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Manuscript title: Microplastics in aquatic environments – a review of recent advances

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Abstract

Global production and usage of plastics have skyrocket to 368 million tons in 2019, resulting in increasing amounts of plastic waste concentrating in our natural and urban ecosystems (especially rivers and oceans), via landfill, incineration, or illegal disposal. As highlighted herein, due to the production and degradation of larger plastics, micro and nano plastics are introduced to these ecosystems, causing detrimental impact to plants and animals, including humans, through accumulation in living systems. Though toxicity or health impact are not clearly established, long term accumulation of microplastics in living systems can lead to impact on health of such systems. Critically, this review explores state-of-the art physical, chemical, and biological methods to remove and destroy new and legacy microplastics in aquatic ecosystems (natural and urban). Currently, there are no standardised, accepted, and cost-effective methods for complete removal of microplastics from these aquatic ecosystems. Gaps in knowledge and recommendations for future research to help inform practice and legislation are highlighted. A key consideration highlighted through the review is that microplastics cycle through ecosystems – natural and engineered, these do not operate in siloes and waste from treatment processes could be a conduit for (unintended) recontamination of microplastics. Hence there is a need to take a whole systems approach when developing innovative removal or destructive solutions and, ultimately, reducing plastic use remains the best option to best safeguard future environmental and public health.

Introduction

Since the pioneering synthesis of polystyrene in 1839 to the present day, plastics have undergone a phenomenal transformation. Substantial advances in plastic properties and diversification (Alimba and Faggio, 2019; Chen and Patel, 2012; Kroon *et al.*, 2016) have led to a dramatic increase in the world production of plastics from 1.5 million tonnes in 1950 to a staggering 368 million tonnes in 2019 (Alimba and Faggio, 2019; Plastics Europe, 2020). Plastics deliver many benefits and production is expected to double in the coming decades as plastics continue to diversify (MacArthur et al, 2016). However, with this explosion of plastic production and use comes a growing environmental concern: waste plastic pollution.

The construction, building, and packaging industries are the biggest consumers of plastic, with polypropylene (PP), polyethylene (PE), and polyvinyl chloride (PVC) being the most widely used products (Plastics Europe, 2020). Plastic use in these industries is anticipated to continue to increase in coming decades and additives used in plastic manufacture to obtain specific physicochemical properties (e.g.: anti-oxidising and anti-ageing) can include hazardous chemicals, adding complexity to plastic pollution. Often plastics are only cycled through the economy for a short period of time before most (95%) of their value is lost (MacArthur et al, 2016). Local and global initiatives are in place to improve collection and recovery infrastructure (incorporating circular economic principles into the plastics sector) but leakage of plastic remains a significant challenge (Hettiarachchi and Meegoda, 2023).

Plastics that evade collection/recycle systems can accumulate in critical natural ecosystems and urban infrastructure, resulting in substantial environmental and economic costs.

For instance, despite having advanced collection systems, 170,000 tonnes of plastics still escape into the ocean each year in the United States and Europe (Jambeck et al, 2015). Therefore, in addition to preventing unintended plastic disposal, there is a need for innovative solutions to remove or degrade plastics in a biologically benign way when they unintentionally enter the environment. Currently available biodegradable plastics do not truly address this; they often only decompose under controlled conditions (i.e. industrial composting) and additive-mediated fragmentation techniques (e.g. oxo-fragmentation) often lead to fragmentation and the creation of additional microplastics in our oceans (MacArthur et al, 2016).

Plastic debris may exist in various shapes and morphologies including fragments, fibres, pellets, beads, films, which significantly affect their environmental fate (Li *et al.*, 2020). Based on their average size, plastics can be divided into macroplastics (>25 mm), mesoplastics (5-25 mm), microplastics (<5 mm), and nanoplastics (<0.1 μm) (Boyle and Örmeci, 2020).

Microplastics (MPs) and nanoplastics (NPs) have become pervasive in our environments due to anthropogenic activities, wind action, flowing rivers, and ocean currents. Microplastics are emerging as micro-pollutants that pose adverse health effects to aquatic life. Their persistence and ability to adsorb organic pollutants from water results in transportation and bioaccumulation of pollutants in aquatic environments (Bandow et al, 2017). Low trophic level animals and other organisms can ingest MPs and NPs, leading to irreversible accumulative damages to the food chain, where over 690 marine species have already encountered plastic debris (Burgess and Ho, 2017). Additionally, when MPs and NPs are degraded (physically or

chemically) the adsorbed organic pollutants are released back into the environment resulting in unexpected mixtures of persistent organic contaminants even in pristine environments (Bandow *et al.*, 2017). Hence MPs and NPs present both direct and indirect environmental risks, a major concern for environmentalists and policymakers alike (Meegoda and Hettiarachchi, 2023).

Legislation to control plastic contamination is being developed, in response to the growing threat of plastic pollution. The United States passed the Microbead-Free Waters Act (MFWA) in 2015, prohibiting the manufacture, packaging, and distribution of cosmetics containing plastic microbeads. The EU has also taken steps to address MPs, requesting the European Chemical Agency (ECHA) to evaluate regulatory actions throughout the region. In 2019, ECHA proposed an extensive restriction on "*microplastics in products placed on the EU/EEA* (European Union/ European Economic Area) *market to avoid or reduce their release to the environment*," essentially banning the addition of MPs to products such as cleansers, cosmetics, fertilizers, and others (Metzler and Simbeck, 2022). However, these are just some of the sources of MPs and such legislation is only addressing the release/leakage of new MPs into the environment. Management of the legacy MPs and NPs already within our natural and urban aquatic environments is critical and requires innovative removal and destruction approaches. Consequently, this review aims to: (*i*) critically evaluate the state-of-the art of currently available engineered separation and degradation technologies, (*ii*) outline current gaps in knowledge, and (*iii*) suggest useful recommendations for further advances in removal of MP and NP from aquatic environments (including freshwater, seawater, drinking and

wastewater).

Sources and occurrence of microplastics in aquatic environments

Whilst MPs are present in different media, they are most abundant in aquatic ecosystems, particularly oceans due to the large body of water. MPs may be introduced into the environment from diverse sources, with specific plastic types having different origins. For instance, PE, PP, and polystyrene (PS) are produced from typical plastic products (e.g. plastic cutlery), polyamide (PA) and polyethylene terephthalate (PET) originate from the textile industry. Polyethersulfone (PES) is produced when tires and plastic particles in roads are crushed or fragmented (Liu *et al.*, 2021). Sources are typically classed as primary (where MPs are directly discharged into the environment) or secondary (where bigger particles fragment and degrade into micro-particles in the environment (Yusuf *et al.*, 2022). Understanding sources and the contamination route is important to mitigate exposure and manage plastic pollution in aquatic systems.

Freshwater (rivers, lakes, reservoirs, groundwater) is the main source of raw water for agriculture, industry, energy production, and human consumption, but in areas where freshwater is scarce, seawater may be used. These sources are vulnerable to MP contamination from agricultural and industrial activities, as well as animal farming discharges (Shen *et al.*, 2020) within the context of drinking water. The abundance of microplastics in freshwater environments varies greatly (from several to millions of tons) due to location, natural conditions, anthropogenic factors and the use of different quantification methods.

Primary sources

Plastics that are manufactured to be of a microscopic size for commercial use are defined as primary MPs. These plastics are typically used in facial-cleansers and cosmetics, while their use in medicine as vectors for drugs is also reported. In addition, the textile industry is a significant source of primary MPs in aquatic systems, with microfibers released through physical agitation in washing machines, contaminating the environment through sewage systems, or settling on the surface of water bodies after being released to the air (Zhang *et al.*, 2021). Similarly, resin powder and pellets, discharged to air during the manufacturing of plastic, have been reported to settle in oceans (Yusuf *et al.*, 2022). Mortula *et al.*, 2021 demonstrated that many plastic materials found in landfills can leach primary MPs, especially polycarbonate (PC), possibly contaminating the groundwater. Manufactured MPs have even been detected in the influent of some drinking water treatment plants (DWTP). One study has shown that whilst the raw water sample from DWTP contained low concentrations of MPs, household drinking water contained greater concentrations, suggesting that the drinking water treatment or distribution systems (DWDS) may be sources of MPs contamination (Shen *et al.*, 2020).

Secondary sources and conduits

Secondary MPs are formed through the (unintentional) fragmentation of larger plastic particles. Secondary MPs may enter surface water environments through wastewater discharges, decomposition of plastic waste in the environment, abrasions of plastic products, and atmospheric deposition. This can occur when plastic products are manually crushed, subjected

to mechanical agitation or UV light, or chemical processes such as oxidation, causing degradation into smaller particles.

Drinking water is treated to a high standard and many MPs are removed, but wastewater treatment plants (WWTP) are reported as a major conduit of MPs in seawater and aquatic environments (Liu *et al.*, 2021; Murphy *et al.*, 2016). MPs in WWTP originate from sources such as textile microfibers, tire dust from roads, paint, microbeads in cosmetics and exfoliants (Yusuf *et al.*, 2022; Zhang *et al.*, 2021). Treatment in WWTP do not entirely remove MPs from the effluent and can cause degradation of MPs into smaller particles, which are then harder to remove during downstream treatment. For instance, Liu et al (2021) reported that the proportion of smaller particles in the effluent of WWTPs (81%-91%) was higher than that of the influent (65.0-86.9%). Conversely, MP removal has been reported to increase from 75% to 91.9% and >98% across the primary, secondary and tertiary (coagulation, ozonation, sand filters) stages of wastewater treatment. Despite this, the concentrations of MPs released into the environment could be substantial; for instance, Bayo *et al.*, 2020 reported wastewater treatment effluent ranges from 5900 MPs/m^3 to 3000 MPs/m^3. Murphy *et al.*, 2016 studied a UK wastewater treatment plant and showed that MP removal efficiency exceeded 98%, yet despite this high efficiency the effluent still released 65 million MPs into the receiving water body. Additionally, a significant proportion of MPs may remain in sludge (and other waste products from waste and drinking water treatment plants). Sludge is often disposed on land and return MPs to the natural environment (Zhang *et al.*, 2020a), including aquatic bodies where they remain in high concentrations. Indeed, MPs in soil ecosystems can be traced back to

wastewater treatment plants (sewage sludge), as well as solid waste (leaching from landfills) and organic fertilizers (Zhang et al, 2021).

MP loads could increase throughout the drinking water system if the treatment, distribution and point-of-use infrastructure (which includes various plastic components) act as sources of MPs (Ding *et al.*, 2021; Shruti et. al., 2020). For instance, Ding *et al.*, 2021 demonstrated that membrane filters were effective in removing MPs but those same organic membranes can be a source of MPs/NPs. No whole-system evaluation has been undertaken to determine the contributions of different water system components to MP loading of drinking water.

In summary, MP contamination of aquatic environments can occur from the synthetic MPs as well as use and disposal of plastics. Hence there is a need to better understand the consequences of the potential return conduit on MP accumulation in soils, watercourses and produce from land animals, which present other exposure pathways.

Exposure and toxicity of microplastics in aquatic environments

MPs have become a major environmental concern due to their persistence in the environment and the potential harm they can cause to organisms. Health risks from MPs in any environment may stem from physical, chemical or microbiological characteristics or behavior of particles. While several studies have investigated the chronic toxicity of MPs, but there is limited information on their acute fatal effects (Li *et al.*, 2018). Evidence on the exact implications is lacking in part due to difficulty in assessing long-term exposure and compounding effects, in combination with the diverse and complex blend of chemicals that comprise MPs and NPs.

Toxicity of MPs and NPs depends on their chemical structure and additives used, such as polymerization linkage (Sussarellu *et al.*, 2016). Polystyrene MPs in particular can accumulate in blood and are recognized as causing reproductive disruption in marine filter feeders (Law *et al.*, 2014). Critically, most MPs and NPs are resistant to bio-/photo- degradation in the natural environment, making them a growing environmental threat.

Direct exposure of MPs

Studies on MP toxicity have mainly focused on their ecotoxicological impacts in natural environments, with little attention given to engineered ecosystems such as drinking water treatment and distribution systems. Risk is dependent upon toxicity and exposure; if drinking water is contaminated it has the potential to impact vast numbers of people. However, drinking water is of low risk to public health within the context of MPs (WHO, 2019) and the main source of MP human intake (12K – 204K MPs per person per annum) is reported as being from food (Kirstein *et al.*, 2021). MPs have been detected in a drinking water supply from source water to end point of use (Koelmans *et al.*, 2019; Li *et al.*, 2018) but most studies have focused on MPs in freshwater sources used as influent for drinking water treatment. Koelmans *et al.*, 2021 and Kirstein *et al.*, 2019 found just 9 and 26 studies respectively, on MPs specifically in drinking water. The characteristics of MPs in drinking water systems are still debated, but fragments and fibers are reported as the dominant particle types (WHO, 2019) and concentrations are typically low based on the available evidence. For instance, the following concentrations were reported: 0.06±0.04 MPs/L from a Spanish drinking water system (Dalmau-Soler *et al.*, 2021), 0-8 MPs/m^3 in a study of a Swiss system (Negrete Velasco *et al.*, 2022) and <0.5 MPs/L in

potable water from a study of eight treatment works in England and Wales (Johnson *et al.*, 2020). It should be noted that quantification of MPs in freshwater systems is still primarily based on modelling with little empirical evidence.

Inconsistencies in studies of exposure to MPs from aquatic environments exist due to differences in sampling, isolating, and analysis methods (Koelmans *et al.*, 2019; Kirstein *et al.*, 2021; Oßmann, 2021) or unreliable data (Koelmans *et al.*, 2019). Koelmans *et al.*, 2019 proposed an assessment of quality across 50 studies of MPs in water sources, from which only four received a positive score with respect to quality assurance of sampling and analysis of MPs. Hence, there is a need for standardized methods before conclusions on MP toxicity and exposure as a consequence of water source, treatment and quality can be justified.

The daily intake of MPs in mineral water was estimated to be 40.1 µg/kg/bodyweight/day and 87.8 µg/kg/bodyweight/day for adults and children, respectively (Zuccarello *et al.*, 2019) and MPs have been detected in various human tissues (Wang *et al.*, 2019), with their adverse effects on human health increasingly recognized. The impacts of MPs on water quality and public health are not well studied. The greatest health risk is perhaps most likely from the smaller NPs than MPs because they can escape water treatment (Li *et al.*, 2021).

Indirect exposure of MPs

MPs may indirectly impact water safety by acting as “transport systems” for microorganisms (including potential pathogens) or other pollutant (Ateia *et al.*, 2020). Biofilm formation in some freshwater studies has illustrated the potential for MPs to act as a transport system of

pathogens, possibly increasing transfer of antimicrobial resistant genes. Whilst there is currently no evidence to suggest that the same would occur within operational drinking water systems, bench-top studies of stagnant drinking water bottles have shown to form biofilms on MPs (Chen *et al.*, 2021). Recent studies have shown that microbial communities associated with microplastics in river (Wang *et al.*, 2020a), estuary and marine waters (Wang *et al.*, 2019; Wang *et al.*, 2020b; Li. *et al.*, 2022) and have a greater occurrence of opportunistic pathogens, antibiotic resistance genes and metal resistance genes. However, Li, *et al.*, 2022 concluded that the structure of bacterial community on MPs within 1L bottles of filtered river water (used as a drinking water source) was determined by the bulk-water quality rather than MP characteristics.

MPs may also release toxic chemicals into water (e.g., phthalates, hydrocarbons, heavy metals) (Cheng *et al.*, 2020) and can be a source of organic carbon (Chen *et al.*, 2021; Lee et. al. 2020), impacting water quality by increasing bacterial growth and reducing biostability (Chen *et al.*, 2021). Additionally, leaching of plastic-associated carbon in bench-top beaker tests has been shown to increase disinfection by-products (Ateia *et al.*, 2020), which are potential carcinogens. MPs have also been shown to impact microbial growth and microbiome composition on ultrafiltration membranes used in drinking water treatment (Xiong *et al.*, 2022).

It is possible that MPs not removed from drinking water by upstream treatment processes (or that contaminate in the distribution system, post-treatment) could be reservoirs and vectors of microbial contaminants. The bench top tests are substantially different from the scale of

operational systems and do not replicate all the physical, chemical and microbiological interactions that occur within real systems. Consequently, the impact or risk that such MP-associated processes would contribute to water quality degradation would likely be insignificant compared to the microbial accumulation and mobilization of biofilms that form on the vast internal surface areas of drinking water infrastructure.

Microplastic removal/destruction from water

MPs cycle through our environments, current and emerging removal methods include physical (coagulation and flocculation methods, electro-sorption, membrane technologies), chemical and biological treatments (as summarized in Figure 1). With respect to water treatment, large size plastics are expected to be removed by screen mesh (6 mm size) and grit processes and MPs largely removed during the sedimentation, activated sludge, membrane bioreactor or sand filtration processes (Bayo *et al.*, 2020; Talvitie *et al.*, 2015). Talvitie *et al.*, 2015 showed that treatment had a significantly higher fiber removal efficiency (92%), than secondary treatment (0.2%) which did have a substantial removal rate of synthetic particles (52%). WHO (2019), highlights that the greatest risk to freshwater and drinking water from MPs is most likely where water treatment is unavailable - 67% of low/middle income countries lack sewage connections and 20% of household wastewater is not treated by at least secondary level treatment. Whilst MP contamination does not present the greatest drive for improving water treatment and safe distribution, achieving this will simultaneously improve MP and NP removal.

Physical methods of MP removal from water

Coagulation and flocculation methods

Drinking water treatment plants (DWTP) use coagulation, flocculation, and sedimentation (CFS) to enlarge particles, improving filtration. Inorganic and organic coagulants used in CFS can bind MPs to waste particles facilitating their removal. CFS has proved effective in removing detectable microplastic in aquatic environments (Li *et al.*, 2022, Rajala *et al.*, 2020, Li *et al.*, 2021). Table 1 show that the based-on laboratory-scale studies CFS methods are effective in removing MPs. The exact removal efficiency depends on several factors, including coagulant type and dose, water sample characteristics and MP properties (Table 1).

Several studies have investigated effectiveness of different coagulants and their optimal doses for MP removal. The properties of MPs, particularly their size, have been found to impact removal efficiency. In laboratory studies using alum and polyaluminum chloride (PAC) have shown the effectiveness of removing MPs from water, with aluminum chloride more effective than iron chloride for MPs of size 10-90 µm where the optimal dose reported was 10 mg/L at a pH of 6 (Na *et al.*, 2021) and higher alum doses improved removal of MPs <90µm (Xue *et al.*, 2021). For removal of smaller MPs from municipal wastewater, iron chloride showed a greater removal efficiency than PAC and polyamine (Rajala *et al.*, 2020). The use of PAC in combination with polyamine-coated sand was found to be effective in removing more MPs than PAC alone, with an optimal alum dose of 20 mg/L and 92.7% of MP particles removed (Shahi et al, 2020). Magnetic magnesium hydroxide (MMHC) has been investigated as a coagulant for the removal of MPs from wastewater, and Mg to OH ratio of 1:1 was found to be

ideal for preparing the most effective MMHC (Zhang *et al.*, 2021). The highest removal efficiency was obtained at a pH range of 5-9 with 93.8% of MPs removed using 200 mg/L of magnesium hydroxide, 120 mg/of iron oxide, and 5 mg/L of polyacrylamide (PAM). Whilst the coagulant type clearly impacts MP removal, the combination of coagulant(s) used are also important. Interactions between these need to be better characterized to obtain optimum chemical coagulant and the operation conditions for most efficient MP removal.

Water characteristics also impact the removal efficiency of MPs, particularly the pH, which influences the floc characteristics through hydrolysis of the coagulant. The optimal pH for the removal of MPs has been reported as 6-8 depending on the coagulant used (Zhang et al 2021; Rajala *et al.*, 2020). Xue *et al.*, 2021 showed that the removal efficiency of carboxylated MPs (simulating those found in aquatic systems) using CFS and found higher removal in water samples collected from Lake Erie water than those from Grand River, possibly due to the higher turbidity of Lake Erie promoting flocculation. In water samples from both locations CFS was effective in removing MPs, with the addition of alum removing more MPS of 45-90 μm (removal of MPs ≤25 μm was unimpacted). These studies demonstrate the need for research to better understand the interactions between physical and chemical characteristics of water and coagulant agents in order to develop best practice for MP removal depending on the matrix of polluted water.

Size of MPs has been extensively studied but the existing literature rarely describes the impact of shape of MP on removal efficiency. Shahi *et al.*, 2020 found that MP removal efficiency in treatment plants is also influenced by shape and surface morphology. Elongated

MPs and those with rougher surfaces were removed more efficiently than spherical MPs or those with smoother surfaces. Elongated smooth MPs were easily removed when compared to spherical rough MPs. These findings suggest that there is a need for further research on the effect of MP characteristics on their removal from DWTPs.

Chemical methods of MP removal from water

Electrochemical methods for microplastics removal

Electrolysis degrades or separates MPs from the wastewater through sorption using an electrolyte. Several studies have demonstrated promising potential to use a range of electrolysis-based methods for MP removal from wastewater. Perren *et al.*, 2018 investigated the effectiveness of electrocoagulation (EC) for removing MPs from wastewater. Perren *et al.*, 2018 found that 90% removal efficiency of MPs under optimal pH of 7.5, NaCl concentration of 2 g/L, and current intensity of 11 A/m^2. Shen *et al.*, 2022 studied various factors and determined that an electrolyte concentration of 0.05 M, pH of 7.2, and applied voltage of 10V resulted in an MP removal efficiency of 91.7%. Fiber MPs and Al anode were preferred to granular MPs and Fe anode, respectively. Xu *et al.*, 2022 used interpenetrating bipolar plate EC to remove mixed pollutants of MPs and heavy metals from wastewater, achieving a high removal efficiency of around 96%. These studies demonstrate that EC is a clean technology capable of reducing the potential harm of MPs and heavy metals to aquatic organisms.

Electro-sorption (ES) is another potential-induced adsorption on the surface of charged electrodes. Following polarization of the electrodes, ions are removed from the electrolyte solution by the imposed electric field and adsorbed onto the surface of the electrodes. Xiong *et*

al., 2020 investigated the efficiency of ES process in treating water contaminated with MPs. The capacity was found to be 0.707 g nano-polystyrene/g AC (70.7 %). This suggested that electro-sorption to be a feasible tertiary treatment method of water contaminated with MPs. Moreover, the laboratory scale study conducted by Mantel *et al.*, 2021 demonstrated high efficiency of removal of MPs using ES. Similarly, electro-oxidation (EO) offers another established electrochemical treatment method; the chief reagent used is an electron, which eliminates the organic matter present in the electrolyte. Developed along similar lines as EC, EO is anodic oxidation on the anode surface that is employed in place of direct chemical oxidation. Its an alternative variation and involves only the generation of an oxidizing species using the chemical pathway being accompanied by direct oxidation of generated oxidizing agents. Critical parameters for EO efficiency include design of electrochemical cell, selection of electrodes, operational parameters and cost of power. A study by Kiendrebeogo *et al.*, 2021 focused on the degradation of synthetic suspension of polystyrene microbeads (26 µm in size). The study employed EO and found that the process can degrade 58 ± 21% of MPs in one hour. Furthermore, current intensity, anode material, electrolyte type and electrolyte concentration are functions of MPs removal efficiency. It was also discovered that MPs did not break into smaller particles but were degraded directly into gaseous products. This suggests that EO is a safe process for degrading MPs in wastewater. As a credit to EO, other electrochemical methods like EC and ES cannot confirm the long-term sustainability due to the formation of secondary MPs. EO ensures complete MPs mineralization by breaking down olefin chains of MPs through generating oxidizing radicals. Eventually, less harmful products (H_2O and CO_2)

are produced (Ranade and Bhandari, 2014).

Membrane Technologies for microplastics removal

Advanced membrane technologies during tertiary treatment can achieve reduction of microplastics in wastewater. Talvitie *et al.*, 2017 investigated the removal of MPs from effluent in WWTP using different advanced treatment technologies including disc filters, sand filters, dissolved air floatation and membrane bioreactor (MBR). The MBR removed 99.9% of MPs during the treatment (from 6.9 to 0.005 MP L^{-1}), rapid sand filter 97% (from 0.7 to 0.02 MP L^{-1}), dissolved air flotation 95% (from 2.0 to 0.1 MP L^{-1}) and disc filter 40–98.5% (from 0.5 – 2.0 to 0.03–0.3 MP L^{-1}) of the MPs during the treatment. Similar findings were reported using a pilot-scale MBR that removed 98.4% of MPs compared to the conventional activated sludge process (Lares *et al.*, 2018).

Removal efficiency of MPs from wastewater treatment plants is also high when advanced filtration methods are employed. Drinking water treatment works are engineered to produce high quality water, part of this includes the output of low turbidity water, which requires the removal of particles that are similar to, or smaller than, MPs in their physiology (size, density) and detected in far higher concentrations. Dalmau-Soler *et al.*, 2021 demonstrated that for PS and PP dominated intake water in drinking water treatment plant MP removal efficiency was 93%, with sand filtration as a critical MP removal stage (78% efficiency). Similarly, Negrete Velasco *et al.*, 2022 reported a decrease in MPs within a pilot drinking water treatment station from 19.5-143.55 MPs/m^3 in raw water to 0.0-8.0 MPs/m3 in drinking water, although this study stressed the importance of coagulation in removing MPs (89% and 97% removal

efficiency without and with coagulant, respectively). The low MP concentrations reported in drinking water (Dalmau-Soler *et al.*, 2021; Negrete Velasco *et al.*, 2022; Johnson *et al.*, 2020) are indicative that current treatment processes are effective at removing MPs from drinking water.

Dynamic membrane (DM) filtration is an emerging technology in the WWTP based on the formation of a cake layer (secondary membrane) formed during the filtration of wastewater through the supporting membrane (Ma *et al.*, 2013). Li *et al.*, 2021 used DM and reduced the effluent turbidity to < 1 NTU (Nephelometric Turbidity Unit) after 20 minutes of filtration an indicator of the effectiveness of the approach. The DM performance was shown by the correlation between the transmembrane pressure and total filtration resistance increasing linearly with filtration time, important considerations to ensure are included in operational practice guidance.

Reverse osmosis is also an advanced system used to water using nanofiltration membranes under high pressure (10-100 bar). It is used in municipal water treatment to recover wastewater by applying high pressure to a concentrated solution forcing the water through a semipermeable membrane leaving all the salts, pollutants and heavy metals in the concentrated solution (Ashfaq *et al.*, 2019). Cai *et al.*, 2019 used an integrated membrane system (IMS) in a coastal reclaimed water plant to remove microplastics. The removal rate of microplastics in the IMS system was 93.2% after membrane bioreactor (MBR) treatment while that further increased to 98.0% after the reverse osmosis (RO). However, a breakthrough for small scale fibres (< 200 μm) was measured in the RO system. Many microplastics (< 10 μm) were still

measured in the WWTP effluent with polyethylene (PE) the most common polymer identified.

Advanced oxidation for microplastics removal

Advanced oxidation processes (AOPs) for chemical degradation of contaminants were introduced in the 1980s (Kim *et al.*, 2022). Advanced oxidation processes (AOPs) are oxidative methods based on the generation of reactive oxygen species (ROS) have been successfully applied for decomposition of different refractory contaminants in the environment. The fact that AOPs could effectively degrade a wide spectrum of pollutants prompted researchers to investigate several AOPs for decomposition of microplastics (MPs). These include ozone, UV photolysis, UV/H_2O_2, UV induced photo-catalysis, Fenton process, photo-Fenton process, and persulfate (PS) treatment. AOPs relies on the formation of reactive oxygen species (ROS) by employing light, plasma, heat, and catalysts. Due to their high redox potentials, the formed radical species (e.g., hydroxyl radicals, 1.89–2.72 V) can attack and degrade several refractory pollutants or at least increase their biodegradation before biological treatment stages. Recently, several studies dealing with the treatment of MPs by AOPs such as ozone, heat/persulfate, UV photolysis, UV/H_2O_2, UV/catalysts, and plasma have been reported (Kim *et al.*, 2022), highlighting the potential options for consideration at full-scale.

Ozone (O_3) is a strong oxidant extensively investigated within AOPs (Kim *et al.*, 2022). Ozone can irreversibly impair the physico-chemical properties of MPs and NPs. Zafar *et al.*, 2021 studied the PE degradation by ozone. PE breakdown was carried out by varying the ozone dosages in the range 4–7 mg/min for 60 min, 120 min and 180 min. Hydroxyl index (HI) and carbonyl index (CI) were recorded throughout the experiments to evaluate MPs

degradation. CI and HI values increased with increasing ozone dosage and reaction time suggesting an enhanced oxidation. Hidayaturahman and Lee, 2019 combined the use of ozone with established wastewater treatment for MPs degradation. Nearly 99% MPs were removed when sequentially applying O_3 treatment and coagulation, indicating the importance of employing multi-step treatment and potentially the specific sequence of those steps which could improve removal.

Persulfate (PS, $S_2O_8^{2-}$) maybe thermally activated to produce sulfate radicals ($SO_4^{\bullet-}$) for destroying refractory contaminants. Liu *et al.*, 2022 investigated polystyrene and PE MPS degradation through heat-activated PS. Significant changes in physico-chemical properties of PE and polystyrene were observed. In particular, CI and surface roughness increased with increasing the reaction time. On the other hand, average particle size decreased due to MPs breakdown of polystyrene and PE during the experiments. Moreover, several functional groups were formed on the surface of the polystyrene and PE MPs, thus resulting in increased hydrophilicity and O/C ratios. This important change in surface morphology led to enhanced adsorption capacities of MPs subject to ageing in natural environments. Therefore, aged MPs may adsorb hazardous pollutants and cause irreversible effects to ecosystems. Wang *et al.*, 2017 and Ding *et al.*, 2022 reported that peroxymonosulfate (PMS) treatment could provide insight into the ageing of MPs. The active oxidizing agents produced in AOPs are typically free radicals like HO• for Fenton and SO4• for PS (Jiang *et al.*, 2021). Furthermore, Fenton and persulfate (PS) treatment could provide insight into the aging of MPs (Wang *et al.*,2017; Ding *et al.*, 2022). Another promising method, the photo-Fenton process has been discussed by

Ricardo *et al.*, 2021 to be effective in MP degradation. Kida *et al.*, 2019 employed the photo-Fenton process to remove a recalcitrant fertilizer. The modification entails iron (II) being replaced by iron (III) as well as improving the process using a chelating agent.

Due to light irradiation, reactive species are formed during UV photolysis (e.g., $^{\bullet}OH$, $^{3}OM^{\bullet}$, $O_2^{\bullet-}$, $ROO^{\bullet}$) and may decompose several refractory pollutants (Stefan, 2017). Suhroff and Scholz-Böttcher, 2016 reported that UV oxidation could alter physicochemical properties of plastics. The possibility of employing natural sunlight irradiation represents a great potential advantage of photolytic treatments of plastics in the environment. According to Suhroff and Scholz-Böttcher, 2016 UV irradiation could affect physico-chemical features of polyvinylchloride (PVC), polyethylene (PE), polystyrene, and polyethylene terephthalate (PET), thus leading to their degradation according to complex reaction pathways. At the end of the photolytic treatment, the samples were broken into nanometers and micrometers sizes. Further studies recoded plastics degradation by UV photolysis under wavelengths of 200–280 nm (UV-C), 280–315 nm (UV-B) and 315–400 nm (UV-A) (Kim *et al.*, 2022). The UV method is a potent method but for considerable impact high energy content is desired.

The use of $^{\bullet}OH$, UV/H_2O_2 has also been extensively investigated in AOPs. Liu *et al.*, 2021 showed UV/H_2O_2 treatment induced irreversible physico-chemical changes to polystyrene, including modification of surface morphology, CI, crystallinity, hydrophobicity and embrittlement. In particular, $^{\bullet}OH$ oxidation led to an increase in surface roughness, CI, crystallinity, and embrittlement increased due to chemical oxidation by • OH. On the other hand, the formation of various surface functional groups and chain scission reactions resulted

in decreased hydrophobicity and average molecular weight. The combination of UV/H_2O_2 can substantially improve. the destruction of MPs.

Fadli *et al.*, 2021 employed UV/photocatalysis with TiO_2 photocatalyst doped with Ag or Ag and reduced graphene oxide (RGO) to enhance PE MP removal by reducing electron-hole recombination on the TiO_2 surface. PE particles with an average size of 100–150 μm were employed for the photocatalytic tests. Nearly 76% PE was decomposed with 3% Ag/TiO_2-1% RGO photocatalyst after 4 h of UV illumination. However, as UV radiations accounts for only 5% of the solar spectrum, visible light-activated photocatalytic processes for MPs and NPs should be extensively investigated. In this regard, Pt-ZnO nanorods were employed by Tofa *et al.*, 2019 to oxidize under visible light low-density PE with a thickness of 50 μm. Pt deposition on ZnO markedly enhanced the visible light absorption of the photocatalyst. Increased surface oxidation of LDPE, CI and vinyl index were recorded during the photocatalytic experiments over time. In addition, SEM analysis revealed cavities and cracks on the LDPE surface. Nabi *et al.*, 2020) successfully employed activated titanium dioxide (TiO_2) film to generate ROS such as OH• and O2•- to decompose PE and polystyrene under UV illumination. Faldi *et al.*, 2021 used a similar approach. However, they modified TiO_2 using Ag and reduced graphene oxide (RGO) to improve the results. Hence the UV treatment can be enhanced by photocatalysis.

Free electron rich plasma may generate reactive oxygen species (ROS) and O_3 from O_2 and air with potential application for the degradation of MPs in water. Electrical discharge plasma was employed by Zhou *et al.*, 2020 for decomposing PVC MPs. Various functional

groups were detected on the surface of MPs. In addition, surface area, CI, hydrophilicity, crystallinity, and increased. On the other hand, plasma treatment lowered MPs sizes. However, plasma method is an emerging method for destruction of emerging contaminants.

Advanced oxidation processes (AOPs) are effective methods of decomposing pollutants, including microplastics (MPs). AOPs generate reactive oxygen species (ROS) through light, plasma, heat, and catalysts, which attack and degrade pollutants with high redox potentials. AOPs, including ozone, UV photolysis, UV/H_2O_2, UV-induced photocatalysis, Fenton process, photo-Fenton process, and persulfate (PS) treatment, have been reported to effectively degrade MPs. Ozone irreversibly impairs the physico-chemical properties of MPs and NPs, while PS, thermally activated to produce sulfate radicals (SO_4•−), can break down refractory contaminants. Reactive species are also formed during UV photolysis, and natural sunlight irradiation is an advantage of photolytic treatments of plastics. The use of UV/H_2O_2 induces irreversible physico-chemical changes in MPs, while UV/photocatalysis with TiO_2 photocatalyst doped with Ag or Ag and reduced graphene oxide (RGO) enhances PE MPs removal. Overall, AOPs will soon have a crucial role in green and sustainable MPs removal in the environment. However, considerable research efforts should be made for complete removal of MPs by employing AOPs. Bench-scale technologies for MPs and NPs removal based on AOPs should be optimized at a large scale with the aim of developing industrial implementation.

Biological methods (biodegradation) for microplastics removal

Biological methods for micro and nano plastics (MNPs) removal from ecosystems remain

increasingly attractive research. These technologies are not only well known due to their low-cost, eco-friendly performance, but also by the capacity of providing in-site and ex-site remediation. Biotechnology to remove MNPs includes complex processes, such as biotransformation, biodegradation, biofilm formation, biosorption, etc. Different microbes have different interactions with MP in the MNP removal. Many researchers are searching for possible solutions for the removal of MPs with different biological organisms. Since biological organisms are sensitive to several factors, such methods require more effort and research for efficient implementations on a larger scale (Badola *et al.* 2022).

Microplastics remain longer in the environment and in food chain because of their low degradability, persistent and cumulative nature (Wahyuningtyas *et al.*, 2017; Urbanek *et al.*, 2017), posing a major risk to human health (Tachibana *et al.*, 2013; Harmaen *et al.*, 2015; Calabrò, and Grosso, 2018 and Suzuki *et al.*, 2018). Also, microplastics have large surface areas and adsorption capacities. This makes them prone to sorb thereby accruing harmful and toxic waterborne pollutants which they usually transport and desorb into new habitats (Rios *et al.*, 2007; Mohsen *et al.*, 2020). Microplastics are quickly colonized by microorganisms such as fungi and bacteria as soon as they are released to the environment (Delacuvellerie *et al.*, 2019; De Tender *et al.*, 2017). This is because, microplastics surfaces provide aquatic and terrestrial microhabitat that support the growth of unique and distinctively different microbial communities (De Tender *et al.*, 2015; Amaral-Zettler *et al.*, 2015; Oberbeckmann *et al.*, 2014; Wu *et al.*, 2019). Certainly, many microorganisms have evolved strategies to survive on the microplastic laden environment where they break them down enzymatically and utilize them as

source of energy (Zeenat *et al.*, 2021).

Biological methods used to eradicate microplastics from the environment, make use of microorganisms and their hydrolytic and proteolytic enzymes with ability to biodegrade various types of microplastics (Pathak, 2017; Danso *et al.*, 2019; Peng *et al.*, 2022). Biodegradation is a process by which plastic debris are reduced into metabolizable harmless products through the aid of microbial digestion (Pathak, 2017). The biodegradation of plastic materials can be influenced by the microorganism species involved, sources of carbon, size, and types of plastic materials (Allouzi *et al.*, 2021). The factors of degradation are categorized into two main groups: biotic and abiotic. Biotic factors refer to microbial growth rate, enzyme activity, culturing feasibility, etc. Abiotic factors include MNPs structure, morphologic surface, functional groups, pH, temperature, level of electronic acceptors such as oxygen, etc. Both aerobic and anaerobic methods can be applied.

Microplastics are mostly made up of non-hydrolysable, hydrophobic and chemically inert groups. This makes them hard to undergo biodegradation unless they are first weakened by abiotic factors that will have to oxidize them first (Yoon *et al.*, 2012; Temporiti *et al.*, 2022; Hakkarainen M, Albertsson, 2004; Arutchelvi *et al.*, 2008; Bryant *et al.*, 2016). Biodegradation of microplastics is therefore reliant on the physical and chemical properties of the constituent microplastic polymers. Some of these physical properties include hardiness and complexity. Polymers that are soft, degrade quickly than hard ones (Corcoran, 2021). Other factors affecting biodegradation include the density and molecular weight of the polymer (Zambrano *et al.*, 2020), hydrophobicity of polymers, which increases available functional groups (Bildik

Dal *et al.*, 2021) together with the nature and type of the bonds where amide and ester bonds break easily (Mahesh *et al.*, 2018). Abiotic factors such as light, oxidative stress, thermal heat, pH, ultraviolet rays, atmospheric humidity, hydrolysis, and oxidation also influence the rate of biodegradation (Teixeira *et al.*, 2021). Microplastics exposed to abiotic factors decrease in strength, flexibility and increase in brittleness (Garrido and Costanzo, 2022). Exposure of microplastics to abiotic factors such UV light, contribute to the initial disintegration of plastic polymers by initiating photo-oxidation (Sen and Raut, 2015; Shah *et al.*, 2007). Photo-oxidation modify plastics physically and chemically leading to weight loss, increased viscosity, bond breaking and molecular weight reduction which together promote biodegradation (Koutny *et al.*, 2006; Da Luz *et al.*, 2015). Therefore, biological methods of removing microplastics, utilize the biodegradation capacity of microbial enzymes from bacteria such as actinomycetes (Gram-positive mycelial bacteria) and fungi (Tamoor *et al.*, 2021; Alshehrei, 2017). For this reason, there are several bacterial, fungal, and algal based methods used bio-remediate the environment of microplastics.

Bacterial-enzymatic processes

The mechanisms of MNPs biodegradation by bacteria remain unclear. However, based on multiple studies of bacterial degradation of plastics, Mohanan *et al.*, 2020 and Tiwari *et al.*, 2020 summarized the biodegradation mechanism by bacteria which includes four stages: (1) biodeterioration of plastic surfaces, conducted by changing the chemical and physical properties of plastics by oxidative enzymes released by bacteria; (2) bio-fragmentation of complex polymer into simpler forms via hydrolysis and release of intermediate products; (3)

bio-assimilation of small fragments generated from the stage 2 by bacteria up take or mineralization; (4) mineralization of metabolites into microbial biomass with associated release of CO_2, CH_4, and water.

Several studies show that suitable bacteria strains aim to selective target micro plastics to degrade the MPs by 6% - 36% from 40 days to 16 weeks under the controlled environmental conditions, such as pH, temperature, salinity, etc., using isolated and cultured pure bacterial strains in the laboratory (Muhonja *et al.*, 2018; Auta *et al.*, 2017; Habib *et al.*, 2020). The advantage of using pure strains in MP degradation studies is that it is a convenient way to investigate metabolic pathways or evaluate the effect of different environmental conditions for MP degradation (Yuan *et al.*, 2020). Park and Kim 2019 isolated mesophilic mixed bacterial from an abandoned landfill and showed efficient degradation of PE. Habib *et al.* (Habib *et al.*, 2020) used the strains from the Antarctic region to degrade PP. These bacteria represent the cells living under the cold weather and poor nutrient levels. Interestingly Liu *et al.* (Liu *et al.*, 2021) proposed a proof of concept with 'trap and release' of MNPs. In this study, instead of degrading the MNPs into smaller and simpler products, genetically modified strains *P. aeruginosa* bio aggregated MNPs by a forming biofilm. Followed by a dispersal mechanism, these trapped aggregated MNPs were released for a further collection and recovery.

Table 2 highlights some of the recent research on the removal of MNPs by bacteria. Bacteria are ubiquitous in the ecosystem and can be most widely applied for the biodegradation of MNPs pollution in environment.

In further studies, species of bacteria such as *Pseudomonas aeruginosa*, have been shown

to colonize microplastics in the environment (Ayush at al., 2022). Two bacillus bacterial strains; *Bacillus cereus* and *Bacillus gottheilii,* isolated from mangrove sediments, have been shown to have the capacity to remove different microplastics composed of polyethylene, polystyrene, polyethylene terephthalate and polypropylene (Auta *et al.*, 2017). In the study, the rate of biodegradation was assessed via measuring the microplastics weight loss using electron microscopy and FTIR analyses. Comparatively, *B. gottheilii* was found to be a better potential microplastic degrader. Other microbial species that are associated with degradation of polyethylene and polypropylene have been identified as Streptococcus, Klebsiella, Micrococcus Staphylococcus, Pseudomonas (Zeenat *et al.*, 2021). Unfortunately, there is no research work conducted at the nano level of plastic biodegradation, hence this will be a future research topic.

The biodegradation begins by the formation of a biofilm where bacteria adhere on the surfaces and cracks of microplastics due to microplastics contact with water (Jacquin *et al.*, 2019; Auta *et al.*, 2018). Hydrophobicity microplastic polymers hinders the attachment of bacteria thereby preventing the formation of a biofilm and decelerating the onset of biodegradation (Pinto *et al.*, 2019; Hossain *et al.*, 2019). Bacterial adhesion is important in microplastic biodegradation, and the ability of bacterial adhesion has been investigated among four types of plastics; polyethylene, polypropylene, polyethylene terephthalate, and polyvinyl chloride (Cai *et al.*, 2019). In this investigation, surface hardness of the plastics was identified as a major factor dominating the adhesion of bacteria onto plastic surfaces in contrast to the other factors (Elbourne *et al.*, 2019). Intrinsic surface properties such as surface roughness,

topography, surface free energy, surface charge, electrostatic interactions, and surface hydrophobicity play an important role in the biofilm attachment (Persat *et al.*, 2015; Pinto *et al.*, 2019). The biofilms on microplastics are then depolymerized, catabolised and eventually bio-mineralized into organic matter (Artham and Doble, 2008; Sen and Raut, 2015; Mercier *et al.*, 2017). Indeed, some members of the bacterial species colonize around and trap microplastics creating biofilms (Yang *et al.*, 2019). In some cases, especially on the water bodies, colonization ad formations of biofilms increase the density of microplastics. This makes microplastics aggregate and sink into the depths and thus decreasing prior photolytic degradation caused by the sun's UV radiation (Robyn *et al.*, 2020). Plastic related bacterial biofilms are more efficient in degrading plastics when compared to planktonic biofilms (Wilkes *et al.*, 2017). Several genera of hydrocarbon-degrading bacteria have been shown to play a vital role in plastic degradation (Delacuvellerie *et al.*, 2019).

Biofilm formation is a multistage process mediated by several factors inter alia temperature, light, surface properties, nutrient solution, and pH (Renner and Weibel, 2011). For instance, exposure of MPs to UV light, enable microbial enzymes to easily attach on the carbonyl group of plastic polymers (Kotova *et al.*, 2021; Taghavi *et al.*, 2021; Vedrtnam *et al.*, 2019; Montazer *et al.*, 2018). Exposure of micro-plastics to conditions such as UV radiation, thermal treatment, and pro-oxidants, generate more polar groups thereby decreasing hydrophobicity and accelerating biodegradation process (Auta *et al.*, 2017; Arkatkar *et al.*, 2009; Auta *et al.*, 2018; Arutchelvi *et al.*, 2008). By secreting various enzymes such as proteinases, bacteria can catalyse and hydrolyse plastic polymers into oligomers, monomers,

and other metabolic intermediates that can be easily mineralised into water and carbon dioxide (Siracusa, 2019).

Although more plastic-degrading bacteria have been found in the environment (Syranidou *et al.*, 2019; Mercier *et al.*, 2017; Auta *et al.*, 2017; Yoshida *et al.*, 2016; Yang *et al.*, 2019), no single strain was found suitable for commercial application to date, but a few studies have focused on the degradation of microplastics by microbial consortia (Lin, 2022). A recent study has shown that, degradation of micro-plastics using more than two bacteria living symbiotically, promotes more efficient plastic biodegradation as compared to pure strains (Oliveira *et al.*, 2020). This is due to creation of stable microbial community and elimination of the effects of toxic metabolites produced by some strains present in the community (Hibbing *et al.*, 2010). The toxic metabolites produced by one strain can be used as a substrate by another strain within the bacterial community (Singh and Wahid, 2015).

Biodegradation is typically an enzyme driven mechanism involving physico-chemical transformation of polymers into smaller units mediated by microorganisms that colonize on the surface of microplastics to form a plastisphere (Shah *et al.*, 2008; Dudek *et al.*, 2020). Several microorganisms that include bacteria, algae, fungi, and bryozoa have been observed within the plastisphere (Alshehrei, 2017; Amaral-Zettler *et al.*, 2020). The colonizing microbiota release extracellular enzymes which bio-fragment and depolymerize microplastics into oligomers, dimers, and monomers (Arkatkar *et al.*, 2009; Siracusa *et al.*, 2008). Once the microplastics have been disintegrated into smaller molecules, they pass through the microbe's cell wall, get assimilated and utilized as carbon and energy source (Da Luz *et al.*, 2015; Corcoran, 2021).

Microbial enzymes that breakdown microplastics, typically bio-fragment them into low molecular weight substrate biomass (Sengupta and Pike, 2015). The resultant bio-fragmented microplastic substrates, are transported into microbial cells via cell membranes where they are mineralized, oxidized, and used for energy production (Gu, 2003; Da Luz *et al.*, 2015; Adele Folino *et al.*, 2020). Within the microorganisms' cells, mineralized microplastics are broken down further aerobically using oxygen as an electron acceptor while producing carbon dioxide, water, biomass, and energy as by-products (Lucas *et al.*, 2008; Ayodeji Amobonye *et al.*, 2021). Biodegradation can also take place anaerobically whereby iron, nitrate, manganese, sulphate, and CO_2 serve as electron acceptor instead of oxygen while producing products such as CH_4 and H_2S (Shen *et al.*, 2020). The energy produced from biodegradation process is eventually utilised for biomass production (Lucas *et al.*, 2008; Osman *et al.*, 2021). Mineralisation yield atoms that are assimilated into microbial cells for complete degradation. Secondary metabolites are transported outside the cells or transferred to other microbes that utilize them (Mohapatra and Balaram, 2021). The secreted extracellular enzymes bio-deteriorate and bio-fragment microplastics into assimilable low molecular weight monomers and oligomers (Rhodes, 2019). Extracellular enzymes such as laccase from *Staphylococcus epidermis* and PETase from *Ideonella sakaiensis* have been shown to have the capacity to disintegrate microplastics (Yoshida *et al.*, 2016; Hiraga *et al.*, 2019; Hou and Majumder, 2021). Other oxidative enzymes such as monooxygenase, peroxidase, manganese, dehydrogenase, and oxidase, can degrade non-hydrolysable polymers e.g., polyethylene and polypropylene (Pathak *et al.*, 2017; Danso *et al.*, 2018). Moreover, bacterial enzymes such as oxygenase have the capacity to break down

carbon chains into environmentally harmless products such as alcohol and peroxyl (Mohanan *et al.*, 2020; Pathak *et al.*, 2017). Likewise, enzymes lipases, proteases, ureases, esterases and endopeptidases, bio-fragment microplastics by catalysing carboxylic groups into amide groups (Amelia *et al.*, 2021; Zeenat *et al.*, 2021; Temporiti *et al.*, 2022). The reaction of such enzymes is normally optimized on those microplastic polymers that have been exposed to the action of both biotic and abiotic factors (Oliveira *et al.*, 2020). Indeed, some microbial enzymes such as laccase, lignin degrading enzymes, lipase, protease, and urease can attack the plastic polymers after such polymers have been exposed to the action of both biotic and abiotic factors (Maity *et al.*, 2021). Beside use of enzymes, micro-organisms use other compounds such as acids and peroxides which will change the surrounding pH and facilitate biodegradation process (Karigar and Rao, 2011). Microorganisms can also chemically weaken plastics by changing the pH of the surrounding microenvironment (Karigar and Rao, 2011).

Fungal-enzymatic processes

Terrestrial and marine fungi biodegrade plastic polymers and utilise them as source of nutrients (Russel *et al.* 2011; Zengal *et al.*, 2021). Fungi can promote the formation of different chemical bonds in micro plastics, such as carbonyl, carboxyl, and ester functional groups, decreasing their hydrophobicity (Yuan *et al.*, 2020). Fungi exhibit extraordinary strategies including adsorption ability, natural biosurfactant production, and extremely unspecific oxidative enzymes which allow them to oxidize numerous substrates that could attack polymers by the co-metabolic process and use plastic particles as carbon, energy, and electron sources (Bacha *et al.*, 2021).

Paço *et al.*, 2017 used a naturally occurring marine fungus *Zalerion maritimum* to degrade PE micro particles of the size range between 250um – 1000um over 28 days. The results showed that starting at the 14th day, the biomass weight increased 82% and PE weight loss 57% which corresponds to the removal rate of 43%. Until the 28th day, when the PE micro plastic weight loss was 73%. The FTIR-ATR and NMR results of *Zalerion maritimum* indicated the obvious reduction of this fungus lipidic and protein content when exposed to microplastics. Meanwhile, the FTIR spectra of PE showed the presence of oxidation functional groups which implied the degradation of microplastics.

Zhang *et al.*, 2020 isolated a PE-degrading fungus *Aspergillus flavus* named PEDX3 from the gut contents of wax moth *Galleria mellonella*. Their results indicated that HDPE of the size less than 200□m degraded into the microplastic particles with a lower molecular weight by PEDX3 after 28 days of incubation. The HDPE mass loss was 3.9% and the molecular weight decreased from 220,003 Da to 89,801 Da which corresponds to a 59% loss. This proved that HDPE produced microplastic particles were converted to smaller molecular weight and were more concentrated after exposure to strain PEDX3, which suggested that the depolymerization of HDPE long chain structure occurred and that lower molecular weight fragments were formed.

Some plastic degrading fungi from the Ascomycete phylum in which *Fusarium*, *Penicillium* and *Aspergillus* belong, have been identified (Tachibana *et al.*, 2010; Raghavendra *et al.*, 2016). Fungal strains belonging to *Penicillium sp* such as *P. chrysogenum, P. oxalicum* and *P. simplicissimum* are potential plastic degraders (Sowmya *et al.*, 2015; Ojha *et al.*, 2017

and Alshehrei, 2017). The genus *Aspergillus* possess several species that include *A. flavus, A. niger* and *A. terreus* that have been found to possess the abilities of degrading plastics (Zhang *et al.*, 2020; Yamada-Onodera *et al.* 2001). Other fungi such as *Myceliophthora* sp., *Phanerochaete chrysosporium*, and *Trametes versicolor*, exhibit the ability to degrade plastics especially those of polyethylene origin (Khalil *et al.*, 2013; Ojha *et al.*, 2017). Plastic wastes can also be degraded by fungal mushrooms which convert microplastics into fungal biomass (Da Luz *et al.*, 2012). Chemical and physical properties such as high molecular weight, hydrophobicity and insolubility of microplastics, make them difficult to biodegrade (Wei and Zimmermann, 2017; Kumar *et al.*, 2020; Amobonye *et al.*, 2021). However, use of filamentous fungi as means of removing microplastics, can circumvent this problem. This is because, filamentous fungal hyphae have penetrative apical growth which allows them to extend their mycelial networks into different kinds of materials. Besides, fungal hyphae secrete biochemical such as hydrophobics which promote adhesion to hydrophobic substrates (Daccò, *et al.*, 2020; Sánchez, 2020). Fungi production of fungal enzymes that can biodegrade microplastics polymers (Temporiti *et al.*, 2022). Fungal exoenzymes such as hydrolases (lipases, carboxylesterases, cutinases and proteases) modify the plastic surface, increasing its hydrophilicity and facilitate the biodegradation PET (Vertommen *et al.*, 2005; Krueger *et al.*, 2015; Webb *et al.*, 2013). Moreover, oxidoreductases (laccases and peroxidases) are involved in plastic degradation into micromeres such as oligomers, dimers and monomers (Álvarez-Barragán *et al.*, 2016; Gómez-Méndez *et al.*, 2018). These enzymes can depolymerize, biodegrade and thereby bioremediate different plastic polymers with higher

stable carbon–carbon (C–C) bonds but after such polymers have undergone oxidation (Da Luz *et al.*, 2019; Restrepo-Florez *et al.*, 2014). The fungus *Pestalotiopsis microspore* (an endophytic fungi), use its hydrolase enzyme to decompose microplastic polymers and utilise them as source of carbon (Russell *et al.* 2011). Furthermore, the enzymes serine hydrolases from fungi, have been shown to degrade micro-plastics in the presence of optimally controlled culture conditions (Ahmaditabatabaei *et al.*, 2021). Enzymes depolymerase, esterase, Manganese peroxidase and lignolytic enzymes produced by white rot fungi; *Phanerochaete chrysosporium*, split and degrade plastic polymers into small chain compounds especially those of nylon and polyethylene origins (Shimao, 2001; Da Luz *et al.*, 2012; Nunes *et al.*, 2014; Sato *et al.*, 2017; Oliveira *et al.*, 2018). Low-density polyethylene-based plastics can be easily deteriorated by fungal enzymes; laccase, manganese peroxidase and lignin peroxidase from *P. ostreatus* (Gómez-Méndez *et al.*, 2018). Moreover, Luccase enzyme produced by the fungus *Cochliobolu*s sp has the capacity to degrade polyvinyl chloride-based plastics with *Myceliophthora* sp also being able to degrade polyethylene and polyurethane-based plastics (Loredo-Treviño *et al.*, 2011; Sumathi *et al.*, 2016; Moreno-Bayona *et al.*, 2019).

Penicillium sp produce the enzymes laccase and manganese peroxidase that have been shown to degrade polyethylene and natural rubber-based plastics (Santo *et al.*, 2013; Nayanashree and Tyhippeswam, 2015). However, degradation of microplastics by the fungi community is usually a slow process over many years, it would become unavoidable to subject microplastics to pre-treatments such as ozonolysis, photo-oxidation and solvolysis to initiate polymer degradation prior to fungal biodegradation (Dey *et al.*, 2021). Besides, efficient fungal

biodegradation sometimes requires specific external conditions to be optimal. For instance, the efficiency of plastic biodegradation by marine fungi *Zelerion maritimum* is increased under dark environment and at 25^{o}C (Paço *et al.* 2017).

There are many research gaps of MP and NP removal by fungi. For example, most research work has been based on the visible polymer plastics and not on the micro plastic size. Only a few papers published mentioned research based on the micro-plastics. None of them studied nano plastics. On the other hand, MP and NP type is limited to PE, as no other types of micro plastic have been studied.

Green algae bioremediation

Edible marine microalgae, seaweed; *Fucus vesicu-losus* has been demonstrated to have the high capacity of approximately 94.5% to adsorb polystyrene microplastics (Sundbaek *et al.* 2018). The microalgae can sorb tiny plastic particles due to the surface charge of plastics (Bhattacharya *et al.* 2010; Nolte *et al.* 2017). However, the sorption of microplastics on algae surface largely relies on surface charge of particles (Lionetto *et al.*, 2021). Positively charged microplastics have higher tendency to be sorb more efficiently, due to the presence of an anionic polysaccharide in the algal chemical structure (Mohsen *et al.*, 2020). For instance, unicellular green algae, *Pseudokirchneriella subcapitata* adsorb positively charged polystyrene microplastics more efficiently as opposed to those that are negatively charged (Nolte *et al.*, 2017).

Planktonic crustacean bioremediation

Antarctic Krill can ingest polyethylene microplastics fragments and alter their size (Dawson *et al.* 2018). However, the degradation of microplastics by zooplankton, is reliant abiotic and biotic factors that must first fragment microplastics into smaller sizes (Ter Halle*et al.*, 2016). Marine communities of *Agios consortium* and *Souda consortium* have the capacity to remove high-density polyethylene (Cocca *et al.*, 2017). In the process of degrading Microplastics, these marine organisms, first adhere to microplastics surfaces, which is the first step in microbial polymer degradation, before breaking them down and utilize them as the source of carbon (Sivan 2011; Cocca *et al.*, 2017). Biodegradation of microplastics by marine organisms is unlikely to be efficient. This is due to microplastics' small size and weight that make them carried to and from by the movement of ocean winds thus reducing physical contact with possible degraders (Ivleva *et al.* 2017).

Concluding remarks and perspectives

There is limited information on drinking water contaminated with high concentration of MPs, also the available evidence suggests that they do accumulate in the human body with limited health risks. Hence human intake of MP from drinking water and the public health risk from micro and nano plastics cannot yet be fully confirmed due to the limited research and variation in sampling and analysis methods and lack of research in this area. However, based on the available evidence and the reported low MP concentrations in treated drinking water, microbial and chemical risks to drinking water quality appear to be greater than those presented by MPs.

There are several physical, chemical, and biological methods to remove and destroy micro and nano plastics in the ecosystem, however, there are not widely accepted and there are no cost-effective methods for complete removal of micro and nano plastics from the ecosystem. Also, there is a need to consider microplastics like other contaminants, cycle through ecosystems – natural and engineered, they do not operate in siloes and waste from treatment processes could be a conduit for recontamination of microplastics. Hence reducing plastic use would be the best option.

Within the context of drinking water, some key knowledge gaps and needs are:

- Standardized methods to enable reliable MP assessment and comparison between studies.
- Simulated environmental or field studies to consider transport, fate and removal of MPs (bench top studies do not replicate the multitude of interactions in operational systems or at the appropriate scale)
- Improved detection, characterization and quantification of microplastics in drinking water systems from source to human. intake to ascertain sources and sinks throughout the system (freshwater and drinking water)
- Epidemiological studies to determine the impact of microplastic characteristics (e.g. size, composition) on uptake into tissues following ingestion to inform which particles (if any) in water supplies are most likely to be toxic.

The research on removal of MPs is still limited. However, it is imperative to be cognizant of the following major considerations:

- Identification of transformation products, elucidation of degradation mechanisms, and toxicity assays, as well as computational tools, are needed to improve the assumption of these processes as feasible treatment options.
- Most current studies demonstrating the MP degradation have been performed in bench-scale investigations. It is indeed important to conduct pilot-scale experiments to scale-up for industrial level applications.
- From a microbial perspective, increased understanding of the environmental variables that influence:
    - Formation of biofilm on microplastics (and other plastic surfaces)
    - Composition of micro-plastic associated biofilms
    - Interactions between the microbiome of microplastics and water quality/safety
    - Transporting capacity/degradation of microplastics (in comparison to other particles in water sources)

In conclusion, the presence of microplastics in drinking water systems is a growing concern, and there are significant gaps in our knowledge of their transport, fate, and potential impacts on human health. Addressing these gaps will require a concerted effort from scientists, regulators, and water system operators, and will likely involve the development of standardized methods for assessing and comparing microplastic contamination, as well as the conduct both simulated environmental and field studies to better understand how MPs behave in real-world settings. Moreover, there is a need for improved detection and characterization methods, as well as epidemiological studies to determine risk assessments and identify potential toxicity.

Finally, the role of microplastics in shaping microbial communities and influencing water quality and safety requires further study to better understand their impacts on both human health and environmental sustainability.

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Table 1. Review of key aspects (MP description, treatment process) of studies evaluating efficiency of MPs removal via coagulation method

Reference	Size of MP	Coagulant(s)	Optimal conditions	Removal efficiency
Zhang *et al.* (2021)	< 270 micrometers	Magnetic magnesium hydroxide	200 mg/L of $Mg(OH)_2$, 120 mg/L of Fe_3O_4 (to synthesize magnetic magnesium hydroxide), 5 mg/L of PAM	93.8%
Zhang *et al.* (2022)	50-1000 nm	PAC and PAM	400 mg/L of PAC, 20 mg/L of PAM, and pH of 8	98.5%
Na *et al.* (2021)	10-90 micrometers	Aluminum chloride and iron chloride	Aluminum chloride as coagulant	100% for MP > 20 micrometers
Rajala *et al.* (2020)		Ferric chloride, PAC and PAM	Ferric chloride and PAC ideal coagulants	99.4%
Xue *et al.* (2021)	3-90 micrometers	Alum		> 80% for MPs < 25 micrometers
Shahi *et al.* (2020)	10-100 micrometers	Alum with PC sand	20 mg/L of alum and 500 mg/L of PC sand	92.7%

Table 2. Overview of Micro and Nano Plastics (MNPs) reported as being biodegraded by bacteria

MNPs	Shape & size	Bacteria strain	Sample type	Analysis method	Test duration	Weight loss %	Reference
Low density polyethylene (LDPE)	Sheets	*Bacillus cereus* strain A5,a, *Brevibacillus borstelensis* strain B2,2,	Soil	Weight loss%, FTIR, GC-MS	16 weeks	*Bacillus cereus*: 35.7% *Brevibacillus borstelensis*: 20.3%	(Muhonja *et al.* 2018)
Polyethylene (PE), Polyethylene terephthalate (PET), Polypropylene (PP), Polystyrene (PS)	Grating and cutting of commercial plastics, <250um	*Bacillus cereus, Bacillus gottheilii*	Sediment	Weight loss%, FTIR, SEM	40 days	*Bacillus cereus*: PE 1.6%, PET 6.6%, PS 7.4% *Bacillus gottheilii:* PE 6.2%, PET 3.0%, PP 3.6%, PS 5.8%	(Auta, Emenike, and Fauziah 2017)
Polyethylene (PE)	Granules, 40 – 600um	Mesophilic mixed bacterial: *Bacillus sp.* and *Paenibacillus sp*	Landfill	Weight loss%, FTIR, FE-SEM, GC-MS, TGA	60 days	Weight loss 14.7%, Mean particle size decrease 22.8%	(Park and Kim 2019)
Polypropylene (PP)	Grating and cutting of commercial plastics, 1mm	*Pseudomonas sp*. ADL15, *Rhodococcus sp*. ADL36	Antarctic soil	Weight loss%, FTIR	40 days	ADL15 17.3%, ADL36 7.3%	(Habib *et al.* 2020)
Polyvinyl chloride (PVC)	By cooling grinding obtained 106–300 µm or < 106 µm particles	Engineered *P. aeruginosa*	Artificial seawater freshwater	Weight loss%, SEM, Raman micro-spectroscopy	24hrs	80% trapped PVC by biofilm	(Liu *et al.* 2021)

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Figure 1. Schematic of processes and key stages in the cycling of (micro)plastics within aquatic environments, including key state-of-the-art removal or destruction methods reviewed herein