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MICROPARTICLE POLLUTION IN THE LIGHT OF POLYMER MATERIALS RECYCLING

Polymer plastics have established a major sector in the global economy, fulfilling the needs of various industries, including packaging, construction, automotive, electronics, and healthcare. Due to a unique set of physical properties, those materials are durable, versatile, and enable large-scale low--cost production. However, as environmental awareness rises, the raw materials sector is encouraged to increase the contribution of sustainable products with a lower environmental impact. It is being conducted through various activities such as recycling, introducing biodegradable alternatives, and/or diversifying sources of raw materials. In this short review, a holistic approach to analyse current knowledge regarding conventional plastic microparticle emissions into the environment is presented, with a special regard to the correlation with the existing plastic waste recycling technologies. The review takes into account current legal provisions regarding increasing the use of recycled plastics and highlights the risks resulting from the increased release of microparticles after reprocessing, washing, use, etc. The presented research results encourage consideration of the possible consequences of polymer reuse.

1. INTRODUCTION

Plastics have been used in human life due to their unique set of properties since the early 50's of the 20th century. Composing different polymers with various additives provides an excellent platform for designing materials with application-oriented properties. Among many advantages, elements made of plastics are light (compared to glass or metal-based products), resistant to external factors (like UV radiation, oxidation/weathering), and extremely easy (relating to process machinery/equipment costs and conditions) to produce in large quantities [1, 2]. Unfortunately, as time went by,

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people discovered that due to those properties, a large amount of plastic waste has been created and penetrated the natural environment. However, plastic materials are too high extend resistant to external factors, but when turned into waste, they undergo a process that induces noticeable changes in physical properties. Different degradation mechanisms (hydrolysis, photodegradation, thermooxidative degradation) [3] act with considerable kinetics when plastic waste is exposed to typical environmental factors, such as UV radiation, moisture, and elevated temperature. However, their degradation time is expected to be measured in decades or centuries, which, in comparison to the yearly global production quota, is much too slow in order to effectively reduce the overall waste amount. That is why plastics are considered to be non-degradable, but can gradually fragment into smaller particles in conventional environmental conditions, forming synthetic polymer microparticles (SPM) more frequently referred to as "microplastics" (MP) [3].

Table 1

Exemplary locations of microplastics in different environments

Country	Location	Mean number of items	Size and shape	Types of main polymers	Ref.
India	beach	183.32/kg dry sediment	60% fragment, 17% film, 12% fibre, 11% sphere	PE, PP, PS	[8]
El Salvador	beach	48–300/m^2	85% fibre, 8% fragment, 5% film, 5% foam	PE, PP, PS, PET, latex	[9]
Poland	soil	112±62\50g of soil	37–91% fragment, 2–18% films, 3–19% fibre, 0–42% sphere	PE, PP, PVC, PU	[10]
Republic of Korea	soil	700±75/kg soil	66% fragment, 20% film, 15% fibre	SBR, PP, PE, EPS, PS, PET	[11]
Poland	soil	1540±912, 383±188, 933±682/kg soil	60% fibre, 40% mixture of fragments and pellets	PET, HDPE, LDPE	[12]

LDPE – low-density polyethylene, PP – polypropylene, PS – polystyrene, PET – poly(ethyl terephthalate), PVC – poly(vinyl chloride), PA – polyamide, PU – polyurethane, SBR – styrene-butadiene rubber, EPS – expanded polystyrene.

The term was introduced in 2004 [4], but well before that, there had been reports of man-made plastic particles swallowed by albatrosses in 1965 [5]. In the following years, the problem became more and more evident, although it was mainly related to the fishing industry and marine litter [6]. At the time, it was already obvious that non-biodegradable, slowly decaying plastic particles can be related to the contamination of living organisms in the marine environment, although it was perceived more in terms of physical irritation/entanglement than material toxicity.

Based on the source of where the microplastics originate, they can be divided into two distinctive groups, primary and secondary microplastics. Primary, i.e., industrially produced small spheres or pellets that can be found in toothpaste, all sorts of cosmetics with increased abrasive properties, body cleaners, and others. The secondary originates from the previously mentioned degradation products of the plastic waste [7]. The presence of MPs in the environment can be described as omnipresent. They have been identified in many different environments to varying degrees, as presented in Table 1.

The presence of MP in soil can be related to several reasons. Littering, car tyres wear and tear, fallout from the atmospheric emissions, along with other inputs such as contamination by sewage sludge and wastewater [13]. Marine contamination and shoreline accumulation are also evident. MPs are being washed away and end up on the beaches, tending to amass along the vegetation lines. Most of those particles are composed of polymers with densities lower than that of water, but in a given time, they can be populated with microorganisms, therefore increasing the density and providing sedimentation [14].

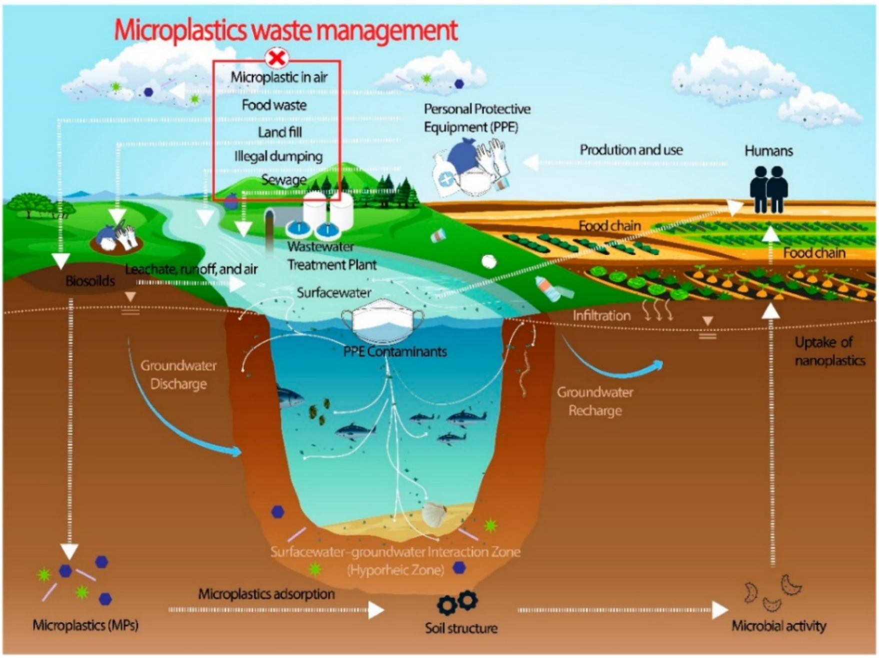

Fig. 1. Sources and routes of propagation of MP in the environment (adopted from [15])

In recent years, during the COVID-19 outbreak, there was a sudden buildup in production and consumption of plastic-based materials due to several factors. Quick and cost-effective production, excellent barrier properties, easy-to-sterilize products (especially for the personal protection equipment (PPE)), etc. The increase in plastic waste was also fuelled by lockdowns in which the takeout and delivery services were often the only available option for acquiring food and necessary goods [16]. Summary of the MP sources and routes of propagation regarding the PPE waste are presented in Fig. 1.

2. TYPES OF POLYMERS IN PACKAGING APPLICATIONS

Global production of plastics has constantly been increasing in recent years and in 2023 had reached 413.8 million tonnes according to Plastics Europe [17]. Since plastics provide an inexpensive alternative to other conventional materials, with a decent strength/density ratio and excellent barrier properties, they have been found suitable for a variety of applications. Since the 50's of the 20th century, they have been replacing more conventional materials, such as wood, glass, metals, and others. Among different applications, the packaging industry is one of the most prominent, as plastics, due to their previously mentioned properties, add the ability to implement various designs and a large-scale production with relative ease [18]. Packaging, especially made of plastic, can be characterized by a very short life span, as in most cases it becomes waste almost immediately after the protected goods have been unwrapped. The same can be said about all kinds of single-use products that by definition are to be replaced and themselves discarded [19]. Among many different polymers used in plastics nowadays, in packaging, there are several large groups of materials that can be distinguished by their main component. Polyethylene – low and high density (LDPE and HDPE respectively), polypropylene (PP), polystyrene (PS), poly(ethyl terephthalate) (PET) and poly(vinyl chloride) (PVC), and polyamide (PA). Due to their large production quota and large usage volume, they are likely to end up in the environment and ultimately become waste [20]. Although all of those plastics are considered non-degradable, they will undergo a fragmentation process along with the degradation mechanisms mentioned earlier. Some are more prone to the hydrolytic degradation, some to photodegradation, but usually it is the combination of several different factors that provides consequent breakdown of the plastic-made elements [21].

One other component, or rather a huge group of substances present in plastics, are the additives. Such as plasticizers that increase their softness and elasticity by weakening forces between the polymer chains, ensuring better processability. Flame retardants that provide better heat resistance, which is a requirement for most polymers, as only a few of them are self-extinguishing, and for selected applications (e.g., insulation, construction, electronic components), such a property is mandatory to prevent flame propagation. Stabilizers that decrease the degradation ratio and ensure the overall longevity of plastics. Antioxidants that play a similar role, but especially prevent oxidative degradation. And many others (e.g., colorants, antistatic agents, blowing agents, impact modifiers, etc.). It is estimated that plastics can contain ca. 10 000 different chemicals [22]. Said chemicals can migrate to the plastic surface and into the surrounding environment. That, combined with the defragmentation, which leads to a larger outer surface and furthermore increases the leaching process, can be especially concerning when investigating the possible health risks from the MP. The additives migration process is, in most cases, associated with a diffusion that depends on several factors, such as plastic thickness, its porosity, molecular weight, the characteristics of the surrounding environments, temperature, moisture content, etc. Therefore, apart from the material's properties, the weathering also has a strong influence on the leaching process [23].

Polymers, due to their diverse structure and sensitivity to various factors, including oxygen, temperature, UV radiation, water, and other substances, undergo degradation processes at different rates. Polymer materials possess diverse properties and, therefore, are used for different packaging purposes (Table 2). Single-use products made of polymeric plastics, such as beverage bottles, straws, cutlery, coffee cups, and bags, have been identified as a significant source of environmental plastic pollution [24]. PE is widely used as a packaging material in the form of films, bags, bottles, cups, etc. It is the most common type of plastic, and upon fragmentation, can be found in forms of both microbeads and fragments. PP is used in bottle caps, straws, and food containers. It is also commonly found in MP pollution due to its widespread use and durability. PS is found in packaging products, including foam products such as disposable cups, food containers, and insulation materials. Polystyrene microplastics often appear as foam fragments and particles. PET is common in beverage bottles, clothing fibers, and food packaging. PET MP particles can appear as fibers from synthetic textiles or fragments from bottles.

Table 2

Commonly used polymers for various types of food containers [25]

Polymer type	Packaging application
Polypropylene (PP)	food packaging, sweet and snack wrappers, hinged caps
High-density polyethylene (HDPE)	milk bottles
Low-density polyethylene (LDPE)	food packaging film, food containers, and trays
Polystyrene (PS)	dairy and fishery food packaging
Polyethylene terephthalate (PET)	water bottles, soft drink cartons, and juice containers

Although many factors cause degradation of plastic materials, aging, temperature, and external mechanical forces are mainly responsible for abrasion and flaking of packaging materials [26].

3. POSSIBLE SIDE EFFECTS OF PLASTIC PACKAGING REUSE

In the interests of the environment, single-use bottles are being increasingly replaced with those that we can reuse many times without increasing the amount of plastic waste. However, there are reports indicating that the number of microplastics in reusable bottled water is significantly higher than in single-use bottled water, because the age of these reusable bottles affects the release of microplastics into the water [27, 28]. Schymanski et al. [28] noted that the number of microplastics in water from reusable or returnable bottles was 8 times higher than in water from single-use plastic bottles and about 10 times higher in comparison with beverage cartons. Oßmann et al. [27] examined both new and older reusable bottles and found approximately 2689±4371 microplastics/dm^3 in new reusable bottled water, which was similar to the number of microplastics found in single-use bottled water. In contrast, older reusable bottled water contained 8339±7043 microplastics/dm^3, approximately 3 times more than new bottles. The study indicates a significant effect of bottle age on the release of MP into water.

Other researchers have investigated the possible correlations between the methods of manufacturing packaging materials (processing technology) and the impact of conditions during storage or transport of food in these packages, as well as their washing, and the potential release of microparticles [29–33]. Du et al. [33] analyzed commonly used takeaway containers made of different plastics. Among the tested materials, PS packaging released the highest amount of microplastics, exceeding those from PE, PP, and PET containers, which may indicate different material properties caused by different production processes (e.g., injecting gas into PS masterbatch melts, pressurized injection of the melted PE, PET masterbatch into mold, lamination in the case of PE). The resulting structure (more or less compact) and type of surface (rough, smooth) may affect the amount of microplastics released from food packaging. Experiments on rinsing the inner surface of containers with water and pouring hot water on them simulated the conditions during storage of hot food in takeaway boxes (temperature, friction). Presented significant differences in the recorded microplastics content from different materials, which indicates that it is important to select a specific material for safe heating of food, e.g., in a microwave oven, and its storage or transport. Visible surface changes were noted in PP and PE containers compared to PS after hot water treatment. Although surface changes in PP and PE containers did not increase the abundance of microplastics in their corresponding containers, they may result in the release of some additives.

The amount of microparticle release from different take-away packaging, such as boxes, bags, bowls, and cups made of polymers such as PS, HDPE, and PP were also investigated by Hee et al. [32]. It was found that repeated washing and treatment of the packaging with hot and cold water caused flaking of the packaging surface, thus producing microplastics for every type of packaging investigated. The highest amount of microparticle release recorded was from the PS box after the hot water treatment, which could be due to a less compact structure providing an increased surface area that allowed greater contact with the liquid. For the HDPE bag, the results varied depending on the water temperature; for hot water, a release of MP was approximately twice as high as for cold water treatment. It was shown that abrasion caused by repeated washing of melamine bowls leads to the release of MPs through flaking due to degradation processes, as washing makes the surface of the bowls more glassy and brittle.

Hernandez et al. [31] conducted studies on the release of MPs from plastic tea bags and found that elevated tea brewing temperature can make polymeric materials brittle, which increases the release of MPs into the beverage. Other studies conducted on infant feeding bottles and paper cups also showed the effect of water or beverage temperature on the release of microplastics from their inner layer into the respective beverages [29, 30]. Li et al. [44] estimated about 16 200 000 MP items/dm^3 released from PP infant feeding bottles after sterilization at 95 °C and incubation with deionized water at 70 °C. The authors concluded that the release of MP from PP bottles is mainly promoted by high temperatures. Additionally, the migration of chemicals such as additives or oligomers from bottles may also increase at higher temperatures.

Another issue is the effect of mechanical stress on polymeric materials and the impact of these actions on the generation of microparticles. Sobhani et al. [34] found that microplastics can be generated by simple activities in our daily lives, such as cutting with scissors, tearing with hands, cutting with knives, or by manual twisting to open plastic containers/bags/tapes/caps. These processes can generate about 0.46–250 MP items/cm. The amount depends on conditions such as stiffness, thickness, anisotropy, and density of plastic materials. Winkler et al. [35] performed repeated capping and uncapping experiments on PET bottles using HDPE caps and investigated the effect of abrasion of the polymer surface on the amount of microparticles released. Such opening and closing procedures repeated 100 times showed a strong impact on the HDPE material with significant signs of mechanical stress, such as abrasions and deep grooves. It was found that reuse of PET bottles and especially repeated cap-bottle friction can release up to millions of microplastic items on the inner surface of the cap and up to 148±253 MP items/dm^3 in water. Giese et al. [36] documented significant microplastic emissions exceeding 130 items/dm^3 after unsealing of screw-capped plastic bottles. The higher incidence of PS items (cap material) than PET items (bottle material) was correlated with the known higher hardness of PET compared to PS, which means that the hardness of the material influences the release of microplastics under mechanical stress [37]. Other researchers in their studies [38] considered the correlation between the place of production of packaged beverages and the abundance of microplastics, indicating that mechanical shocks occurring during transport from production to sales locations, such as shaking and vibration, can cause beverages to rub against the inside of containers, potentially leading to the release of microplastics. In line with this observation, studies conducted in Australia confirm that imported bottled water had an average concentration of microplastics four times higher than locally sourced bottled water [39].

4. LEGAL REGULATIONS

Although the term “microplastics” has already been well established in colloquial speech, newspaper headline clips, TV broadcasts, and even professional reports, one can struggle to precisely define its meaning. In fact, it may be even more confusing when the discussion comes to biopolymers – materials originating from renewable resources. So what form of matter stands precisely for the term “microplastic”?

A precise, most up-to-date definition can be found in Commission Regulation (EU) 2023/2055 dated 25th September 2023, introducing changes in Annex XVIIth REACH directive [40]. Synthetic polymer microparticles (SPM) are solid materials meeting both of the following conditions: are contained in particles and constitute at least 1% of those particles, or build a continuous surface coating on particles, and follow the dimensions requirement. The latter one in particular means that all of them are less than 5 mm, or 15 mm long for particles with a length to diameter ratio of more than three.

Abovementioned Regulation contains several exclusions, not taking into consideration polymers that are the result of a natural polymerization process and are not chemically modified, degradable polymers (following Appendix 15 “Rules on proving degradability” in quoted master document) or medical/vet medicinal products and food additives.

Although the REACH Regulation seals the market, preventing an excessive emission of SPM into the environment, it has to be clearly stated that “microplastic ban” (how the Regulation is named informally) is not a huge constrain for R&D entities or industrial plants working with composites (containing SPM permanently enclosed in the structure), fertilizers/agriculture chemicals or for own use. The Regulation mostly affects the cosmetic industry, preventing SPM from being introduced into the market, subsequently excluding their specific forms following an agreed timeline (Table 3).

Table 3

SPM in cosmetics phase-out schedule [54]

Product type	Phase out deadline
Exfoliating microbeads, plastic loose glitter	October 17, 2023
Rinse-off products	October 17, 2027
Waxes, polishes	October 17, 2028
SPM for encapsulation of fragrances, leave-on products	October 17, 2029
Lip products or make-up products	October 17, 2035

Obviously, the discussed Regulation does not apply to lowering the SPM emission from secondary plastics, but it was never intended to. Processes such as abrasion (footwear products, fabrics, bicycle tires, and soft covers) or textile washing produce a significant amount of contaminants. Although the exact numbers are hard to determine, scientific data vary from several thousand to millions of particles per milliliter, depending on the type of fabric [41, 42], detergent type, and washing conditions, it can be assumed that each washing cycle releases 0.1–0.3 g SPM per kg of fabric [42]. However, the numbers describing discharged SPM to wastewater are impressive, but, on the other hand, sewage plants are equipped with treatment technologies separating SPM from the liquid phase and depositing it into sludge [43], making drinking water safe for consumption purposes. Dewatered and dried sewage sludge has been perceived as a valuable source of organic particles due to its significantly high nutrient content [44, 45]. Recently, its reuse on the field has raised a concern, according to the possible direct introduction of SPM directly into the environment from the concentrate [46]. It seems this issue needs to be addressed immediately in the discussion on the pros and cons of the circular economy.

As possible hazards in using sludge as a crop fertilizer have been identified and described very clearly [47, 48]. It seems that the side effects of technologies applied to maintain the water supply network are still under investigation in terms of SPM. Sanitation is a process in which ozone is used as an active medium to clean up the piping system. Long-term exposure to this highly reactive agent also hurts the piping surface, due to changes induced in the surface area, bulk of the material, and thus mechanical properties [23, 49], finally leading to SPM discharge. Zhang et al. [50] investigated four types of plastic frequently used in the piping industry (PE, PP, PVC) and validated the number of released SPM after 4, 10, and 20 h ozone exposure. Results showed almost twice as much SPM concentration after 20 h treatment in HDPE and PP samples. Authors also identified additional features supporting this process, like wall porosity, shear forces acting during medium flow, and residual stress after pipe extrusion. Those observations are critical when taking into consideration the fact that ozone disinfection processes are carried out in residential areas, in the piping network system, long after water treatment plants. SPM discharged into piping systems is capable of directly migrating into the last chain of the whole network – the point of use. This secondary generated impurities are a significant concern when more and more frequently tap water direct usage is on the public discussion.

The problem with synthetic polymer microparticles has also been detected in bottled water. Several works [28, 27] showed that SPM concentration in bottles made of recycled PET is greater compared to those fabricated from pure original PET. Also worth noting is the fact that the concentration of SPM in returnable plastic bottles was ca. 10 times greater compared to single-use plastic bottles [51, 52]. On the other hand, following the principle rule of circular economy: the more recycled material in use, the better. Increasing the volume of recycled plastic in single-use plastic packaging has been widely described in official documents like the last one: Regulation (EU) 2025/40 of the European Parliament and of the Council of 19 December 2024 (published on 22 January 2025, entered into force on 11 February 2025). This Regulation, abbreviated to PPWR (*Packaging and Packaging Waste Regulation*), will officially apply from 12th August 2026. The question that needs to be stated at this point is how this Regulation complies with scientific data, when it comes to recycled polymer plastics and SPM discharge? The undeniable fact is that, according to Articles 7 and 8 of the aforementioned regulation, from 2030, the minimum amount of recycled material in plastic packaging will be 30% rising to at least 50% in 2040. Do not we then welcome more problems related to SPM discharge, which, moreover, would be directly distributed into the beverages? Obviously, Regulation also excludes specific types of packaging like medical applications (0% of recycled material) or packaging for sensitive materials, where the amount of recycled material is no more than 10%. But again, looking at the whole, the packaging market covers around 40% of total world plastics production, making it the biggest branch for plastic processors [17] before building and construction (18%) and automotive (8%).

5. CONCLUSIONS

As described above, synthetic polymer microparticle pollution (called in short: microplastic) is an emerging issue to be solved for modern societies. According to its specific properties, SPM is an abundant material easily distributed by all means of transportation, with air and water environments being the most popular ones. Legal regulations, introduced in developed countries, solved a problem with primary SPM, reducing their application and (over)usage. The main issue that is not trivial is pollution by secondary SPM. Not only mention particles being released into the environment due to (photo/oxidative) degradation, abrasion of plastic elements. Also, those threats arise from limitations arise from the perception of waste management processes. To name them – intensive SPM discharge by the introduction of waste products again into the life cycle loop. Sewer sludge used as a fertilizer and an increasing amount of recycled material in polymer packaging are the most glaring examples. Comparing scientific data materials with law regulations officially entered into force, it is clearly seen that some aspects of the circular economy policy line have to be renegotiated.

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