



DEGRADATION OF POLYMER MATERIALS IN ENVIRONMENT

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Abstract

Environmental occurrence of the polymer-based materials (PBMs) and their associated chemical additives have been recognized as an emerging worldwide problem, and their impacts are now a subject of wider scientific and social interest. Production of PBMs have increased substantially over the last few years due to their flexible nature, making them compatible for use in domestic, industrial and developmental purposes. PBMs can be released into the environment from a variety of sources. The principal introduction routes being general littering, dumping of waste materials, migration from landfills, emission during refuse collection and air-blasting technologies. Once in the environment, PBMs can be broken down by thermal, biological and chemical processes. A number of literatures to date have addressed the impacts of PBMs in the environment and eventually on human health. Bulk PBMs are well documented as entanglement and ingestion hazards. Interaction of PBMs with persistent organic pollutants (POPs) have been shown to biomagnify in food webs causing traumatic injury and cardiovascular diseases in human body. This review summarizes up-to-date findings about the source and occurrence of PBMs in both aquatic and terrestrial environments, various degradation processes of PBMs and most important, the harmful effects of PBMs on environment and human.

Keywords: Aquatic, environment, food webs, organic pollutants, terrestrial.

Introduction

Polymer-based materials (PBMs) are a complex mixture of their respective polymer/s and their various additive compounds (Plastics Europe 2021). The first synthetic mass produced polymer was Bakelite, developed by the Belgian chemist Leo Baekeland in 1909. Later in the 1900s the modern form of PVC was created in 1926, followed by polyurethane (PUR) in 1937, a more processable polystyrene in 1938, and high-density polyethylene (HDPE) and polypropylene (PP) in 1951 (Hammer *et al.* 2012). More recently, technological advances have seen the development of polymers produced from the bacterial fermentation of sugars and lipids, and include polyhydroxyalkanoates (PHA), polylactides (PLA), aliphatic polyesters and polysaccharides (Martinho *et al.* 2022).

Different types of polymers have different properties due to their variations in molecular structure, size of the molecule, density of the polymer and functional groups. In general, polymers consist of a range of organic and

inorganic molecules formed from elements, such as carbon, silicon, hydrogen, nitrogen, oxygen and chloride (Martinho *et al.* 2022). These elements are joined together in different bond combinations to achieve the differences in polymer properties. Polymers in their pure states are not generally usable as a commercially viable material. Therefore, polymers are processed with a range of compounds to adjust their characteristics which make them suitable for their intended purpose.

Increasing human population and associated economic growth has led to an increase in the demand for consumable goods such as those made from PBMs (i.e., plastics and elastomers). During their lifecycle PBMs can be released into the environment from a variety of sources. Once in the environment, PBMs are exposed to a variety of mechanical and chemical weathering processes. This causes a change to the PBM structure and facilitates the disintegration of the PBM into increasingly smaller fragments (Martinho *et al.* 2022). Furthermore, these materials are now thought to be contributing to the build-up of chemicals in the environment via the leaching of chemical additives that

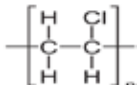
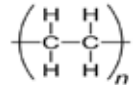
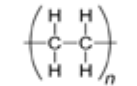
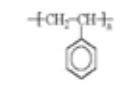
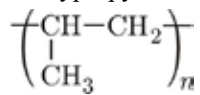
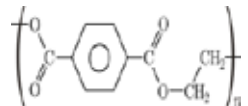
are used in the manufacturing process (Erren *et al.* 2009). The majority of physical effects data regarding bulk PBM items identifies them as presenting a hazard to mammals and birds as they can become entangled and/or mistake PBMs as a food source (Mao *et al.* 2022). The majority of ecotoxicity data regarding PBM additives has focused on the effects of compounds that are generally referred to as having endocrine disruptive potential, such as the phthalates (Oehlmann *et al.* 2009). However, receiving environments are potentially exposed to a combination of both these physical and chemical components, as well as substances produced during degradation processes. Therefore, PBMs and their associated degradation products may compromise the viability of organisms at all trophic levels. At the base of the food chain primary producers may be more sensitive to substances that have a biological action. Non-selective and filter-feeding consumers could be susceptible to ingesting both bulk

PBMs and fragmented particles, leading to the potential passage up the food chain to secondary and tertiary consumers (Mao *et al.* 2020, Oehlmann *et al.* 2009). The purpose of this review is to shed light on the release, occurrence and degradation of PBMs and their associated chemical additives in aquatic and terrestrial environments.

Types and production of PBMs

There are currently twenty different groups of PBMs, each with numerous grades and varieties. The PBMs used in society today are made from a broad class of materials that are both natural and synthetic in origin (Table 1). Production of polymers has increased substantially over the last sixty years from around 1.5 million metric tons in 1950 to approximately 367 million metric tons in 2020 (Dehaut *et al.* 2016). Plastics Europe (2021) have predicted that the net polymer production by 2050 will be around 34 billion tons.

Table 1. Characteristics and uses of major polymer types in Europe

Polymer type	Global polymer usage (%)	Characteristics
Polyvinylchloride 	10.3	Rigid; can be blended with other materials; readily available; good tensile strength.
Low density polyethylene 	17.2	High tensile strength; high impact and puncture resistant; good electrical properties.
High density polyethylene 	12.1	Large strength to density ratio; strong intermolecular force and tensile strength; can tolerate higher temperatures.
Polystyrene 	7.0	Thermoplastic; brittle; poor barrier to oxygen and water vapour; low melting point.
Polypropylene 	19.2	Low density; high stiffness; heat resistance; chemical inertness; steam barrier properties; good transparency; good hinge property.
Polyethylene terephthalate 	7.0	Absorbs little water; gas barrier properties; chemical resistant, transparency.

Releasing phenomena of PBMs to environment

Polymer-based materials (PBMs) may enter the environment from both ocean- and land-based sources. Ocean-based sources include items lost or discarded from commercial fishing vessels, offshore oil or gas platforms and waste dumped by recreational boat users. Losses of cargo can also occur from shipping during bad weather events or accidents and items lost from improper loading, unloading or on-board storage (Liang *et al.* 2021). In the past, pre-production polyethylene (PE) and polypropylene (PP) pellets have reportedly been used on the decks of ships to reduce friction when moving large objects; as such, many of these pellets are washed from the deck and are dispersed by winds and ocean currents (Borrelle *et al.* 2020).

On land, general and accidental littering are important routes of environmental entry of PBM debris. General littering is the direct dropping of litter and dumping of items; for example, illegal dumping of waste that can then be transported by wind or from drainage and storm water runoff to ocean sinks. Littering at festival sites is noted as an issue; especially from sites that have inadequate waste management systems. Accidental littering, results from windblown debris from bins, or from recycling and landfill facilities (Cierjacks *et al.* 2012). Landfills are a major end-of-life disposal route for PBMs (Schyns and Shaver 2021). In most developed regions of the world, waste is collected, transferred to landfills and is typically covered with soil daily (Liang *et al.* 2021). However, in many developing regions waste materials are often disposed of in areas lacking adequate infrastructure, and are rarely or inadequately covered with soil. This increases the likelihood of windblown debris migrating from landfill sites. PBMs associated with personal hygiene products and microscopic polyethylene beads found in some hand cleaners can constitute a portion of this waste stream. Larger items are generally removed by screening methods, but may enter the environment during sewage overflow events that occur during periods of heavy rainfall. The ability of sewage treatment works to process microscopic beads and fibres has now been questioned.

Industrial sources of PBM waste include air-blasting technologies that use microscopic beads to strip paint from metallic surfaces and for cleaning engine parts; when

discarded, they enter the environment through foul-water, or via transfer through sewage treatment processes (Mao *et al.* 2020). Low density polyethylene (LDPE) films constitute a large-volume use of PBMs in agricultural crop production, and consequently they have become an important agricultural emission. Their application is thought to be one of the most important sources of PBM contamination of soils, because they become brittle and easily disintegrate rendering their recovery difficult. Agriculture films can also contain light-sensitive additives, such as ferric and nickel dibutyldithio-carbamates, the ratio of which can be adjusted so that the film is usable during a specific growing season, after which the product begins to photo-degrade. This ultimately results in disintegration of the materials (Lozano and Rillig 2020, Schyns and Shaver 2021).

Occurrence of PBMs in oceans

Large item of PBM debris, known as ‘macro-plastics’ (>5 mm in diameter,) provides an opportunity to assess markings to trace an object to its origin. PBMs are believed to contribute up to 80% of all anthropogenic debris in the oceans (Mao *et al.* 2020). A well-documented example are pre-production PE and PP (polypropylene) pellets that are transported from manufacturing plants to plastic injection factories, where they are melted and molded into consumer products. These pellets have been reported floating in coastal surface waters, and in the world’s oceans (Lozano and Rillig 2020, Moore 2018). Lightweight items, such as PE bags, polystyrene (PS) foam items and polymer bottles, inappropriately disposed of on land, can be readily transported long distances via a windblown route or carried by freshwater to eventually accumulate in the oceans (Ryan *et al.* 2009).

Shorelines around the world have been found to accumulate debris, including island shorelines far from any centres of human activity. Ana *et al.* (2023) surveyed beach litter on Ducia Atoll in the south Pacific and found 953 items of debris over a 1.5 miles survey transect. Evidence of the increasing occurrence of PBMs is provided from Scotland, where Duis and Coors (2016) found 0.35 items of litter/m², with plastics accounting for 29% of items found. Ten years later the same area of beach was surveyed and the density of litter was found to have increased to 0.8 items of litter/m², with PBMs accounting for 37% of

items found (Ding *et al.* 2020). Particles <5 mm, known as microplastics, formed as a result of the breakdown of larger materials, are now found floating on the ocean surface, mixed into the water column, and embedded in bottom sediments and beach sands (Ding *et al.* 2020). In 2004, the CPR survey, the longest running plankton monitoring program in the North Sea and North Atlantic, added micro-plastic as their first non-biological marine entity to their recordings (Richardson *et al.* 2006). One area that has received particular attention is the subtropical accumulation zone in the North Pacific gyre. In this area, debris has accumulated at such high densities as a result of high atmospheric pressure and the clockwise rotation of ocean currents that forces debris into a central area where strong winds and currents diminish. Neuston sampling at 11 sites, using a mantra trawl, estimated a mean PBM abundance of 334,271 pieces km² (Moore 2018). Infra-red spectroscopy techniques have been utilized to identify fragment PBMs in the microscopic range by comparing spectra to those in a database of common polymers. This technique was principally pioneered by Thompson *et al.* (2004), whose research identified synthetic fibres (PE, PP, PS) in samples of beach sand and sub-tidal sediments from around the UK.

Nature of degradation of PBMs in environment

Once in the environment PBMs are degraded through abiotic or biotic factors working together or in sequence; these processes cause the polymer matrix to disintegrate, resulting in the formation of fragmented particles of various sizes and leached additives. There are now a number of studies whose authors have investigated the degradability of a range of PBMs under a range of exposure conditions (Table 2).

In the aquatic environment the disintegration of PBMs is facilitated by wave action and grinding with sediment particles, whereas changes in chemical functionality are driven by UV exposure. Floating debris has a greater exposure to sunlight and the oxidative properties of the atmosphere, which act alongside the hydrolytic properties of water to cause the material to become brittle and fragment. In the deep ocean environment where sunlight and oxidative processes are missing, the rate of degradation is extremely low (Mao *et al.*, 2020). Degradation in these environments is considered minimal, due to the reduced

diversity and density of microbial communities. Therefore, PBMs do not readily biodegrade but rather disintegrate, breaking into smaller and smaller pieces.

Soil burial studies have been used as a method to evaluate the degradation of PBMs in the terrestrial environment. Soil type is an important factor affecting degradation; under laboratory conditions, polycaprolactone (PCL) degraded to a greater extent in clay soils than in sandy soils, owing to the great density of microbial communities associated with the clay soils (Yu *et al.*, 2022). However, when compared to solar exposed samples, buried samples degraded at a much slower rate. The combined effect of multiple degradation processes has also been studied. For example, Oehlmann *et al.* (2009) used polyhydroxybutyrate (PHB) films with a 0.1 – 0.12 mm thickness and found samples exposed to 9 h UV radiation showed ~52% weight loss after 28 days soil burial, compared to ~32% weight loss for samples without pre-UV exposure. These studies suggest that abiotic pre-treatment acts as a first step in weakening the polymer structure. This initiates the formation of oxygenated compounds and low molecular weight hydrocarbons, which are recognized by microbial communities and can be utilized as a food source (Yu *et al.* 2022). Biodegradation studies have tended to deal with the use of concentrated microbial cultures, with the aim of assessing a particular strain's ability to degrade a particular PBM. Actinomycetes are reported to be the main group of NRL degrading microbes, with *Bacillus* sp. Erren *et al.* (2009) studied strains of *Nocardia* and observed that they only slightly degraded strips of tread cut from truck tires, when used as a sole carbon source. However, degradation of the tire was enhanced by the addition of more easily accessible carbon sources in the form of latex glove and unvulcanized rubber materials, which were readily utilized by the bacteria (Erren *et al.* 2009). Biological processes are affected by the amount and type of microorganisms present, their sensitivity to associated environmental parameters and the adaptability of the microbiota. Martinho *et al.* (2022) isolated bacterial strains from forest soils, most belonging to different genera of the proteobacteria group and three *Rhodococcus* strains, and showed that commonly found bacteria were capable of adhering to and growing on the surface of oxidized LDPE film (Kapanen *et al.* 2008).

Table 2. Polymer degradation studies in various environmental matrices

Type of material	Length of study	Main findings
Aquatic		
HDPE, LDPE, PP (1.5 mm thick)	6 months	Samples were immersed in the seawater at a depth of 3 m. Weight loss was greatest in LDPE sheets (2.5%), followed by HDPE (0.75%) and then PP (0.5%).
PE glycols (PEGs) water soluble	135 days	Greater degradation was observed in freshwater media when compared to seawater media.
PHA	42 days	Species of bacteria belonging to the phylogenetic groups of <i>Cytophaga-Flavobacterium-Bacteroides</i> , <i>g-Proteobacteria</i> and <i>b-Proteobacteria</i> were found to utilize PHA in a eutrophic reservoir.
PUR	12 months	The degree of cross-linking effects PUR degradability in sea water. PUR with a heavily cross-linked network was very resistant to degradation.
Soil		
PE	10 years	Photooxidation processes produced carbonyl groups which were utilised by microorganisms to degrade the shorter segments of the PE chain.
LDPE	12 months	After 12 months it was impossible to separate film residues from soil. No change in diversity of ammonia-oxidizing bacteria was detected.
PP	12 months	Thermally pre-treated PP showed greater weight loss, greater loss of tensile strength and greater changes in crystallinity than non pre-treated PP.

*HDPE=high density polyethylene, LDPE=low density polyethylene, PP=polypropylene, PHA=polyhydroxyalkanoates, PUR= polyurethane

Polymer-based materials (PBMs) with a starch component are effectively hollowed out when exposed to microbial communities; this increases the surface to volume ratio allowing for higher oxygen and moisture permeability, enhancing both oxidative and hydrolytic processes (Yu *et al.* 2022). In theory, the released polymer fragments will have a greater surface area than the original polymer, allowing them to be further degraded by the micro-biota. However, in the case of PE, microorganisms have been found to utilize the starch component, but are unable to utilize the remaining PE fragments, which remain non-degradable. The starch is utilized by microorganisms, leaving behind a lace-like structure with reduced physical integrity. However, the molecular weight of the remaining material was not reduced sufficiently for permanent assimilation into the microbial biomass (Rutkowska *et al.* 2002). Therefore, the remaining polymer matrix was no more biodegradable than the untreated polymer. This causes the disintegration of the polymer matrix, which generates many smaller particles and produces a wider

distribution of polymer particles in the environment (Kurkcu *et al.* 2012).

Polymer characteristics play an important role in the degradation rate of PBMs. Those PBMs that contain ester linkages are reported to be readily biodegraded by the action of esterases (Arkatkar *et al.* 2009). The molecular composition of a PBM also affects the hydrophobicity of the polymer surface, which in turn affects how easily microorganisms can attach themselves. Complexity of a specific polymer structure and composition (co-polymers) can affect overall degradability by directly influencing the accessibility of enzymes (Orhan and Buyukgungor 2000). PBMs with short and regular repeating units that have high symmetries and strong inter-chain hydrogen bonding often limit accessibility and are less susceptible to enzyme attack. Under ambient conditions, photo-degradation is one of the primary means by which PBMs are damaged (Richardson *et al.* 2006). The main processes involved are chain scission and cross-linking reactions, when exposed

to ultra-violet (UV) radiation (290-400 nm) or visible radiation (400-700nm) (Al-Salem 2009). Most polymers tend to absorb high-energy radiation, which activates their electrons to higher reactivity and foments oxidation, cleavage, and other forms of degradation. The most damaging UV wavelength for a specific material depends on the bonds present; for PE this is 300 nm and for PP 370 nm (Singh and Sharma 2008). Thermal degradation is the molecular deterioration of a polymer as a result of over-heating, which causes bond scissions of the main polymer chain and results in a change in properties. This process affects the entire polymer and not just the polymer surface, and results in changes to molecular weight, loss of tensile strength, changes in crystallinity, reduced durability, embrittlement, changes in color and cracking (Arkatkar *et al.* 2009). Thermal degradation of polyolefins at temperatures of 673, 773, 873 and 973 K were found to form tar-containing paraffinic structures in PP and LDPE, while aromatic structures were produced by pyrolysis of polyethylene terephthalate (PET) (Zhou *et al.* 2020). Typically chemical degradation processes are oxidation and hydrolysis leading to a reduction in the molecular weight and degree of polymerization of the polymer (Lucas *et al.* 2008). Oxidation processes can be photo or thermally induced and are considered important, especially for non-hydrolyzable materials such as PE (Rutkowska *et al.* 2002). The introduction of O₂ into the polymer matrix leads to the formation of OH and CO functional groups, which aid subsequent breakdown by biotic processes. The presence of O₃ in the atmosphere, even in small concentrations, accelerates the ageing process of PBMs, because O₃ attacks covalent bonds to produce cross-linking reactions and/or chain scissions producing free radicals (Bernhard *et al.* 2008). The rate of hydrolysis is dependent on the presence of hydrolyzable covalent bonds in the polymer (Lucas *et al.* 2008). PBMs with these functionalities are able to absorb moisture which then promotes hydrolytic cleavage of the polymer chain. Hydrolytic degradation occurs when positively charged hydrogen ions in acidic or negatively charged hydrogen ions in alkaline media attack the ester linkage, thus breaking the polyester chain (Zhou *et al.* 2020). Biological processes involved in PBM disintegration

start outside the microbial cell, with the secretion of extracellular enzymes (Singh and Sharma 2008). These enzymes are too large to penetrate deep into the polymer, so act on the surface by cleaving the polymer chain via hydrolytic mechanisms. Biological processes are further enhanced by the formation of the utilizable functional groups in the polymer chain (Zhou *et al.* 2020).

Impacts of PBMs pollution on environment and human

Once the PBMs enter the environment they have the potential to mimic natural food sources (Table 3). Researchers identified 135 species of marine vertebrates and 8 species of invertebrates that are susceptible to entanglement; and 111 species of seabirds that are known to ingest PBMs. Researchers also reported PBM packing loops as a threat to sea lions in California and fur seals in Australia. Petry *et al.* (2009) identified PBMs in 77% of the stomachs of Cory's Shearwater, *Calonectris diomedea*, a pelagic seabird that winters in the waters off the state of Rio Grande do Sul in Southern Brazil.

Entanglement and ingestion of PBMs in the terrestrial environment is not as well documented in the literature as it is in the marine environment; however, livestock are known to consume PBMs. In a recent study, PBMs were identified as the dominant foreign item consumed by livestock (Andrady and Neal 2009). The ingestion by a variety of organisms of micro size PBM particles has been reported (Table 3). Thompson *et al.* (2004) exposed amphipods (detritivores), lugworms (deposit feeders), and barnacles (filter feeders) to microscopic plastic particles and found all three species ingested them within a few days. Barnes *et al.* (2009) found microscopic polystyrene fragments (2 µm in diameter) were ingested by the mussel *Mytilus edulis* under laboratory conditions; these particles were then translocated from the gut to the circulatory system. Researchers have suggested that ingesting PBM particles could present a potential physical hazard leading to the following effects: intestinal blockage in fish, hindering formation of fat deposits, blocking gastric enzyme secretion, feeding stimulus diminution, lowering steroid hormone levels, and delaying ovulation that may cause reproductive failure (Ryan *et al.* 2009).

Table 3. Potential impacts produced during the degradation of PBMs

PBMs	Behavioral effects	Morphological effects	Reproductive effects
Macro	Blockage of digestive tract and internal injury; diminish food consumption and loss of vigor; starvation	Intestinal blockage; hinders fat formation; blocks gastric enzyme secretion; lowers steroid hormone levels; diminishes feeding stimulus	Delayed ovulation
Meso			
Micro	Mobility effects; reduced vigor		
Nano			
Sub-lethal		Increase/ decrease in heart rate; induced malformation; neurotoxicity; pericardial edema	Inhibition of growth; delay in maturity
Lethal			

The Ingestion of PBMs could provide a novel route of expose for chemicals that adsorb to the PBM surface. Persistent organic pollutants (POPs) have been shown to biomagnify in food webs, mimic natural hormones to cause reproductive disorders, and possibly increase the risk of disease. If the PBMs are taken up by organisms, the polymer-associated chemicals would also be transported into the organisms, possibly leaching into tissues and leading to long-term toxicity issues. Since then, polymer particles have increasingly been investigated as a vector for hydrophobic contaminants to enter the food-web (Abdullah *et al.* 2023). Mato *et al.* (2001) found that PE and PP pellets (1–5 mm diameter) accumulated polychlorinated biphenyls (PCBs) at concentrations up to 106 times that of the surrounding environment, while Liang *et al.* (2021) found a positive correlation between ingested PBMs and PCB concentration in seabirds, indicating transfer of these contaminants to the organisms. It has recently been suggested that sorption behaviour of POPs to polymer surfaces is driven by polymer characteristics such as polymer type and density (Abdullah *et al.* 2023). In their study, authors found LDPE had higher diffusion coefficients than did HDPE, meaning shorter equilibrium times for low density polymers. The knowledge that chemical contaminants adsorb to PBMs creates the potential for novel uptake exposure routes, with the potential for indirect effects on PBM debris consumption.

It has been emphasized that PBMs may provide a substrate for fouling organisms to be transported long distances, thereby contributing to species dispersal (Lozano and Rillig 2020). Barnes *et al.* (2009) reported sightings of an exotic species of barnacle, *Elminius modestus*, on debris in the northern Pacific. However, Majer *et al.* (2012) do highlight temperature as a limiting factor with the geographical range of this species, as low water temperatures would prevent their full development.

The PBMs can impact human health at every stage of the plastic lifecycle through exposure to PBMs themselves and/ or associated chemicals. Over 170 chemicals that are used to produce the main feedstocks for PBMs have known human health impacts, including cancer, neurological, reproductive, and developmental toxicity and impairment of the immune system. These toxins exert documented impacts on skin, eyes, nervous, liver and brain (Lee *et al.* 2023). Production of PBMs releases carcinogenic substances into the environment eventually entering human bodies through ingestion and/or inhalation. Effects of exposure to these substances include impairment of the nervous system, chronic inflammation, cancer, leukemia, endocrine-disrupting, reproductive and developmental problems (Zhao *et al.* 2021, Lee *et al.* 2023).

Conclusions

Polymer-based materials (PBMs) can be released into the environment from a variety of sources. The principal introduction routes being general littering, dumping of

waste materials, migration from landfills and emission during refuse collection. Once in the environment, PBMs are primarily broken down by photodegradation processes, but due to the complex chemical makeup of PBMs, receiving environments are potentially exposed to a mixture of different sized polymer fragments, leached additives and subsequent degradation products. The majority of literature to date that has addressed the environmental contamination of PBMs has focused on the marine environment. This is because the oceans are identified as the major sink for macro PBMs, where they are known to present a hazard to wildlife via entanglement and ingestion. The published literature establishes the occurrence of microplastics in marine environment and beach sediments, but is inadequate as regards contamination of soils and freshwater sediments. There is currently a need to establish appropriate degradation test strategies consistent with realistic environmental conditions to evaluate the impacts of PBMs in soil and freshwater environments.

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