Microplastics Derived from Food Packaging Waste

Subjects: Polymer Science

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Plastics are commonly used for packaging in the food industry. The most popular thermoplastic materials that have found such applications are polyethylene (PE), polypropylene (PP), poly(ethylene terephthalate) (PET), and polystyrene (PS). Unfortunately, most plastic packaging is disposable. As a consequence, significant amounts of waste are generated, entering the environment, and undergoing degradation processes. They can occur under the influence of mechanical forces, temperature, light, chemical, and biological factors. These factors can present synergistic or antagonistic effects. As a result of their action, microplastics are formed, which can undergo further fragmentation and decomposition into small-molecule compounds. During the degradation process, various additives used at the plastics' processing stage can also be released. Both microplastics and additives can negatively affect human and animal health.

Keywords: polymer ; plastic ; degradation ; nanoplastic ; food safety ; human health ; microplastic

1. Food Packaging

Products made of plastics have gained popularity due to their low production costs, light weight, ease of use, and durability. It is estimated that about 39.6% of such materials are used for packaging [1]. The purpose of food packaging is protection, encapsulation, convenience, and communication with consumers. Packaging protects food from mechanical damage and microbiological and chemical contamination [2] and facilitates food storage, handling, and transportation [3].

Currently, there are several regulations on plastic products intended for food contact. Unfortunately, the topic of MPs is not explicitly addressed in them. They refer to polymers, plastics, and additives used at the processing stage. European Commission Regulation No. 10/2011 on plastic materials and articles intended to come into contact with food states that substances with a molecular weight of more than 1000 Da cannot be absorbed in the body, and possible health risks may be caused by unreacted monomers or said additives, which are transferred to food through migration from the material [4]. According to the legislation, the released substances must not adversely affect the organoleptic characteristics of food and exceed the permissible limits of global and specific migration. Global migration is understood as the mass of residues of all substances released from the product into food simulants. The global migration limit is equal to 10 mg per 1 dm^2. Specific migration, on the other hand, refers only to the specific substance released from the article into the model fluids under the test conditions. The specific migration limit from plastic products was set for selected elements, i.e., Ba (up to 1 mg/kg), Co (up to 0.05 mg/kg), Cu (up to 5 mg/kg), Fe (up to 48 mg/kg), Li (up to 0.6 mg/kg), Mn (up to 0.6 mg/kg), and Zn (up to 25 mg/kg).

In addition, European Commission Regulation No. 10/2011 states that the risk assessment for substances released from packaging should include the substance itself and the degradation products arising from the intended use [4]. This statement, therefore, eliminates secondary MPs arising from the degradation of plastic packaging waste in the environment from the area of concern.

The most popular packaging for food protection and storage are containers, bottles, films, pouches, and cups [1]. They are usually made of high-density polyethylene (HDPE), low-density polyethylene (LDPE), polypropylene (PP), polyesters (such as poly(ethylene terephthalate) (PET)), and polystyrene (PS) [3][5].

Each type of polymer is characterized by different properties, and thus, they find various applications. Polyethylene is mainly dedicated to film and bags. Water bottles are made of PET, and caps are usually made of PP [6]. The release of MPs from plastic bottles and cartons was investigated. Most of the particles in water from returnable bottles were identified as PET (84%) and PP (7%), while in water from beverage cartons, other MPs, such as polyethylene (PE). This can be explained by the fact that the cartons are coated with PE film on the inside. In both situations, the particles are smaller than 20 µm [6]. The last of the polymers described—PS, is most often applied in the foamed form. Until recently, PS was used for disposable food packaging with heat-insulating properties. However, according to Directive 2019/904 of

the European Parliament and the Council, the marketing of food and beverage containers made of expanded PS has been restricted since 2021 [7].

2. Plastics Degradation

2.1. Mechanical Degradation

Mechanical degradation refers to the breakdown of plastics due to external forces, collision, and abrasion of materials [8][9][10]. Microplastics can be introduced into food during its preparation. It has been estimated that 100–300 MPs/mm are formed on the cutting board when cuts are made during food preparation. On the other hand, in an aqueous environment, the freezing and thawing of plastics can also cause the mechanical degradation of polymers [11].

The effect of external forces depends on the mechanical properties of the materials [12]. Plastics with a low elongation value at break are more prone to fragmentation under external tensile forces. This leads to the tearing of polymer chains [13]. As a result of the mechanical degradation of primary and secondary MPs, smaller plastic particles (e.g., NPs) can be obtained [14].

2.2. Thermal Degradation

In addition to mechanical grinding, the temperature can also affect the course and efficiency of plastic degradation [15]. When enough heat is absorbed, long polymer chains can be broken, generating radicals [16]. These can react with oxygen and produce peroxides, which decompose to form free hydroxyl radicals and alkoxy radicals. The reaction can proceed spontaneously until the energy supply ceases or inert products are formed by the collision of two radicals. The temperature required for thermal degradation is related to the thermal properties of the plastics and the availability of oxygen [12]. Singh et al. concluded that the decomposition of PE occurs in one stage, between 230 and 510 °C [17]. Polypropylene has an onset degradation temperature of 286 °C [18], while PS has an onset degradation temperature of 370 °C [19]. However, the pyrolysis process of PET starts sharply and occurs at around 427–477 °C (around 90% of the process) [20].

2.3. Photodegradation

Photodegradation of plastics involves reactions initiated by solar radiation. As a result of the changes, plastics are gradually destroyed, with fragmentation into smaller particles and the formation of MPs [21]. As a result of UV radiation, new functional groups are formed, and the crystallinity, thermal and mechanical properties, and surface morphology of MPs change [22]. In the environment, during solar radiation, plastic waste is also affected by atmospheric oxygen, so the process is often referred to as oxidative photodegradation. Thermal oxidation of plastics occurs in conjunction with photodegradation, especially on beaches or sidewalks that are exposed directly to sunlight [23][24]. Polymers containing aromatic rings in their structure (PS and PET) were found to be more susceptible to oxidation compared to polymers formed by aliphatic chains (PE and PP) [25].

Currently, plastics in which photodegradation is an intended feature are gaining popularity. These are photodegradable materials containing sensitizers that degrade when exposed to UV light in the presence of oxygen. Polyolefins, intended for the manufacture of disposable packaging, are the largest contributor.

2.4. Chemical Degradation

The most important chemical factors affecting the degradation of plastics in an aqueous environment are the pH value and salinity of the water. High concentrations of H^+ or OH^- in the aqueous environment can catalyze the degradation of plastics that are susceptible to hydrolysis, such as polyamide (PA) [26]. These two factors can also affect the surface of MPs, their properties in aqueous environments, and their affinity for other contaminants. Polyethylene and PS in the form of MPs were studied by Liu et al. [27]. In the mentioned work, it was found that the presence of NaCl and $CaCl_2$ increases the sorption of both diethyl phthalate (DEP) and dibutyl phthalate (DBP) [27].

2.5. Biological Degradation

The biological degradation of plastics is determined by organisms (e.g., bacteria, fungi, and insects) that can destroy materials physically through biting, chewing [28], or biochemical processes [12][29]. The ingested plastics can be retained in the stomach, where fragmentation will occur, subsequently releasing particles [30]. This process can be accelerated by abiotic degradation, resulting in the formation of low molecular weight degradation products and the formation of cracks and pores on the surface of the plastic [31]. The biological degradation of PP with *Bacillus* sp. strain 27 and *Rhodococcus*

sp. strain 36 made it possible to conclude that this is a process dependent on the type of microorganisms. In the first case, the weight loss was found to be 4.0%, and in the second, 6.4% [32]. In terms of degradation potential, the type of polymer is also important. It was found that bacteria degraded PP more easily than PE, while fungi degraded PE more easily than PP [33]. However, among synthetic polymers, aliphatic polyesters are the most susceptible to microbial degradation. It is widely believed that the ability of microorganisms to degrade synthetic polyesters is due to their chemical similarity to natural polyhydroxybutyrate (PHB), which is the backup material of many bacterial strains. Depending on the absence or presence of ester and amide groups, plastics can be attacked by various extracellular hydrolases. It is assumed that polyesters are degraded by enzymes, such as proteases, esterases, lipases, and cutinases.

The degradation of polymers that do not contain ester and amide groups by extracellular enzymes is a very complicated process. These polymers can be oxidized by O_2 with hydrolase catalysis, resulting in low-molecular-weight degradation products. Laccase enzyme has played a major role in PE degradation by *Rhodococcus ruber*. The activity of laccase is improved by the presence of copper. Hydroquinone peroxidase, on the other hand, was found to be responsible for PS degradation by *Azotobacter beijerinckii* HM121 [34]. The biological degradation of plastics can also be caused by algal enzymes [35].

Degradation with the participation of extracellular enzymes breaks polymer chains with the yield of shorter-chain polymers as well as oligomers, dimers, and single molecules [36]. Ultimately, plastics can be mineralized to CO_2 and H_2O under aerobic conditions and to CH_4, CO_2, organic acids, H_2O, and NH_4. Degradation of plastics under anaerobic conditions is energetically disadvantageous compared to degradation under aerobic conditions, and complete mineralization can take much longer [37].

3. Packaging Waste Dump

Packaging waste is ubiquitous. It can be transported from land to water and from water to land [38][39][40][41]. Microplastics formed in land and water can also move between different ecosystems [42][43]. These environments are commonly thought of as independent, but in fact, they are closely interconnected [44].

3.1. Terrestrial Environment

Microplastic in the terrestrial environment is formed by the fragmentation of larger plastics into smaller pieces due to exposure to UV radiation, wind action, agricultural activities, oxidation processes, and chemical and biological interactions [43][45][46]. The combined effects of the aforementioned factors can accelerate the aging of MPs, manifested by changes in color, crystallinity, chemical composition, and surface properties [47]. Microplastic in the terrestrial environment affects soil quality and biota [48][49]. For example, it has been found that the presence of MPs can significantly reduce the volume of phosphates available in the soil [50].

3.1.1. Sources and Transport of Microplastics in the Terrestrial Environment

Significant amounts of MPs are generated in landfills, peri-road areas, and agricultural areas [51][52]. Soil contamination can come from many sources, including compost [53], mulch film [54], greenhouse materials, irrigation tools [55], plant protection products, fertilizers [56], municipal solid waste, sewage treatment plants [57], used tires [9][58], and precipitation [59]. The presence of plastic particles in soils from China [60][61][62], Iran [63], Brazil [64], and Spain [65] was confirmed. It was found that the distribution of MPs in soils showed differences not only regionally but also in-depth [66]. The movement of MPs with groundwater can cause pollution of freshwater ecosystems, also contributing to marine pollution [67].

Plastic particles that reach the soil surface are transported to deeper layers of the soil through cultivation, infiltration, and animal activity [45][55][68]. Polyethylene beads can be transported from the soil surface down the soil profile by *Lumbricus terrestris* [68].

Polyethylene is the most commonly used polymer to study the degradation of plastics in soil. The degradation of this material was found to be increased by elevated pH and humidity. Polyethylene bags buried in soil for 2 years showed an increase in surface roughness. A nearly 5% decrease in weight was found for commercial carrier bags made of PE stored in mangrove soil over 8 weeks. This was due to the action of heterotrophic bacteria capable of producing hydrolytic enzymes [69]. In an experiment on the degradation of plastics buried in soil for 32 years, significant bleaching of LDPE film was observed, but no evidence of PS degradation was observed [70]. Thus, further studies are needed to determine the effects of individual polymers on soil properties and functions. These analyses should consider a wide range of particle sizes and shapes, as well as different types of substrates.

3.1.2. Impact of Microplastic on the Terrestrial Environment

MPs-induced changes affect soil function and the soil microbial community. The presence of the described particles can directly or indirectly affect chemical processes in the soil environment and the circulation of water, nutrients, and geochemical elements. The size distribution of soil aggregates changed when MPs were present, suggesting potential changes in soil stability. It was found that the PE film increased the evaporation rate of water by creating channels for moving water [52]. Biogenic transport of MPs in the soil can lead to groundwater contamination [71][72], results in uptake by plants [73], and causes changes in root biomass. Microplastics have the potential to affect plant growth and can accumulate in plants [45]. A reduction in seed germination and shoot length of *Lolium perenne* was found after exposure to poly(lactic acid) PLA. In contrast, the biomass of *Aporrectodea rosea* was significantly reduced as a result of HDPE exposure compared to control samples [54].

In addition, MP present in the soil affects invertebrates living in this environment and can penetrate the intestinal walls of soil nematodes, causing oxidative stress and affecting gene expression [74]. It was found that the moist soil environment had a pronounced effect on the release of plasticizers [75].

3.2. Aquatic Environment

Aquatic ecosystems are very diverse chemically, physically, and biologically. Microplastic research in aquatic environments includes analysis of pollution from rivers and lakes [76][77][78] to seas and oceans [79][80][81][82] and the Arctic ice shelf [83]. The freshwater environment can differ from the marine environment in several aspects, including the intensity of sunlight, the physicochemical properties of the water, and its biological properties. The results of plastic degradation indicate different rates of this process depending on specific environmental conditions. Microplastic accumulates both on the water surface and in sediments [84][85].

3.2.1. Sources and Transport of Microplastics in the Aquatic Environment

Plastic packaging waste can be transported downwind and downstream. It is believed that most MPs in the aquatic environment are formed by weathering of plastic waste [86]. This process consists of photodegradation and mechanical, chemical, and biological degradation. Plastics on the surface or in the photic zone of water can undergo photodegradation under UV radiation. It is responsible for the initial degradation of plastics floating on the surface of seawater. Mechanical degradation is caused by waves. Chemical degradation in the aquatic environment mainly involves the breakdown of chemical individuals under the influence of various compounds [87]. Plastics in water can be colonized by microorganisms that form a biofilm and break down organic matter. Biofilm formation will limit light transmission [44]. Plastics can be very persistent on the seafloor and in sediments due to UV protection and low oxygen content [50]. Biological degradation by microorganisms in the biofilm is the main cause of plastic degradation in seawater in the aphotic zone. The rate of plastic degradation can be reduced by low water temperatures [86]. As a result of degradation in the aquatic environment, weight loss, changes in the appearance and structure of plastics, and deterioration of mechanical properties will be observed [88][89][90].

The degradation of plastics in marine, river, and lake ecosystems was investigated. Poly(ethylene terephthalate) was found to be the dominant polymer in the coastal waters of Hainan Island in China (South China Sea), occurring as white-black linear or fragmented particles [91]. It was observed that PET bottles collected from the seafloor were found to remain robust for about 15 years, after which a significant decrease in native functional groups was observed [92]. However, in the case of LDPE, rapid initial decomposition within the first week, followed by little further loss of tensile strength over 4 months, was established [93]. The total PE, PP, and PS mass, with a particle size of 32–651 µm suspended in the upper 200 m of the Atlantic Ocean, was determined to be 11.6–21.1 million tons [83]. In addition, PE and PS were shown to be present in the Wei River Basin, which is located in northwest China [71]. The occurrence of PE and PP-sized MPs was confirmed in the Thames River [78]. Particles of this type were also present in the waters of another European river, the Rhine [77], and in the surface waters of the Laurentian Great Lakes in the United States [76].

3.2.2. Impact of Microplastic on the Aquatic Environment

Microplastics have a very long degradation cycle. As a result, they can enter natural ecosystems and accumulate. It is estimated that the maximum degradation time of PE in the deep sea is about 292 years [94]. The phenomenon of MPs accumulation has been proven in more than 200 species of freshwater fish [95][96] and is linked to adverse effects on fish digestion [97][98], reproduction, and development. Microplastics can cause histopathological damage to the fish liver [99]. In addition, the adverse effects of PS and PP on *Danio rerio* fish embryos were evaluated. A reduction in body length and heart rate was observed [100].

4. Biodegradable Plastics

Biodegradable plastics constitute interesting types of materials. This group includes natural polymers, polymers obtained by modification of natural polymers, and polymers produced by various chemical synthesis methods and biotechnological processes. The degradation time depends on the conditions and the types of polymers. They can, in a relatively short time, be degraded to H_2O, CO_2, and CH_4 [101]. The rate of degradation of biodegradable polymers depends on the environment in which they are stored [101]. For example, it was found that poly(butylene succinate-co-butylene adipate) (PBSA) degrades at 37 °C, with the generation of CO_2 for 40 days [102]. The complete degradation of PLA can be achieved at composting temperatures of 60 °C or higher [103][104], but it does not degrade in seawater [96]. Itävaara et al. found that 90% mineralization degree of PLA was reached at 60 °C within 120 days [105].

However, laboratory-defined degradation conditions are virtually unattainable under natural ones. The degradation of such materials will take longer in the environment compared to the degradation time in the laboratory. Some biodegradable plastics will fragment and slowly accumulate in the environment in the form of MPs and NPs [106].

Microplastic derived from biodegradable plastics may show similar or even higher harmfulness to organisms compared to conventional materials. For example, PLA in the form of MP has higher toxicity to *Chlorella vulgaris* compared to PA and PE [107]. Moreover, it shows a higher adsorption capacity of tetracycline (TC) and ciprofloxacin (CIP) compared to poly(vinyl chloride) PVC [108].

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