Eco-Environment & Health 4 (2025) 100138

Contents lists available at ScienceDirect

Eco-Environment & Health

journal homepage: www.journals.elsevier.com/eco-environment-and-health

Review

The wheel of time: The environmental dance of aged micro- and nanoplastics and their biological resonance

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ARTICLE INFO

Keywords:
Microplastics
Nanoplastics
Aging effects
Environmental behavior
Biological effects

ABSTRACT

The aging of micro- and nanoplastics (MNPs) significantly affects their environmental behavior and ecological impacts in both aquatic and terrestrial ecosystems. This review explored the known effects of aging on MNPs and identified several key perspectives. Firstly, aging can alter the environmental fate and transport of MNPs due to changes in their surface properties. This alteration accelerates their accumulation in specific habitats like oceans and soils, resulting in increased bioaccumulation by organisms. In addition, aged MNPs interact differently with living organisms than their pristine counterparts by influencing the attachment of biofilms and other microorganisms in aquatic ecosystems. Moreover, the aging processes of MNPs exhibit adverse effects on aquatic and terrestrial organisms via increasing the bioavailability and potential toxicity of MNPs as degradation products are released. Last but not least, the biodegradation potential of MNPs can be altered by the aging process, thus affecting their degradation rates and pathways in the environment. However, there are still knowledge gaps regarding the natural aging behaviors of MNPs, such as the aging mechanisms of different types of plastic, the influence of environmental factors, the release of pollutants, and even the effects of aging on their transformation in different ecosystems. Therefore, a great contribution can be made to sustainable plastic use and environmental preservation by studying the natural aging of common MNPs and their subsequent biological effects.

1. Introduction

Plastic pollution is currently a significant environmental issue, with plastic debris persistently appearing in the environment [1]. It is estimated that by 2050, plastic products will generate 2.5×10^{11} tons of waste [2]. Microplastics (MPs), defined as plastic particles with diameters ranging from 1 μm to 5 mm, and nanoplastics (NPs), typically sized between 1 and 1000 nm, represent the two primary forms of plastic pollution [3–6]. Primary micro- and nanoplastics (MNPs) originate directly from plastic products, while secondary MNPs are generated through the degradation and fragmentation of larger plastic items [7–9]. These particles have been found in various environmental compartments, e.g., air, ocean, soil, sediment, surface water, and groundwater worldwide [10–19].

As reported in published literature, MNPs show negative impacts on organisms [20–23]. For instance, exposure to MNPs could cause lung and intestinal injuries in organisms, which are commonly accompanied by oxidative stress, cell damage, and inflammation [24–28]. Moreover, considering their small size, MNPs have been reported to penetrate the cell membrane and the blood–brain barrier, subsequently resulting in migration and accumulation in different tissues, organs, and even offspring [29–32]. In addition, the uptake of MNPs alters the reproductive capability of organisms, contributing to the reduced offspring number and size, as well as the increased abnormality frequency [33]. Generally, plants can absorb NPs rather than MPs, and the accumulation of NPs in plants is influenced by the surface charge of the NPs [34]. It was reported that NPs could induce the accumulation of reactive oxygen species (ROS) in plants, which could account for the inhibition of growth and seedling development [35,36].

In the natural environment, plastics undergo abiotic and biological aging, leading to progressive fragmentation and production of MNPs [37]. It is estimated that a single plastic item with a diameter of 200 mm and a thickness of 0.20 mm can produce approximately 6.25×10^4 microparticles of ~0.80 mm in diameter [38]. Moreover, biodegradable

Given his role as an Editor, Cheng Gu had no involvement in the peer review of this article and has no access to information regarding its peer review.

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https://doi.org/10.1016/j.eehl.2025.100138
Received 26 June 2024; Received in revised form 1 December 2024; Accepted 11 February 2025
Available online 14 February 2025
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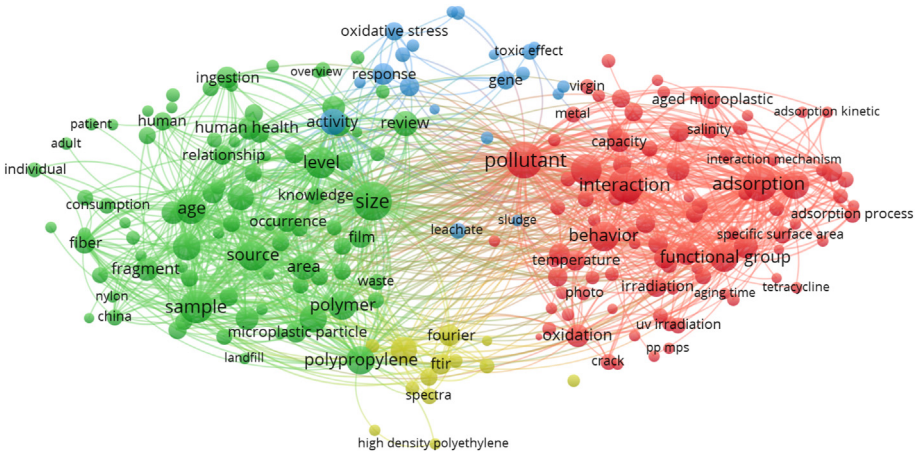

Fig. 1. Keyword network in articles about aging MNPs.

plastics exposed to UV radiation were reported to produce more MNPs than non-degradable plastics [39]. Subsequently, MNPs in the environment undergo continuous aging processes, which inevitably alter their physicochemical properties, including changes in surface morphology, particle size, oxygen content, and surface charge [40,41]. These variations subsequently affect their mobility, adsorption, and colloidal stability [42–46]. Aging can also promote or suppress the aggregation of MNPs, depending on various factors, such as surface charges and the presence of organic matter [47]. Moreover, surface oxidation and microcrack formation are common outcomes of the natural aging process, which significantly influence the adsorption behavior of MNPs [48,49]. Due to the formation of interfacial hydrogen bonding and photo-generated ROS, aged MNPs can act as catalysts to influence the transformation of coexisting emerging contaminants [50–52].

In addition to the physicochemical properties of MNPs, the aging processes can also change their toxicity. It was reported that aged MNPs exhibit increased toxicity due to the changes in surface functional groups that affect their interactions with algae [53]. Moreover, the exposure risks of aged MNPs would be strengthened with their enhanced adsorption capacities for coexisting contaminants [54–56]. However, a recent study provided an opposite conclusion that the aging process could mitigate the cytotoxic effects of MNPs for the different adsorption of plasma proteins and the reduced cellular uptake induced by the oxygen-containing moieties on the aged MNP surfaces [57]. Therefore, the effects of aging on the biological impacts of MNPs remain unexplored, presenting a significant research opportunity.

In order to systematically summarize the related research on aging MNPs, the Web of Science database was used to retrieve the relevant publications from 2021 to 2025 with the following keywords: aging micro- and nanoplastics, resulting in 933 records that were analyzed for keyword networks. As shown in Fig. 1, the two major research hotspots are the interactions between MNPs and pollutants (primarily focusing on adsorption) and the MNPs' occurrence and distribution. However, the keyword analysis indicates relatively little research activity on the biological effects of aging MNPs. Furthermore, the keywords “bioavailability” and “biodegradable plastic” appear only 28 and 20 times, respectively, highlighting limited studies on the biodegradation and aging processes of biodegradable MNPs. This suggests that these topics need further exploration and more attention.

The objectives of this review are to (i) summarize the common aging modes of MNPs considering their crucial roles in comprehending the transformation of MNPs; (ii) introduce the effects of natural aging on the environmental behavior of MNPs, e.g., migration ability, adsorption/desorption capability, and catalytic degradation performance; (iii) provide an overview of the effects of aging on the toxicity and biodegradability of MNPs. By summarizing the current states of research and knowledge gaps, this review serves as a reference to guide future studies about the effects of aging on the environmental behavior and biological impacts of MNPs.

2. Nonbiological aging mode of MNPs

Similar to large-sized plastics, MNPs undergo various nonbiological aging processes when exposed to the environment. As shown in Fig. 2, these processes are typically categorized into mechanical aging, photoaging, and thermal aging based on different mechanisms [8,58,59]. Mechanical aging of MNPs refers to the effects of environmental forces, including wind, waves, and sand abrasion [49,59,60]. Photoaging occurs when MNPs are exposed to sunlight or artificial light sources [61]. Meanwhile, thermal aging involves the impact of elevated temperatures

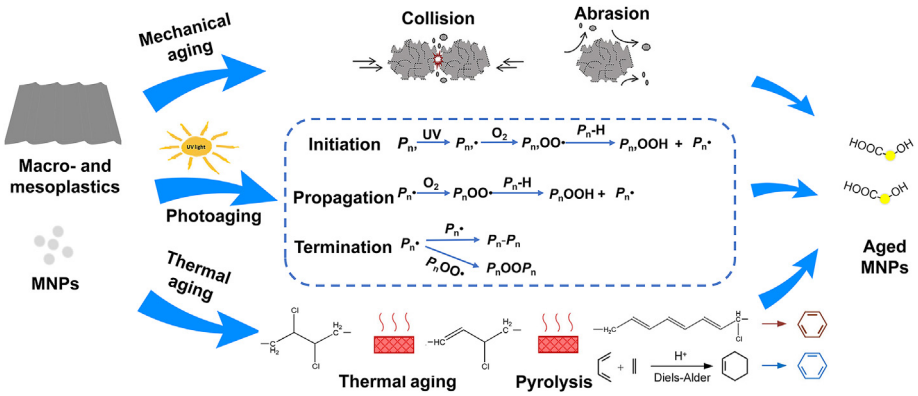

Fig. 2. Summary of the mechanical, photo-oxidative, and thermal aging processes of macro- and mesoplastics and MNPs.

on MNPs, accelerating their degradation and inducing structural and property changes [62]. Thus, understanding the various aging mechanisms is crucial for comprehending the transformation and behavior of MNPs in aquatic and terrestrial environments. These aging processes significantly affect the physical and chemical properties of MNPs, ultimately influencing their interactions with organisms and ecosystems.

Mechanical aging is a prominent process in the environmental fate of MNPs. When these particles enter the environment, they are exposed to various mechanical forces that contribute to their aging. Environmental forces, e.g., pebbles, waves, tides, and contact with other plastic debris, subject the MNPs to constant physical stress [62]. In aquatic environments, the primary environmental forces are the friction and disintegration forces between MPs and sediments, pebbles, waves, and tidal actions [62]. In terrestrial environments, these forces may arise from the collision and abrasion of MNPs with soil and sand or be driven by human activities such as soil tillage and crop rotation [63,64]. Under the influence of these mechanical actions, the particles can undergo cracking, breakage, and fragmentation, leading to the generation of smaller-sized particles. This process increases the overall number of plastic particles in the environment as the larger MPs are fragmented into multiple smaller fragments. The generated small particles exhibit enhanced mobility and can disperse more widely within the ecosystem [46]. Moreover, smaller particles generally have a higher specific surface area, which can result in increased interactions with other environmental components, such as organic matter, nutrients, and pollutants [59,65]. Furthermore, the increased number of smaller particles poses challenges for remediation and removal efforts, as they become more difficult to manage and remove from the environment.

Photoaging is a crucial process for the degradation of MNPs [61]. It can be divided into three stages: initiation, propagation, and termination [66–69]. As shown in Fig. 2, during the initiation stage, ultraviolet (UV) light is absorbed by the unsaturated chromophoric groups in the polymer structure, breaking C–H bonds and generating polymer radicals (i.e., $P_n\cdot$). Then, in the propagation stage, $P_n\cdot$ reacts with oxygen to form peroxy radicals ($P_nOO\cdot$), triggering a series of complex free radical reactions that lead to autoxidation, chain scission, or chain crosslinking. Finally, the chain reaction terminates when free radicals combine or the polymer chain cleaves, forming inert products, e.g., olefins, ketones, and aldehydes.

Interestingly, it has been reported that photoaging affects MPs and NPs to varying degrees. For example, pristine polystyrene (PS) NPs produce more ROS upon light irradiation compared to PS MPs [70]. This may be attributed to the larger specific surface area of NPs, which provides more reaction sites and surface defects for UV absorption and radical formation [70]. In the photodegradation process of PS NPs, smaller particles exhibit higher reactivity, resulting in the production of formic acid, benzoic acid, lactic acid, and benzaldehyde compounds [71]. Additionally, photoaging alters the colloidal stability of NPs by modifying the surface charge density [47]. Therefore, the photoaging process of MNPs is essential for their transformation, transport, and potential biological impact.

Thermal aging of MNPs is an oxidation reaction that occurs when the particles are subjected to high temperatures. The elevated temperature enables the polymer structure to surpass the dissociation energy of the chemical bonds between the molecular chains, resulting in the random breaking of these chains and the shedding of branched chains [72]. This process generates alkyl radicals (R·) that follow a similar autoxidation cycle to that observed in photoaging [62]. In a terrestrial environment, dark dry soil can reach temperatures as high as 90–100 °C [73]. Consequently, MNPs may undergo thermal aging on land due to the extreme surface ground temperature. In addition to high-temperature thermal aging, the freeze–thaw cycle is also a crucial natural aging process in the environment [74]. During this cycle, MNPs experience changes in surface morphology, hydrophobicity, and crystallinity [75]. Sun et al. [76] have shown that freeze–thaw cycles disrupt the amorphous regions of polyethylene (PE), reduce the number of micropores on its surface, and decrease the adsorption of atrazine by PE. It is important to note that unlike photoaging, which primarily affects the surface of the particles, thermal aging occurs throughout the pellet of MNPs in a high-temperature environment. Interestingly, MNPs rarely undergo a purely thermal aging process in natural settings. Thermal aging often coexists with photoaging, and the increased temperature accelerates the rate of free radical generation under UV light irradiation, thereby expediting the aging of MNPs [77]. Currently, there is still a knowledge gap regarding the specific interaction between the thermal and photoaging processes of MNPs.

3. Effect of aging on the environmental behavior of MNPs

As shown in Table 1, the aging process profoundly affects the physicochemical properties of MNPs, such as surface roughening [42,78], fragmentation [46,49,79], changes in crystallinity [80] and color [49,81], increased oxygen-containing functional groups [44,82,83], enhanced hydrophilicity and surface electronegativity [61,84], formation of environmentally persistent free radicals (EPFRs) [85], and enhanced fluorescence [86]. As the physicochemical properties of MNPs are altered during aging, several key aspects of their behavior undergo significant changes. One important aspect affected by aging is the

Table 1
Role of aging in affecting MNPs' properties and environmental behavior.

Type of MNPs	Aging process	Changes in the physical and chemical properties of MNPs	Environmental behavior of MNPs	Ref.
PBAT, LDPE	UV	Smaller size	Promote the diffusion of MNPs in aqueous environments	[93]
PS	UV or O_3	Surface oxidation, enhanced surface charge negativity & hydrophilicity	Accelerate MNP transport in saturated loamy sand	[46]
PS	UV	Increased O-containing functional groups	Inhibit PS aggregation in NaCl but promote it in $CaCl_2$ solutions	[47]
PS and PVC	UV	More oxygen-containing functional groups	Enhance the affinity of hydrophilic contaminants	[94]
PS	UV	Formation of cracks & surface oxygen-containing functional groups	Inhibit the adsorption of hydrophobic pollutants	[95]
PBAT, PS, PP, and PE	UV & potassium persulfate thermal activation	Generated functional groups, enhanced electrostatic interactions & hydrogen bonds	Improve adsorption of polar contaminants	[96]
PS	High temperature	Formation of oxygen-containing surface groups & hydrogen bonds with surrounding water molecules	Inhibit the adsorption of non-polar pollutants	[83]
PET	UV	Increased specific surface	Improve adsorption of heavy metal ions	[97]
PS and PF	Light irradiation	Formation of EPFRs	Enhance the formation of ROS & facilitate the degradation of coexisting pollutants	[85]
PP, PE, PS and PVC	UV	Enhanced fluorescence	Render aged MNPs efficient fluorescent tracers & enable their fluorescent visualization in various organisms	[86]

EPFRs, environmentally persistent free radicals; LDPE, low-density polyethylene; PBAT, poly(adipate-co-terephthalate); PE, polyethylene; PET, polyethylene terephthalate; PF, phenol-formaldehyde resin; PP, polypropylene; PS, polystyrene; PVC, polyvinyl chloride; ROS, reactive oxygen species; UV, ultraviolet.

migration capacity of MNPs. Aging can lead to increased surface roughness and fragmentation, which can enhance the mobility of MNPs in the environment [87]. The adsorption property of MNPs is also influenced by aging. Surface roughening and the formation of oxygen-containing functional groups during aging can change the adsorption capacity of MNPs for various organic and inorganic contaminants [88,89]. Moreover, aging processes can affect the release of additives from MNPs, e.g., plasticizers, flame retardants, and stabilizers, which can leach out over time [90]. Furthermore, aging shows great potential to impact the catalytic activity of MNPs. The presence of cracks, defects, and oxygen-containing functional groups on the aged MNPs' surfaces can promote catalytic reactions, thus influencing environmental processes, e.g., oxidation, degradation, and transformation of coexisting contaminants [91,92]. Overall, the aging processes of MNPs have far-reaching consequences on their environmental behaviors. Understanding these changes is crucial for assessing the fate of transport and potential risks associated with the aged MNPs in the environment.

3.1. *Effect of aging on the migration ability of MNPs*

As illustrated in Fig. 3, the aging process of MNPs can significantly impact their interaction with different environmental compartments, particularly in terms of migration ability and stability. For instance, the particle size of MNPs has been found to influence their penetration rate in porous media [98]. The accumulation of cracks from aging has the potential to cause fragmentation of MNPs into smaller particles [59,98], thus enhancing the penetration of MNPs in saturated quartz sand media [98]. Additionally, the presence of oxygen-containing moieties generated during aging treatment enhances the surface charge negativity and hydrophilicity of MNPs, which could accelerate their transport in saturated loamy sand [46]. It was reported that environmental factors, such as flow rate, ionic strength, and cationic valence, can further influence the mobility of aged MNPs in saturated porous media [5,99]. By comparison with their pristine counterparts, the mobility of aged PS NPs could be greatly enhanced in the presence of Mg(II) and Ca(II) cations, which could be attributed to the lower zeta potential of the aged PS NPs than the pristine PS NPs in the presence of Mg(II) and Ca(II), resulting in the increased repulsion with the quartz sand. However, Al(III) cations could significantly inhibit the transport ability of PS particles in the sand column by forming complexes with oxygen-containing moieties on aged PS NPs' surfaces, which enhance the interparticle interactions and further increase the size of PS NP aggregates [100]. Therefore, aging can significantly alter the transport characteristics of MNPs in soil and soil-water media.

Moreover, the stability of MNPs in solution also varies with the aging of MNPs. Liu et al. [47] demonstrated that the aging process inhibited PS NPs aggregation in NaCl solution by increasing the negative charge on the PS NP surface and the organic matter content in solution while promoting PS NPs aggregation in $CaCl_2$ solution due to the interaction between Ca(II) and carboxyl groups formed on the aged PS NP surface. This aggregation process would lead to the deposition and accumulation of MNPs in sediments of aquatic environments. Interestingly, Wei et al. [93] conducted a comparative study on the formation of MNPs from biodegradable poly (adipate-co-terephthalate) (PBAT) and the non-biodegradable low-density polyethylene (LDPE) in various aquatic environments. The study revealed that PBAT yielded a greater number of plastic fragments/particles than LDPE in all tested aquatic environments following UV aging [93]. Moreover, MNPs derived from biodegradable plastics (bioMNPs), including PBAT, polylactic acid (PLA), polyhydroxyalkanoate (PHA), and poly (3-hydroxybutyrate) (PHB), possess densities higher than that of water, while several non-biodegradable MNPs have densities that are lower than or equivalent to that of water [101]. Furthermore, compared to non-degradable MNPs, bioMNPs exhibit higher polarity, which enhances microbial colonization on their surface, thus promoting their suspension in water [102]. Consequently, the risk posed by bioMNPs is considerable and necessitates further evaluation over extended time periods, considering the biological fate of intermediate and final products in freshwater, estuarine, and marine ecosystems.

3.2. *Effect of the aging process on the adsorption performance of MNPs and the release of additives from MNPs*

Aging processes lead to various changes in the physicochemical properties of MNPs, which subsequently influence their interactions with contaminants and other environmental components (Fig. 4). As mentioned above, the generated oxygen-containing functional groups after aging treatments can increase the hydrophilicity of MNP surfaces [103], which can inhibit the adsorption of hydrophobic pollutants while enhancing the affinity of hydrophilic contaminants [68,94,95]. Additionally, the generated functional groups on the aged MNP surfaces are able to influence the polar interactions, e.g., intermolecular hydrogen-bonding force, which plays a crucial role in the adsorption of some polar compounds. It was reported that the presence of oxygenated portions on MP surfaces of aged PS, polyvinyl chloride (PVC) and PBAT could form hydrogen-bonding with ciprofloxacin and tetracycline,

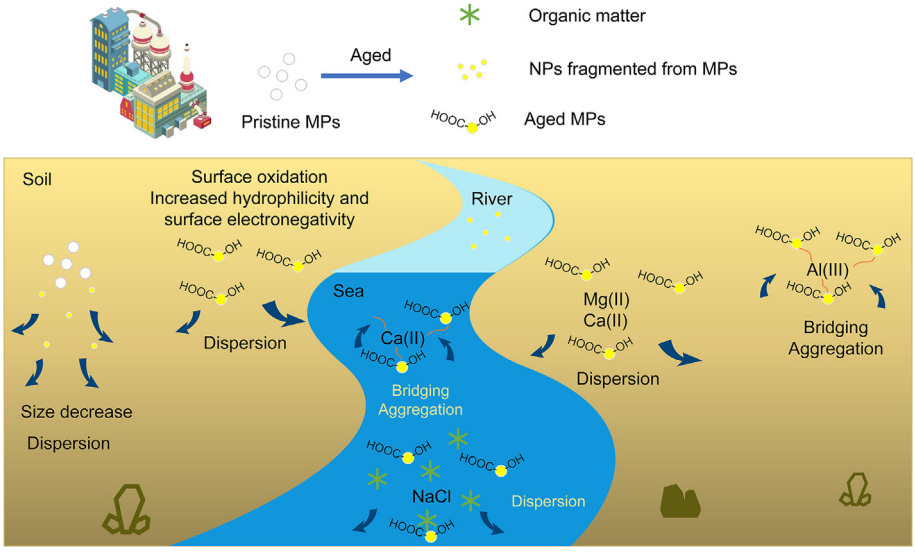

Fig. 3. The effect of aging on the mobility and migration of MNPs.

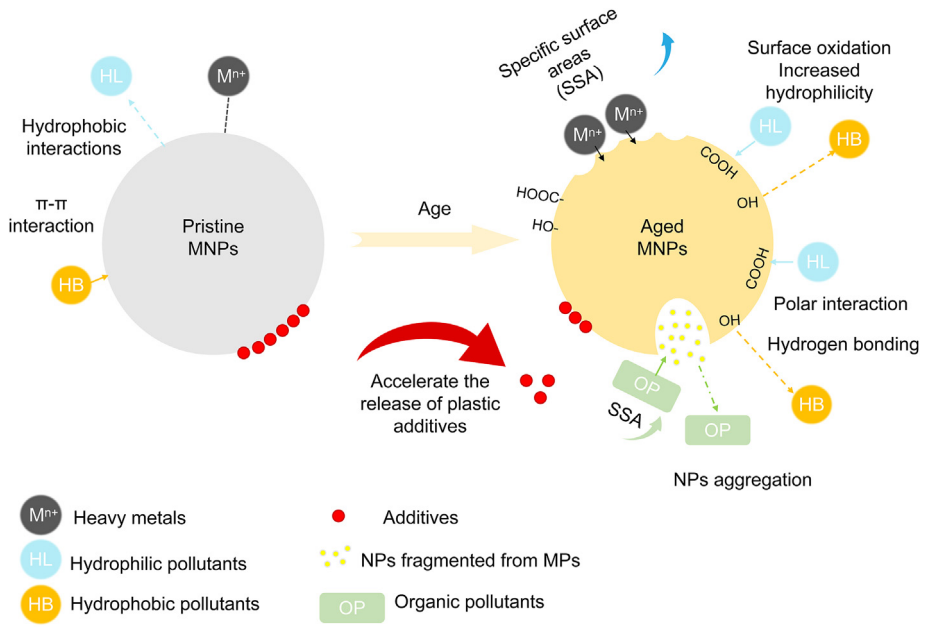

Fig. 4. The effect of aging on the adsorption of contaminants by MNPs and the release of additives from MNPs.

respectively, which improved drug adsorption onto MPs [94,96]. However, the enhanced hydrogen-bonding force between aged MPs and water molecules inevitably inhibits the adsorption of polycyclic aromatic hydrocarbons, which represent non-polar pollutants, with decreased adsorption sites on the MP surfaces [83].

Moreover, material fragmentation during aging increases the specific surface areas of MNPs, thus providing more adsorption and reaction sites for coexisting substances [104]. Wang et al. [97] found that the adsorption of heavy metal ions [Cu(II) and Zn(II)] on the aged MPs underwent a significant enhancement due to the increased specific surface area after UV irradiation. Similar phenomena were also reported for NPs that oxygen-containing moieties generated during aging could increase the adsorption of Pb(II) and Cd(II) on NP surfaces and facilitate their transport in porous media [105]. Therefore, the oxygen content on NP surfaces plays a crucial role in facilitating heavy metal transport. Interestingly, the aging degree also exhibits a significant effect on the interaction between NPs and minerals present in the environment. Zhang et al. [106] demonstrated that positively charged minerals possessed greater adsorption capacities for aged NPs due to the enhanced electronegativity induced by oxygen-containing moieties. It is worth noting that there can be differences in the adsorption behavior of MPs and NPs towards organic compounds. Generally, the specific surface area and the number of adsorption sites increase with decreasing particle size, which enhances the adsorption capacity. The adsorption capacity of NPs was reported to be higher, even 1–2 magnitudes higher than that of MPs [107]. Zhang et al. [108] found that the sorption capacity of polypropylene (PP) MPs for 3,6-dibromocarbazole and 1,3,6,8-tetrabromocarbazole increased with decreasing particle size, which was attributed to increased specific surface area of the MPs. However, some studies have reported that smaller sizes, especially at the nanoscale, enhanced particle aggregation and thus reduced the specific surface area and the effective adsorption sites [109]. Wang et al. [109] reported that the adsorption of phenanthrene and nitrobenzene by nano-PS (50 nm) was less than that of submicron-PS (235 nm) due to aggregation of NPs.

Furthermore, the aging process can influence not only the adsorption properties of MNPs for pollutants but also the release of additives from them. Although the additive release from MNPs is fairly slow, the aging effect exhibits great potential to accelerate the release of these chemicals by altering the physical and chemical structures of MNPs [90]. For instance, the simulated solar irradiation has been widely reported to induce the alteration of MP surfaces, including fragmentation and oxidation, which significantly increase the release of brominated flame retardants, hexabromodiphenyl ether, and phthalate from MPs [110–112]. In addition, the release behavior of additives is strongly dependent on reaction conditions, e.g., light irradiation and microbial activity [82,113]. Compared to a single factor, the total amount of phthalate released from PVC increases up to 5-fold when exposed to a combination of light and bacteria [113]. Therefore, the leaching of additives from MNPs can be promoted by the aging process, which will further increase the biological risk associated with plastic pollution. Moreover, the presence of additives also potentially affects the aging rate of MNPs. By comparison with pure plastics, PS particles containing brominated flame retardants are more facile to photoaging, which is attributed to the enhanced light absorption induced by chromophores in flame retardants [114]. Moreover, the oxidation of MNPs with brominated flame retardants can also be enhanced by reactive bromine radicals that are generated from the photodegradation of brominated additives [110].

However, so far, there is limited research available on the release of additives from NPs. Considering the nanoscale size, large specific surface areas, and high instability, additives are more facile to be released from NPs into the environment and even into organisms that uptake NPs.

3.3. *The effect of aging on the role of MNPs as catalysts for coexisting contaminant degradation*

As shown in Fig. 5, artificial aging treatments by means of simulated solar light sources are able to induce the formation of EPFRs on MNP surfaces, which are subsequently transformed into ROS via electron transfer [85,115]. As reported in recently published literature, aged PS and phenol-formaldehyde resin (PF) MPs could act as the sources for EPFRs, which were strongly correlated with the production of ROS [e.g., singlet oxygen ($^1O_2$), superoxide radicals ($O_2^{\cdot-}$) and hydroxyl radicals (·OH)] [85]. The ROS generated during the photodegradation of MNPs have dual effects. On the one hand, they can react with the surrounding MPs, promoting the aging process by inducing oxidative degradation [116,117]. On the other hand, the ROS released during MNPs aging processes can also contribute to the degradation of coexisting contaminants [91]. In addition to ROS, interfacial interactions can also render MNPs as a catalyst to accelerate the removal of coexisting contaminants [50].

As a highly oxidative species, ·OH generated on MNP surfaces is able to enhance the degradation of decachlorobiphenyl, tetracycline, and

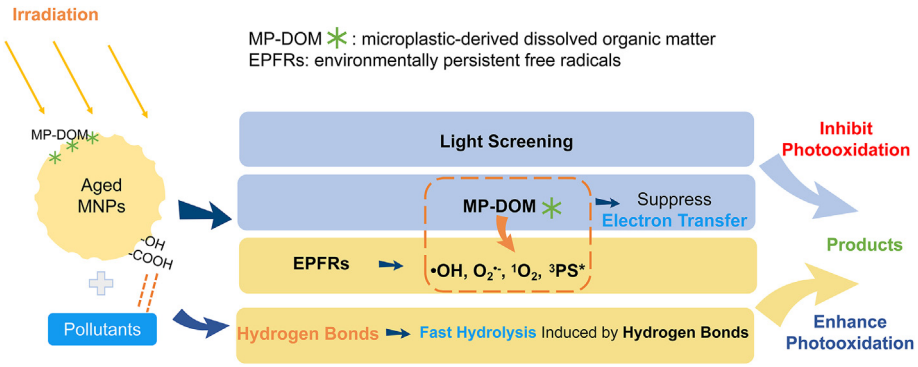

Fig. 5. The effect of aging on the role of MNPs as a catalyst for the degradation of coexisting contaminants.

organotin via bond cleavage and ring–opening reactions [118–120]. Also, the presence of hydroxylated products resulting from hydroxyl addition indicates the involvement of ·OH in the photodegradation of contaminants mediated by MNPs [121]. Wu et al. [120] demonstrated that ·OH produced by irradiated PS MPs was the main ROS responsible for the erosion of decachlorobiphenyl with the formation of hydroxyl substitution product. Additionally, chlorine and pentachlorobenzene radicals were released during the degradation process, highlighting the crucial role of ·OH in the enhanced photodegradation of decachlorobiphenyl in the presence of PS MPs [120]. In addition to ·OH, $^1O_2$ and $O_2^{\cdot-}$ also exhibit great potential to accelerate the transformation of coexisting organic contaminants. Wang et al. [52] demonstrated that the presence of PS MPs could promote the photo-transformation of atorvastatin (ATV), in which $^1O_2$ generated through PS MPs photosensitization played a dominant role in the enhanced degradation. Ding et al. [119] also reported that $^1O_2$ and $O_2^{\cdot-}$ could act as major contributors to the enhanced degradation of tetracycline.

Moreover, ^{3}PS* produced during PS photosensitization also affects the degradation of ATV. Wang et al. [52] showed that a large number of ROS produced by aged PS MPs were involved in ATV photolysis, including ^{3}PS* and $^1O_2$. ^{3}PS* can not only participate in the photodegradation of ATV by directly reacting with the pharmaceutical molecule but also transfer energy to dissolved O_2 with the generation of $^1O_2$, thereby further promoting ATV phototransformation [52]. Similarly, Wang et al. [122] found that the enhanced photodegradation of cimetidine in the presence of aged PS MPs, compared to pristine PS MPs, was attributed to the generation of $^1O_2$ and ^{3}PS*. Interestingly, PS MPs can reduce Cr(VI) to Cr(III) under sunlight irradiation, and the reduction of Cr(VI) is mainly dependent on $O_2^{\cdot-}$ produced on PS surface, but is inhibited by $^1O_2$, ^{3}PS* and ·OH [123].

In addition to the generation of ROS, the surface interaction between MNPs and coexisting contaminants can also contribute to the transformation of pollutants. Wang et al. [50] observed the accelerated hydrolysis of cephalosporin antibiotics in the presence of photoaged PVC MPs. The results showed that photoaged PVC MPs exhibited greater adsorption capacity for cephalosporin antibiotics compared to pristine MPs. The enhanced adsorption was attributed to the intermolecular hydrogen-bonding between the β-lactam carbonyl group in cephalosporin and the hydroxyl group on the aged PVC MP surface, which increased the positive charge on the carbon atom of the β-lactam carbonyl. This increased positive charge would facilitate the subsequent nucleophilic attack by water molecules and reduce the energy gap required for the hydrolysis of cephalosporin antibiotics. This research highlights the importance of not only ROS generation but also surface interactions in understanding the mechanisms of pollutant transformation mediated by MNPs.

Indeed, the presence of MNPs shows different effects on the photodegradation of various pollutants, as demonstrated in several studies. Zhang et al. [91] found that the presence of PS MPs inhibited the photodegradation of antibiotic sulfamethoxazole (SMX), and the aging degree of PS MPs was negatively correlated with the degradation rate of SMX, which might be attributed to the light screening effect of aged MNPs. During the aging process, PS MPs underwent the generation of unsaturated chromophores and the redshift of absorbance, leading to an increase in UV-visible light absorbance and subsequent enhancement in the light screening effect [91]. Furthermore, the effect of MNPs on photodegradation can also be influenced by the structures of target molecules. For instance, PS MPs can promote the photodegradation of anthracene while inhibiting pyrene decomposition, which may be explained by their individual reactivity for $^1O_2$ and $O_2^{\cdot-}$ [124,125]. PS MPs can promote the production of $^1O_2$ upon light irradiation, thus accelerating the photodegradation of anthracene [121]. Additionally, PS MPs were found to suppress the electron transfer from the excited pyrene to oxygen, thus reducing the production of $O_2^{\cdot-}$ and subsequently inhibiting the photo-transformation of pyrene [121].

When exposed to light, PS MPs can release a range of chemicals, including MP-derived dissolved organic matter (MP-DOM), plasticizers, chain-cutting products, and oxidation products (e.g., phenols and ketones), into the surrounding water bodies [126]. The chromophore-containing organic compounds are able to absorb UV energy and generate ROS [127]. Chen et al. [128] investigated the direct photolytic inhibition of SMX by MP-DOM from PS MPs and PE MPs and found that PS-DOM exhibited high oxidation resistance and inhibited the photodegradation of SMX by suppressing the electron transfer to SMX. Liu et al. [129] demonstrated that PS MPs could accelerate the photodegradation of coexisting PP MPs, in which ·OH derived from PS MPs and the associated PS-DOM was identified as the key ROS. Notably, in addition to MP-DOM, other substrates in natural aquatic environments may compete with ROS and inhibit the degradation process [127,130]. For example, nitrate photolysis produces nitrite ion, which in turn can eliminate ·OH [131]. In addition, background inorganic ions in water (e.g., chloride ion and phosphate ion) can act as ROS quenchers, depleting ROS and competing for light absorption [132,133]. Further research should be conducted to investigate the effects of natural substances in actual aquatic systems on the environmental behaviors of MNPs and the coexisting contaminants.

Numerous studies have focused on the photochemical behaviors of virgin MNPs or MNPs aged for a short period, which may not fully represent the conditions of MNPs in natural environments. Upon release into the environmental matrices, MPs would undergo photoaging for years, resulting in the generation of carbonyl and hydroxyl groups with great light absorption capacity [134]. This may have implications for the generation of ROS and their subsequent effects on the associated contaminants in the environment. Additionally, the presence of MNPs in sewage and sludge treatment systems induces microorganisms to generate ROS, leading to oxidative stress. This stress reduces the abundance of key bacteria involved in aerobic digestion, thereby inhibiting the aerobic digestion of waste-activated sludge [135]. However, little attention has been focused on the specific effects of ROS induced by MNPs in these systems. Therefore, there is indeed an urgent need for

further studies to investigate the role of MNPs-generated ROS across a broader range of environmental conditions and scenarios.

4. Impacts of aging on MNPs' biological effects

As shown in Fig. 6, aged MNPs can impact various organisms, including microorganisms, algae, zooplankton, aquatic animals, and terrestrial plants, thereby potentially affecting ecosystems and human health [136]. These problems arise as organisms ingest MNPs, which subsequently results in the diffusion of MNPs to cellular boundaries and their accumulation/transfer in food chains [137–139]. Notably, the toxic effects of MNPs can also be altered by aging processes. Changes in surface properties and the release of additives during aging can modify the bioavailability and interactions of MNPs with organisms. Generally, MNPs exhibit weak biodegradability, while aging treatments have been reported to enhance their susceptibility to microbial degradation by breaking polymers into smaller fragments [140]. The presence of oxygen-containing moieties on aged MNP surfaces provides numerous reactive sites for microbial attack and enzymatic degradation [53]. In addition, aged MNPs can serve as carriers for some harmful pollutants and transport pollutants into ecosystems through the food chain [141]. Thus, aged MNPs increase the bioavailability of coexisting pollutants.

4.1. *Effects of aging processes on the interaction of MNPs with organisms*

Aged MNPs may exhibit different interactions with living organisms compared to their pristine counterparts. On the one hand, the roughness and hydrophilicity of MNPs increase after aging, which facilitates the adhesion of microorganisms to the surface of MNPs and promotes the colonization of certain hydrophilic microorganisms [72,142]. On the other hand, aging can promote the release of additives from MNPs, which may serve as a nutrient source to promote microbial growth [143]. The adherence of biofilms to the surfaces of MNPs results in biofouling, which increases their density and diminishes their buoyancy, thereby accelerating the sinking rate of MNPs in aquatic environments. This, in turn, augments their ingestion by benthic organisms [144,145]. Furthermore, biofilms can amplify the role of MNPs as vectors for certain pollutants. Bhagat et al. [146] found that the adsorption of methylene blue and phenanthrol by MPs increased with the biofouling duration of the MPs, indicating an affinity between these pollutants and the biofilm biomass on the particles. Furthermore, studies have reported that biofilm formation can accelerate the development of antibiotic resistance genes (ARGs) and the accumulation of antibiotics [147]. The aging process can transform MNPs into attachment sites for ARGs, thereby exacerbating their toxic effects and contributing to the emergence of fish diseases [147,148]. Sun et al. [147] and Tavsanoglu et al. [148] revealed that biofilms on MNPs exhibit a higher enrichment capacity, with a more stable and tightly linked ARG-bacteria coexistence network on MNP surfaces. Additionally, biofilm-coated MNPs can serve as substrates for pathogenic microorganisms. Research has demonstrated that potential pathogenic bacteria, such as *Vibrio*, *Bacillus*, *Escherichia coli*, *Enterococcus faecalis*, as well as harmful algae, accumulated on MNP surfaces [148,149]. Therefore, these findings highlight the potential risks associated with MNPs and biofilm formation for environmental and public health.

The toxicity of aged MNPs differs significantly from their pristine counterparts with the changes in their physicochemical properties. For example, aged MPs undergo surface modification, such as the formation of carbonyl groups on their surfaces and the increase in zeta potential. These changes contribute to the toxic effect of aged MPs on algae. Wang et al. [53] demonstrated that both pristine and photoaged PVC MPs negatively affected the growth of green algae in freshwater, resulting in reduced chlorophyll-a levels in cells and increased superoxide dismutase and malondialdehyde enzyme activities in algae. In addition, the toxicity of photoaged PVC MPs is significantly higher than that of pristine PVC MPs due to the decreased repulsive force between photoaged PVC MPs and the anionic cellulose in the cell wall with the decrease in surface charge of photoaged PVC MPs. Moreover, the increase in surface roughness, as well as oxygenated functional groups after aging, improves the adsorption of PVC MPs to microalgae. Thus, aging affects the interaction of PVC MPs with algae and increases their toxicity.

The aging of MPs not only affects their physical and chemical properties but also has implications for the biological effects of associated chemicals, including heavy metals and organic compounds [54]. For example, studies have demonstrated that the leachate of aged MPs had a more pronounced inhibitory effect on cell growth and photosynthesis of *Microcystis aeruginosa*, a common freshwater cyanobacterium, compared to pristine MPs [150]. Additionally, the toxicity assessment of commercial bags fabricated from PLA and PBAT to the sea urchin *Paracentrotus lividus* in the marine environment, post-UV exposure, showed that the observed toxicity was attributed to leached additives from the plastics rather than the plastic polymers themselves. This inference is supported by the dissipation of toxicity after 7 d, which coincides with the rapid

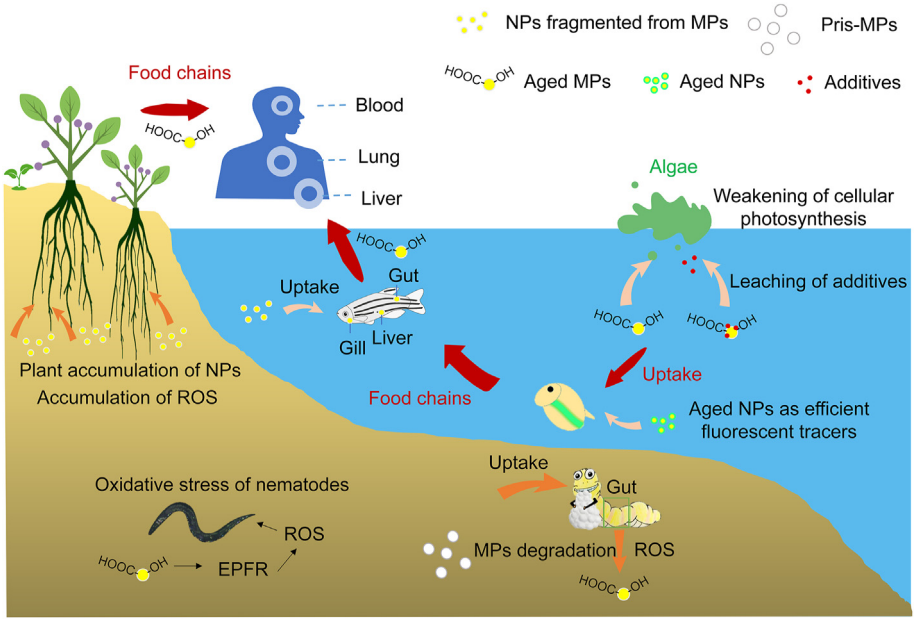

Fig. 6. The effects of aging processes on the toxicity of MNPs.

leaching of additives into the water [151]. This finding implies that aging processes may augment the release of compounds from MNPs, which could potentially exert more adverse effects on the growth and metabolic processes of aquatic organisms than the MNPs themselves. Furthermore, the leachate from aged MPs has been found to contain phthalate esters that are known to act as endocrine disruptors and pose potential risks to organisms, while no additives were detected in the leachate of pristine MPs [152–155]. In addition, we have already discussed above that aged MPs could adsorb heavy metals. It was reported that PS NPs could serve as the carrier of Cd for bio-enrichment in wheat, and the interaction between them significantly affected the activities of wheat tissues [156]. Similarly, Wang et al. [157] found that Cd levels increased from 2 to 10 mg/L in the presence of PS NPs, which jointly entered the maize root system and transported upward through the vascular bundle. It is worth noting that MPs can transfer metal cations from the residual form to the exchangeable/mobile form, thus posing a translocation risk for uptake by plants and soil organisms through the food chain [158].

Moreover, the aging of MPs leads to the formation of EPFRs [85], which has been discussed above. When organisms ingest MPs containing EPFRs, the ROS derived from EPFRs induce oxidative stress and even cause damage to cells and organisms [159,160]. Cao et al. [161] investigated the formation of EPFRs on UV-aged MPs and their toxic effect on nematodes, indicating that ROS were produced in nematodes, causing reduced locomotor behavior, body length, and brood size. Studies have shown that smaller particle size and higher surface oxygen-containing groups of MNPs after aging increase the hydrophilicity and stability of the MNPs, which are more easily ingested by organisms, particularly invertebrates, such as mussels and oyster larvae [140]. Lu et al. [162] investigated the exposure of pristine PS MPs with different sizes (70 nm, 5 μm, and 20 μm) to zebrafish and found that PS MPs with a diameter of 5 μm were mainly accumulated in the gills, liver, and viscera of the fish, while PS MPs of 20 μm were only accumulated in the gills and intestine. Moreover, both 5 μm and 70 nm PS MPs induced inflammation and lipid accumulation in the fish liver [162]. Therefore, different particle sizes lead to variations in uptake and accumulation sites, with smaller particles exhibiting higher toxicity to zebrafish.

Generally, most MPs are unable to penetrate plant cell walls due to their large size and high molecular weight. However, prior studies showed that NPs could be accumulated and translocated in plant tissues [163,164]. NPs, with smaller sizes, exhibit higher toxicity towards plants, and their toxicity can also be influenced by their surface charges. A recent research demonstrated that both positively and negatively charged NPs could be accumulated in *Arabidopsis thaliana*, a commonly used model plant [34]. Interestingly, the positively charged NPs induced a higher accumulation of ROS and had a more pronounced inhibitory effect on plant growth and seedling development compared to negatively charged NPs [34]. These results suggest that surface charge plays an important role in the toxicity of NPs towards plants. Furthermore, NPs with diameters of 38.3 and 34.4 nm have been observed to enter the cytoplasm of wheat plants [165]. Once accumulated in plant cells, NPs may persist in plant tissues for extended periods, potentially exerting negative impacts on plant growth and development, which in turn poses risks to human health if these plants are consumed.

A recent study also reported that NPs posed greater threats to both ecological and human health than MPs [166]. This is primarily attributed to the superior capability of NPs to cross cell membranes, enter cell organelles, translocate within the circulatory system, and accumulate in various tissues and organs [166]. Once ingested, NPs have been found to induce lung and intestinal damage, commonly accompanied by oxidative stress, cellular damage, and inflammation [24–26]. Wang et al. [167] demonstrated that upon exposing *Daphnia magna* to environmentally relevant concentrations of MNPs, rapid acidification of the intestinal environment and a decrease in esterase activity were observed, indicating that MNPs could affect intestinal metabolism and disrupt normal physiological processes. The study also showed that NPs apparently induced intestinal inflammation compared to MPs, suggesting a size-related effect of oxidative stress [167]. Lin et al. [166] investigated the effects of NPs on human liver and lung cells, and demonstrated that NPs could enter cells and cause mitochondrial damage, subsequently resulting in excessive ROS production, mitochondrial membrane potential alteration, and mitochondrial respiration inhibition. The blood analysis further confirms that the mean total concentration of MNPs can even reach 1.6 μg/mL [168]. Additionally, several studies have reported that PS and PP NPs with sizes of <240 nm could enter the human placenta and pose significant threats to fetal health [169,170].

Once NPs enter the bloodstream, they interact with plasma proteins in the blood [172]. This leads to the formation of a protein corona around the NPs, rendering them different from their original forms [171]. The protein corona plays a crucial role in mediating the recognition [172], uptake [173], transfer [174], and elimination [175] of NPs at the cellular level. Prior studies have provided strong evidence for the influence of protein corona on NPs behaviors and cytotoxicity [176]. For instance, Du et al. [79] investigated the changes in the protein corona composition formed on photoaged PS NPs in human bronchoalveolar lavage fluid (BALF) and observed a significant increase in the adsorption of negatively charged hydrophilic proteins by aged PS NPs. This, in turn, enhanced the uptake of aged PS NPs by lung macrophages, suggesting that aging-induced changes in the protein corona of BALF could influence NPs' uptake by cells [79]. However, another study found that the aged PS NPs showed reduced cellular internalization and cytotoxicity compared to their pristine counterparts, which might result from the reduced stability of PS NPs after aging [57]. These contrasting results highlight the complexity of the interactions between NPs, protein corona, and cellular processes.

To date, only a limited number of studies have explored the toxicity of aged NPs. The aging process of NPs significantly alters their physicochemical properties and modulates their interactions with plasma proteins and cellular responses. Gaining an understanding of these changes is crucial for the comprehensive assessment of the potential risks associated with aged NPs. Further studies are greatly needed to shed light on the underlying toxicological mechanisms of aged NPs and help develop strategies to minimize their adverse effects on living organisms.

4.2. *Biodegradation of biodegradable MNPs induced by aging*

The biodegradation of MNPs involves the conversion of the polymer into its monomer form, followed by mineralization through the action of microorganisms such as bacteria, fungi, and algae [177]. Microorganisms initially adhere to the surfaces of MNPs, where they subsequently establish colonies. The extracellular polymeric substances secreted by these microorganisms form a viscous matrix that facilitates their anchoring and colonization of the MNPs' surfaces [178]. Subsequently, biological deterioration of the MNPs' surface ensues, culminating in the physical disintegration of the MNPs [179]. Thirdly, the secretion of enzymes by microorganisms catalyzes the depolymerization of MNPs, resulting in the breakdown into smaller molecular intermediates, including dimers and monomers, and concurrently releasing any embedded additives [177]. Ultimately, these monomers undergo mineralization by microbial action. Under aerobic conditions, microorganisms can mineralize organic matter, resulting in the production of CO_2 and H_2O [180]. However, in the absence of oxygen, CO_2, and CH_4 are generally produced [180]. MNPs that enter soil and aquatic environments are inevitably subjected to biodegradation. Microorganisms, such as fungi and bacteria, primarily modify the structure of MNPs through enzymatic actions and metabolic processes [181]. Typically, under composting conditions or in microbially rich environments, enhanced biodegradation rates are observed at elevated temperature, humidity levels, and with the facilitation of specific microorganisms [182]. Zhang et al. [183] identified several functional bacteria species, including *Sphingomonas*, *Bacillus,* and *Streptomyces*, which were found to

degrade PBAT and PLA in two distinct natural soil samples. It is noteworthy that environmental factors in natural settings have the potential to influence microbial activity, thereby affecting the biodegradation of bioMNPs [184]. For example, under anaerobic conditions within the soil profile, the erosion rate of bioMNPs is slower than that at the soil surface due to reduced oxygen availability [185]. While soil microorganisms influence the degradation of MNPs, MNPs also impact soil characteristics and microbial communities [186]. Zhou et al. [187] demonstrated that incubating PBAT MPs in soil at 30 °C for 100 d significantly increased dissolved organic carbon content and microbial biomass and significantly reduced available nitrogen (NH_4^+-N and NO_3^--N) content, potential nitrification rate, and β-glucosidase activity. This treatment also increased the relative abundance of certain Proteobacteria members, such as Ramlibacter, Bradyrhizobium, Ellin6067 and Pedomicrobium, while decreasing fungal community diversity and altering community structure, with a notable enrichment of potential pathogens or bioMNPs degraders from the genera *Acrophialophora* and *Tetracladium* [187]. Furthermore, biodegradation of MNPs also occurs in aquatic ecosystems. Yamano et al. [188] demonstrated that polyamide 4 degraded in marine environments, with a weight reduction of 70% over 6 weeks, primarily due to the activity of the MND-1 strain belonging to the *Alteromonadaceae* family. Meanwhile, Bagheri et al. [189] investigated the biodegradation of poly (lactic-co-glycolic acid) (PLGA), polycaprolactone, PLA, and PHB in artificial seawater and freshwater for one year, finding that only PLGA was completely degraded.

However, the extent of biodegradation is somewhat limited due to the high molecular weight of the polymer chains and the scarcity of functional groups that can serve as reaction sites for microorganisms [190,191]. To achieve significant biodegradation, polymers need to be initially fragmented into smaller pieces through abiotic degradation [67]. This process can generate oxygen-containing moieties that provide reactive sites for microbial attack [67]. Volova et al. [192] found no differences in the biodegradation rates of various types of PHAs in marine environments; instead, the size of the polymer fragments accounted for the differences. Biodegradation primarily occur on the surface of MNPs, where smaller plastic particles have a higher specific surface area, resulting in greater biodegradability [193]. Moreover, during the aging processes, the surfaces of aged MNPs undergo oxidation reactions induced by ROS and generate polar carbonyl and hydroxyl groups, which subsequently result in enhanced hydrophilicity of the aged MNPs [194,195]. Generally, the presence of polar functional groups on the aged MNPs' surfaces enhances their interactions with biological systems, facilitating their breakdown and transformation in aquatic and terrestrial environments. Therefore, the increased hydrophilicity makes the surfaces of aged MNPs more susceptible to biological attacks than their pristine counterparts [196]. Balasubramanian et al. [197] demonstrated that the degradation of high-density polyethylene in soil by *Aspergillus terreus* MF12 was also greatly enhanced by UV and $KMnO_4$/HCl pre-treatment, which could be explained by the ability of photo-oxidative treatments could increase the oxygen-containing functional groups on the surface of high-density polyethylene and improve its hydrophilicity, thus making it more compatible with microorganisms and more susceptible to biodegradation [197]. However, aging may inhibit the efficiency of biodegradation due to the formation of cross-links in aged MNPs, the generation of ROS, and the leaching of toxic additives [198–200]. For instance, UV exposure alters the cross-link density in biodegradable plastics, limiting the movement of plastic segments and their interaction with water and microbes, which slows down biodegradation [198]. Therefore, the aging of MNPs significantly influences their biodegradation by altering their physicochemical properties and interactions with microorganisms. Future studies should systematically investigate the mechanisms of aged MNP-microbe interactions, explore diverse aging pathways such as photo-oxidation and thermal aging, and assess the long-term ecological impacts of degradation products from aged MNPs in natural environments.

5. Conclusions and future perspectives

In conclusion, this review has summarized the effects of natural aging on the environmental behavior and biological effects of MNPs, revealing several key perspectives:

(i) Altering environmental fate and transport: Aging processes can change the surface properties of MNPs, such as increasing their hydrophilicity and polarity, which promotes the adsorption of hydrophilic contaminants and polar pollutants. This can affect the fate and transport of MNPs in the environment, potentially leading to their accumulation in specific habitats such as oceans and soils or their uptake by organisms.

(ii) Enhancing interactions with living organisms: Aged MNPs may exhibit different interactions with living organisms compared to their pristine counterparts. For example, the surface modifications during aging influence the attachment of biofilms or other microorganisms, potentially leading to the altered colonization patterns and ecological consequences within aquatic ecosystems.

(iii) Increasing bioavailability and toxicity: Aging processes result in the release of degradation products from MNPs, such as smaller particles or leached chemicals. These aged MNPs and their associated products may exhibit increased bioavailability and potential toxicity to organisms, as they can be more readily taken up and accumulated in tissues, potentially leading to adverse ecological effects.

(iv) Altering biodegradation potential: Aging makes MNPs more susceptible to biodegradation. Changes in surface properties, such as increased hydrophilicity or the presence of reactive functional groups, can enhance the microbial colonization and enzymatic degradation of MNPs. It can lead to the release of breakdown products and potentially influence the biodegradation rates and pathways of MNPs in the environment.

However, there are still knowledge gaps and challenges in understanding the aging processes of MNPs. Future research is needed to explore the mechanisms of natural aging for different types of MNPs, the influence of environmental factors on aging, the release of associated chemicals and their toxic effects during the aging processes. In summary, MNPs aging is a complex and important issue that has significant implications for environmental protection and ecosystem health. By delving into the mechanisms and biological effects of MNPs aging, more scientific foundations for future management efforts can be provided, thus achieving sustainable plastic use and environmental preservation goals.

CRediT authorship contribution statement

Hongjian Li: Writing – original draft, Visualization, Investigation, Conceptualization. **Lihua Bai:** Writing – review & editing, Conceptualization. **Sijia Liang:** Writing – review & editing. **Xiru Chen:** Writing – review & editing. **Xinyue Gu:** Writing – review & editing. **Chao Wang:** Writing – review & editing, Supervision, Funding acquisition, Conceptualization. **Cheng Gu:** Writing – review & editing, Supervision, Project administration, Funding acquisition, Conceptualization.

Declaration of competing 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.

Acknowledgements

This work was financially supported by National Natural Science Foundation of China (22241601, 22176092, 22276091), Open Project of

State Environmental Protection Key Laboratory of Soil Environmental Management and Pollution Control (SEMPC2023001), Fundamental Research Funds for the Central Universities (0211/14380204) and International Institute for Environmental Studies.

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