal. Furthermore, organosilanes combined with polysaccharides can be used to agglomerate and fix MPs, enabling their removal from water.<sup>245</sup> Biochar, which can be derived from polysaccharide-rich biomass, has shown potential for MP removal through mechanisms such as “Stuck,” “Trapped,” and “Entangled” interactions. However, hybrid biochar-sand filters have shown promise as low-cost systems.<sup>246</sup> The chemical structure of polysaccharide chains is shown in Fig. 16. Furthermore, acrylamide (AM) cross-linked psyllium polysaccharide (PLP-AM) has been synthesized and used as a flocculant for removing PS, PVC, and PET MPs from water. Under optimal conditions, PLP-AM achieved removal percentages of 92.55% for PS, 93.85% for PET, and 94.31% for PVC.<sup>247</sup> Polysaccharides can be extracted from *spirulina platensis* using hot water, followed by chitosan flocculation treatment to remove impurities.<sup>248,249</sup>

#### 4.6 Lignin

The complex structure and abundant functional groups of lignin make it a viable option for adsorbing pollutants, including MPs, from wastewater.<sup>250,251</sup> Studies have explored the direct application of lignin as an adsorbent for MPs. For example, organosolv lignin derived from *Miscanthus* sp., pine bark, and solid anaerobic digestates has been evaluated as an adsorbent for various types of MPs.<sup>199</sup> Additionally, lignin-based materials, especially when processed into activated carbons or porous structures, offer a high surface area and porosity.<sup>252,253</sup> MPs can become entrapped within these pores through physical adsorption.<sup>253</sup> Cationic lignin polymers can neutralize the negative surface charges commonly found on MPs in wastewater.<sup>254</sup> This charge neutralization reduces repulsive forces between MP particles, allowing them to aggregate.<sup>255</sup> Furthermore, lignin polymers can act as bridges between MP particles, linking them together to form larger flocs.<sup>254</sup> These larger flocs

are then easier to remove *via* sedimentation or filtration.<sup>256,257</sup> Kaur *et al.*<sup>258</sup> demonstrated that lignin can degrade MPs under light irradiation. Lignin can be chemically modified with [2-(methacryloyloxy)ethyl]trimethylammonium chloride (METAC) or acrylic acid to improve its charge density, water solubility, and molecular weight, improving its flocculation performance.<sup>254,255,259</sup> For example, introducing cationic groups through METAC can enhance the ability of lignin to flocculate negatively charged MPs.<sup>255</sup> The underlying polymeric structure of lignin is demonstrated in Fig. 17.

## 5. Coagulation and flocculation mechanisms

Coagulation and flocculation are crucial processes for removing MPs from wastewater.<sup>260</sup> These processes involve destabilizing suspended particles, including MPs, and aggregating them into larger flocs that can be easily removed through sedimentation or filtration.<sup>261,262</sup> Coagulation involves neutralizing the surface charge of the suspended particles, allowing them to aggregate.<sup>262,263</sup> Common coagulants such as aluminum-based salts (such as alum) and iron-based salts (such as ferric chloride) destabilize the MPs by reducing the repulsive forces between them, promoting initial aggregation.<sup>264,265</sup> Flocculation, on the other hand, involves the addition of polymers that bridge the destabilized particles, forming larger, more settleable flocs.<sup>262,266</sup> These polymers, often polyacrylamides, improve the aggregation of MPs, leading to improved removal efficiency.<sup>267</sup> A research by Awan *et al.*<sup>268</sup> illustrated that high dosages of coagulants can lead to the formation of a precipitate that enmeshes MPs, facilitating their removal. This is often referred to as sweep flocculation. Fig. 18 presents the coagulation–flocculation strategy for MP elimination.<sup>269</sup> Flocculation thus complements both adsorption and filtration methods, indicating that multi-stage or hybrid systems may provide the most scalable and sustainable solutions.

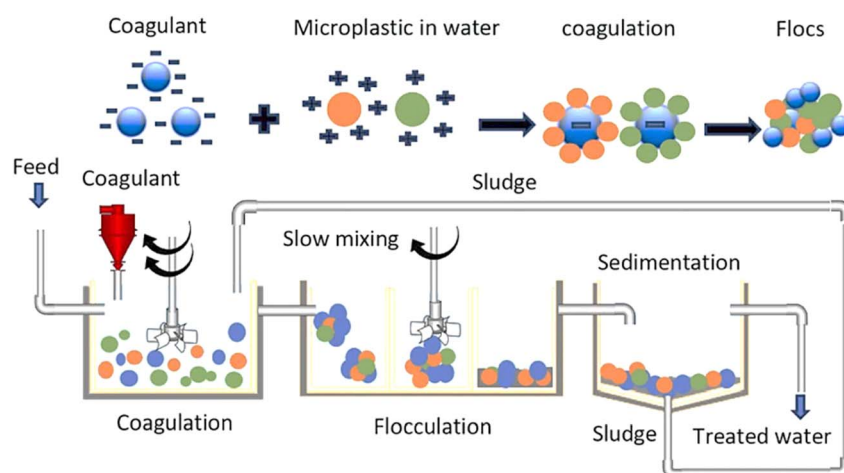


Fig. 18 Schematic overview of coagulation–flocculation in MP treatment. Reproduced with permission from ref. 269. Copyright 2024, Institution of Chemical Engineers. Published by Elsevier Ltd.





Table 6 Overview of treatment agents: removal efficiency and associated limitations

Coagulant/flocculant	Dosage (mg L <sup>-1</sup> )	MP type(s)	Removal percentage	Drawbacks	Ref.
Alum (aluminum sulfate)	50–100	HDPE, general MPs	HDPE: 86.6; PS: 67	High sludge, pH reduction	277 and 278
Ferric chloride	50–100	HDPE, PS	PS: 48	Corrosive, heavy metal risk, sludge management	277 and 278
PAC	0.4 mmol L <sup>-1</sup>	PS, PP, PVC, PA, PE, PU	PS: 97	Cost, residual Al, pH sensitivity	278
PAC + chitosan	—	PET	PET: up to 90; improved for others	Cost, limited scale, stability issues	279
Chitosan	10–40	PET	PET: up to 90	Higher cost, limited scale, pH sensitive	279
<i>Moringa oleifera</i> seed extract (MOCP)	100–150	HDPE, PE, pristine & weathered MPs	PE, HDPE: 70–87	Variable quality, shelf life, less effective for some MPs	277, 278 and 280
<i>Benincasa hispida</i> extract	100	HDPE, general MPs	HDPE: 83.7	—	277
Protein-coated sand (f-sand)	—	PE (weathered)	60	Lower efficiency, charge reversal issues	278 and 280

However, pH affects the surface charge of MPs and the speciation of the coagulants used in flocculation.<sup>270</sup> A research conducted by Ummalyma *et al.*<sup>271</sup> have shown that the surface charge of algal cells changes with pH, which affects flocculation efficiency. For instance, maximum flocculation efficiency (94%) was achieved by changing the medium pH from 8.5 to 12 through addition of NaOH. Similarly, in Yellow River water treatment, the pH influences the coagulation behavior and floc properties when using ferric-based coagulants. The pH also affects the structure of humic acid flocs, where flocs formed at pH 5 with low coagulant dosage exhibited a compact structure and high strength.<sup>272</sup> Ionic strength also influences the electrostatic interactions between MPs and flocculants by affecting the electrical double layer surrounding the particles.<sup>273</sup> Higher ionic strength can compress the double layer, reducing the repulsive forces between particles and promoting aggregation.<sup>274</sup> However, excessive ionic strength can lead to destabilization of the suspension and hinder flocculation.<sup>275</sup> Studies have shown that increasing ionic strength accelerates aggregation.<sup>274</sup> In a similar way, the ionic strength affects floc formation and growth in polymer-clay flocculation.<sup>273,275</sup> The effect of total hardness and ionic strength on coagulation performance has been investigated using titanium tetrachloride, showing that these parameters influence the removal of organic matter.<sup>276</sup>

The combined effect of pH and ionic strength determines the overall efficiency of flocculation. For instance, in aluminum sulfate-induced microalgae flocculation, changes in pH and ionic strength influence algal flocculation by altering the zeta potential of cells. This relationship is described by the classical DLVO theory, where cells with lower total DLVO interaction energy exhibit higher flocculation.<sup>270</sup> Table 6 provides a summary of coagulant and flocculant performance for MP removal, including dosage, target MP types, efficiency, and key limitations.

## 6. Adsorption mechanism

Adsorption mechanisms are complex and depend on several factors, including the properties of the MPs, the adsorbent

material, and surrounding environmental conditions.<sup>281–283</sup> Different MP types have varying chemical compositions and surface properties, which can affect their affinity for pollutants like 4-nonylphenol.<sup>284</sup> Additionally, the surface area,<sup>284</sup> pore size,<sup>285</sup> surface charge,<sup>286</sup> and chemical functional groups of the adsorbent material determine its capacity to bind MPs. For example, the linear mycelium of *Ganoderma lucidum* shows high adsorption efficiency due to its morphology, achieving equilibrium adsorption capacities of 102.92 mg g<sup>-1</sup> for polyethylene, 156.39 mg g<sup>-1</sup> for polypropylene, and 311.76 mg g<sup>-1</sup> for polystyrene.<sup>287</sup> Narwal *et al.*<sup>282</sup> showed that interactions between  $\pi$  electrons in aromatic rings can improve if both the MP and adsorbent have aromatic moieties. Another study by Yuan *et al.*<sup>288</sup> showed that 3D RGO has adsorption capacity of polystyrene MPs, showing a distinct porous spatial structure beneficial for adsorption. As shown in Fig. 19, various physicochemical interactions contribute to the adsorption of contaminants by MPs.<sup>289</sup>

To understand the interaction between absorbers and adsorbents, adsorption isotherm models are important. They provide a mathematical description of the adsorption process, essential for predicting the behavior of adsorption systems. The Langmuir isotherm is one of the models that assume monolayer adsorption onto a homogeneous surface with identical adsorption sites and no interaction between adsorbed molecules.<sup>290–292</sup> The Langmuir model is expressed as:

$$q_e = \frac{q_m K_L C_e}{1 + K_L C_e}$$

where  $q_e$  is the equilibrium adsorption capacity (mg g<sup>-1</sup>),  $q_m$  is the maximum adsorption capacity (mg g<sup>-1</sup>),  $K_L$  is the Langmuir adsorption constant (L mg<sup>-1</sup>), and  $C_e$  is the equilibrium concentration of the adsorbate in solution (mg L<sup>-1</sup>)

Several studies have utilized the Langmuir isotherm model for adsorption processes. For example, the adsorption of Cd and landfill leachate on wood-derived biochar was analyzed, and the Langmuir model was one of the isotherms applied.<sup>293</sup> Similarly, the adsorption of nitrate onto solid olive mill residues was examined using the Langmuir isotherm, yielding the equation  $y = 0.007x + 0.4576$  with  $R^2 = 0.9787$ , indicating a good fit.<sup>294</sup> In



another study, the adsorption of benzene onto grass-derived biochar was modeled using the Langmuir isotherm, resulting in  $K_L = 0.008$ ,  $q_m = 238.9$ , and  $R^2 = 0.934$ .<sup>292</sup> Another mathematical model is the Freundlich isotherm, which is an empirical model that describes adsorption on heterogeneous surfaces, where adsorption energy varies across different sites.<sup>290–292</sup> The Freundlich model is expressed as:

$$q_e = K_F C_e^{1/n}$$

where  $q_e$  is the equilibrium adsorption capacity ( $\text{mg g}^{-1}$ ),  $K_F$  is the Freundlich adsorption constant,  $C_e$  is the equilibrium concentration of the adsorbate in solution ( $\text{mg L}^{-1}$ ), and  $n$  is the Freundlich exponent, reflecting the intensity of adsorption or surface heterogeneity.

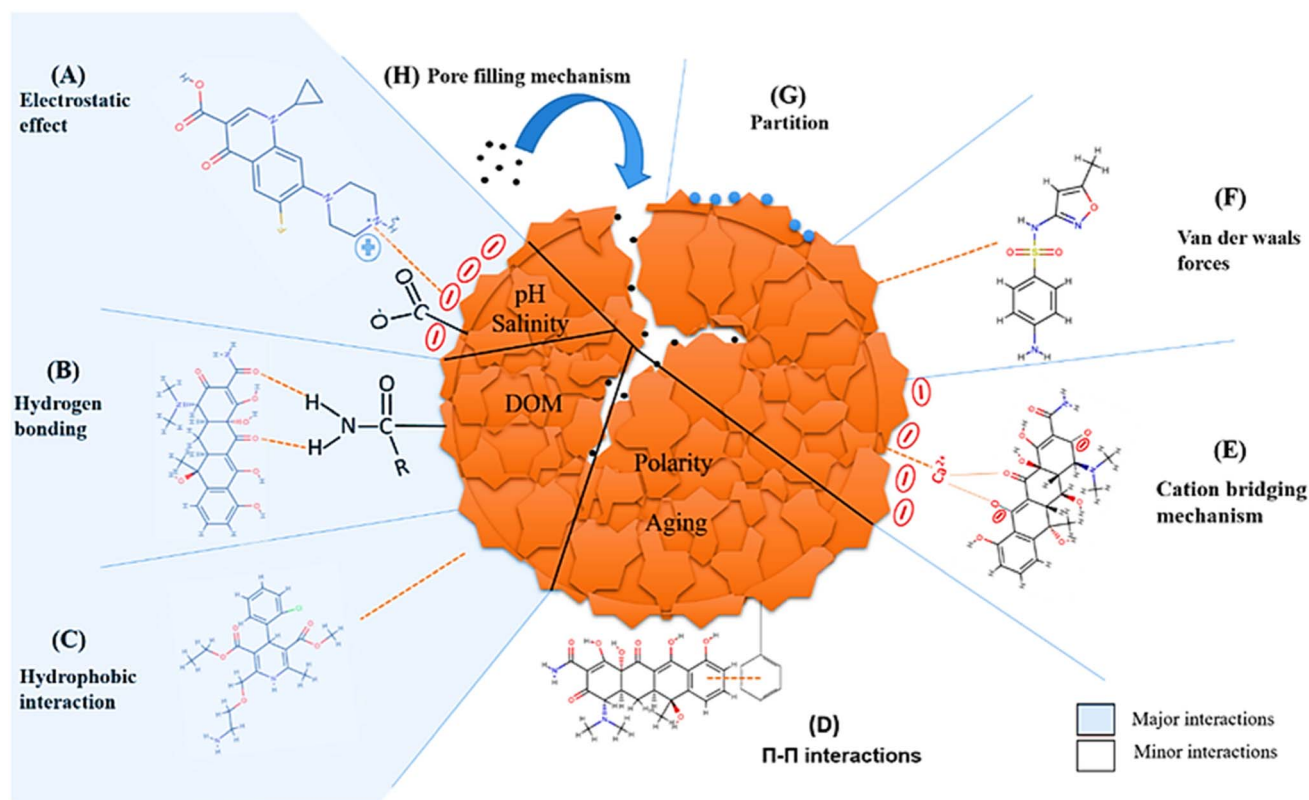


Fig. 19 Illustration of interactions involved in the adsorption of pollutants onto MP surfaces, including electrostatic forces, hydrogen bonding, hydrophobic interactions,  $\pi$ - $\pi$  stacking, cation bridging, van der Waals forces, partitioning, and pore-filling. Published under the CC-BY License.<sup>289</sup> Copyright 2022, The authors. Springer Nature Switzerland AG.

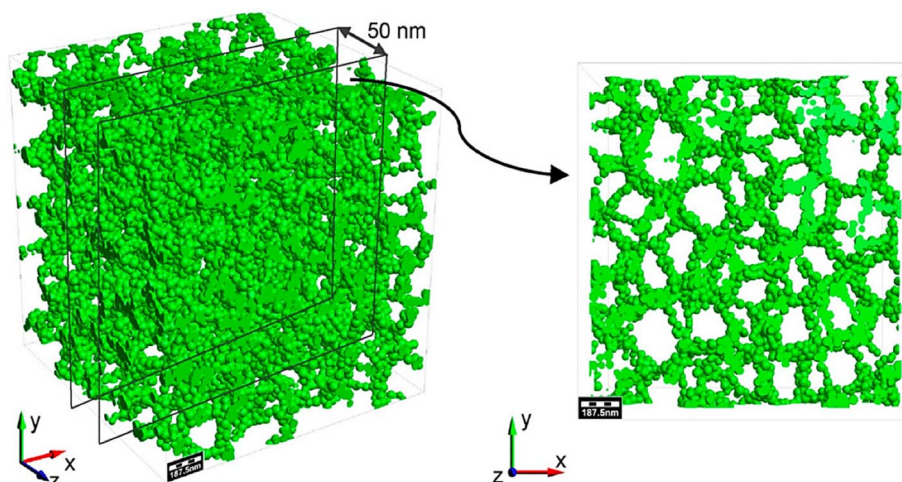


Fig. 20 Aerogel structure comprises atom or nanoparticle clusters forming a solid network (green), with interstitial spaces representing its pores. Published under the CC-BY License.<sup>303</sup> Copyright 2018, The authors. Published by MDPI.



Zand and Abyaneh<sup>293</sup> found that the adsorption of Cd from landfill leachate and the adsorption of nitrate onto solid olive mill residues fit the experimental data with the equation  $y = 0.3888x + 2.5552$  and  $R^2 = 0.9816$ .<sup>294</sup> The adsorption of benzene onto grass-derived biochar was also modeled using the Freundlich isotherm, resulting in parameters  $K_F = 8.065$ ,  $n = 1.778$ , and  $R^2 = 0.963$ .<sup>292</sup> While Langmuir and Freundlich isotherms are commonly used, other models such as the Temkin, Redlich–Peterson, Sips, Toth, Dubinin–Radushkevich, and Koble–Corrigan isotherms can also be employed to describe adsorption processes, especially when the assumptions of Langmuir and Freundlich models are not fully met.<sup>291,292,295–297</sup>

## 7. Synergistic application of NC and biopolymers

Biopolymers combined with NC form composite materials with enhanced properties for MP removal.<sup>298</sup> For instance, the integration of NC and chitosan results in synergistic effects, increasing overall adsorption capacity and selectivity for MPs.<sup>299</sup> Wu *et al.*<sup>300</sup> developed a sustainable, environmentally adaptable adsorbent through the supramolecular self-assembly of chitin and cellulose. This biomass-based fibrous framework exhibited excellent adsorption performance across various MP types. Similarly, Mok *et al.*<sup>301</sup> demonstrated that reusable polyvinyl alcohol/chitin/NC biopolymer composite films, crosslinked with maleic acid, effectively removed both MPs and methylene blue dye. Another renewable and abundant biopolymer, starch, when combined with NC, shows improved mechanical properties and enhanced MP capture and retention capabilities.<sup>302</sup>

Additionally, composite aerogels made from activated carbon and NC blended with cross-linked biopolymers such as hydroxypropyl methylcellulose and chitosan have been evaluated for their ethylene gas adsorption capacity, as illustrated in Fig. 20.<sup>303</sup> Although this study focused on gas removal, the materials' high porosity and surface area suggest strong potential for MP adsorption applications as well.<sup>298</sup> The synergistic application of NC and biopolymers is commonly used for those MPs that cannot be eliminated through the sole biopolymer or NC. Fig. 21 shows the removal efficiency of NC, biopolymer, and synergistic materials.<sup>304</sup>

Despite the advantages, NC and biopolymers face compatibility challenges. Many biopolymers are hydrophilic, while some NC modifications can introduce hydrophobic characteristics, leading to poor compatibility.<sup>179</sup> Poor interfacial bonding can degrade the mechanical properties of the composite material, reducing its overall effectiveness in MP removal.<sup>305</sup> Additionally, NC has a tendency to aggregate due to strong hydrogen bonding between individual fibers.<sup>306</sup> Uniform dispersion on the NC matrix is important for achieving optimal functional properties.<sup>307</sup> Incomplete dispersion can lead to stress concentration points and reduced performance of the composite material.<sup>308,309</sup>

## 8. Sustainability of NC and biopolymer-based treatment systems

The advancement of renewable, sustainable, biodegradable, environmentally sound, safe, and sustainable materials depends on the fabrication of NC-based products.<sup>124</sup> NC can be

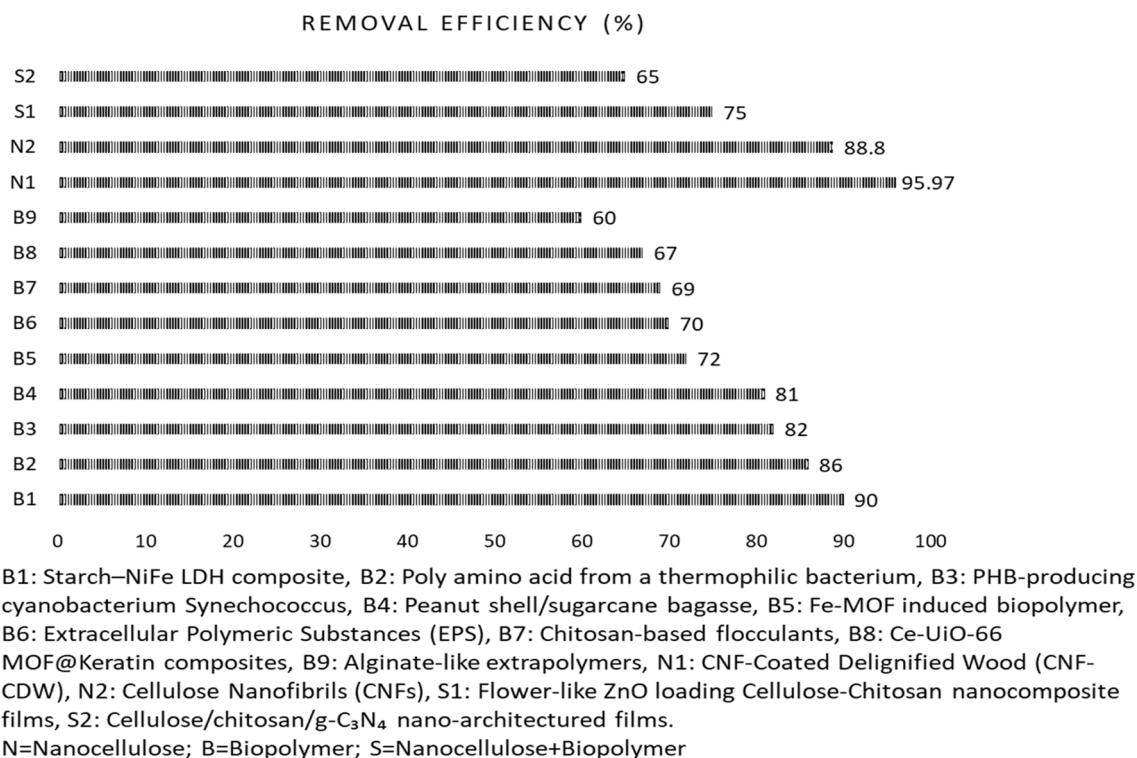


Fig. 21 Removal efficiency comparison across NC, biopolymer, and synergistic materials (created with MS Excel).





created from bacteria, plants, or biomass using relatively straightforward, scalable, and effective isolation methods. The most common nanostructured component found in wood, cotton, hemp, flax, and other plant-based materials is cellulose, which is biodegradable and non-toxic. One can extract NC from renewable resources *via* mechanical, chemical, enzymatic, or a combination of these methods. Source, isolation method, and possible surface changes all affect the characteristics of NC.<sup>310</sup> Ali *et al.*<sup>311</sup> demonstrated that materials based on NC work well as adsorbents for pollutants in water. By enriching the surface of materials like NC, water contaminants can be eliminated. Water treatment methods, especially adsorption, are required to meet the growing demand for clean water and to mitigate pollution caused by bottled water waste. However, adopting the right adsorbent is an important issue for water treatment since it necessitates striking a careful balance between natural components, high removal effectiveness, ease of separation from treated water, environmental friendliness, and economic viability. As a result, there is a lot of interest in creating innovative adsorbent films, such as bacterial NC-clay film, for use in water treatment.<sup>312</sup> NC is a prime contender to enable novel, sustainable, and affordable water purification technologies because of its special physicochemical characteristics, which include high surface area, several functions, nontoxic and biodegradable characteristics, and scaffolding stability and versatility.<sup>313</sup> Academic research discussing the cutting-edge advantages of using NC-based membrane filtration processes has grown exponentially, reflecting the predicted success of NCs in the field of improving water quality through membrane filtration processes.<sup>314</sup> This move to a bio-based solutions solve our worldwide plastic pollution problem and offers a chance for a circular economy, which would decrease reliance on fossil fuels and create a cleaner environment.<sup>315</sup>

In the case of biopolymers, the most prevalent kind of biobased polymer, polyhydroxyalkanoate (PHA), is made by a variety of microorganisms and acts as a carbon and energy storage medium.<sup>316</sup> PHA is a viable substitute for non-biodegradable petro-based plastics due to its biodegradability and advantageous material properties.<sup>317</sup> Prior research explored the ability of synthetic biomaterials to absorb or immobilize pollutants, as well as their adsorption capabilities for eliminating a variety of impurities and enhancing water quality.<sup>318</sup>

However, carbon footprint analysis plays a crucial role in assessing the overall environmental impact of NC and biopolymer production and application.<sup>319,320</sup> This analysis involves quantifying greenhouse gas emissions throughout the entire life cycle, from raw material extraction to end-of-life disposal.<sup>321,322</sup> In this context, particular attention should be given to electricity and chemical consumption, which are critical sub-aspects influencing carbon footprint growth.<sup>319</sup> To enhance sustainability, the choice of disposal methods for NC and biopolymer-based products is also significant.<sup>323</sup> Therefore, sustainable end-of-life management should align with circular economy principles, emphasizing the safe and economical recycling and reuse of composite plastics.<sup>324</sup> Additionally, employing energy-efficient manufacturing processes, such as

chemo- and bio-mechanical methods, can substantially reduce the energy requirements for NC production.<sup>325</sup>

Moreover, several Life Cycle Assessment (LCA) studies have compared synthetic polymers with biopolymer/NC-based materials to evaluate environmental trade-offs.<sup>148</sup> For instance, the LCA of garbage bags made from polyethylene (PE), biomass polyethylene (Bio-PE), and poly(butylene adipate-co-terephthalate)-starch blends (PBAT/starch) illustrates the potential benefits of bioplastics in waste management scenarios.<sup>326</sup> Supporting this, a review of LCA studies on NC-based adsorbents highlights their effectiveness in environmental remediation applications.<sup>327</sup> Furthermore, an LCA conducted by Hervy *et al.*<sup>328</sup> confirms that NC materials typically exhibit a lower environmental footprint, particularly concerning resource consumption.

## 9. Regulatory framework and policies

Mitigating MP contamination in emerging nations requires an understanding of the laws and policies governing plastic waste in the coastal environment.<sup>329</sup> Many countries implemented laws and regulations to reduce plastic pollution or are in the process of engaging in stakeholder discussions, as demonstrated in Table 7. These laws and regulations differ in their approach (*i.e.*, traditional viewpoint (top-down) methods, market-oriented approaches, and voluntary efforts) because African nations have different customs for the consumption, manufacture, and disposal of plastics.<sup>330</sup> Aquatic systems were mostly exposed to MPs from WWTPs. However, a sustainable governance framework—such as the one proposed by the European Parliament in its review of the Urban Waste Water Treatment Directive (TA/2019/0071)—is useful to ensure the removal of MPs at WWTPs. This framework also calls for the adoption of multiple technologies along the value chain.<sup>331</sup> To control the growth of oyster aquaculture, the Chinese Taipei Shallow Sea Oyster Aquaculture Management Autonomous Regulations have been implemented. Specific clauses of the regulation encourage oyster farmers to use non-styrofoam buoys by providing subsidies, even though its main purpose is to handle the oyster aquaculture zoning system. The government took this step to stop styrofoam buoys from shedding MPs.<sup>332</sup> A regional action plan was set up by HELCOM in 2015 to address marine litter and secondary MPs by 2025. By enhancing stormwater management and increasing the number of WWTPs, assessing primary sources and legal mechanisms to address them, promoting MP-free formulae, putting certification programs (like EU Ecolabel and Blue Angel) into place, encouraging no littering policies, supplanting primary MPs in personal care products, and increasing public awareness, this plan suggests ways to combat primary MPs.<sup>333</sup> To lessen the negative environmental effects of plastic waste, the European Union has established in place an extensive set of policies.<sup>334</sup> The European Union has banned single-use plastics and set broadened manufacturer responsibility and recycling standards for its member states.<sup>335</sup> In Vietnam, several measures accompanied by particular laws are used to reduce the diffusion of



**Table 7** National strategies and regulations aimed at mitigating MP pollution, including plastic categories, policy actions, implementing countries, and targeted environmental goals<sup>337</sup>

Plastic category	Policy	Country	Goal
Aquatic MPs	Microbead-free waters act 2015	USA	<ul style="list-style-type: none"> <li>• Ban the production and sale of wash-off cosmetic products</li> </ul>
Plastics	The break free from plastic pollution act 2023		<ul style="list-style-type: none"> <li>• Shift the financial responsibility of plastic waste management to producers of plastics</li> <li>• Ban single use of plastic products</li> <li>• Prohibit the export of plastic waste</li> </ul>
Aquatic MPs	Circular economy law (waste prevention and management) 2018	France	<ul style="list-style-type: none"> <li>• Ban cosmetic products containing plastic particles</li> </ul>
Microfibers, microbeads	Draft law on combating plastic pollution (adopted 2022)		<ul style="list-style-type: none"> <li>• Regulate the loss and leakage of industrial granules, prohibit intentional usage of microbeads in detergent, and provide impact assessment of plastic fibers on the textile industry</li> </ul>
Larger plastics	Single-use plastics prohibition regulations (2022)	Canada	<ul style="list-style-type: none"> <li>• Prohibit the manufacture, importation, and distribution of single-use plastic products</li> </ul>
Aquatic MPs	Microbeads in toiletries regulations (2017)		<ul style="list-style-type: none"> <li>• Reduce the amount of plastic microbeads entering Canadian freshwater and marine environments</li> </ul>
Larger plastics	Plastic bag control and management regulations (2018)	Kenya	<ul style="list-style-type: none"> <li>• Reduce usage, manufacture, and importation of plastic bags</li> </ul>
	The wildlife conservation and management act 2020		<ul style="list-style-type: none"> <li>• Ban single-use plastic products</li> </ul>
Aquatic MPs	The plastic reduction and circular economy act 2021	Australia	<ul style="list-style-type: none"> <li>• Ban the distribution of wash-off personal care products</li> </ul>
Aquatic MPs	Waste minimization act through waste minimization (microbeads) regulations 2017	New Zealand	<ul style="list-style-type: none"> <li>• Prohibit plastic beads as an ingredient in personal care products</li> </ul>
Larger plastics, aquatic MPs	Environmental permitting regulations 2018	UK	<ul style="list-style-type: none"> <li>• Ban cosmetics and cleaning products containing microbeads</li> <li>• Charge levies on single-use carrier bags</li> <li>• Ban single-use plastics</li> </ul>
Aquatic MPs	The environmental protection (microbeads) (Northern Ireland) regulations 2018	Northern Ireland	<ul style="list-style-type: none"> <li>• Prohibit the use of plastic beads</li> </ul>
Aquatic MPs	2019 industrial catalogue	China	<ul style="list-style-type: none"> <li>• Ban the production and sale of cosmetics containing microbeads</li> </ul>
Aquatic MPs	The single-use plastics directive 2019	EU	<ul style="list-style-type: none"> <li>• Target eradicating the 10 most common single-use plastics found on the beaches and seas in Europe</li> </ul>
Aquatic plastics	Clean up	The ocean cleanup	<ul style="list-style-type: none"> <li>• Developing technologies to reduce plastics in the ocean by 90% by 2040</li> </ul>
Aquatic MPs, larger plastics	Thailand ministry of public health (2019) through roadmap on plastic waste management (2018–2030)	Thailand	<ul style="list-style-type: none"> <li>• Ban the production, sale, and distribution of cosmetics with microbeads as an ingredient</li> <li>• Ban single use of plastics</li> </ul>
Plastics waste	Environmental management act (the commodities act decree)	The Netherlands	<ul style="list-style-type: none"> <li>• Control packaging and consumer products</li> </ul>
Aquatic MPs	The microbeads (prohibition) act 2019	Ireland	<ul style="list-style-type: none"> <li>• Regulate single-use plastics</li> <li>• Ban the use of plastic beads in households and industrial cleaning products</li> </ul>
Larger plastics	Plastic waste management (amendment) 2022	India	<ul style="list-style-type: none"> <li>• Phase out single-use plastics</li> </ul>
Larger plastics	The German ordinance on single-use plastics 2021	Germany	<ul style="list-style-type: none"> <li>• Reduce the impact of plastic waste on the environment</li> <li>• Ban some single-use plastic products</li> </ul>
Larger plastics	The national environmental management waste act 2008 (amended 2014) through national waste management strategy 2020	South Africa	<ul style="list-style-type: none"> <li>• Reduce production of single-use plastics destroying the marine environment</li> </ul>
Larger plastics	Tax/levies on single-use plastics	Wales, Ireland, Scotland	<ul style="list-style-type: none"> <li>• Discourage the single use of plastic products to reduce waste</li> </ul>
Larger plastics	The single-use foodware and litter reduction ordinance (2022)	Berkeley, California	<ul style="list-style-type: none"> <li>• Reduce plastic waste in the environment</li> </ul>





MPs into the aquatic environment, particularly the riverine environment.<sup>336</sup>

In addition to legislative measures, voluntary ecolabeling schemes such as the EU Ecolabel and Blue Angel play a significant role in promoting environmentally responsible technologies. These certifications assess products based on their entire life cycle—from raw material sourcing to end-of-life disposal.<sup>338</sup> NC and biopolymer-based MP removal systems align with the criteria of these ecolabels, as they are derived from renewable resources, are biodegradable, and pose minimal toxicity risks to ecosystems.<sup>339,340</sup> Also, these materials ensure that they break down naturally in the environment, minimizing long-term pollution.<sup>339</sup> NC and biopolymers are generally less toxic compared to synthetic materials.<sup>341</sup> Their production can also be optimized to reduce energy use and waste generation,<sup>342</sup> thereby supporting circular economy goals.<sup>343</sup> Importantly, alignment with such ecolabels may unlock policy-driven incentives, including subsidies, grants, or preferential procurement in public and private sectors. For example, ecolabel-certified technologies can benefit from EU-level funding schemes (e.g., Horizon Europe, LIFE Programme) and national green procurement strategies, encouraging broader adoption of natural MP removal methods.<sup>344,345</sup>

## 10. Conclusion and future outlook

Microplastics (MPs) are commonly found in water, posing risks to the environment and human health, which makes it crucial to find practical and sustainable methods for their removal. This review highlights the key role that natural compounds (NCs) and biopolymers can play in water treatment due to their unique properties. NC has a large surface area that effectively captures MPs and is biodegradable, making it an eco-friendly choice compared to synthetic materials. It works by sticking to, filtering, and clumping together pollutants. Biopolymers also help gather and separate MPs. When NC and biopolymers are used together, they often enhance the effectiveness of each other, resulting in better removal of MPs than when used separately. Even with these improvements, there are still significant areas of research that need to be addressed before these materials can be effectively utilized in real-life situations. First, improving the production, surface treatment, and blending of nanocomposites and biopolymers is essential to ensure strong performance across different types of MPs, particularly those with varying shapes and chemical compositions. It is essential to modify the surface properties of these materials to enhance their interactions with specific substances. In addition, limited research has been conducted on their long-term environmental behavior and potential ecological effects after use. Comprehensive assessments of their entire life cycle and environmental impact are necessary to ensure safe and sustainable large-scale application.

There is a significant gap in research regarding the identification, quantification, and understanding of nanoplastics. These tiny plastic particles can enter into our bodies and may be harmful. Unlike larger MPs, nanoplastics are more challenging to study because they are small, contain a mixture of materials,

and their signals can be weak and difficult to detect. Some methods, like Raman spectroscopy and nanoscale FTIR, can help, but they need to be improved and made more sensitive. Clear guidelines are needed to ensure that results can be replicated and compared across different studies. Additionally, developing improved tools for imaging and analyzing nanoplastics in complex environments is crucial for accurately tracking these particles. Many studies to date have focused on specific areas of the environment, such as oceans, rivers, land, or the air, without adequately considering how MPs and nanoplastics move between these areas. Future research should adopt a broader perspective to understand how these particles travel and transform throughout the environment. This means investigating how MPs move from wastewater systems to soil when sludge is applied, from surface waters to living organisms, and through the interactions between air, soil, and water under different environmental conditions. It is also important to conduct experiments that consider various factors, such as pH, salt levels, organic matter, and temperature, to understand how these factors influence the removal of MPs and their behavior. Studies in regions with poorly managed plastic waste are critical, as these areas are major spots for MP pollution. Conducting pilot and large-scale studies is essential to bridge the gap between laboratory findings and real-world applications. Such studies should involve collaboration among researchers, industry professionals, policymakers, and local communities to jointly identify problems, design treatment systems, and share knowledge. Working together in this way not only makes the technology more relevant but also helps the community accept it and ensures it aligns with policies. It is also essential to test these treatment systems on a larger scale with various types of wastewaters, including those from cities and industries, to determine if they are practical and cost-effective.

The use of nanocellulose and biopolymer technologies in wastewater treatment plants presents exciting possibilities; however, careful consideration is required regarding their integration with existing systems. There are challenges, including the risk of clogging, increased operational complexity, and managing or recycling materials. A modular approach, which may include additional filtration steps utilizing these new materials, could provide flexibility while ensuring the primary treatment processes run smoothly. Future research should focus on developing scalable production methods, enhancing material durability under real wastewater conditions, and implementing long-term performance monitoring. Additionally, emerging technologies such as machine learning could enhance traditional treatment processes, potentially optimizing the management of wastewater through the use of these advanced materials.

The implementation of new ideas in real-world settings requires addressing several challenges, including financial constraints, technological limitations, and regulatory hurdles. Some of these challenges involve the high costs of producing bioplastics, differences in the sources of materials such as biomass, energy-intensive processing steps, and the lack of clear safety standards for waste materials. Policy-driven plans and cross-sector collaboration are essential to address these



issues. Increasing production by utilizing eco-friendly chemistry, leveraging agricultural waste as raw materials, and exploring methods to reduce energy consumption in production can help lower costs and make processes more sustainable. At the same time, regulatory agencies should create clear guidelines to assess the safety and effectiveness of bioplastics and related technologies in environmental applications. Nanocellulose and biopolymers hold great potential for removing microplastics; however, effective utilization requires focused attention on several key areas. This includes improving materials for enhanced performance, developing more effective tools for detecting tiny plastics, and conducting real-world pilot studies to assess their effectiveness. It is also necessary to implement innovative monitoring systems that provide quick feedback during use. On the policy front, reducing microplastics can be achieved by linking wastewater management with solid waste processes, ensuring high standards in recycling, regulating new sources of microplastics, and raising public awareness. Addressing these challenges will be essential for making these solutions practical and environmentally friendly in tackling microplastic pollution in our water systems.

## Author contributions

Sayam conceived the study, developed the methodology, wrote the original draft, created visualizations, and performed proofreading; Tarikul Islam conceived the study, developed the methodology, contributed to writing, editing, and reviewing the manuscript; conducted the investigation; prepared visualizations; supervised the project; curated data; validated results; and provided resources; Tasnim Hanan Tusti contributed to writing the original draft, prepared visualizations, edited the manuscript, curated data, and conducted formal analysis; Joyjit Ghosh contributed to writing and editing the manuscript and assisted with the literature review.

## Conflicts of interest

The authors declare that they have no conflict of interest.

## Data availability

All data related to the research are included in the manuscript.

## Acknowledgements

The authors declare that they did not receive any internal or external funding for the research.

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