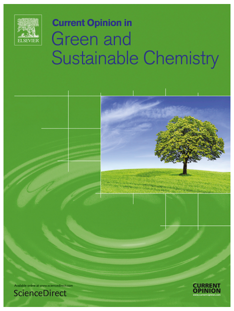

New frontiers in remediation of (micro)plastics

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New frontiers in remediation of (micro)plastics

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Abstract

Plastic pollution is increasing daily, raising social, environmental, and economic concerns. Along with the reduction policies on plastic use and consumption, and improvement of waste management systems, it is of utmost importance to develop and implement remediation and emission control measures. Focused on the most recent literature, this paper provides a critical overview and in-depth discussion on breakthrough technological and biotechnological research that may sustain an effective and efficient (micro)plastic remediation in the near future.

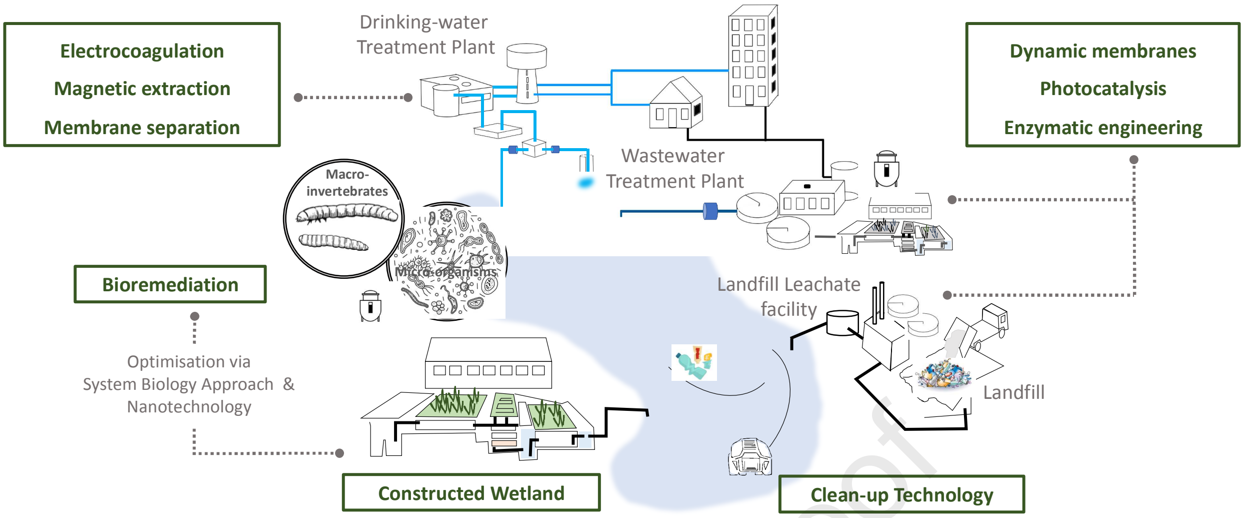
Keywords

Plastic pollution; Cleanup technologies; Biotechnological approaches; Microbial consortium Systems Biology

Highlights

- Plastic pollution calls for mitigation and remediation strategies
- Membranes and enzymatic engineering offer efficacy in microplastics removal
- Biotechnological approaches enhance plastic/microplastic bioremediation
- Combining multiple strategies may solve plastic pollution and microplastic contamination

Graphical abstract



1. Introduction

Plastics (synthetic or semi-synthetic polymers) are essential to modern society, with a wide range of applications in food packaging, medical appliances, electronics, consumer products, among others [1]. While the benefits of plastic use are far-reaching, plastic waste mismanagement and improper disposal are escalating plastic pollution worldwide. The accumulation of plastic waste in natural environments is threatening both animal (e.g., through ingestion, suffocation and entanglement) and human health (e.g., by limiting livelihoods, affecting food security and quality, providing suitable habitats for zoonotic diseases), while imposing an increasing economic pressure to overcome losses in tourism, fisheries and habitats. However, the problem does not stop here, since plastic waste does not biodegrade in open environments due to their resilient polymeric composition. Instead, they undergo deterioration and fragmentation into micro- and nanoplastics by physicochemical processes [2]. Such small plastic debris knows no boundaries and can be easily transported and even carry a panoply of hazardous chemicals and pathogens to a wide range of environments, potentially affecting biota at different biological systems, and consequently threatening ecosystem functioning and services [3,4].

To mitigate the above mentioned social, economic, and environmental impacts of plastic pollution and reduce plastic leakage, several international agreements (e.g., UNCLOS, MARPROL), strategic frameworks (e.g., EU 1st plastic strategy), and enterprise alliances (e.g., the Alliance to End Plastic Waste) have been established; along with the implementation of national or state-wide plastic restriction policies (e.g., SUP ban, deposit-refund, Recycling Act and Compulsory Trash-sorting Policy) (as reviewed by [5]). Although these strategies have resulted in a significant decrease in the use and consumption of plastics and plastic waste generation at regional/local levels, they are still unable to compete with generalized increasing quantities of plastic entering the environment [2,6]. This fact is claiming an urgent need for remediation actions to restore and protect threatened ecosystems by plastic pollution while ensuring their ecological services and functions.

Scientists have been developing different technological and biotechnological approaches to mitigate plastic pollution and reduce environmental contamination by microplastics. With increasing evidence on the successful application of these (bio)technologies in controlled conditions, has increased the need to summarize and critically analyze their potential for field application. Several interesting and detailed reviews have been recently released (e.g., [7-14]), but each one has specific focus (e.g., cleanup technologies, WWTP, WTP, LLT). This paper intends, therefore, to provide a broader overview on the latest technological and biotechnological advances that can pave the path towards an effective and efficient (micro)plastic environmental remediation, and it critically addresses main challenges and potential solutions to overcome them.

2. Technological approaches for plastic debris removal

The presence of plastic debris in aquatic and terrestrial ecosystems worldwide has been highlighted in a significant number of studies (as reviewed by [15,16]). As an attempt to reduce plastic pollution, several cleanup campaigns have been encouraged and implemented, such as the Clean Up The World [17] and The Clean Seas Plastic Challenge [18]. However, cleaning campaigns revealed to be expensive and time-consuming, also requiring a high number of volunteers and specialized personnel (for operative machinery). The application of cleanup technology such as the Interceptor 2.0 (launched in 2019, it uses aquatic surface currents and barriers to redirect plastic trash waste onto its conveyor belt [19]) and Urban

Rivers Trash Robot (launched in 2018, it also uses conveyor belts to move plastics in the hold [20]) revealed to be more effective and efficient in the removal of large plastic items than cleaning campaigns, although mostly designed for aquatic environments [21] (Table I, for more examples see [13]). Nevertheless, the successful implementation of such cleanup technologies relies on their technical optimization to reduce potential adverse environmental impacts such as habitat damage, accidental capture of aquatic fauna and flora, and restriction of organic matter between compartments. The reduction of such environmental impacts can be achieved if cleanup technologies only skim surface plastics (although a compromise, as highly dense plastic debris will not be collected), which will presumably affect the upper few meters of the water column, and thus have a lower encounter rate with aquatic life. In addition, their action could be space-temporarily reduced by targeting specific areas and times where and when plastic densities are high [6]. As examples, existing barriers in freshwaters such as dams and dikes can accumulate high levels of plastic pollution during rainy seasons (personal observation), and tropical gyres during anticyclone eddies [39], thus being interesting spots for plastics collection/removal. The successful application of such cleanup technologies with concomitant waste management improvement, plastic reduction policies, and individual behaviour change towards zero-waste/circular economy was predicted to stabilize the ocean plastic stock in the upcoming years, with less pressure on low- and middle-income countries [6].

These cleanup technologies fail, however, in removing plastic debris of smaller sizes such as microplastics (MPs), demanding for complementary approaches. MPs can result from the abrasion and fragmentation of polymeric materials or being produced at the microscale (e.g., microbeads) [22]. With a significant portion coming from inland activities, these MPs can end up in landfills and wastewater streams, or find their way into freshwater systems (directly or indirectly via soil leachates). Some traditional operations, such as ozonation, coagulation/flocculation, rapid sand filtration and dissolved air floatation commonly carried in wastewater treatment plants – WWTP and in landfill leachate facilities (LLF) are able to retain part of MPs (particularly of higher size and density [23]). However, small-sized MPs can pass untreated through both WWTP and LLF, being released to natural environments [14, 24].

Several innovative technological approaches have been developed with the intuit of reducing the release of these particles to natural environments and consequent involuntary consumption by organisms and humans, by improving WTP, WWTP, and LLF [8, 14]. Designed as an "add-on" technology or "secondary and tertiary treatment facilities", recent innovative technologies proved efficiency but still present challenges than need to be overcome in the near future (Table I). For instance, membrane bioreactor technology proved their ability to remove a considerable amount of MPs (> 90%) from aqueous solutions than conventional methods in WWTP [15,25], but such filtration modules are relatively expensive, and can easily blocked, resulting in higher operating costs. Nevertheless, the recovery of MPs from tertiary treatment employing bioreactor technology are practically clean (as most adsorbed matter would have been removed in previous steps); thus, they have the potential to be revalorized (upcycling).

Dynamic membranes are also considered to be a promising technology in removing MPs particularly of low density, but its performance is also delimited by several factors such as membrane materials, pore size, deposited materials and operating conditions (pressure, cross-flow velocity, hydraulic retention time, temperature, among others) [23]. Nevertheless, with breakthroughs in membrane technology and membranes for nanofiltration, a blend of reverse osmosis and ultrafiltration is currently emerging, which promises to remove MPs more effectively and without clogging [25].

The application of electrochemical techniques (e.g., electrocoagulation, where the liberation of metal ions from electrodes will form coagulants that destabilize the surface of MPs) [24], or the application of magnets or adsorbates that will adhere and cluster MPs for removal (e.g., magnetic carbon nanotubes and inorganic-organic hybrid silica gels – organosilanes), seem to be efficient and eco-friendly alternatives in WTP or WWTP, particularly in the removal of persistent polymers such as polyethylene, polyethylene terephthalate, and nylon (up to 90% removal efficiency) [26]. However, they require optimization (from application to end-products treatment) and plea competitive costs before their widespread implementation. The combination of granular activated carbon filtration, high doses of $FeCl_3.6H_2O/AlCl_3.6H_2O$ coagulants, and ultrafiltration through polyvinylidene fluoride (PVDF) membranes have been highlighted as vital treatment methodologies for removing small-sized MPs, which is of high interest particularly for WTP [27].

Photocatalytic degradation (e.g., with zinc oxide) also stands out as a promising, viable, and energy-efficient method for MPs degradation in WWTP and LLF [28]. During the photocatalytic degradation process, the MP surface will face a direct attack by highly oxidizing radicals that will cause polymeric chain oxidation and breakage. The decomposed chains are leached from the bulk as an intermediate and simpler compound that can be further used as raw materials for organic synthesis. The major drawback of this procedure is related to the maintenance costs, limiting the widespread practical application of these technologies. Furthermore, some end-products from photocatalytic degradation may impose a risk to both animal and human health, limiting their application to WWTPs or LLFs. Nevertheless, the application of highly efficient sources of UV radiation, the improvement of the reactor design itself, and the use of catalysts that absorb radiation from the visible spectrum may overcome these challenges.

Another promising approach recently used as a barrier for MPs leakage is horizontal subsurface flow constructed wetlands (CW) (a.k.a., engineered treatment wetlands, which consists of a horizontal subsurface flow that uses vegetation, soil, and organisms to treat the wastewater) (Table I). CW is not new as it has been used as a substitute for WWTP (in small villages) or as a tertiary treatment facility in municipal WWTPs (in city-capitals). The application of CW has proved efficiency in processing total suspended solids and remediating hazardous chemicals; thus, it may also contribute to MP removal or prevent their leakage to natural environments. A recent study carried out by Wang et al. [29] revealed that CWs indeed has the potential to efficiently reduce MPs concentration (88% efficiency, as MPs were ingested and accumulated by the macroinvertebrates [specifically by Tubificidae annelids] with negligible effects), preventing them from entering vulnerable aquatic systems. CW can even be improved if their macro- and microorganisms are capable not only to ingest but to biodegrade MPs, without toxic effects on their fitness (as further explored in section 3). Considering the use of macroinvertebrates, it remains unclear if the depuration of the organisms in controlled conditions would be feasible (at first sight, it might), to avoid their sacrifice.

3. Plastic-degrading organisms for *in-situ* remediation purposes

Some macroinvertebrate species proved to be able to degrade plastics. The mealworms (*Tenebrio molitor*) and larvae of the greater wax moth (*Galleria mellonella*) seem to chew and digest the plastic, such as low-density polyethylene plastic bags [30]. Such capacity relies on their gut microbiota [30]. The biotechnological potential of these macroinvertebrates (or others under investigation) and their microbiota represents an open window to develop biotechnologies with the potential for bioremediation or to expedite the development of biochemical recycling technologies. However, in the current status and from the technological point of view, the application of macroinvertebrates is uncertainty, as raising larvae can be too slow and costly (i.e., requiring regular maintenance for habitable conditions) when compared with their effectiveness on plastics degradation. Furthermore, it remains unclear if they contribute for MPs production rather than biodegradation (which is problematic and studies to date do not exclude the possibility of this unintended consequence) [30].

Various microorganisms inhabiting soils, landfills, aquatic environments, and wastewater sludge, also proved to be able to degrade plastics (i.e., use them as a substrate and source of nutrients) as a result of their capacity to degrade natural organic polymers. For instance, the fungal genera *Aspergillus* and *Penicillium* and the bacterial genera *Pseudomonas* and *Bacillus* have been involved in the degradation of more than ten types of plastics, including recalcitrant ones such as polyethylene, polystyrene, polyethylene terephthalate [31,32]. Isolated strains are capable of biodegrading plastics, but bacterial consortia or biofilm (which combines, among other organisms, bacteria and fungi) offer higher efficiency in the biodegradation processes – where some strains are involved in the deterioration and degradation process, and others are responsible for eliminating toxic metabolites excreted by the counterparts – ensuring complete mineralization [25, 31, 33].

The success of plastic biodegradation by microbes rely, however, on their metabolic processes and enzymatic system (i.e., on both extracellular and intracellular enzymes). The amount and type of enzymes present in microorganisms vary with species and even between strains of the same species, and they are very specific in their action on substrates; thus, effective biodegradation implies the action of multiple enzymes. So far, 79 extracellular enzymes have been involved in polymers chain-scission (into oligomers-dimers-monomers), such as: lignin-degrading enzymes (laccase, manganese-dependent peroxidase); lipase, esterase, and cutinase; depolymerase (generally followed by the action of proteases, such as protease K and trypsin); hydrolase (urease, protease, and lipase); and oxygenase (monooxygenases and dioxygenases) [31,32]. Recently, an enzyme highly homologous to several cutinases – PETase, was isolated from the bacteria *Ideonella sakaiensis* 201-F6 and displays outstanding performance in hydrolyzing PET into monomers, holding an excellent potential for the bioconversion of plastics [34].

After the action of the extracellular enzymes, monomers sufficiently small will cross the microbial cell membrane, where they will be oxidized via the catabolic pathway (i.e., the action of intracellular enzymes), and the energy produced will be used to form new cell biomass [31]. The incorporation of the atoms into the microbial cell (i.e., assimilation) and the secondary metabolites formed as a result of this process will be excreted and probably used by other microorganisms. When plastics biodegradation occurs in the presence of oxygen (i.e., aerobically), it releases carbon dioxide and water, whereas, in the absence of oxygen (i.e., anaerobically), the main end products generated are the same nature along with the additional generation of methane, while in sulfidogenic condition, the formation of hydrogen sulfide [35]. Yet, some secondary metabolites with potential for environmental hazardousness can be released; thus, the application of microorganisms as bioremediators must be carefully evaluated (as they must constitutes a lower risk than the primary contaminants).

Although biodegradation of plastics by natural microorganisms seems to offer a low-cost and eco-friendly remediation approach, it remains a slow process as it is highly dependent on several factors such as polymer properties and surface area, and environmental factors (biotic and abiotic) [33]. One approach to promote plastic bioremediation is through biostimulation (with the application of growth supplements, fertilizers, natural surfactants, and nanoparticles, along with the optimization of environmental requirements) and/or bioaugmentation [36]. Another approach includes applying modern biotechnological techniques such as protein/enzyme engineering (for purification/stabilization and industrial production). For instance, the engineered PETase mutants R61A, L88F, and I179F (created through the rational protein engineering of key hydrophobic sites) increased PET degradation by 1.4-fold, 2.1-fold, and 2.5-fold, in comparison to wild type strain [35]. A modified cutinase enzyme Tcur1278 (the modified version has an anchor peptide Tachistain A2 – that acts as an adhesion promoter for Tcur1278) significantly increased the degradation of polyester-Polyurethane [37].

The development of optimized or synthetic microbial consortium, the application of genetic engineering, systems biology (i.e., application of multi-omics – genomics, proteomics, metabolomics), and the development of genetically modified organisms are also under study/consideration as potential solutions to improve plastics biodegradation processes [31]. As a simple example, an optimized/tailored microbial consortia revealed the ability to thrive in the presence of mixtures of plastics and participate more efficiently in their degradation compared with the original consortia [38,39].

The use of genetically modified organisms or tailored microbial consortia, along with the discovery of new enzymes, metabolic pathways, and metabolically active molecules with improved functionalities (via system biology approach), will boost the advance of modern biotechnological tools for plastic remediation and, potentially, recovery [31]. However, the implementation of such novel approaches is not without challenge. The introduction of modified strains or organisms to the ecosystem may cause an unmeasurable adverse effect on the natural structural and functional microorganism's community composition and occurrence. Therefore, these processes must be carefully considered to be applied in bioreactors (ex-situ bioremediation) instead of natural environments. However, when *in situ* is required, the use of modified strains or organisms must be firstly optimized (in a laboratory, micro, and mesocosms conditions) and planned (i.e., considering controlling measures) before their application in impacted environments.

4. Main conclusions and future directions

Plastic pollution is imposing a severe threat to the environment, and it is escalating year after year, calling for remediation strategies. Several innovative cleanup technologies prove their efficiency in removing plastic debris from aquatic environments (and aqueous solutions) to be further applied as add-ons or additional treatments to WTP, WWTP, and LTF. Besides, biotechnological approaches such as genetic engineering, systems biology, and the synthetic microbial consortium have been developed and implemented to enhance plastic biodegradation and have been raised as a promising solution for *in-situ* or *ex-situ* bioremediation. Nevertheless, they all need optimization to avoid increased environmental problems and need to improve their cost-effectiveness.

While recognizing the positive impacts of each strategy/approach, none offers a "fit-of-all solution" due to the complexity of factors intimately related to plastic debris/microplastics removal/biodegradation. The combination of membrane and enzyme technology, along with biotechnological techniques such as genetic engineering, systems biology, and the development of synthetic microbial consortium, will provide better results than individual solutions and should be put in place in a short-term scenario.

However, these innovative remediation approaches do not solve plastic pollution and must be accompanied by effective mitigation strategies that focus on source reduction to avoid continuous cleanup and remediation efforts. This could be achieved by i) straightening plastic reduction policies emphasizing a decreasing use and consumption of plastics (along with extended producer responsibility, shared environmental responsibility principle, and the implementation of incentives for recycling and redesigning [38]; ii) optimizing waste management infrastructures, iii) seeking sustainable plastics by scaling up in innovation to ensure their environmental friendliness and their integration in the circular economy; iv) increasing public awareness on plastic pollution along with a behavioural-shift to entail the implementation of good and sustainable practices.

Conflict of Interest

The author declares no conflict of interest.

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(*) references indicated as being papers of special interest.

Table I: An overview of main processes for plastics and microplastics removal or elimination found in literature.

	Macropastics (aim at preventing and collection)		**Microplastics (aim at removing)**			
	Surface waters (as reviewed by [13])	**Terrestrial environments**	**Wastewater treatment plants** (as reviewed by [15, 23])	**Landfill leachates facilities** (as reviewed by [14])	**Treatment water facilities** (as reviewed by [11])	**Ex-situ/in-situ bioremediation** (as reviewed by [25, 31, 33]
Traditional approach	Boats & nets	Cleanup campains (collection)	Chlorination disinfection (7%) Coagulation/flocculation (47-82%) Rapid sand filtration (45-97%) Dissolved air floatation (up to 95) Oxidation dicth (up to 97%, size > 25 µm) Ozone (90%, particularly at temperatures > 35 ºC) Conventional activated process (up to 95.6%, size 20-5000 µm)	Artificial soil filtration (~98%, size 50-500 µm) Sand bed filtration (70%, size 50-500 µm) Electrochemical oxidation*	Coagulation/flocculation + sand filtration (70%) Coagulation/flocculation + sedimentation + sand filtration + activated carbon filtration (81%) Coagulation/ flocculation + flotation + sand filtration + activated carbon filtration (83%)	Biostimulation* Bioaugmentation*
Existing advance approaches	Stormwater and wastewater filters (prevention: e.g., StormTrap TrashTrap) Laundry balls (Prevention; e.g., coral ball) Drones and robots (collection, e.g., FRED) Boats and wheels (collection; e.g., Interceptor) Detection aids (detetion/collection; e.g., NetTag)		Sequencing batch reactor (up to 98%) Reverse osmosis (90% removal) Membrane biological reactor (up to 99%, size > 2 Dissolved air flotation (up to 95%) Ultrafiltration (42%) Microscreen filtration with disc filters (DFDS) * Electrocoagulation (up to 99% at pH 7) Coagulation/floculation (47-82%, size < 1.2 µm)	Sequencing Batch Biological Reactor (100%; size 50-500 µm) Reverse osmosis* Advanced oxidation processes(e.g., photo-Fenton, O3/UV, H2O2/UV, ultrasound (US), UV/US, and H2O2/US)	Electrocoagulation* Magnetic extraction (up to 98%, size 200-1 Membrane separation*	Fungi and bacteria species (e.g., *Ideonella sakaiensis degrades 0.13 mg PET·cm−2·day−1*) Microbial consortium (e.g., Bacillus cereus, Bacillus pumilus and Arthrobacter; up to 22% PE weight loss in 14 days)
Emerging approaches		Bioremediation (elimination)	Blend of reverse osmosis and ultrafiltration Dynamic membranes (99%) Photocatalysis (e.g., 98.40% for 400-nm OS) Elimination with fats* Constructed wetlands (88%) Biological degradation	Thermochemical technologies using supercritical water* Biological degradation*	Granular activated carbon filtration, high doses of $FeCl_3.6H_2O/AlCl_3.6H_2O$, and ultrafiltration through polyvinylidene fluoride (PVDF) membranes (up to 40%)	Multiomics - System Biology Approach* Nanotechnology (e.g., Nano barium Titanate, Fullerene 60, Super magnetic iron oxide + bacteria interaction; higher efficiency degrading PE)

* identified as potential (bio)technological approach, although not tested under the target circumstances.

Declaration of interests

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

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: