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Review

Weathering of microplastics and interaction with other coexisting constituents in terrestrial and aquatic environments

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ABSTRACT

Weathering of microplastics (MPs, < 5 mm) in terrestrial and aquatic environments affects MP transport and distribution. This paper first summarizes the sources of MPs, including refuse in landfills, biowastes, plastic films, and wastewater discharge. Once MPs enter water and soil, they undergo different weathering processes. MPs can be converted into small molecules (e.g., oligomers and monomers), and may be completely mineralized under the action of free radicals or microorganisms. The rate and extent of weathering of MPs depend on their physicochemical properties and environmental conditions of the media to which they are exposed. In general, water dissipates heat better, and has a lower temperature, than land; thus, the weathering rate of MPs in the aquatic environment is slower than in the terrestrial environment. These weathering processes increase oxygen-containing functional groups and the specific surface area of MPs, which influence the sorption and aggregation that occur between weathered MPs and their co-existing constituents. More studies are needed to investigate the various weathering processes of diverse MPs under natural field conditions in soils, sediments, and aquatic environments, to understand the impact of weathered MPs in the environment.

1. Introduction

With the wide application of polymer, more and more particulate plastics have been detected in the environment due to the degradation of bulk plastic polymers and discharge from plastic products (Chen et al., 2019b; Dawson et al., 2018). It has been estimated that the mass of plastics released to land annually (4.73×10^8 - 9.1×10^8 kg) may be 4 to 23 times higher than that released to oceans in the European Union (Horton et al., 2017). Particulate plastics with sizes smaller than 5 mm are defined as microplastics (MPs) (Arthur et al., 2009). By 2100, it is speculated that 2.5×10^7 to 1.3×10^8 tonnes of MPs float on the ocean surface (Everaert et al., 2018). Sediments are a long-term sink for MPs in deep-sea areas and river-estuary regions (Xu et al., 2020b; Zhang et al., 2020b). Some MPs were found to pose negative impact on organisms, because they may induce oxidative stress and adsorb pollutants, such as heavy metals or organic matters (Ho et al., 2020; Wang et al., 2021b).

The weathering processes of MPs mainly include mechanical fragmentation, photo-degradation, thermal-degradation, and biodegradation (Chen et al., 2019b; Resmeriță et al., 2018; Tu et al., 2020). These abiotic and biotic processes for MP degradation can take hundreds, and even thousands, of years (Iñiguez et al., 2018), depending on the physicochemical properties of MPs and environmental conditions (Chen et al., 2020; Mei et al., 2020; Tian et al., 2019; Turgay et al., 2019; Wang et al., 2020a). However, the combined effects of these factors on various MP weathering processes are complex under natural environmental conditions, which results in unpredictable lifetimes of MPs in the environment. Meanwhile, weathering of MPs influences their interactions with coex-

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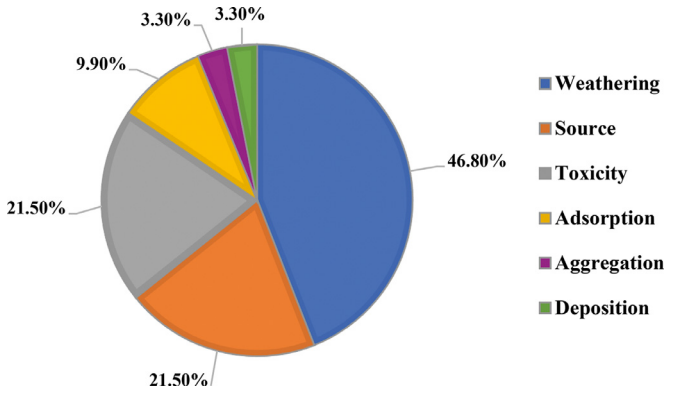


Fig. 1. The proportion of research papers up to February 2021 investigating the transport and transformation of microplastics.

isting pollutants (Liu et al., 2019b, 2019d; Wang et al., 2020b), and these interactions need to be summarized to understand the distribution of MPs in the environment and the transport of constituents associated with the MPs.

In order to systematically summarize the related research on MP transport and transformation, the Web of Science database was used to retrieve the relevant publications up to February 2021 with the following keywords: microplastics, weathering, source, toxicity, adsorption, aggregation, and deposition. The number of publications that investigated transport and transformation of MPs was 5311. As shown in Fig. 1, 46.8% of the research focused on the weathering of MPs, which was more than the research in other areas. Currently, most reviews of MPs have concentrated on the environmental behavior of pristine MPs in water (Mei et al., 2020; Wang et al., 2021a), but have ignored the interaction between weathered MPs and their coexisting constituents in aquatic and terrestrial environments. Additionally, most of these reviews only have summarized a single weathering process (Yuan et al., 2020), without focusing on the influence of multiple weathering processes that affect the fate of MPs.

This review provides an overview of recent progress in studying weathering of MPs in terrestrial and aquatic environments. To understand fully the weathering processes of MPs and associated factors influencing the weathering in natural environments, the sources of MPs are first described to provide their types, concentrations, and particle sizes in natural waters or soil. Next, the mechanisms of the weathering processes and the main factors that influence them, including the physicochemical properties of MPs and environmental conditions, are summarized. Then how the physicochemical properties are changed during the weathering process is discussed. The section also reviews the interaction between weathered MPs and other coexisting constituents in natural waters or soil. Finally, the knowledge gaps and future recommendations regarding the weathering of MPs are discussed.

2. Sources of particulate plastics enter the environment

The sources of MPs in natural waters or soil are reviewed ahead of the description of their weathering processes to give their types, concentrations, and particle sizes in natural environments. This provides fundamental information for researchers to investigate the weathering processes of MPs under environmentally relevant conditions. Table 1 shows that most MPs are linked to anthropogenic activities. The common types of MPs consist of polyethylene terephthalate (PET), polypropylene (PP), polyethylene (PE), polyamide (PA), and polystyrene (PS).

Table 1
The types, sizes and concentrations of MPs from different sources in terrestrial and aquatic environments.

Environment	Source	Sample site	Type	Size	Average concentration	Reference
Terrestrial environments	Landfill refuse	Laogang landfill, Shanghai, China	PP, PE, PS, PEUR, EPM	0.23–4.97 mm	62 items g^{-1} (dw)	(Su et al., 2019)
	Landfill leachate	Laogang landfill, Shanghai, China	PP, PE, PS, cellophane	0.07–3.67 mm	8 items L^{-1}	(Su et al., 2019)
		Landfills, Shanghai, Wuxi, Suzhou and Changzhou, China	PP, PE	0.1–1 mm	5.08 items L^{-1}	(He et al., 2019)
		Landfills, Shanghai, China	PP, PA, Rayon	0.02–0.1 mm	291 items L^{-1}	(Xu et al., 2020c)
	Biosolids	Dewatered sludge of WWTPs, Guilin, China	PP, PE	0.2–5 mm	2.2–5.2 items g^{-1} (dw)	(Zhang et al., 2020d)
		Agricultural fields underwent sludge applications, Mellipilla, Chile	N.D.	N.D.	0.6–10.4 items g^{-1} (dw)	(Corradini et al., 2019)
	Plastic mulches	Agricultural lands, Xinjiang, China	N.D.	N.D.	502.2 kg ha^{-1} (maximum)	(Zhang et al., 2016)
		Agricultural lands, Yunnan, China	N.D.	0.05–5 mm	18.8 items g^{-1} (dw)	(Zhang and Liu, 2018)
Aquatic environments	Effluent of WWTPs	Glasgow, UK	PP, PE, PET, PA, PS, PES	0.6–1.6 mm	0.25 items L^{-1}	(Murphy et al., 2016)
		Sydney, Australia	PP, PE, PET, PA, PS	0.025–5 mm	0.21–1.6 items L^{-1}	(Ziajahromi et al., 2017)
		Australia	PP, PE, PET, PA	0.025–5 mm	0.18–1.91 items L^{-1}	(Ziajahromi et al., 2021)
		Daegu, South Korea	N.D.	0.03–1.5 mm	33–297 items L^{-1}	(Hidayaturrahman and Lee, 2019)

Note: Ethylene-propylene copolymer (EPM), Polyether urethane (PEUR), Polyester (PES), Polypropylene (PP), Polystyrene (PS), Polyethylene (PE), Polyethylene terephthalate (PET), Polyamide (PA), Wastewater treatment plants (WWTPs). N.D. represented no data. [a] represented plastic mulches residual rather than MPs.

2.1. Terrestrial environments

Major sources of terrestrial plastic fragments are from refuse landfills, sewage sludge and plastic films. Solid-waste landfills accumulate 21–42% of global plastic wastes (Nizzetto et al., 2016). Landfill leachate from a municipal solid waste landfill in China had concentrations of MPs with 235.4 ± 17.1 item L^{-1}, and the percentage of tiny MPs smaller than 50 μm was over 50% in landfill leachate (Sun et al., 2021). The sewage sludge retains more than 90% of the MPs that are in the influent wastewater (Zhang et al., 2020a). The recurrent application of the biosolids to agricultural lands (e.g., compost) results in small particles that tend to form secondary MPs and nanoplastics (NPs), especially from PE, due to mixing, abrasion, and milling (Kumar et al., 2020). Plastic films have been extensively utilized in agriculture, and the covered agricultural area worldwide is estimated to be around 180,000 km^2 and 13,000 km^2 for plastic mulches and greenhouses, respectively (Xu et al., 2020a). Plastic mulches left on the ground are prone to weathering due to UV light, microbes, and tillage, and they become brittle enough to fall into fragments (Astner et al., 2019). The concentration left in 60% of the agricultural areas in China has exceeded the national film residual standard (75 kg ha^{-1}).

2.2. Aquatic environments

Drainage from domestic and industrial wastes is received by wastewater treatment plants (WWTPs), becoming an essential source of MPs in aquatic environments (Da Costa et al., 2018; Li et al., 2020). Household wastewater contains heavy loads of MPs from cosmetics, personal hygiene products, and synthetic fibers from garments. Cosmetic exfoliants release about 4500 to 94,500 microbeads in a single rinse, of which 90% consist of PE (Napper et al., 2015). Primary and secondary treatment of wastewater at WWTPs can eliminate 70% to 99% of the MPs (Prata, 2018). At a treatment plant in Scotland, even at 98.4% removal efficiency, 65 million particles were discharged into the river Clyde per day with 0.25 items L^{-1} in the final effluent (Murphy et al., 2016). Pedrotti et al. (2021) estimated that 4.3 billion synthetic microfibers were released daily into the marine environment from the Haliotis treatment plant, despite its high removal efficiency (87.5% to 98.5%). In several wastewater treatment facilities across the U.S., the daily number of microplastics discharged into freshwater was estimated to be in the range of 50,000–15,000,000 (Mason et al., 2016). Usually, PS, PE, PET, PA, and PP polymer types dominate in the wastewaters (Murphy et al., 2016; Ziajahromi et al., 2017).

2.3. Particulate-plastics transfer between the terrestrial and aquatic environment

Vertical transfer of MPs leads to their distribution at various depths in the soil. A greater abundance of MPs has been reported in shallow soil than in deeper soil (Liu et al., 2018). Ploughing, soil cracking, and bioturbation caused by earthworms can incorporate MPs in topsoil layers to the 25-cm depth in the soil matrix (Bläsing and Amelung, 2018; Rillig et al., 2017). Soil pores filter out relatively large particles that remain in the surface or near-surface soil, whereas smaller particles (0.1–6.0 μm) presumably pass through the pore channels, and possess greater mobility downwards (Rillig et al., 2017). However, even larger particles (< 1.5 mm) have been found to reach groundwater aquifers due to migration along fractures and crevices in the ground. The occurrence of 12 particles L^{-1} in groundwater has confirmed the vertical transfer in soil (Panno et al., 2019).

Erosion can translocate MPs incorporated in the surface or subsurface soil over the ground towards surface water bodies. In particular, arable lands are often vulnerable to the movement of MPs by erosion due to high soil permeability, intensive drainage, loss of vegetative cover, and heavy applications of biosolids and compost (Bläsing and Amelung, 2018). In northeast China, the translocation of MPs was claimed to be 96% due to surface-soil water loss (Zhang et al., 2020e). During floods, high tides, and extreme wind conditions, MPs in water can move landwards and become deposited on terrestrial soils. Suspended MPs in rivers and lakes tend to become deposited on floodplains, shorelines, and even remote mountainous areas due to aeolian transport (Scheurer and Bigalke, 2018).

3. Weathering of particulate plastics in terrestrial and aquatic environments

In terrestrial and aquatic environments, MPs will undergo different weathering processes (Auta et al., 2018; He et al., 2018; Iñiguez et al., 2018). The weathering mechanisms based on physical, chemical, and biological reactions have been systematically summarized for the first time in Section 3.2. Additionally, these weathering processes are affected by physicochemical properties of the MPs (e.g., size, structure, and crystallinity) and environmental conditions (e.g., oxygen, water, temperature, and biofilms) (Chen et al., 2020; Mei et al., 2020; Tian et al., 2019; Turgay et al., 2019; Wang et al., 2020a), and they are rarely considered in previous reviews. We compared the effect of physicochemical properties of MPs and environmental conditions on different weathering processes, as discussed in Sections 3.3.1 and 3.3.2, respectively. Recent studies regarding MP weathering processes are summarized in Table 2, which shows that most experiments have been conducted under laboratory conditions and are mainly focused on the aquatic environment.

3.1. Weathering processes

3.1.1. Mechanical fragmentation

Mechanical breakdown originates from abrasion and disintegration forces that are a result of the interaction of MPs with sediments, pebbles, waves, and tide action in aquatic environments. In the terrestrial environment, it can result from human activities (e.g., soil cultivation and crop rotation) (He et al., 2018). As shown in Table 2, mechanical fragmentation of MPs has a great possibility of producing more small-sized MPs, even NPs (<10 nm) by simulated sand or wave action. However, the potential disintegration processes, such as freeze-thaw cycles and rainstorm events, are rarely researched. Furthermore, the rate of mechanical fragmentation was promoted by UV irradiation. Song et al. (2017) reported that pristine PP MPs only produced 10.7 ± 0.7 particles $pellet^{-1}$ under 2 months of mechanical abrasion, while UV-aged MPs for 12 months, after 2-month mechanical abrasion, formed significant amounts of PP fragments (6084 ± 1061 particles $pellet^{-1}$). Therefore, MP accumulation in natural environments will be accelerated under prolonged UV exposure and frequent mechanical wear.

3.1.2. Photo-degradation

Photo-degradation is the major weathering process for most MPs. Table 2 summarizes recent research about photo-degradation of MPs. The UV fraction of light irradiation plays a key role in MP weathering. UV radiation with a wavelength of 290–400 nm in sunlight has enough energy (299–412 kJ mol^{-1}) to break the C–C bond (284–368 kJ mol^{-1}) and the C–H bond (381–410 kJ mol^{-1}) for most plastics (Kholodovych and Welsh, 2007). Usually, the photo-weathering process mainly occurs on the outer layer of MPs with a μm-range depth, because the high crystallinity of MPs results in light scattering and reflection, which reduces the

Table 2
Review of recent studies regarding the weathering processes of MPs in-situ or in laboratory.

Weathering process	MPs	Experimental conditions				Key conclusions	Reference
		Size	Time	Matrix	Method		
Mechanical fragmentation	Polyolefin	398 ± 54 nm	180 s	Water	Broken by mechanical impeller, horn sonicator, hydraulic pump	Water shear forces promote nanoplastic (< 10 nm) generation mainly by crack propagation and crushing mechanism.	(Enfrin et al., 2020)
	PE, PP, expanded PS	26 ± 0.8 mm^3, 19 ± 0.9 mm^3, 20 ± 2.2 mm^3	2 months	Sand	Ground by roller mixer	Expanded PS pellets are more susceptible to fragmentation than PE and PP, due to the lower mechanical strength of Expanded PS.	(Song et al., 2017)
	PE	6 mm × 2 mm	30 d	Simulated seawater	Ground in a wave tank with UV light	Secondary MPs (1–2.4 μm^2) account for an increasing proportion with the increasing of weathering time.	(Resmeriță et al., 2018)
Photo-degradation	LDPE	< 500 μm	42 d	Air, 69% humidity	Xenon lamp (120 mW cm^{-2})	Aged-MPs had a rougher surface, lower glass transition temperature, and higher surface stiffness.	(Luo et al., 2020c)
	PS	150 μm	150 d	Ultrapure water	Simulated sunlight (68.25 mW cm^{-2})	Reactive oxide species played a major role in the photodegradation process of PS MPs in their aqueous suspension.	(Zhu et al., 2020a)
	^{14}C-PS	250 ± 88 nm	2 d	Distilled water/air	UV-254 nm (0.7 mW cm^{-2})	The mineralization efficiency of PS in water were higher (17.1 ± 0.55%) than in air (6.17 ± 0.1%).	(Tian et al., 2019)
	PP, PS, PE	N.D.	3 months	Simulated seawater/ultrapure water/air	UV 340 nm	The weathering degree of MPs decreased in the order of air < ultrapure water < seawater, depending on the irradiation time, oxygen content, and water salinity.	(Cai et al., 2018)
	PE	< 500 μm	56 d	Air, 50% humidity	Xenon-lamp (120 mW cm^{-2})	Aged MPs resulted in color change, decrease of high temperature resistance, and increase of CI value.	(Luo et al., 2020d)
	HDPE, PA, PP, HIPS	2 mm	5 d	Simulated seawater	253.7 nm	UV-induced shape (fiber and particle) of secondary MPs depends on plastic types. HDPE and nylon 6 mainly generated MP fibers, while HIPS and PP were more resistant to photo-transformation.	(Naik et al., 2020)
Thermal- degradation	PS	< 500 μm	56 d	Compost suspension	70 °C	High temperature (70 °C) caused the formation of cracks, whereas no obvious change occurred at 40 °C.	(Chen et al., 2020)
	PS	1 μm	3 months	Simulated seawater/ ultrapure water/air	75 °C	The aging mechanisms of PS were different in the three aging conditions according to sequence of aged functional groups.	(Ding et al., 2020)
	PP	5 mm	0.5 h	Purified water	121 °C	The leaching rate of BPA increased with higher temperature.	(Zhou et al., 2018)
Biodegradation	UV-aged PP	2.4 mm	40 d	Liquid medium	Isolated microorganism from sediments of mangrove	The weight losses of PP after biodegradation by *Rhodococcus* and *Bacillus* were 6.4% and 4.0%, respectively.	(Auta et al., 2018)
	PE *	3–5 mm	135 d	Seawater from Yellow Sea	Biofilm	The thickness of biofilms on PE surface increased with prolonging of exposure time (30, 75, and 135 days), but decreased with deeper natural coastal water (2 m, 6 m, and 12 m).	(Tu et al., 2020)
	HDPE	< 200 μm	28 d	Liquid medium	Isolated microorganisms from wax moth guts	*Aspergillus flavus* was isolated, and the fungus degraded PE with the mass loss percentage of 3.90 ± 1.18%.	(Zhang et al., 2020c)
	LDPE	< 150 μm	21 d	Sandy soil	Isolated microorganisms from earthworm gut	Gram-positive bacteria, including phylum Actinobacteria and Firmicutes, were isolated from the earthworm's gut, and they resulted in weight loss of 60%. Furthermore, several long-chain alkanes were detected in the bio-treatment.	(Huerta Lwanga et al., 2018)
	MPs *	N.D.	45 d	Sludge	Composting with high temperature (70–90 °C)	The degradation efficiency (weight loss percentages) of MPs was 43.7%.	(Chen et al., 2020)
	PS	< 500 μm	56 d	Liquid medium	Isolated microorganisms from sludge	The degradation efficiency (weight loss percentages) of PS MPs was 7.3% and 1.1% at 70 °C and 40 °C after 56 days, respectively.	(Chen et al., 2020)

Note: Bisphenol A (BPA), Carbonyl index (CI), High impact PS (HIPS), High density polyethylene (HDPE), Low density polyethylene (LDPE), Polypropylene (PP), Polystyrene (PS), Polyethylene (PE), Polyamide (PA), Polyethylene terephthalate (PET). * represented the *in-situ* experiment of MP weathering process. N.D. represented no data.

light penetration distance (ter Halle et al., 2017). To be further oxidized, the weathering layer needs to be removed via mechanical force and a new non-oxidized layer needs to be formed (ter Halle et al., 2017). Moreover, the weathering layer increases surface hydrophily, which enhances microbial adhesion and mineralization rate (Chamas et al., 2020).

3.1.3. Thermal-degradation

Thermal-degradation of MPs leads to bond breakage as a result of overcoming the bond dissociation energy (Pielichowski and Njuguna, 2005). The theoretical maximum temperature of darkish and dry soils has been estimated to be between 90 and 100 °C in natural ground surfaces (Mildrexler et al., 2011; Tang et al., 2019). Therefore, MPs may undergo thermal-degradation process on land due to the extreme maximal surface ground temperatures. On the contrary, water dissipates heat better and has a lower temperature than dark and dry soils. Thus, thermal-degradation seems to be more important in land than in aquatic environments. It has been demonstrated that thermal weathering (80 °C, 3 months) of the PE strips was equal to approximately 270 days of UV irradiation at 43–45 °C in terrestrial environments (Erni-Cassola et al., 2020). Most studies about MP thermal-degradation kinetics have been conducted at temperatures higher than 300 °C in a nitrogen atmosphere (Luo et al., 2020d). The testing environment is greatly inconsistent with the natural environment (e.g., oxygen and temperature). Further studies are needed to explore long-term MP thermal-degradation in natural aquatic or terrestrial systems with environmental temperatures.

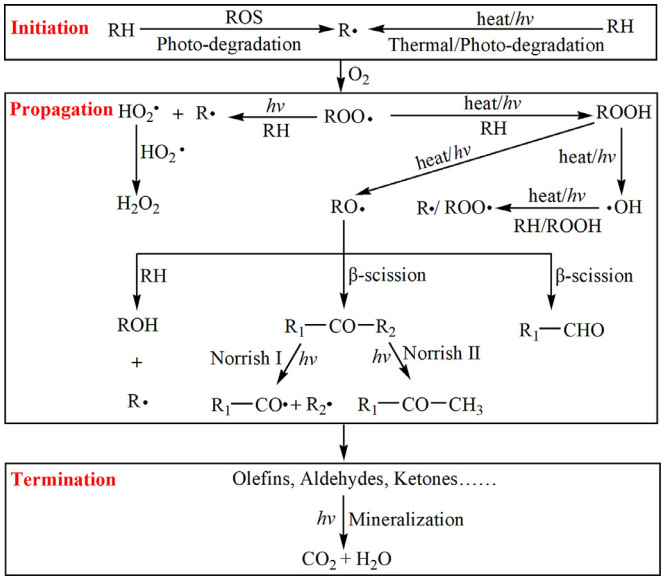
3.1.4. Biodegradation

Microbial degradation, and biological ingestion and digestion are main pathways of MP biological weathering. Many plastic-degrading microbial strains have been identified (Chen et al., 2020; Zhang et al., 2020c). As shown in Table 2, the degradation efficiency of MPs has ranged from 3.9% to 60%, depending on the species and the diversity of the microbial community. In natural environments, symbiotic and synergistic interactions among microorganisms in bacterial consortia play a major role in MP biodegradation (Yuan et al., 2020). For example, toxic metabolites produced by one microorganism may be used as substrate by another microorganism, thus reducing the influence of the toxic metabolites on MP-degrading bacteria (Yuan et al., 2020). Huerta Lwanga et al. (2018) found that biodegradation of low-density polyethylene (LDPE) MPs by bacterial consortia (phylum Actinobacteria and Firmicutes) separated from the earthworm gut caused a weight loss of 60% and formation of NPs after 21 days in soil. Compared with bacterial consortia, a single bacterium has low biodegradation efficiency of MPs (< 15%) after 28–40 days (Auta et al., 2018), indicating bacterial consortia may provide potential solutions for improving biodegradation efficiency of MPs, even though the role of each bacterium in the consortia is unclear. In addition to microbial degradation, PE MPs could rapidly be broken into smaller particles by freshwater amphipod *Gammarus duebeni*, and the fragments accounted for 65.7% of all observed MPs in digestive tracts (Mateos-Cárdenas et al., 2020).

3.2. Weathering mechanisms

3.2.1. Physical mechanisms

Physical weathering of MPs results in their fragmentation by crack propagation and crack failure under localized shear forces (Enfrin et al., 2020; Julienne et al., 2019a). The defective structures, such as microcracks, are responsible for the initiation of MP fragmentation (Enfrin et al., 2020). Enfrin et al. (2020) used Grady's model regarding the theory of solid failure through crack propagation and fracture to demonstrate that the existing cracks played a determinant role in breaking of MPs into NPs in freshwater. They revealed that longer cracks reduced the minimum stress of crack propagation, thereby resulting in crack failure under further shear forces, which produced a planar exfoliation and NP fragments. Therefore, the generated cracks of MPs during manufacturing or other weathering processes enhance the probability for further fragmentation (Enfrin et al., 2020). Particularly, the destructive effect by crack propagation and failure is more obvious in brittle materials (e.g., PS) in comparison to materials with toughness (e.g., LDPE). It has been demonstrated that 99.8% of PS debris was fragmented into MPs and NPs within 24 h by simulating the breaking of waves (Irina et al., 2018). On land, the continuous frictional stresses between tire treads and road surface are mainly responsible for MP generation. When the stress achieves the limiting strength of the rubber material, the tire will be cut or scratched slightly, producing smaller-size particles (Zhang et al., 2021).



Scheme 1. Thermal-degradation and photo-degradation pathways of microplastics (RH represents microplastics; R_1 and R_2 represent different polymer chains of variable lengths).

3.2.2. Chemical mechanisms

The chemical weathering processes, including photo-degradation and thermal-degradation, can lead to MP chain scission, branching, and generation of oxygenated intermediates via similar radical-based weathering mechanisms (Gardette et al., 2013; Liu et al., 2020c). As shown in Scheme 1, the radical-based chemical weathering process is generally divided into three steps, including initiation, propagation, and termination reactions (Yousif and Haddad, 2013). The difference for the two chemical weathering processes is the initiator species and weathering products due to the different oxidation efficiencies (Gardette et al., 2013; Liu et al., 2020c).

In the initial step, alkyl radicals (R·) are considered as important initiating species (Yousif and Haddad, 2013). For the thermal-degradation process, high temperature overcomes the energy barrier, and causes R· generation via random chain scission at weak sites or chain-end scission of C–C bonds (Singh and Sharma, 2008). For the photo-degradation process, the chromophore of MPs absorbs the energy of UV irradiation to become an excited singlet state, which then is transformed into an excited triplet state by intersystem crossing (Yousif and Haddad, 2013). The energy of triplet state of MPs is transferred to the nearest C–C/C–H bonds by intramolecular energy transfer processes, subsequently resulting in

chain scission and formation of R·. For example, the excited state of the benzene ring on PS (443 kJ mol^{-1}) has enough energy to cause C–C/C–H bond (< 410 kJ mol^{-1}) rupture (Waldman and De Paoli, 2008). Recent research proved reactive oxide species (ROS) involving the hydroxyl radical (·OH), superoxide radical ($O_2^{\cdot-}$), and singlet oxygen ($^1O_2$) generated by PS MPs played a vital role in their photo-aging (Zhu et al., 2020a). They might promote R· generation via H abstraction from plastic molecule (RH), C–C scission, or phenyl ring-opening to accelerate weathering process of MPs (Cho and Choi, 2001).

In the propagation step, high-activity radicals, such as ·OH, R·, alkoxyl (RO·), and peroxyl (ROO·), can promote self-catalyzed reactions. First, R· react with O_2 and form ROO·, then ROO· abstract the hydrogen atom from another RH or the media (e.g., H_2O) to form hydroperoxide (ROOH), which will be decomposed into ·OH and RO· by absorption of light and heat energy (Tian et al., 2019; Yousif and Haddad, 2013). Subsequently, ·OH attack ROOH and RH to form ROO· and R·, respectively (Zweifel, 1999). Moreover, the combination of ROO· and RH can form charge transfer (CT) complexes, which through photolysis change into the hydroperoxyl radical (HO_2·) and R·. The radical (HO_2·) could subsequently form hydrogen peroxide (H_2O_2) (Gugumus, 1990). These reactions provide high-activity radicals to keep chain propagation going. The alkoxyl radical is a key intermediate in the reaction and undergoes several reaction pathways, including hydrogen atom abstraction from RH to produce alcohol and β-scission to form ketones or aldehydes (Gewert et al., 2015). Under UV irradiation, ketones undergo successive reactions to produce R· and acyl radicals (R-CO·) by the Norrish I reaction, and to produce the end carbonyl group (R-CO–CH_3) by the Norrish II reaction (Gardette et al., 2013).

The termination step mainly results in recombination between bimolecular or low molecular radicals, with the main products of ketones, olefins, and aldehydes (Gewert et al., 2015). Additionally, a recent study demonstrated that photo-degradation could result in PS MPs to be completely mineralized into CO_2 under simulated solar irradiation with light intensities of 3 and 10-fold higher than those of natural solar at 0° and 50° north latitude, respectively (Ward et al., 2019). However, the mineralization mechanism of MPs during the photo-aging process needs to be further verified.

3.2.3. Biological mechanisms

For microorganisms (mainly bacteria and fungi), the degradation process of MPs is divided into four steps, as shown in Fig. 2. First, the microbes adhere and subsequently colonize onto the MP surface. The extracellular polymeric substances (EPS) secreted by microorganisms provide a sticky matrix for colonization (Ganesh Kumar et al., 2020; Michels et al., 2018). Second, biodeterioration occurs on the MP surface, resulting in physical disintegration of MPs (Lucas et al., 2008). Third, enzymes secreted by microorganisms facilitate the depolymerization process of MPs, which transform MPs into intermediates with smaller molecules (e.g., dipolymers and monomers) and release additives (Yuan et al., 2020). Finally, these small molecular substances and additives, which are used as carbon and energy sources by microorganisms, undergo assimilation, subsequent mineralization, and generation of metabolites (e.g., CO_2, CH_4, and H_2O) (Jacquin et al., 2019).

For crustaceans and omnivores, biological ingestion and digestion are considered to be potential biodegradation mechanisms (Dawson et al., 2018; Mateos-Cárdenas et al., 2020). MPs were cut and ground into NPs by mandibles for mastication, and then they were transported into the stomach and gastric mill, which resulted in the fragmentation of plastic particles (Dawson et al., 2018; Mateos-Cárdenas et al., 2020). The digestive enzymes in the stomach might catalyze hydrolytic cleavage of MPs and participate in MP degradation (Mateos-Cárdenas et al., 2020). It is speculated that several enzymes (e.g., amylase, cellulose, esterase, protease, and lipase) from intestinal tracts might be responsible for MP degradation (Song et al., 2020). The contribution of the enzymes to breakdown of MPs and the mechanism of action of the enzymes remain elusive. Furthermore, the interaction between ingested MPs and sharp edges, such as triturated algae, have been shown to accelerate the fragmentation of MPs (Dawson et al., 2018).

3.3. Factors affecting weathering processes

3.3.1. Physicochemical properties of microplastics

3.3.1.1. Size. Smaller-sized MPs have a higher specific surface area, and thus provide larger reaction areas for accelerating the fragmentation rate of MPs and leaching rate of chemicals (Luo et al., 2020b; Wang et al., 2020a). In a photo-aging experiment of PVC MPs, the dechlorination efficiency of PVC MPs had a negative correlation with particle size (Wang et al., 2020a). The concentrations of released Cl^- from 2, 10, 25 and 150 μm PVC MPs in water were 21.4, 13.2, 10.6 and 7.2 μM after 96 h xenon lamp exposure, respectively (Wang et al., 2020a). However, for semi-crystalline PE particulate plastics with sizes < 500 nm, smaller particles have higher crystallinity and more compactness of polymer chains, thus exhibiting lower thermal-degradation rates (Paik and Kar, 2009). For this reason, the weathering rate of MPs not only depends on the specific surface area, but also crystallinity and chain compactness resulting from smaller sizes.

3.3.1.2. Chemical structure. The chemical structure of MPs is one of the most essential factors determining the weathering rate (Gewert et al., 2015). Chromophore-induced light absorption is the prerequisite for the plastic photo-aging reaction (ter Halle et al., 2017). Thus the dissolved organic carbon (DOC) production and carbon mass loss of PS MPs were faster than other MPs without a chromophore (PP and PE) in seawater (Zhu et al., 2020b). Polyethylene terephthalate MPs are difficult to be biodegraded directly compared to other polyesters, and the main reason for the difficulty is that the aromatic terephthalate units limit the chain mobility, resulting in a low hydrolysis rate of the backbone ester linkages by enzymes (Webb et al., 2012).

In addition, the weathering degree is limited by the rate of hydrogen abstraction from plastic molecules or the media (e.g., H_2O) by R· and RO· (as shown in Scheme 1), which is related to the stability of C–H bonds (Hujuri et al., 2008; Song et al., 2017). The dissociation energy of C–H bonds decrease in the order of primary (418 kJ mol^{-1}) > secondary (402 kJ mol^{-1}) > tertiary (389 kJ mol^{-1}) hydrogen (Wang and Brown, 2004). Thus, MPs with tertiary hydrogen (e.g., PS, PP, and PVC) have lower weathering resistance, but MPs without tertiary hydrogen (e.g., PE) are highly stable. For example, the oxidation degree of PP film was approximately 1.75 times faster than that of PE under xenon lamp irradiation (Julienne et al., 2019b). The temperature required for 100% weight loss of PE MPs (495 °C) was found to be higher than that of PP MPs (471 °C) (Hujuri et al., 2008). To reduce the accumulation of polymer with high stability such as PE in environments, it is necessary to investigate their fates to provide an effective remediation strategy.

3.3.1.3. Crystallinity. Crystallinity of MPs reflects the order degree of the chain structure (Andrady, 2017). The crystalline region has a more tightly and ordered structure than the amorphous region (Mei et al., 2020). Therefore, MPs with high crystallinity have a limited crushing area compared to those with amorphous regions, in which crack propagation, chain scission, and mechanical breakdown occur preferentially (Julienne et al., 2019a). In addition, the direction of crystallite alignment of pristine plastics is in the direction of crack propagation, thereby affecting the shape and number of formed fragments in water under xenon lamp irradiation

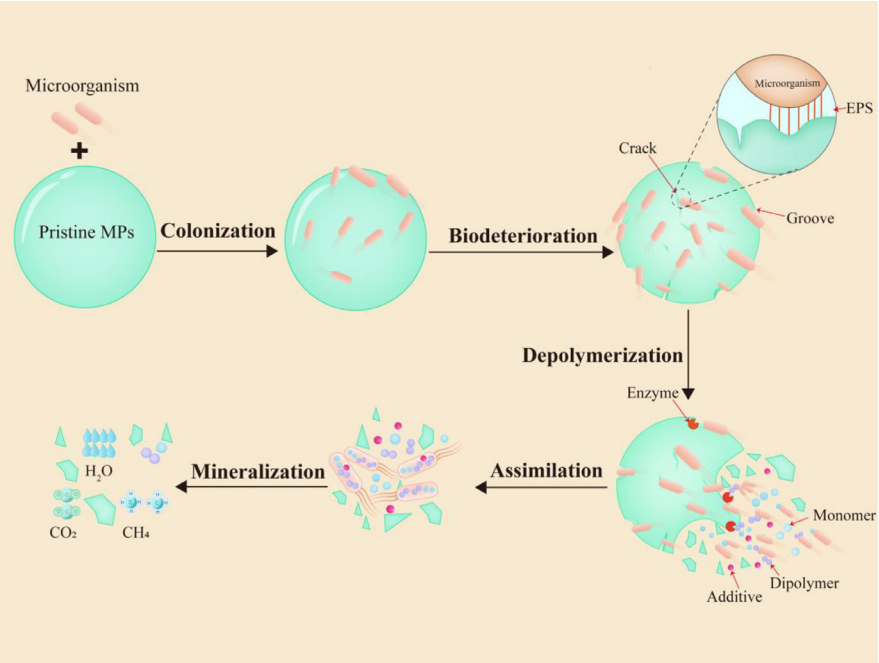


Fig. 2. Degradation pathway of microplastics by microorganisms (EPS presents extracellular polymeric substances).

(Julienne et al., 2019a). Both LDPE and PP MPs had linear crystallite structures perpendicular to the direction of extrusion lines, which caused the direction of crack propagation also to be perpendicular to the extrusion direction, and elongated shapes were formed. Besides, PP had a spherulite structure to allow the propagation of cracks along the radial direction. Eventually, the cracks caused by the linear crystallite and spherulite structures coalesced together, producing fewer elongated and smaller PP fragments than LDPE (Julienne et al., 2019a).

3.3.2. Environmental conditions

3.3.2.1. Oxygen. Oxygen plays a dominant role in the photo-degradation, thermal-degradation, and biodegradation of MPs in water and soil. Wang et al. (2020a) confirmed that oxygen might contribute to the photo-dechlorination of PVC MPs. They found that the dechlorination rates of PVC MPs (2 μm) under oxic conditions were 2.8 and 1.8 times higher than those under anaerobic conditions in oxalate and citrate aqueous solutions, respectively (Wang et al., 2020a). Additionally, oxygen can change the metabolic pathway of microorganisms. Under aerobic conditions, microorganisms use oxygen as an electron acceptor, while under anaerobic conditions, they use sulfates, nitrates, carbon dioxide, or metals as electron acceptors (Priyanka and Archana, 2011). Thermodynamically oxygen is a more effective electron acceptor than other substances (Gu, 2003), and, thus, anaerobic conditions may not be conducive to biodegradation.

3.3.2.2. Water. Water can either promote or inhibit the weathering process of MPs. On the one hand, water could limit oxygen content and prevent UV penetration. Consequently, the photo-aging rate of MPs in pure water is lower than that in air (Cai et al., 2018; Mao et al., 2020; Resmeriță et al., 2018). On the other hand, water plays a prominent role in light scattering and crack propagation. The light scattering of water in a suspension of PS MPs (250 ± 88 nm) increased the light exposure area of the MPs, which promoted the generation of ROO· or R·. They can abstract hydrogen atoms from H_2O to generate ROOH, subsequently decomposing into RO· and ·OH and providing more free radicals for the oxidation process (Tian et al., 2019). In addition, water is an essential substance for metabolism, growth, and reproduction of microorganisms; thus, suitable moisture could accelerate the rate of MP biodegradation, especially in the terrestrial environment, by promoting biofilm formation (Grima et al., 2000).

3.3.2.3. Temperature. Temperature strongly affects mechanical weathering of MPs and the biodegradation rate due to the change of microbial community structure and metabolic activities. High temperature could destroy surface mechanical properties and accelerate the movement of molecules inside MPs and promote the release of additives and monomers (Zhou et al., 2018). Meanwhile, high temperature improved enzymatic activity. Chen et al. (2020) found the mass loss of MPs in sewage sludge was 43.7% after 45 days of composting it using hyperthermophilic composting technology (*h*TC, 70 °C), whereas the mass loss was only 4.5% with conventional thermophilic composting (*c*TC, 40 °C). The predominant genera during the biodegradation of MPs during the *h*TC process were *Thermus* (54.2%), *Bacillus* (24.8%), and *Geobacillus* (19.6%) (Chen et al., 2020). It should be noted that higher temperature (85 °C) might result in inactivation of most enzymes and decrease the bacterial abundance and diversity, and, thus, it may inhibit the biodegradation of MPs (Chen et al., 2020).

3.3.2.4. Organic matter. Dissolved organic matter (DOM) in the aquatic environment and soil organic matter (SOM) in the terrestrial environment play important roles in MP photo-degradation and biodegradation (Cai et al., 2018; Liu et al., 2020c). Due to their abundant chromophores (e.g., aromatic rings and carboxyl groups), DOM and SOM serve as photosensitizers and produce hydrated electron, excited triplet states, $O_2^{\cdot -}$, ·OH, and $^1O_2$ (Li et al., 2015), which promote the MP photo-aging process. Liu et al. (2019c) suggested that DOM in the Taihu Lake and Yangtze River might be an important factor leading to different photo-aging rates of PP and PS MPs. Furthermore, DOM and SOM are considered

as some of the most important carbon sources for microorganisms in water and soil environments (Xue et al., 2012). However, Blöcker et al. (2020) found that PP and LDPE MPs were barely biodegradable in soil. The results might be attributed to the short incubation time (28 days) and low mobility of DOM and SOM in soil. Future work should determine the relationship between DOM/SOM and MP biodegradation at long-term research sites.

3.3.2.5. Salinity and ion species. Salinity and ion species affect the photo-degradation rate of MPs in the aquatic ecosystem by influencing depth of UV penetration and radical reaction. Increasing salinity results in a high refractive index of water and the formation of attached salt crystals on the MP surface, which, thus, protect MPs from photo-degradation by decreasing the absorption efficiency of light (Cai et al., 2018; Ranjan and Goel, 2019). HCO_3^- and Cl^- are considered as the main ions in freshwater and seawater, respectively (Chen et al., 2019a; Hansard et al., 2011), which can scavenge ·OH with reaction constants of 4.3×10^9 M^{-1} s^{-1} and 8.5×10^6 M^{-1} s^{-1}, respectively (Liao et al., 2001). Therefore Cl^- and HCO_3^- in natural water might weaken the promotion effect of ·OH on photo-aging. However, the common trace metal ions (e.g., iron and manganese) in natural water act as catalysts to oxidize and break MP chains under light irradiation (Leonas, 1993).

3.3.2.6. Biofilm formation. The surfaces of MPs with various chemical compositions, roughness, and densities can serve as substrates for biofilm formation, especially in aquatic environments (Turgay et al., 2019). Biofilms can either promote the deterioration of the structure of MPs by secreting enzymes and slime matter or inhibit photo-aging and the mechanical breakdown process, because the formed dense layer can shield the surface from light irradiation and shear forces (Yuan et al., 2020). At the same time, biofilms can increase the density of MPs, such as PE MPs, and result in their sinking to the bottom of the water column (Bråte et al., 2018). Settlement further leads to the exposure of MPs to low temperatures and weak light, which may, consequently, inhibit the photo-aging process or thermal-degradation.

4. Impact of weathering on properties of microplastics

4.1. Surface functional groups

In general, weathering promotes the generation of different oxygen-containing functional groups, mainly C = O, O–H, and C–O on the surface of MPs (Ding et al., 2020; Zhang et al., 2020c). Most studies have used the carbonyl index (CI) and the hydroxyl index (HI), determined by FTIR, to evaluate the weathering degree of MPs (Wu et al., 2020a). The types and generation order of oxygen-containing functional groups depend on the weathering process and environmental conditions. In an aqueous environment, the phenolic hydroxyl group was preferentially formed on the surface of PS MPs, because of excessive hydrogen atoms, while, in a dry environment, the C = O group was more likely to be formed under UV irradiation (Mao et al., 2020). Moreover, under UVB irradiation for 12 months, the surface of PP MPs evolved into O–H and C = O groups, but only the O–H group was observed after thermal weathering at 50 °C for 12 months or at 100 °C for 6 months (Tang et al., 2019).

4.2. Color change

Most MPs can undergo visible color changes, which is considered as an intuitive indicator of weathering behavior (Luo et al., 2020d). A colorimeter is applied to measure the color intensity of pristine and weathered MPs based on the CIE 1976 L*a* b* color system (Robertson, 1977). An increase in the value of L* indicates that the color of MPs is lightening (Robertson, 1977). The color coordinates a* and b* represent the red/green coordinate and the yellow/blue coordinate, respectively (Robertson, 1977). The total color change (ΔE^*) of MPs is calculated and considered as an index of color change according to Eq. (1) (Robertson, 1977):

$$\Delta E^* = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2} \quad (1)$$

where ΔL^*, Δa^*, and Δb^* are the differences between pristine and weathered values of L*, a*, and b*, respectively.

A laboratory study demonstrated that the ΔE^* value of pristine PE MPs increased drastically from 0 to 8.4 after a 6-week exposure to a xenon lamp (Luo et al., 2020d). This phenomenon indicated that the chromaticity of MPs was primarily correlated with the oxidation reaction (Luo et al., 2020d). During the chemical weathering process, yellow discoloration is a common aging result for most of the white, off white, or translucent MPs, which is attributed to the formation of chromophores during the aging process (Battulga et al., 2020). For example, phenolic antioxidants in MPs are oxidized into by-products with quinoidal structures that result in a yellow discoloration of MPs (Battulga et al., 2020).

4.3. Size and surface morphology

The decrease in particle size and increase in surface roughness are frequently observed for weathered MPs (Liu et al., 2019c; Song et al., 2017). The mechanical forces from wave, wind, and sand abrasion lead to the loss of mechanical stability, in which the tensile strength of MPs is an important factor (Liu et al., 2019c; Song et al., 2017). As shown in Fig. 3a-c, the photo-aging process causes the formation of a brittle surface and cracks on the MP surface, which can accelerate the fragmentation rate after the action of physical forces. After exposure to UV (12 months) and subsequent mechanical abrasion (2 months), the fragment number of LDPE, PP, and expanded PS was 699, 569, and 2 times more than those with only mechanical abrasion for 2 months, respectively. Moreover, SEM images of aged PP MPs showed obvious pits, microcracks, grooves, or broken edges on their surfaces, as shown in Fig. 3d (Song et al., 2017). Correspondingly, the mean particle size of MPs decreased along with the increase in the number of fragments (Zhu et al., 2020a).

4.4. Crystallinity

An increase in crystallinity can be considered as an index of the weathering of MPs (Wu et al., 2020a). Weathering preferentially degrades the amorphous portion of plastics, thus increasing the percentage of the crystallization region (McGivney et al., 2020). The shorter chains from chain scission can crystallize more readily than longer chains due to their higher mobility, and this accelerates the re-crystallization process in later stages of the weathering process (Andrady, 2017; Julienne et al., 2019a). The degree of crystallinity of MPs is measured by an X-ray diffractometer (XRD) and differential scanning calorimetry (DSC) (McGivney et al., 2020; Wu et al., 2020a). The XRD patterns shown in Fig. 4 indicated that the weathering process improved the crystallinity of PS MPs, which increased in the order of pristine PS (62.5%) < seawater-aged PS (63.9%) < UV-aged PS in seawater (65.0%) < UV-aged PS (66.8%) (Wu et al., 2020a). Moreover, the DSC exotherms revealed that the presence of biofilms increased the crystallinity of PE MPs by 3.2% (McGivney et al., 2020).

4.5. Leaching of chemicals

Weathering of MPs can leach chemicals, such as additives, monomers, and oxygenated intermediates, as shown in Table 3

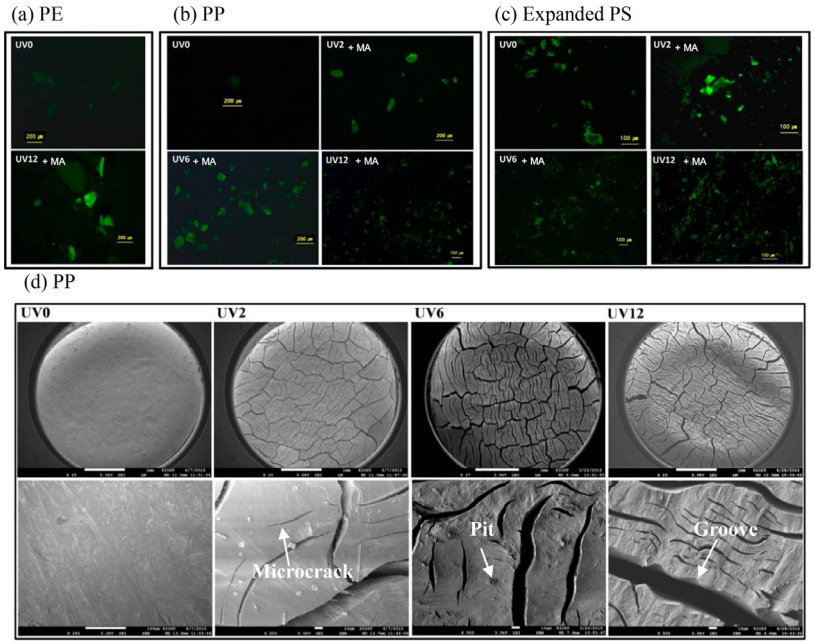


Fig. 3. Fluorescent images of fragment of (a) polyethylene (PE), (b) polypropylene (PP), and (c) expanded polystyrene (PS) MPs after different UV exposure time (0, 2, 6, 12 months) and subsequent mechanical abrasion (MA) by sand (2 months); (d) SEM images of PP after different UV exposure time (0, 2, 6, 12 months). UV0, UV2, UV6 and UV12 represent UV exposure time for 0, 2, 6 and 12 months. Graph was cited and reproduced from "Combined effects of UV exposure duration and mechanical abrasion on microplastic fragmentation by polymer type" (Song et al., 2017) with permission.

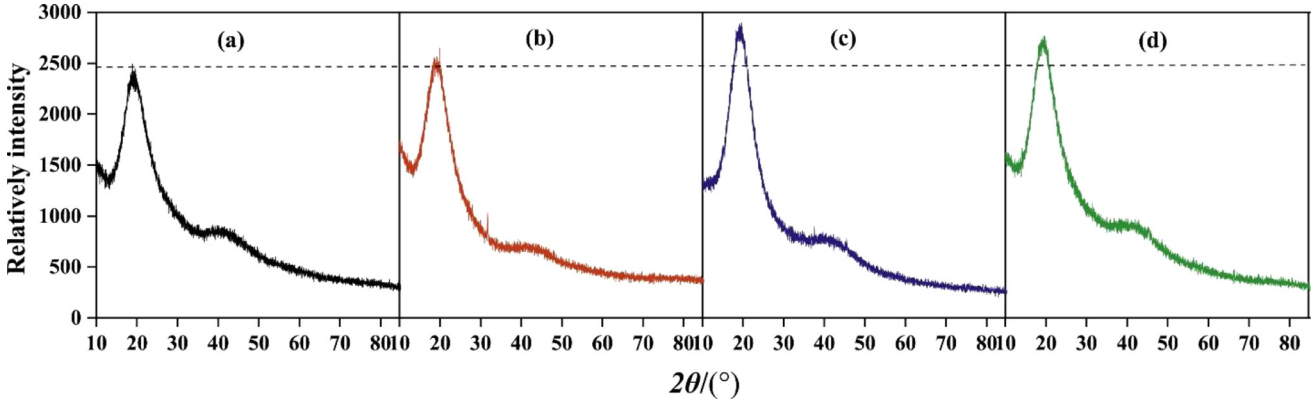


Fig. 4. The XRD patterns of (a) pristine PS, (b) aged PS via seawater soaking, (c) aged PS via UV irradiation and (d) aged PS via concurrent seawater soaking and UV irradiation. Graph was cited and reproduced from "Effects of polymer aging on sorption of 2,2′,4,4′-tetrabromodiphenyl ether by polystyrene microplastics" (Wu et al., 2020a) with permission.

(Luo et al., 2020b; Zhang et al., 2019; Zhu et al., 2020b). The released additives mainly include endocrine disrupting chemicals (EDCs) (e.g., phthalate and bisphenols) and metal ions (e.g., Cr^{6+} and Pb^{2+}) in the aqueous environment. Dissolved organic carbon and CO_2 were major by-products of MPs through chain scission and oxidation reactions in an aquatic environment under UV irradiation (Ward et al., 2019; Zhu et al., 2020b). In addition, light irradiation promotes chemical leaching, because the formation of cracks and fragments in the weathered MPs provides a larger contact area for chemicals with oxygen or for the aqueous solution (Luo et al., 2020b). For MPs sampled from the North Pacific Gyre, the leaching rate of DOC in light (0.235 ± 0.003 mg g^{-1} D^{-1}) was 10 times faster than that in the dark (Zhu et al., 2020b). By contrast, other studies reported that UV irradiation decreased the concentrations of some released chemicals (such as organotin and DOC), because of their photodegradation, volatilization, and reabsorption onto the MP surface (Chen et al., 2019a; Romera-Castillo et al., 2018).

5. Interactions between weathered microplastics and coexisting constituents

The increased surface area and hydrophilicity of weathered MPs are of great importance for the interaction between weathered MPs and other coexisting constituents, which includes adsorption of inorganic/organic contaminants and homo-/hetero-aggregation with other coexisting solid constituents (Liu et al., 2019b, 2020b; Wang et al., 2020b). These weathering-induced environmental behaviors further affect transport and bioavailability to organisms of MPs (Liu et al., 2019b).

5.1. Sorption of inorganic contaminants

Weathered microplastics can promote the absorption of potentially toxic elements including heavy metals and metalloids via ion complexation, hydrogen bonding, and electrostatic interaction forces as shown in Fig. 5 (Dong et al., 2020; Wang et al., 2020b). Weathered MPs have a positive correlation between their adsorp-

Table 3
The types and concentrations of released chemicals from weathered MPs.

Type of chemicals	Released chemicals	Concentration	MP type	Source	Weathering process	Reference
Additive	EDC	1.10 and 0.25 $\mu g\ g^{-1}$ for small (0.5–1.5 mm) and medium (1.5–5 mm) MPs, respectively	PE	The North Pacific Subtropical Gyre	Natural weathering	(Chen et al., 2019b)
	DiBP, DnBP	0.083, 0.120 $\mu g\ g^{-1}$	PE	Plastic garbage bag	Artificial light weathering	(Paluselli et al., 2019)
	BPS, BPAF	0.012, 0.070 $\mu g\ g^{-1}$	PP	Disposable plastic boxes	Thermal weathering (121 °C)	(Zhou et al., 2018)
	DMP, DEP	0.010, 0.069 $\mu g\ g^{-1}$	PVC	Insulation layer of electric cables	Artificial light weathering	(Paluselli et al., 2019)
	DMT, DBT	1.51–14.48, 1.03–4.55 $\mu g\ g^{-1}$	PVC	Thin sheet	UV 365 nm weathering	(Chen et al., 2019a)
	THMs	90–454 $\mu g\ L^{-1}$	PE, PP, PLA, PMMA, PS	Commercial products	UV 340 nm weathering	(Ateia et al., 2020)
	Cr (VI) Pb (II)	12.1, 81.4 $\mu g\ g^{-1}$	PE	Raw plastic masterbatches	Xenon lamp weathering (120 mW cm^{-2})	(Luo et al., 2020a)
	Cd (II)	98.95 $\mu g\ g^{-1}$	PP	Plastic buckets	Xenon lamp weathering (61.5–70.6 mW cm^{-2})	(Liu et al., 2020a)
Monomer	TPA	0.085–75 $\mu g\ g^{-1}$ (dw)	PET	Sewage sludge	Natural weathering	(Zhang et al., 2019)
	BPA	0.0083–2.5 $\mu g\ g^{-1}$ (dw)	PC	Sewage sludge	Natural weathering	(Zhang et al., 2019)
Oxygenated compounds	TOC	7130 $\mu g\ L^{-1}$	PS	Synthetic MPs	UV 254 nm weathering (0.7 mW cm^{-2})	(Tian et al., 2019)
	DOC	2870 $\mu g\ L^{-1}$	PS	Sigma-Aldrich corporation	UVA weathering (4 mW cm^{-2})	(Lee et al., 2020)
	DOC	6.0, 1.12 $\mu g\ cm^{-2}$	LDPE, HDPE	GoodFellow	Artificial light weathering	(Romera-Castillo et al., 2018)
	DOC	1280 $\mu g\ L^{-1}$	PVC	Sigma-Aldrich corporation	UVA weathering (4 mW cm^{-2})	(Lee et al., 2020)

Note: Bisphenol A (BPA), Bisphenol AF (BPAF), Bisphenols (BPS), Dimethyltin byproducts (DMT), Dimethyl phthalate (DMP), Diethyl phthalate (DEP), Di-isobutyl phthalate (DiBP), Di-n-butyl phthalate (DnBP), Dibutyltin (DBT), Dissolved organic carbon (DOC), Endocrine disrupting chemicals (EDC), High density polyethylene (HDPE), Low density polyethylene (LDPE), Polyethylene (PE), Polypropylene (PP), Polystyrene (PS), Polyvinylchloride (PVC), Poly (methyl methacrylate) (PMMA), Polylactic acid (PLA), Polyethylene terephthalate (PET), Polycarbonate (PC), Terephthalic acid (TPA), Trihalomethanes (THMs), Total organic carbon (TOC).

tion capacities for the inorganic contaminants and CI values in aqueous solution, indicating that the degree of weathering of MPs influences the adsorption ability of heavy metal ions and metalloid ions (Mao et al., 2020; Wang et al., 2020b; Yang et al., 2019). A 1.6-fold increase of equilibrium Zn^{2+}adsorption capacity was observed by 300 h UV-aged PET MPs (86.8 $\mu g\ g^{-1}$) over the pristine MPs (54.7 $\mu g\ g^{-1}$) in aqueous solution (Wang et al., 2020b). The higher adsorption capacities of weathered MPs are principally due to the following three reasons: (1) increased chapped, wrinkled, and rough surfaces of weathered MPs provide more active sites than pristine MPs for contaminants (Fu et al., 2019; Wang et al., 2020b), (2) oxygen-containing functional groups strengthen the interaction forces between MPs and metal cations by ion complexation and hydrogen bonding (Brennecke et al., 2016; Mao et al., 2020), and (3) increased electronegativity on aged MP surfaces enables greater electrostatic attraction between metal cations and MPs (Yang et al., 2019).

In addition, biofilms on weathered MPs have been shown to act as vectors of metal cations. The EPS in biofilms include O–H and C = O groups, promoting Cu^{2+} adsorption onto the surface of PE MPs via complexation (Wang et al., 2020d), which highlights the probable effect of biofilms on migration and transformation of inorganic contaminants in the environment. Biofilm coverage can change the mechanism of Cu^{2+} diffusion from intra-particle diffusion of pristine MPs into film diffusion of aged MPs, which facilitates Cu^{2+}adsorption (Wang et al., 2020d).

5.2. Sorption of organic contaminants

As shown in Fig. 5, hydrogen bonding and electrostatic interaction play dominant roles for sorption of hydrophilic organic contaminants (e.g., tetracycline and ciprofloxacin) on weathered surfaces of MPs due to increased oxygen-containing functional groups, high hydrophilicity, and electronegativity (Liu et al., 2019a; Wu et al., 2020b). Compared to pristine MPs, the maximum adsorption capacity of tetracycline and ciprofloxacin on UV-aged PLA MPs increased by 2.2 (from 2.5 to 5.5 mg g^{-1}) and 1.2 (from 3.2 to 3.8 mg g^{-1}) fold, respectively (Fan et al., 2021). On the contrary, a decrease of hydrophobic interaction forces is the primary reason for lower adsorption capacity for hydrophobic organic contaminants (HOC) on weathered MPs. For example, the equilibrium sorption capacity of 2,2′,4,4′-tetrabromodiphenyl ether (BDE-47) on UV-aged PS MPs (3.75 ng g^{-1}) decreased to half of that for the pristine PS (6.16 ng g^{-1}) (Wu et al., 2020a). In addition, π-π interaction was shown to be the key interaction mechanism between pristine PS MPs and aromatic compounds. Although the π-π interaction might be weakened due to the opening of phenyl rings of PS MPs during the UV aging process (Liu et al., 2020b, 2020c), the interaction seems to play a minor role compared with the above interaction forces on weathered MPs.

5.3. Homo-aggregation or hetero-aggregation with other solid particles

5.3.1. Homo-aggregation of weathered MPs

Weathered MPs can influence their aggregation behavior mainly by changing hydrophilic interaction, electrostatic interaction, van der Waals interaction, and biofilm formation (Fig. 6). Ultraviolet irradiation inhibited the homo-aggregation of PS NPs (50–100 nm) in NaCl solutions, which was primarily because the formed oxygen-containing functional groups on the PS NP surface improved the electrostatic repulsion between two approaching particles (Liu et al., 2019d). Similar phenomena were observed in the soil environment (Liu et al., 2019b). Under UV irradiation, the surface oxidation of PS NPs increased their negative charges and hy-

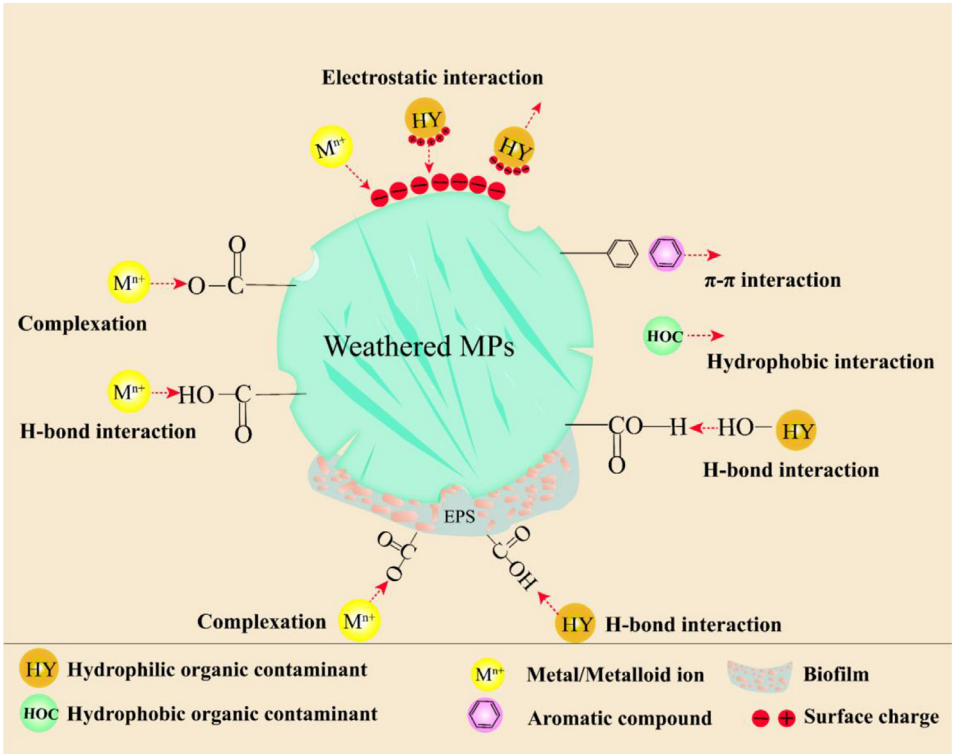


Fig. 5. Interaction between weathered microplastics and organic/inorganic contaminants (the arrows pointing to and away from the weathered microplastics represent the promotion and inhibition of sorption, respectively). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

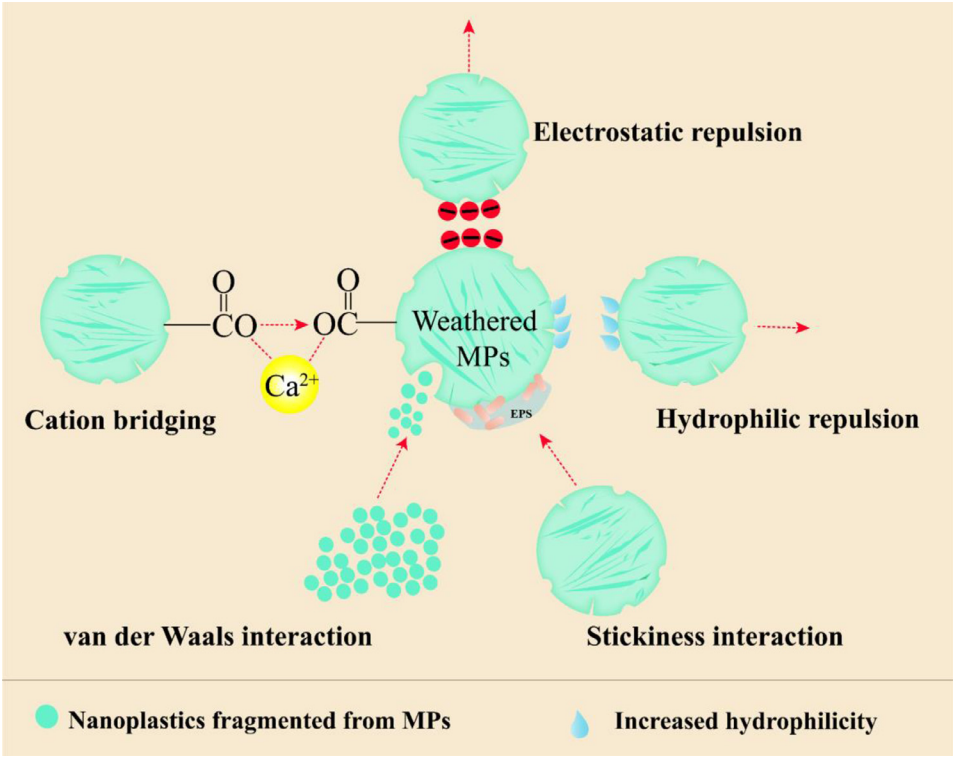


Fig. 6. Homo-aggregation of weathered microplastics (The arrows pointing to and away from the weathered microplastics represent the promotion and inhibition of aggregation, respectively, EPS represents extracellular polymeric substances). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

drophilicity in saturated loamy sand, thus decreasing the aggregation of MPs (Liu et al., 2019b).

By contrast, UV light promoted the aggregation of aged PS NPs in $CaCl_2$ solutions, which resulted from the bridging effect between the formed oxygen-containing functional groups and Ca^{2+} (Liu et al., 2019d). In water environments, most of the MP surfaces were covered by biofilms, which secreted sticky EPS served as tackiness agents, which subsequently accelerating homo-aggregation of MPs (Michels et al., 2018). Additionally, NPs formed by MP fragmentation via mechanical weathering (e.g., shear forces) were prone to homo-aggregation, which was attributed to the increased surface energy. Nanoplastics with high surface energy tended to aggregate to minimize their Gibbs free energy and reached a stable state (Enfrin et al., 2020). However, the aggregates were readily dispersed by repeated shear forces, due to their low cohesion forces (low van der Waals force) (Enfrin et al., 2020).

5.3.2. Hetero-aggregation of weathered MPs with other particles

Weathering of MPs has a greater impact on hetero-aggregation of MPs with other solid particles than homo-aggregation due to the abundance of coexisting solid constituents in the natural environment (Alimi et al., 2018). Michels et al. (2018) investigated the effect of biofilms on the hetero-aggregation between PE MPs and biogenic particles (phytoplankton) in the aquatic environment. With the naked eye, they were able to observe hetero-aggregate formation by pristine PS MPs in seawater with phytoplankton after one day, while biofilm-covered PS MPs hetero-aggregates with biogenic particles were obvious just after a few hours. However, the interaction of weathered MPs with other solid particles (e.g., suspended sediments and microalgae) in water has yet to be elucidated.

6. Knowledge gaps and future recommendations

6.1. Evaluate the weathering process under actual environmental conditions

Most published studies have concentrated on investigating the weathering of MPs under laboratory-simulated environments (Chen et al., 2019a; Enfrin et al., 2020; Luo et al., 2020c), and only a few studies have focused on the weathering of MPs under natural environmental conditions (Chen et al., 2020; Tu et al., 2020). Even though weathering phenomena and mechanisms have been concluded from these studies, it is controversial whether the laboratory-simulated conditions represent environmentally relevant systems. For example, photo-weathering experiments with MPs commonly use a xenon lamp to simulate sunlight. However, the latitude, longitude, solar elevation angle, and meteorological conditions determine the intensity and spectral distribution of sunlight (Casado et al., 2019), which cause the difference between MP weathering with a xenon lamp and natural sunlight. Some potential influencing factors (e.g., ions, natural organic matter, and plant secretions) might exert an influence on the MP weathering. Furthermore, most MPs undergo various weathering processes at the same time in natural environments, which results in complexity and uncertainty of the weathering behavior and mechanisms. Considering the complexity of natural systems and the weathering processes, future studies should explore multiple weathering behaviors and mechanisms of MPs under actual aquatic or terrestrial environmental conditions.

6.2. Consider the effect of microplastic diversity on weathering

Most weathering studies of MPs have used traditional and spherical plastic particles, such as PP, PS, PE, PVC, and PET. Recently, MPs with irregular shape and new plastics, including biodegradable MPs (e.g., polyhydroxy butyrate) and functional MPs (e.g., carboxyl-modified PS), are being released into natural environments (Gonzalez-Pleiter et al., 2019; Wang et al., 2020c), and they have been rarely considered in weathering studies. Meanwhile, different types of MPs co-exist in the field. However, only a single type of MPs is used in weathering experiments, which ignores the mutual influence or competition among MPs. For instance, the excited state of the benzene ring (4.59 eV) on PS could transfer energy to the excited state of the carbonyl (4.09 eV) on PP by an intramolecular energy transfer process. Then the energy can break the C–H bond of PP, generating more high-activity tertiary carbon radicals under UV irradiation (Waldman and De Paoli, 2008). More research should be carried out using new synthetic, irregularly shaped, and mixed MPs as model particles, to provide a more reasonable understanding of MP weathering.

6.3. Improve the understanding of the weathering process in soils or sediments

Factors influencing the weathering processes of MPs have been mainly studied in aquatic environments with little attention paid to terrestrial environments. In addition to biological phenomena, mechanical degradation, photo-degradation, and thermal-degradation may also influence the distribution and fate of MPs in soils. Research investigating the influence of soil conditions (e.g., moisture content, mineral materials, microbial species, and soil physico-chemical parameters) on the MP weathering processes is urgently needed. Previous studies have suggested that sediments in the ocean and river estuaries accumulate MPs and become long-term sinks for MPs (Xu et al., 2020b; Zhang et al., 2020b). The anaerobic environment and special microbial habitat of sediments may result in MPs undergoing anaerobic biodegradation and mechanical fragmentation. It is of great importance to focus on the weathering processes of MPs in soils or sediments, where the low oxygen content and weak light intensity may limit the extent of photo-weathering reactions.

6.4. Strengthen research on the interaction between weathered MPs and coexisting constituents

In water environments, the stability and mobility of MPs change during the aging process, because of altered zeta potentials, surface areas, roughness, and hydrophobicity (Wang et al., 2020b; Wu et al., 2020a). These altered physicochemical properties may also influence the interaction of MPs with other solid particles. Hetero-aggregation of weathered MPs in water remains elusive. How and whether the aging process interferes with homo-aggregation and hetero-aggregation of weathered MPs with other particles in terrestrial environments are unknown, due to the lack of separation, identification, and quantification methods for particle size of MP aggregates. Research regarding the interaction of weathered MPs with other pollutants has mainly concentrated on the sorption of a single heavy metal or organic matter (Dong et al., 2020; Wang et al., 2020b). Competitive adsorption of weathered MPs for mixed pollutants needs to be studied, because many heavy metals and organic pollutants co-exist in natural environments. Weathered MPs can release additives, monomers, or oxygenated intermediates, which may be reabsorbed onto the surfaces of MPs (Liu et al., 2020b). Further work is needed to better understand the effect of the reabsorption process on heavy metals and organic pollutants.

6.5. Concern MP weathering process in waste water

Most previous research has concentrated on the weathering of MPs in natural water, and, sporadically, research has consid-

ered the soil, but ignored the weathering process in waste water (e.g., landfill leachates), which is also a major source of MPs. Large amounts of plastic waste disposed in landfills or dumpsites are subjected to multiple weathering processes, which depend upon leachate pH (4.5–9), salinity, concentration of heavy metals and organic pollutants, and bacterial communities (He et al., 2019; Sun et al., 2021). For example, the transition metals in landfills or leachates (e.g., Fe and Cu) might promote the decomposition of organic molecules (e.g., ROOH), and thus accelerate polymer weathering (Hou et al., 2021). Further research is needed to investigate the weathering processes in waste water under the influence of complex field conditions that are different from those in the natural, ambient environment (e.g., surface water).

7. Conclusions

The weathering of MPs is an important process determining their transport, transformation, and interactions with contaminants and microorganisms in water and soil. Sources of MPs are usually linked to anthropogenic activities including refuse in landfills, biowaste application, plastic film utilization, and wastewater discharge. Microplastics undergo various weathering processes, including mechanical fragmentation, photo-degradation, thermal-degradation, and biodegradation. The physicochemical properties of MPs and environmental conditions affect the degree of their weathering. Generally, MPs, which have a small size and large specific surface area, provide many reaction sites; MPs with a low degree of crystallinity facilitate the diffusion of oxygen, water molecules, and radicals, which accelerate the weathering rate. However, some factors have opposing effects on the weathering of MPs. For example, biofilm formation on MP surfaces promotes plastic biodegradation, but inhibits photo-degradation. The surface-property modification of MPs resulting from weathering strongly affects the sorption or aggregation of weathered MPs and other co-existing constituents by influencing interactive forces, such as hydrogen bond interaction, hydrophilic interaction, electrostatic interaction, and van der Waals interaction. To predict fully the fate and environmental interactions of MPs, more knowledge is needed concerning the *in-situ* weathering behavior of different types of MPs that co-exist in natural aquatic and terrestrial environments.

Declaration of Competing Interest

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

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