

Review



Sustainable Management of Microplastic Pollutions from PET Bottles: Overview and Mitigation Strategies

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Abstract: The widespread use of polyethylene terephthalate (PET) in bottled water packaging remains significant and is expected to increase further in the coming years. This trend raises concerns due to the generation of large amounts of waste. The degradation of PET leads to the release of low-molecular-weight substances and microplastic particles, which contaminate food products and the environment. This work highlights the significance of microplastic pollution, summarizes the mechanisms of PET degradation, and discusses methods for microplastic detection. A key section of the paper explores potential degradation management strategies, focusing on their applications and existing limitations. The study underscores the need for coordinated action among the scientific community, industry, and policymakers to mitigate this pressing environmental challenge.

Keywords: waste management; sustainable development; polyethylene terephthalate; PET bottle; microplastic

1. Introduction

Managing food safety and the quality of food products is a key challenge in the modern world. Food quality is defined as the degree to which consumer expectations are met in terms of sensation, experience, and the impact of food ingredients on human health. Providing access to healthy and safe food is a priority that requires a comprehensive approach that includes strict control and supervision of the entire food chain, i.e., from cultivation and breeding through processing to packaging and distribution. One of the key roles in ensuring safe food is played by packaging, the function of which is, among others, protection against the penetration of various types of contaminants into the product. Modern research shows that plastic packaging can be a significant source of food contamination, especially micro- and nanoplastic particles, so-called MPs, which negatively affect the quality of food products and thus increase the risk to consumer health. These contaminants can be created during the thermal forming process of the packaging, insufficient binding of substances contained in the packaging material, or interactions between packaging components and food that are in direct contact [1]. Under the influence of various environmental conditions, chemicals contained in plastic packaging can migrate into the product and contribute to changing the composition or organoleptic characteristics of packaged food. In the case of bottled drinking waters, where the packaging material is in direct contact with the product, this is a serious quality problem [2]. Moreover, plastic packaging, as a result of degradation, releases MP particles into the environment, which pose a significant threat to human health.



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Copyright: © 2025 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https://creativecommons.org/ licenses/by/4.0/). Exposure to microplastics can be considered a risk factor for cardiovascular disease [3]; MPs exhibit proinflammatory activity [4,5], may cause endocrine disruption and infertility problems [6], and lead to other adverse effects.

The presence of microplastics may hinder efforts towards sustainability, particularly in waste management and renewable energy generation. For example, MPs can negatively affect the anaerobic digestion of organic matter during kitchen waste treatment [7,8]. This example demonstrates the far-reaching consequences of MPs, not only from the perspective of human health but also in the context of developing technologies essential for sustainable development.

The growing popularity of convenience foods, which are primarily sold in disposable containers, has contributed to the steady increase in global packaging production year over year. In 2024 alone, the value of the polyethylene terephthalate (PET) bottle market amounted to USD 45.50 billion and is forecast to grow to USD 60.30 billion by 2033 [9]. In addition, the amount of plastic waste in 2024 was 220 million tons (an increase of over 7% compared to 2021) of which over 6 billion PET bottles were recycled [10].

The growth of PET packaging production may appear inconsistent with the UN sustainable development goals and EU environmental policy. However, in the Single-Use Plastic (SUP) Directive [11] and the Packaging and Packaging Waste Regulation (PPWR) [12], there is no limit to the production volume of PET packaging, but there are guidelines on the amount of recycled PET used in the production of new bottles. In this way, the growth of the market value may result from the development of the recycled PET segment, which is in line with the principles of closed circulation. Considering the above fact and the detrimental environmental impact of increasing bottled water consumption in PET containers, the UN and the EU require PET packaging manufacturers to operate in compliance with sustainable development principles, emphasizing, among other approaches, the circular economy [11,13].

PET bottle producers are working to align their production with sustainability requirements. In 2021, Danone's Aquas brand became the first in the mineral water sector to use 100% recycled PET (rPET) [14] and Coca-Cola switched to rPET bottles in the Netherlands and Norway [15]. Other initiatives include the introduction of a deposit return system in Europe [16,17], which helps increase PET recycling rates, and the use of "super-cleaning" technology for rPET [18], ensuring food safety standards are met. These actions, along with Environmental, Social, and Governance reporting and fiscal policies, support a more sustainable circular economy.

The increasing focus on sustainability also draws attention to the material itself—its origins, structure, and inherent properties. Polyethylene terephthalate is a polymer that is part of the polyester group. The first step in the industrial synthesis of PET involves the prepolymerization of dimethyl terephthalate (DMT) or terephthalic acid (TPA) with ethylene glycol (EG). The reaction leads to the formation of the intermediate compound bis(hydroxyethyl)terephthalate (BHET). Subsequently, polycondensation is carried out to form the polymer. In this process, a catalyst containing elements such as Sb, Ge, or Ti is used [19]. The obtained product exhibits glass-like transparency and good thermal and mechanical properties [19,20]. It is resistant to various compounds, such as oils, fats, aromatic solvents, chlorinated hydrocarbons, most acids, and diluted alkaline solutions [21].

A significant consequence of PET degradation is the generation of microplastic particles. According to EU legislation [22], microplastic is defined as a small solid object, insoluble in water, or wholly composed of synthetic polymers or chemically modified natural polymers. Microplastic particles have dimensions equal to or less than 5 mm and a length-to-width ratio of 3 or less. Additionally, microplastic fibers are defined as objects with a length equal to or less than 15 mm and a length-to-width ratio greater than three. Sometimes a more precise definition is provided, distinguishing between (i) nanoplastic as particles with the largest dimension ranging from 1 to 1000 nm, (ii) microplastic with dimensions from 1 to 1000 μ m, (iii) mesoplastic with dimensions from 1 to 10 mm, and (iv) macroplastic with dimensions of 1 cm and larger [23].

To counteract adverse situations effectively, it is essential to understand the mechanisms of PET degradation. These mechanisms vary depending on the degradation pathway and conditions [24]. As a result of degradation, not only can MPs be formed [25] but also different low-molecular-weight chemical compounds can be released [19]. The degradation of PET depends on various factors, including UV radiation [26], the presence of water [25], the presence of additives enhancing the material's properties [27], and even the specific form in which the PET material appears [25]. Factors occurring simultaneously, such as the presence of water combined with exposure to UV radiation, interact synergistically, accelerating the degradation of the material [28].

Previous studies have reported the presence of microplastics in bottled water [29–33]. However, these findings sometimes differ significantly regarding the composition of the detected microplastic particles. Despite this variability, there is general agreement about their presence. The discrepancies highlight the urgent need for the standardization of detection procedures, covering all stages of analysis—from sample collection and concentration to quantitative determination, shape characterization, and reporting methods [22]. It is also worth noting that matrix effects and the risk of cross-contamination are significant challenges in such analyses. While these issues are less pronounced in the case of bottled water, they still need to be considered carefully. Additionally, the detection of the smallest particles is crucial, yet it continues to pose significant challenges [31]. The fraction of the smallest MPs with sizes below 5 µm is considered to be potentially the most hazardous [34].

The aim of this work is to discuss the problem of degradation of polyethylene terephthalate bottles intended for water. The paper aims not only to raise awareness of the importance of this issue but also to highlight potential management strategies and explore the application of solutions. The situation is critical and demands prompt action across various fields. Increasing product stability before use [35,36] while facilitating waste degradation [37,38] are two seemingly contradictory approaches, yet their application could be a way to minimize the problem. Recycling and the use of reusable plastic bottles are important solutions, but if they lead to a greater release of microplastics due to reduced durability or the release of other contaminants, they may ultimately become counterproductive [32,39]. The discussed topic is complex and requires an integrated approach. Action is needed across overlapping areas, with collaboration between chemists, scientists, industry, and policymakers to implement new technologies and guide the direction of change.

To familiarize the reader with the discussed issue, this paper is divided into several main sections, addressing the significance of microplastic pollution, the mechanisms of PET degradation and the impact of environmental factors on degradation, detection methods for microplastics, and PET bottle degradation management strategies. The final section is the culmination point of the paper. It aims to highlight potential ways to manage the issue and suggests possible directions for minimizing the damage. Given the complexity of the issue, integrated strategies are essential. However, it can be argued that there is significant application potential for solutions, though they will require further development and legislative efforts.

2. Significance of Environmental and Food Pollution by Microplastics

Market globalization contributes to the increase in the pace of life and the emergence of new qualitative factors influencing the lives of consumers around the world. The lifestyle of consumers is changing with an increase in the importance of consumerism, ecological

4 of 30

and health awareness, and the possibility of using new technologies to quickly and safely meet their needs. The fast pace of life is accompanied by an increase in the popularity of convenience foods, mostly available in disposable packaging. A healthy lifestyle is also fashionable among consumers, resulting from the growing awareness of consumers about food safety and the role of water in the body. This contributes to the increase in bottled water sales. In 2022, the average amount of bottled water consumed in EU countries was 121 L per capita. The largest amount of water was consumed in Italy (249 L per capita) [40], and the least in Sweden (10 L per capita) [41]. In 2023, the share of plastic bottles accounted for 68.5% of the market share for water packaging.

In 2024, the size of the European bottled water market was estimated at USD 59,547.8 million. It is forecasted that by 2030, revenue from bottled water sales in Europe will amount to USD 83,302.4 million. The average annual growth rate between 2025 and 2030 is expected to be 5.7%. In 2024, European revenues in the global bottled water market accounted for 17.1%. In 2030, the Asia-Pacific region is expected to be the world market leader, with a value of USD 239,316.1 million. Furthermore, by 2033, the global bottled water market will be worth about USD 551.2 billion, up from USD 328.9 billion in 2023. A compound annual growth rate (CAGR) of 5.3% is forecasted from 2023 to 2033 [42].

Observed trends in bottled water consumption are closely linked to differences in regional environmental policies, infrastructure, and consumer behavior. In countries with well-developed access to safe tap water and a high level of public trust—such as Sweden-per capita consumption remains low. In contrast, in parts of Southern Europe, bottled water is often perceived as superior in terms of quality or taste [43]. The continued dominance of plastic packaging raises legitimate concerns regarding the EU's circular economy goals. In response, the EU is introducing new regulations-including the Single-Use Plastics Directive, the Packaging and Packaging Waste Regulation, and initiatives focused on the development of Deposit Return Schemes (DRSs)-aimed at reducing environmental impact by increasing collection and recycling rates, mandating recycled content, and enforcing Extended Producer Responsibility (EPR) [11,12]. These systemic changes, supported by evolving consumer preferences and the expansion of infrastructure (e.g., water refill stations), are essential for reconciling market growth with sustainability objectives. However, it is important to critically note that without stronger enforcement mechanisms, transparent monitoring, and harmonization across EU Member States, the effectiveness of these policies may remain limited. There is also a risk that responsibility continues to be shifted disproportionately onto consumers, rather than addressing systemic issues within production and supply chains.

The increasing trends in bottled water consumption in countries such as China, Indonesia, and the USA are due to population growth [44]. In developed countries, demand correlates with increasing consumer wealth. In addition, there has been an increase in the consumption of bottled water in tourist countries due to the growing number of tourists. In Italy, however, there is a widespread belief in the health benefits of mineral-rich water from specific regions of the country, which is in line with current consumer preference trends. Moreover, worldwide demand for bottled water is driven by the growing shortage of fresh and clean water (e.g., in India, Thailand, and Brazil) [44]. Another factor is the development of technology, which enables faster acquisition of cheaper quality water and water for special purposes (e.g., water enriched with magnesium ions or iodine). There is also a growing belief among consumers that bottled water is healthier than tap water. That is why it is becoming fashionable among parents to use bottled water intended for children from the first day of their life. Availability and reliability are other determinants of bottled water purchases. The largest share of bottled water sales is represented by PET plastic packaging. The advantage of water in PET bottles is their lightness, durability, flexibility, and widespread availability. PET bottles are designed to be strong, shatterresistant, and economically viable, making them particularly attractive to both bottled water producers and consumers. PET bottles are also highly recyclable, which increases their appeal in environmentally conscious markets [45]. Moreover, in accordance with legal regulations, this material is considered safe for contact with food.

Unfortunately, consumers who choose bottled water contribute to a significant and destructive impact on the natural environment. Increased consumption of PET bottled water results in increased waste. Data from 2021 indicates that PET waste in the United States accounted for 44.7% from single-use bottles and 12% of global solid waste [46]. Plastic packaging waste poses a serious threat to the natural environment due to its very long decomposition time. This is due to the inherent stability of PET, making it highly resistant to environmental biodegradation. It is estimated that PET bottles can take up to 500 years to decompose, contributing to pollution of the terrestrial and marine environment through the accumulation of degradation products [47,48]. In the natural environment, under the influence of atmospheric conditions, PET fragments mechanically and chemically, resulting in the formation of microplastic particles [23,49,50]. Degradation of plastic waste in the environment is a key process responsible for the accumulation of MP in the environment [51,52]. The degradation rate depends on the plastic's characteristics, the degree of weathering, and the conditions of the natural environment [53]. Nanoplastics pose a serious threat because, due to their small size, they are extremely difficult to detect and monitor in the environment.

Studies on the occurrence of plastic decomposition products in the natural environment have shown their presence in the air [54], soil [55,56] and water [57,58]. MPs contamination poses serious threats to the entire food chain. The MPs contamination pathway from bottles is presented using a modified Holdgate model (see Figure 1).

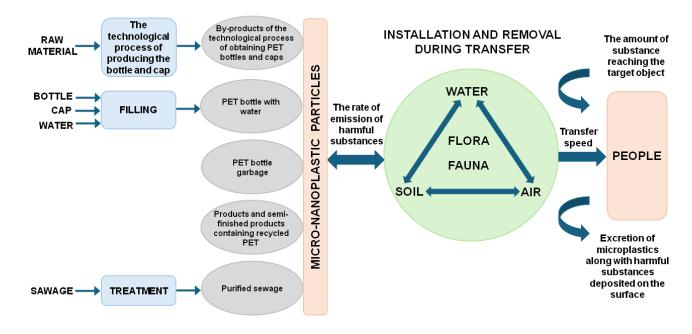


Figure 1. Paths of contamination from PET bottle decomposition products to the target object.

The analysis of the human (target object) contamination path should start with identifying the sources of pollution and the types of harmful substances. In the case of PET bottles, harmful substances released during degradation are micro- and nanoplastic particles and chemicals (including terephthalic acid, ethylene glycol, diethylene glycol, acetaldehyde, antimony, and bisphenol A) along with other harmful substances deposited on the surface. The sources of these contaminants are many, depending on the stage of the packaging life cycle. These contaminants can be created at any stage of the bottle production process, starting with the production of PET granulate, preforms, and the blowing of bottles. The next source of pollution occurs when the empty bottles are filled with water and sealed with plastic caps. During storage, harmful substances migrate from the packaging into water due to direct contact. Another source of microplastic contamination in bottled water is the cap, from which particles escape into the water when unscrewed. Once used, PET bottles become a serious problem as post-consumer waste, ending up in both legal and illegal landfills, both on land and in water. Some of this waste is recycled and used to produce new PET bottles or other products (e.g., clothes), which constitute a chain reaction, becoming both a source and a cause of further pollution. The source of the harmful substance determines the rate of emission of these pollutants. These contaminants can then enter the human body through various routes, which differ in the time it takes to reach the target object and the number of links (objects) in the distribution chain:

- Contamination path I degree—contamination reaches the target object very quickly because the contamination distribution chain consists of object I (e.g., air, water) and the target object.
- Contamination path II degree—contamination reaches the target object at a moderate pace because contamination is transferred to object I, from which it goes to object II (e.g., plants, vegetables, fruit, animals) and reaches the target object.
- Contamination path III degree—contamination reaches the target object slowly because contamination is transferred to object I (e.g., air), then to object II (e.g., soil), from which it gets to object III (e.g., plants, vegetables, fruit, animals) and reaches the target object.
- Contamination path IV degree—contamination reaches the target object very slowly, as the contamination is transferred to object I (e.g., air), then to object II (e.g., soil), from which it passes to object III (e.g., water) and to object IV (e.g., fish, seafood, plants) and reaches the target object.

The amount of harmful substances reaching the human body is determined by the complexity of the distribution chain. When microplastic particles reach the human body without passing through intermediate steps, material loss at each stage is minimized, leading to higher concentrations and increased exposure. It is believed that the shorter the chain, the greater the amount of substances entering the body. Some of the harmful substances are excreted from the body (e.g., through sweat, urine, and feces [59]), and some of them accumulate in the body, contributing to the development of diseases. Figure 2 schematically illustrates the contamination pathways categorized from degree I to IV.

Numerous studies conducted on the accumulation of PET bottle decomposition products in the human body have confirmed the presence of MPs in feces [60], endometrium [61], placenta [62,63], and even in gallstones [64]. The presence of PET microplastic particles was also detected in the bone marrow [65] and blood [66,67].

In patients with cardiovascular problems, it has been found that microplastics accumulating in the main artery are responsible for clogging it [3]. In addition, they also affect blood clotting, cause systemic inflammation, and influence the diffusion capacity of the lungs [68,69]. Nanoplastic particles pose a very serious problem, as they can penetrate biological barriers [70,71] and exhibit a higher level of toxicity compared to microplastic particles [72,73]. MPs from packaging contribute to the development of various serious diseases and infertility issues [6]. Moreover, it is important to consider their accumulation in the body from multiple sources [74].

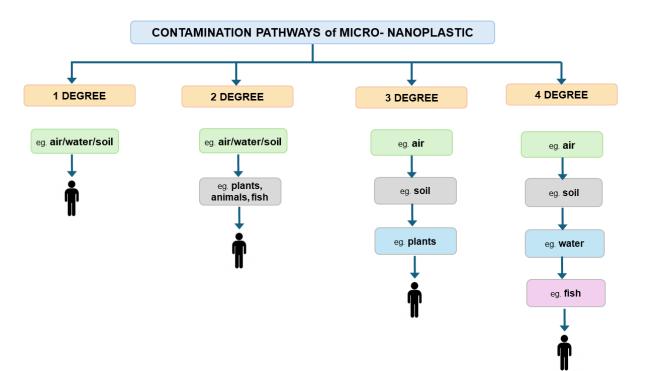


Figure 2. Contamination pathways (degree I to IV) by which harmful substances (e.g., microplastics) can enter the human body.

3. Mechanism and Factors Affecting PET Degradation

According to IUPAC, degradation of polymer materials is an alteration in the chemical structure of these materials that results in unwanted changes of their properties [75]. These changes may include a change in average molecular weight [25], color and transparency (plastics become yellow and opaque), as well as mechanical properties (polymers become brittle) [28]. Degradation, as shown in Figure 3, can be classified based on the factors that cause it [24]:

- Thermal degradation (factor: temperature),
- Photo-degradation (UV radiation),
- Hydrolytic degradation (water),
- Chemical degradation (chemical substances),
- Mechanical degradation (mechanical factors and stress),
- Biochemical degradation (enzymes or microorganisms),
- Combination of factors above.

Thermal degradation induced by high temperature in the absence of oxygen can be observed only in a controlled environment. The process primarily begins at the methylene group in the β -position relative to the carbonyl group, which serves as the primary site for β -scission. This reaction mainly leads to random scission of ester linkages, resulting in the formation of carbonyl-terminated units and carboxylate units terminated with a vinyl group. Intramolecular hydrogen shift leads to the formation of vinyl alcohol, which is unstable and rapidly converts into acetaldehyde through tautomerization. Meanwhile, intermolecular reaction results in the formation of ethylene. Additionally, various other degradation products have been identified, including CO, CO₂, aliphatic aldehydes, tetrahydrofuran, and unsaturated and aromatic hydrocarbons such as butadiene, benzene, and biphenyl [76,77]. It is worth mentioning that Pirzadeh [78] states that the thermal degradation rate is insensitive to moisture content, whereas the hydrolysis rate depends on it.

In the presence of oxygen and at elevated temperature, thermo-oxidative degradation occurs, known as Bolland's cycle. It begins with the formation of side groups in the form of hydroperoxides on the diester linkage of the polyester chain, which is initiated by the extraction of hydrogen by oxygen. Next, the hydroperoxides break down, leading to the formation of macroradicals [24,79].

Photodegradation of PET plastics is initiated by ultraviolet (UV) light. Many studies have proven that photodegradation of PET occurs through several major mechanisms: Norrish type I, Norrish type II, and Photo-Fries reactions [24–28,80–83]. A Norrish type I reaction is a reaction involving radicals in a degradation process. The radicals are created around the ester bond of the PET polymer chain. These radicals are capable of recombination, hydrogen abstraction, or reaction with oxygen [27]. During a Norrish type II reaction, a cyclic intermediate product is created via intramolecular rearrangement of γ -hydrogen. This unstable product breaks down, which leads to the creation of polymer chains ended with carboxyl groups and alkenes. The photodegradation process can also cause crosslinking, which affects the material's structure. The creation of vinyl ester groups can further promote cross-linking, leading to the formation of polyenes and other colored compounds, causing PET to turn yellow [24].

PET is able to undergo hydrolytic degradation in the presence of water at high temperature and pressure. Hydrolysis leads to the formation of terephthalic acid (TPA) and ethylene glycol (EG). Further transformations may result in the formation of benzoic acid from TPA and acetaldehyde from EG [84].

There are many products of PET chain degradation, including carboxylic acids, aldehydes, formates, aliphatic alcohol ending groups, quinones, dimers, anhydrides, mono- and dihydroxyl PET derivatives, CO and CO₂, terephthalic acid, and ethylene glycol [26,27,80,84–86]. Figure 3 presents the types of degradation mechanisms along with some of their products.

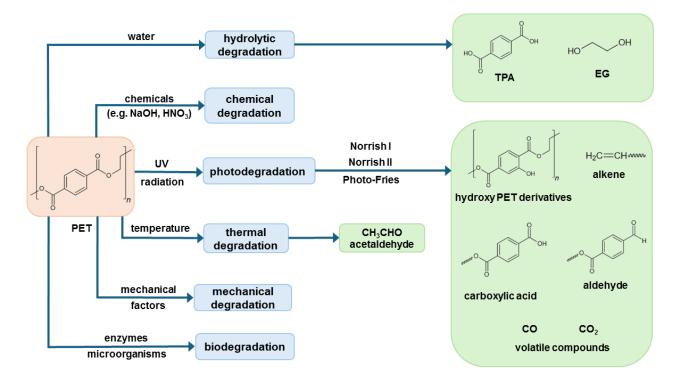


Figure 3. PET degradation mechanisms and some of their products.

The degradation of PET in the natural environment is influenced by multiple factors, including UV radiation, temperature, oxygen, humidity, and mechanical damage. These factors do not act independently; instead, they accelerate one another, leading to the simultaneous occurrence of different degradation mechanisms. PET is known to be relatively resistant to hydrolysis and even more to thermo-oxidative and thermal degradation, which typically does not occur under natural conditions. However, exposure to certain environmental factors can significantly increase the degradation rate [79,87].

The most important factor influencing PET degradation is UV radiation, as its presence significantly enhances the process. Shi et al. [83] showed that PET that is exposed to UV light generates significantly more microplastic fibers (MPFs) than PET that is kept in the dark. PET strongly absorbs short wavelengths of UV light compared to longer wavelengths. According to Horne et al. [26], this polymer shows high absorbance at $\lambda = 302$ nm. Radiation at this wavelength does not penetrate deeper into the plastic; degradation occurs primarily on its surface. In comparison, UV radiation at wavelength $\lambda = 365$ nm is less efficiently absorbed. This property makes long-wavelength light able to penetrate the plastic and cause degradation not only on the surface but also deeper within the material.

The presence of water is another crucial factor that significantly influences the degradation of PET. A high water content accelerates the degradation process of the material [25,28,84]. In a low-humidity environment, PET that undergoes photodegradation does not show any visible signs of damage, such as yellowing or loss of transparency; there are only minimal changes in its chemical composition [28]. In wet conditions, PET degradation undergoes intense degradation. Moisture leads to the hydrolysis of PET, resulting in the creation of TPA and EG as main products [28,84].

It was proven that PET is not resistant to alkaline solutions. The study of Pfeffier et al. [82] shows that PET particles would degrade when extracted using solutions of bases (potassium hydroxide and sodium hydroxide). The strongest degradation effect was observed for a concentrated NaOH solution. On the other hand, alkaline conditions do not have a great impact on the deeper parts of PET plastics, as degradation with NaOH solution is confined to the polymer surface [88].

The way plastics are exposed to water, such as constant submergence or wetting by sprinkling, influences the mechanisms and types of surface defects. Different surface changes are observed depending on the form of PET plastic and its production history [25]. As PET degrades over time, its surface contact angle decreases. This change is expected as the carboxyl groups, which are hydrophilic, are being created by exposure to UV radiation [26]. Interestingly, at a certain point during degradation, the contact angle increases instead of continuing to decrease. The roughness and heterogeneity of the degraded surface proved to be the cause of this change.

The temperature at which degradation takes place influences the rate of the process. A study on degradation at different temperatures (30, 40, and 50 °C) conducted by Rostampour et al. [28] has shown that degradation at 50 °C initiates photodegradation faster. The study has proven that the creation of carboxylic acid is much faster at 50 °C than at 40 °C and 30 °C. The work of Colnik et al. [84] has shown that higher temperatures promote PET hydrolysis to TPA and EG. In this case study, increased temperature to 350 °C also promotes decarboxylation of TPA, leading to the creation of benzoic acid and the degradation of EG to acetaldehyde. Papers [82,86] show that increased temperature accelerates the degradation of PET, especially in NaOH solutions.

The introduction of additional compounds in PET alters its stability. The use of UV stabilizers and different types of coatings on bottles, in general, enhances their mechanical properties and increases the resistance of plastics to harmful factors, such as UV light and

temperature. The impact of different additives on PET bottle properties is described in detail in Section 5.1.

4. Methods for Detecting of Microplastic Particles

The above sections underlined the significance of microplastic pollution. One of the key consequences of this problem is the necessity of determining MP content not only in bottled water but also in other products and environments. Various research teams employ different methods to achieve this goal. A commonly used approach is chemical imaging, which utilizes spectroscopic techniques such as FTIR microscopy (μ -FTIR) and Raman microscopy (μ -Raman) [22,30–32]. These techniques provide detailed information on the number, size, and shape of microplastic particles. Additionally, the spectral data collected for each detected particle enable the identification of polymer type. Standardization of research procedures is certainly justified, creating the need to develop a unified method for MP detection.

Recently, the European Commission published a decision regarding methods for detecting microplastics in water intended for human consumption [22]. This document provides general guidelines for conducting such analyses. The method outlined in the decision involves several key steps. First, water is filtered through cascade filters, with a minimum sample volume of 1000 L, to collect the MPs. Sample analysis can be performed either directly on the filter or after transferring the sample to an alternative support. To identify the particles, microscopic techniques are used to assess their size, shape, and quantity. In addition, spectroscopic techniques such as Raman microscopy or FTIR microscopy are employed to determine the type of material the particles are made of by comparing the recorded spectrum with a spectral library. The results obtained from these analyses are presented by reporting the number of MPs, expressed as the number of particles per cubic meter of water. The particles are categorized by their shape, size, and material. To simplify the identification process, a pragmatic approach is applied, focusing on 10 priority polymers. The main material of interest in this review, PET, is included in this group of priority polymers. A properly conducted measurement should be verified by assessing the recovery of material through the analysis of a sample with a predetermined amount of MPs. Additionally, the analysis of a blank sample is necessary to determine background contamination.

The administrative decision has not yet been implemented, yet it has already faced criticism. Concerns have been raised [31] that due to its restriction to detecting larger particles, the methodology requires refinement. It is designed for the detection of microplastic particles larger than $20 \,\mu\text{m}$, while the majority of MPs found in bottled and tap water are smaller than $10 \,\mu\text{m}$ [30,31] (see Figure 4). It is obvious that this topic requires further research and development of better methodology. Therefore, numerous research teams are actively working on this issue.

The example of the study [30] shows that there is a great need for guidelines not only to standardize detection methods and develop improved techniques but also to establish harmonized reporting criteria [22]. Defining particle size and shape categories is an undeniable necessity. Moreover, even fundamental aspects, such as data presentation and concentration reporting, require standardization. For instance, MP concentrations can be expressed as the number of particles per cubic meter of water [22] or as mass per volume [31]. Guidelines are also essential to prevent fundamental errors, such as reporting an excessive number of significant digits or presenting numerical values for measurements where the uncertainty far exceeds the measured value [30]. Additionally, the lack of basic statistical analysis further undermines data reliability.



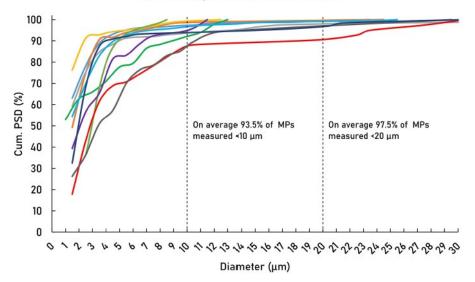


Figure 4. Cumulated microplastic particle size distribution detected in bottled waters and tap water. Each line corresponds to a single water sample. Image from Hagelskjær et al. [31], PLOS, CC-BY 4.0.

When it comes to addressing challenges related to analytical methodology, an interesting and perhaps the simplest approach is to exploit the autofluorescence properties of PET [89]. This method involves either recording the fluorescence spectra of the bulk solution or using fluorescence microscopy. The first approach does not directly provide the exact number of particles but rather a general signal proportional to their concentration. The second allows for the assessment of particle quantity, shape, and size. However, the question remains open regarding the sensitivity of this method, the lowest concentration ranges it can detect, as well as its selectivity. The latter issue is likely still relevant despite the fact that the analyte is dispersed in a relatively simple matrix, such as bottled water. This method appears to be suitable for strictly defined, synthetically produced samples with well-known composition, rather than for the analysis of environmental or real-life food samples.

A variation in the method described above involves using the fluorescent dye Nile Red. The use of this reagent provides a significant degree of specificity [90]. This method is well recognized for such measurements and is sometimes considered a standard approach [91–93]. The procedure involves binding the dye to microplastic particles, followed by microscopic observation. However, the detection of less hydrophobic particles, such as PET, may be somewhat limited [94].

The combination of microscopic observations with spectral techniques offers much greater specificity and the ability to identify the material of MPs compared to simple fluorometric techniques mentioned earlier [89,90,94]. While μ -FTIR and μ -Raman are more complex and expensive, they do have limitations and do not address all the challenges. Standard FTIR microscopy, for example, struggles with detecting particles smaller than 20 µm, and the resulting spectra may be inaccurate [95]. This led to the use of more advanced techniques, such as optical photothermal infrared spectroscopy (O-PTIR) and tunable mid-infrared quantum cascade laser (QCL-IR) technology [96].

O-PTIR is a non-invasive technique that uses the photothermal effect. The sample is illuminated with IR radiation, causing thermal expansion of the material and a change in the refractive index, which can then be measured using a narrow visible light beam [97]. Its advantage is its high spatial resolution, around 450×450 nm [98].

The QCL-IR system includes a set of tunable lasers enabling emission at specific wavelengths, covering the wavelength range from 1800 to 950 cm⁻¹ [99]. The QCL-IR method is faster and covers a larger area of the sample, but it has a lower resolution compared to O-PTIR. The QCL-IR is not suitable for detecting the smallest MPs, ranging from 1 to $3 \mu m$ [96].

A promising modification of Raman microscopy for MP detection is the use of Surface-Enhanced Raman Spectroscopy (SERS) with a Klarite substrate [100]. The substrate, on which the sample is placed, contains a dense grid of inverted pyramidal cavities made of gold, which focus light into hotspots. This structure enables large electric field enhancement. The prepared surface is then subjected to Raman mapping. The aim of developing this method was to create an uncomplicated but reliable analytical technique for detecting particles smaller than 1 μ m. Research has shown that the application of this method allows for the detection and identification of MPs down to 360 nm.

Thermo-analytical techniques, such as pyrolysis gas chromatography-mass spectrometry (py-GC-MS), represent a distinct approach compared to spectral methods [101,102]. This approach was developed in an attempt to address the limitations of IR and Raman imaging. In this method, polymers undergo thermal decomposition (pyrolysis), followed by separation using gas chromatography. The resulting degradation products are then identified based on mass spectrometry analysis. The method requires the prior construction of a reference library by analyzing known polymer types [22]. By design, thermo-analytical methods do not allow for the determination of particle number, size, or shape [101]. However, they enable the assessment of the total mass of the polymer and the identification of its type.

A novel approach for detecting nanoplastics may lie in the application of biosensors, including those focusing on measuring electrochemical properties [103]. Commonly, this type of sensor consists of three main components: a receptor, a transducer, and a reading device. The receptor can be a peptide, a protein (including enzymes), an antibody, or even a whole cell. It interacts specifically with the analyte, in this case a microplastic particle. With the help of the transducer, these interactions generate electrochemical or optical responses.

An example of a sensor from this group is a system with functionalized gold nanoparticles designed to detect PET MPs [104]. In this approach, gold nanoparticles were intended to be linked with a synthetic peptide exhibiting high affinity for PET monomers and PET polymeric ligands. The authors proposed a concept for developing a tool for the instantaneous colorimetric evaluation of microplastic presence. However, despite its promising potential, this concept requires practical validation, as the study was based solely on computational in silico simulations. The microplastic detection methods described in this section are summarized in Table 1.

The degradation of plastic can also be assessed by examining the migration of low molecular weight substances into water. These undesirable compounds include, among others, antimony trioxide, a catalyst used in PET production, as well as formaldehyde and acetaldehyde, which result from the thermo-mechanical and thermo-oxidative degradation of PET [19]. Instrumental analytical techniques, such as liquid chromatography coupled with mass spectrometry, can be employed to detect these substances [105]. The significance of this issue is recognized, particularly in the case of recycled PET [39].

Method	Key Parameters and Notes	Ref.
FTIR microscopy (μ-FTIR)	Microscopic imaging, particle identification based on FTIR spectra. Determination of number, size, shape and material of particles. Problematic detection of particles smaller than 20 µm.	[22,95]
Raman microscopy (µ-Raman)	Microscopic imaging, particle identification based on Raman spectra. Determination of number, size, shape and material of particles.	[22,30–32]
Autofluorescence measurements	Recording of fluorescence spectra of the bulk solution (does not directly provide the number of particles) or fluorescence microscopy. Limited to well defined samples, relatively cheap method but low selectivity.	[89]
Fluorescence measurements with dye Nile Red	Dye binding to microplastic particles followed by microscopic observation. Increased specificity compared to autofluorescence measurements.	[90–94]
Optical photothermal infrared spectroscopy (O-PTIR)	IR-irradiated sample thermally expands, refractive index change detected by visible light. High spatial resolution below 1 µm.	[96–98]
Tunable mid-infrared quantum cascade laser spectroscopy (QCL-IR)	Tunable lasers (1800–950 cm $^{-1}$). Method fast, scans larger area, lower resolution vs O-PTIR.	[96,99]
Surface-enhanced Raman spectroscopy (SERS)	Sample placed on Klarite substrate and analyzed by Raman microscopy. High spatial resolution below 1 µm.	[100]
Pyrolysis gas chromatography-mass spectrometry (py-GC-MS)	Sample undergoes pyrolysis followed by gas chromatography separation and mass spectrometry identification. Determination of polymer type and total mass only; not particle number, size, or shape.	[101,102]
Biosensor with functionalized gold nanoparticles	Gold nanoparticles linked with a synthetic peptide exhibiting a high affinity for PET. Visual or instrumental, colorimetric detection. Theoretical concept has not yet been tested experimentally.	[104]

Table 1. Summary of methods used for microplastic detection, with particular focus on the detectionof PET particles in bottled water.

Summarizing this brief overview of MP detection methods, we would like to draw attention to an important issue. Unexpectedly, data from [31] suggest that polyethylene MPs may, in some cases, be more abundant than PET in these products. However, other reports present different findings [30,32]. This highlights the importance of keeping an open mind, as research findings may not always align with initial expectations.

The methods discussed here focused primarily on analytical techniques for direct detection of microplastics and other degradation products. However, determining the MP content is a broader challenge that encompasses several critical aspects. These include, among others: separation [106,107], concentration, and collection of MPs, matrix removal (for example, H₂O₂ digestion [31], Fenton reaction, or enzymatic treatments [101]) and mitigation of cross-contamination. Filters used for MP collection can themselves introduce microparticles. To ensure the reliability of results, performing appropriate negative and positive controls is absolutely necessary. Ultimately, addressing the issue of microplastic content determination requires a comprehensive approach that covers all stages of the process, rather than focusing solely on the detection step itself.

5. PET Bottle Degradation Management Strategies

The European Union has introduced a series of regulations to address the growing issues of microplastic accumulation in the environment, aiming to reduce plastic waste, improve recycling efficiency, and mitigate the negative impact on the environment and human health. Directive 2018/852 on packaging and packaging waste [108] requires member states to recycle at least 55% of all plastic packaging by 2030. A key element of the European Union's strategy to reduce plastic pollution is Directive 2019/904 on single-use plastics [109], which obliges PET bottle manufacturers to include 25% recycled materials by 2025, and this rate is expected to increase to 30% by 2030. This directive also sets collection targets for plastic beverage bottles: by 2025, at least 77% of PET bottles should be collected, and by 2029, 90%. Additionally, the EU tax introduced in 2021 (the so-called plastic levy) [110] on non-recycled plastic packaging waste aims to motivate producers to take greater responsibility for the lifecycle of their products. Implementing these regulations is crucial in the context of supporting a circular economy, where PET bottles are reused multiple times or recycled. Furthermore, increasing social and industrial awareness regarding proper waste management is an important aspect.

Two key strategies, operating at different stages of the PET bottle lifecycle, distinguish the management of PET bottle degradation, as presented in Figure 5. The first concerns PET bottles in use and involves preventing their degradation and securing the contents of the bottles. The second strategy relates to post-consumer PET bottles, i.e., waste, and focuses on improving the PET degradation process, efficiently processing used bottles, and effectively removing and monitoring microplastics in wastewater treatment plants. Both strategies share the common goal of reducing microplastics in the environment and protecting human health. Although they achieve these goals in different ways, their overarching aim is to promote sustainable development and environmental protection, which is also in line with the existing EU regulations regarding the handling of PET bottles.

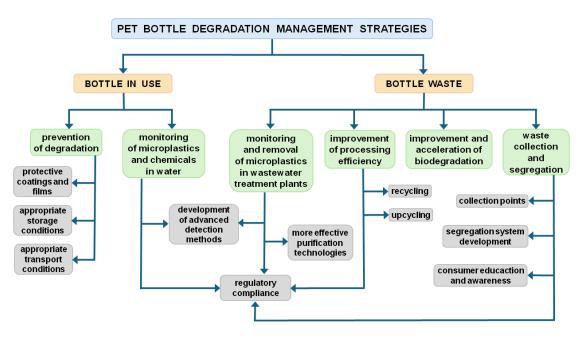


Figure 5. Strategies for managing PET bottled water degradation.

5.1. Preventing PET Bottle Degradation

The use of various coatings and films on PET bottles is one of the most effective ways to increase their durability and reduce degradation. This is particularly important from both an environmental and human health perspective. The degradation of PET during the storage of water in PET bottles can lead to the migration of microplastics and various chemical substances, such as formaldehyde and acetaldehyde, as well as antimony, which is used as a catalyst in PET production [19,111]. Therefore, it is crucial to monitor the presence of microplastics and harmful chemical substances in bottled water, as well as to develop more advanced methods for their detection, which are discussed in Section 4. This approach will allow for more effective monitoring and quicker identification of potential health risks for consumers.

It is worth noting that exposing PET water bottles to sunlight during transport and storage can cause the breaking of polymer bonds in the chemical structure, which promotes the migration of microplastics into the water [33]. Studies indicate that the main source of microplastics in bottled water is the degradation of PET bottles, which occurs at every stage—from production, through transport, to consumption [33]. Additionally, UV radiation and high temperatures can lead to the release of volatile organic compounds (VOCs) from PET bottles, which affects the quality and safety of the water [112]. To minimize these risks, it is necessary to ensure proper storage and transport conditions for PET bottles, particularly avoiding exposure to high temperatures and direct sunlight. Additional protection can be provided by coatings on PET bottles, which limit the contact of PET with water, thereby reducing the risk of harmful substance migration. Moreover, appropriate coatings on PET bottles improve their barrier properties, increase thermal and mechanical resistance, and effectively protect against UV radiation.

Surface modifications of PET to increase their resistance to degradation involve various materials and techniques. A popular approach is the use of polymer coatings. Poly(vinyl alcohol) (PVOH) is one of the materials that significantly improves the durability and barrier properties of PET, particularly in terms of oxygen permeability [113]. Reducing oxygen permeability not only prevents the oxidation of the contents but also reduces the risk of PET degradation caused by contact with oxygen. Similarly, polyacrylates, such as methyl methacrylate-methyl acrylate-diallyl maleate-maleic acid (MMA-MAc-DAM-MA), improve these properties, although too thick a layer can paradoxically worsen them [114]. Another solution is hybrid coatings, modified with OVPOSS (octavinyl-polyhedral oligomeric silsesquioxane), UV-curable [115]. These types of coatings significantly improve the surface properties of PET, making them more durable and resistant to mechanical and chemical damage. These coatings also exhibit good thermal stability.

One of the most effective materials used to provide a gas barrier for PET bottles is silicon-based coatings. Studies have shown that a thin protective layer of silicon oxide (SiO_x) , applied to the inner side of the PET bottle using the plasma impulse chemical vapor deposition (PICVD) process, effectively reduces the permeability of gases such as oxygen and carbon dioxide [36]. The effectiveness of SiO_x coatings in reducing the migration of substances from PET material into food simulants has been quantitatively assessed in studies conducted under realistic storage and transport conditions, such as elevated temperatures (40 °C). Although these studies did not directly cover prolonged UV exposure or variable temperatures, they evaluate the effectiveness of the coating under conditions simulating contact with food. The Plasmax technology [116], used in the coating process of PET bottles, known as FreshSafe and offered by the German PET machinery manufacturer KHS Corpolast, allows for the creation of micro-thin, glass-like SiO_x coatings on PET bottles. FreshSafe PET bottles can be fully recycled. During the recycling process, the coating is hydrolyzed using hot caustic lye, which allows for its removal from the inner wall of the bottle. It is worth noting that the FreshSafe PET protective technology is also suitable for coating PET bottles made from recycled PET (rPET). Another method is coating PET bottles with a thin layer of hydrocarbon silicate (SiOC) using the hot wire CVD (chemical vapor deposition) technique assisted by plasma [117]. SiOC coatings are characterized by

high gas barrier properties against oxygen, carbon dioxide, and water vapor, and unlike SiO_x coatings, they exhibit stability in contact with various liquids, including neutral pH beverages.

Another group of materials used to modify the surface of PET is carbon coatings. Among them, hydrogenated amorphous carbon coatings (a-C:H) [35] and diamond-like carbon (DLC) coatings [118] are of particular importance. The deposition of carbon films on PET bottles is carried out using plasma technologies, which allow for the creation of thin, uniform layers. Carbon coatings on PET bottles significantly enhance their gas barrier properties, as well as improve their resistance to mechanical and chemical damage, increasing their durability. Moreover, the use of carbon coatings does not hinder the recycling process of PET bottles.

Stabilizers, used as additives in plastics, aim to increase the material's resistance to degradation. UV stabilizers are added to improve the durability of the plastic under UV radiation. This effect has been confirmed in studies by Fechine et al. [27]. This study focused on comparing the effects of UV radiation on the degradation of plastic with and without the addition of a stabilizer commercially known as Tinuvin 1577w. The presence of Tinuvin 1577w stabilizer results in a reduction in the formation of carboxyl groups. Data obtained from fluorescence measurements showed the formation of monohydroxyterephthalate derivatives on the surface of PET with the addition of Tinuvin 1577w. This stabilizer primarily acts at the surface level by absorbing UV radiation, thereby delaying the photochemical degradation of the material's outer layers. In the context of UV protection, PET surfaces are coated with titanium dioxide (TiO₂) nanoparticles [119]. TiO₂ in its non-photocatalytic form serves as a physical barrier to UV radiation, while also improving the material's thermal stability and mechanical properties, such as crack resistance. However, it is worth noting that the impact of TiO_2 on the recyclability of PET composites has not yet been clearly determined. Another way to reduce UV permeability through PET and improve the mechanical properties of PET bottles is to produce composite PET bottles using borates such as takedaite $(Ca_3B_2O_6)$ [120] or calcium metaborate (CaB_2O_4) [121]. These compounds act as barriers to UV/visible light, significantly reducing radiation permeability and increasing the material's resistance to environmental stress cracking. Composites of PET with other polymers can also delay the degradation process. Shi et al. [83] conducted a study that compared the photodegradation of pure PET and PET composite with polyurethane (PET-U). The results indicated that the addition of polyurethane (PU) significantly delays the degradation rate of the material. Despite the lower degradation rate, the PU and PET mixture still generates significant amounts of micro- and nanoplastics. Furthermore, this study demonstrated a decrease in the average molecular weight for both pure PET and PET-U composite.

To date, no studies have quantitatively compared the effectiveness of different UV stabilizers—such as Tinuvin 1577w, TiO_2 nanoparticles, and borate compounds—under identical experimental conditions using the same type of PET and testing protocols. Most available studies assess these additives separately, under varying test parameters, which limits the ability to perform reliable comparative evaluations. Therefore, we emphasize the need for further coordinated research encompassing different classes of UV stabilizers, conducted under standardized and realistic environmental conditions while also considering their long-term effects on PET material durability, health and environmental safety, and recyclability.

In summary, although the available literature data indicate the benefits of using protective coatings in terms of improving the barrier properties of PET bottles and their resistance to UV radiation, there is a lack of extensive studies that, under realistic storage and transport conditions (e.g., prolonged exposure to sunlight and temperature fluctuations), would evaluate both their effectiveness and the potential risks associated with the degradation and migration of substances from the coatings themselves into bottled water. Therefore, it is advisable to conduct further research aimed at quantitatively assessing the effectiveness and stability of coatings and identifying potential chemical hazards associated with their use.

Moreover, the use of coatings and additives on PET bottles poses a significant challenge for recycling and upcycling, even though they significantly improve barrier properties, increase resistance to degradation, and provide protection against harmful substances [122]. These coatings can hinder material recovery, making recycling processes more complex and time-consuming. Therefore, despite their positive impact on the durability and quality of the bottles, it is necessary to further develop solutions that will enable easier processing of PET bottles after their use. A summary of the strategies for preventing PET bottle degradation is presented in Table 2.

Table 2. Summary of strategies for preventing PET bottle degradation.

Preventive Measure	Key Characteristics and Potential Drawbacks	Ref.
Proper storage and transport conditions	Avoiding high temperatures and sunlight due to their impact on PET stability.	[28,112]
Surface modifications with polyvinyl alcohol (PVOH)	Improves durability, limits oxygen permeability. Limits PET degradation by oxygen.	[113]
Surface modifications with MMA-MAc-DAM-MA	Limits oxygen permeability, too thick layer has adverse effect.	[114]
Hybrid coating with octavinyl-polyhedral oligomeric silsesquioxane	Surface more resistant to mechanical and chemical damage. Coating has good thermal stability.	[115]
Silicon-based coatings	Protective layer of SiO _x or hydrocarbon silicate applied to the inner side of the PET bottle. Reduced gas permeability, including oxygen.	[36,116,117]
Carbon coatings	PET surface modified by hydrogenated amorphous carbon coatings or diamond-like carbon coatings. Enhancing gas barrier properties, surface more resistant to mechanical and chemical damage. Bottles may be recycled.	[35,118]
TiO ₂ nanoparticles coatings	UV protection, increased thermal stability.	[119]
Addition of UV stabilizers (Tinuvin 1577w)	Improves durability, reduces formation of carboxyl groups.	[27]
PET composites with $Ca_3B_2O_6$ or CaB_2O_4	Improves mechanical properties, reduces UV permeability.	[120,121]
PET composites with polyurethane (PET-U)	Delays degradation but composite still generates microplastic.	[83]

5.2. Improving the Efficiency of Waste Management

PET, as one of the most commonly used polymers for bottle production, is characterized by high strength and stability. However, these same properties make its degradation in the environment a long-term process [48]. As a result, its accumulation in ecosystems poses a significant environmental threat. Improper disposal of this material further exacerbates the problem, making it necessary to implement effective methods to reduce its negative impact on the environment.

To reduce the amount of PET waste and improve its degradation, various approaches have been developed, including biodegradation, biological and mechanical recycling, chemical recycling, and upcycling. Each of these solutions aims to increase the efficiency of material reuse or accelerate its decomposition. The effectiveness of PET bottle recycling and

18 of 30

upcycling processes largely depends on the effective segregation and classification of waste by type of plastic. However, the segregation process poses significant technological and economic challenges, as many products contain various components and types of plastic, which significantly complicates their separation [46,122]. Pinter et al. [123] emphasize that the quality of the input material, including the presence of contaminants or fragments of other polymers, has a significant impact on the final purity of the recycled PET. The effectiveness of recycling can be improved by implementing mono-collection systems, in which only food-grade PET bottles are selectively collected. This solution provides better results than co-collection systems [124]. Additionally, the use of artificial intelligence (AI) in the sorting process allows for precise identification and removal of non-PET containers from the recycling stream, significantly improving the efficiency of segregation and the quality of the obtained rPET [46].

5.2.1. Biodegradation and Biological Recycling

Microorganisms capable of degrading PET, which can even assimilate the products of degradation, represent one of the solutions to the problem of plastic pollution in the environment. Reduced human involvement in the PET biodegradation process is a significant advantage of this approach. This process is natural but usually very slow. Many microorganisms capable of breaking down PET have already been discovered, and some of them have been genetically modified to improve the efficiency of this process.

One of the most recognizable microorganisms degrading PET is *Ideonella sakaiensis*. This gram-negative bacterium was isolated in 2016 in Sakai, Japan, from samples taken from a PET bottle recycling site [125]. This bacterium has a unique ability to break down PET using two enzymes: PETase and MHETase. PETase hydrolyzes PET into mono(2-hydroxyethyl) terephthalate (MHET), which is then converted by MHETase into terephthalic acid and ethylene glycol. These end products can be utilized by the bacteria but also harvested for further applications, such as in industry. Studies have shown that *Ideonella sakaiensis* grows exclusively on amorphous regions of PET while areas with high crystallinity do not support its growth [126]. This indicates the significant impact of PET bottle surfaces used for water packaging are made of highly crystalline PET, which hinders their enzymatic breakdown. In contrast, low-crystallinity PET is much more susceptible to enzyme action [127]. To improve the efficiency of biodegradation and enzymatic recycling of PET bottles, it may be crucial to apply techniques that modify their crystallinity, such as rapid cooling of molten PET, leading to a more amorphous structure [126].

Protein engineering enables the design of modified enzymes that hydrolyze PET, significantly improving the efficiency of its degradation. An example is the engineering of PETase, with modifications aimed at increasing the enzyme's thermal stability and activity. The IsPETase triple mutant exhibits significantly higher thermal stability and degradation efficiency compared to the wild-type PETase [128]. Additionally, Tournier et al. [129] developed an improved variant of leaf-branch compost cutinase (LCC), which achieves at least 90% PET depolymerization into monomers within 10 h at a temperature of 72 °C. This process, based on the enzymatic depolymerization of PET waste, allows for the reuse of monomers to produce new bottles, thereby supporting the concept of a circular economy, where PET can be cyclically used in packaging production. Studies have shown that PET obtained through biological recycling exhibits the same properties as PET produced from petrochemical raw materials.

One of the solutions to increase the efficiency of PET degradation could be enzyme engineering based on machine learning. Using this approach, Gupta and Agrawal [130] designed a mutated PETase enzyme that exhibits activity at a higher optimal temperature and

is characterized by thermostability, enabling more effective PET degradation. Additionally, Lu et al. [37] applied machine learning to design a new PET hydrolase, named FAST-PETase. They found that this variant exhibits significantly higher hydrolytic activity towards PET water bottles compared to wild-type PETase and other modified variants. FAST-PETase is capable of almost completely degrading pre-treated PET bottles at a temperature of 50 °C in less than 2 weeks. Moreover, at this temperature, FAST-PETase can also depolymerize untreated, amorphous parts of commercial water bottles and whole bottles subjected to thermal treatment.

One of the key challenges for enzymatic PET recycling is its variable crystallinity, the presence of contaminants, and additives in actual post-consumer waste streams. Although enzymes such as FAST-PETase, modified LCC, and IsPETase variants exhibit high activity under laboratory conditions, particularly toward amorphous PET, their effectiveness in more complex environments (e.g., highly crystalline PET, laminates, or in the presence of contaminants) remains limited and requires further verification. Thus far, studies on these enzymes have mainly looked at processed samples that are less crystalline, which helps evaluate their potential but does not truly represent real industrial situations.

To accelerate PET biodegradation, it is essential to use enzymatic synergy, where different enzymes work together in the degradation process. In their study, Mrigwani et al. [131] suggested using two different enzymes that work well together: leaf-branch compost cutinase and thermostable carboxylesterase (Thermus thermophilus carboxylesterase). The first enzyme degrades the PET surface, while the second hydrolyzes intermediate degradation products. This approach enables the complete breakdown of PET products into terephthalic acid (TPA) and ethylene glycol (EG).

Combining the action of enzymes and microorganisms can lead to more effective degradation of PET microplastics during high-temperature composting [132]. The presence of microbiota, such as *Acinetobacter* and *Bacillus*, without the addition of enzymes, contributes to a 26% reduction in PET. However, introducing PET hydrolase (WCCG, a quadruple variant of LC-cutinase) into the compost triggers a chemical reaction with PET, reducing the population of plastic-degrading bacteria, leading to a 35% degradation of PET.

Effective degradation of used PET bottles can be achieved by integrating chemical and enzymatic methods [38,133]. Studies have shown that mild alkaline pre-treatment at 25 °C. modifies the PET surface, reduces its crystallinity, and increases PETase accessibility to hydrolyzed bonds, significantly improving degradation efficiency [38]. Additionally, pre-treatment of PET bottles using microwave irradiation supports enzymatic plastic recycling [133]. This process effectively shortens PET polymer chains and induces favorable conformations, increasing their availability for enzymatic hydrolysis.

5.2.2. Mechanical and Chemical Recycling

Mechanical recycling is one of the most commonly used methods for processing PET. In the first stage, bottles are ground into small pieces, which are then washed to remove impurities. The next step is melting the PET flakes, which allows for the production of resin used to create new bottles. This process is energy- and cost-efficient, contributing to the reduction in greenhouse gas emissions and preventing the accumulation of waste in landfills and oceans [134]. Unfortunately, with each recycling cycle, PET loses its original properties and becomes increasingly brittle [135]. Additionally, due to multiple processing cycles, the material may contain more additives, such as dyes, toners, or catalysts. Therefore, repolymerization becomes necessary, which involves additional costs and reduces the overall efficiency of the process [46]. Despite these challenges, the mechanism of mechanical recycling is well understood, and its cost-effectiveness makes it widely used [136].

Chemical recycling involves depolymerizing the PET polymer into its original components, i.e., monomers or smaller oligomers, which can then be repolymerized into a new polymer. This process may also include solvolysis, which involves dissolving the polymer for subsequent purification [46,135]. Several methods of chemical recycling can be distinguished, including hydrolysis, which breaks down PET into ethylene glycol and terephthalic acid under acidic, alkaline, or neutral conditions, elevated pressure, and temperature [46,135,136]; alcoholysis, in which PET reacts with alcohol, most commonly methanol or ethylene glycol, being a commercially used method [46,135,136]; pyrolysis, which leads to the production of fuel compounds [134,136]; and the less studied but promising aminolysis, yielding aminoderivatives of terephthalic acid [46,135,136].

The main advantage of chemical recycling is the ability to obtain pure, high-quality material even from heavily contaminated waste [46], making it easier than mechanical recycling. Unfortunately, this process involves high complexity, resulting in higher energy costs and the need for catalysts [134] and may also lead to the generation of environmentally toxic waste, such as residues from acids and amines [136]. Therefore, chemical recycling is most often used as a complement to mechanical recycling [46,136]. Additionally, in the case of mixed waste, particularly when it is colored, the formation of black residue in the final product may occur [136]. This occurrence is mainly due to the degradation of pigments, dyes, and other additives at high temperatures (e.g., during pyrolysis). Furthermore, impurities like PVC found in mixed waste can cause PET undergo undesirable decomposition reactions, create unwanted byproducts, and leave more black residue [137,138]. These contaminants disrupt the PET decomposition process, reducing the efficiency of recycling and the quality of the obtained products.

5.2.3. Upcycling

Upcycling PET bottles is an effective method of reducing plastic pollution, contributing to the "waste to resource" strategy, and supporting sustainable development. This process involves transforming PET waste into products of higher value and durability. However, it requires advanced technologies and specialized skills, which can be a barrier to its widespread implementation [134]. PET upcycling can lead to the creation of new materials in various industries, including textile production [134]. An interesting example is the conversion of used PET water bottles into high-performance activated carbon, which can be used in electrochemical processes such as water desalination [139]. Using PET bottles to produce activated carbon electrodes reduces plastic pollution and contributes to the development of electrode materials, offering economical and energy-efficient solutions for water desalination. Another approach is converting PET bottles into hydrogel materials through aminolysis, enabling their use in water purification processes, particularly in the removal of industrial dyes [140]. This solution represents an interesting way to recycle plastic waste and transform it into materials with adsorptive properties. In the context of construction, PET bottle waste has also been used as an additive to asphalt pavements [141]. Studies have indicated that the addition of processed PET bottles to stone mastic asphalt (SMA) improves the mixture's properties, including resistance to permanent deformation, contributing to the extended durability of road surfaces. However, when developing such materials, it is important to ensure that they themselves do not become a source of microplastics [142]. Further technological innovations in PET bottle upcycling will contribute to more effective management of plastic waste.

5.2.4. Monitoring and Removing Microplastics in Wastewater Treatment Plants

Microplastics enter wastewater systems in various ways, and one significant source of their presence is the recycling of PET bottles and other plastics. Recycling has a positive impact on waste management, but this process also contributes to the emission of MP pollutants [143]. Wastewater treatment plants play a crucial role in retaining microplastics, serving as the last barrier before they enter the environment [144,145]. Filter-based technologies, such as membrane bioreactors (MBRs) and rapid sand filtration (RSF), are highly effective in removing microplastics, achieving efficiencies of 99.99% and 97%, respectively [146]. Nevertheless, some microplastics remain in sewage sludge, which can become a secondary source of soil and groundwater contamination, posing a threat to ecosystems and public health [143,147,148].

One promising solution to this problem is the application of enzymatic technologies that enable the biodegradation of PET microplastic particles. Particularly effective in this process are *Comamonas testosteroni* bacteria. These organisms can significantly enhance the efficiency of wastewater treatment and reduce the amount of particles accumulating in sludge [149,150].

In addition to implementing new treatment technologies, it is equally important to monitor microplastics in wastewater and sludge. Spectroscopic methods are most often used for this purpose because they accurately identify and analyze microplastics in environmental samples [147,151,152]. Methods for detecting microplastics are described in detail in Section 4.

Due to the growing threat posed by the presence of microplastics in waters discharged into the environment, the European Union adopted Directive 2024/3019 [153], which obliges member states to implement more effective technologies for treating and monitoring microplastics in both wastewater and sewage sludge. According to this directive, wastewater treatment plants should implement additional treatment stages, known as fourth-stage treatment, aimed at removing microplastics from wastewater before it is released into the environment. Compliance with these regulations is crucial for environmental protection and public health.

6. Conclusions

The degradation of PET in bottled water packaging poses a serious threat to both public health and the natural environment. To mitigate the negative impacts of this phenomenon, it is essential to implement two strategies for managing PET degradation: protecting PET bottles in use from degradation and improving the degradation process and efficient processing of post-consumer PET containers. Despite apparent contradictions, these strategies are complementary, operating at different stages of the PET bottle life cycle. Their overarching objective is to minimize the harmful effects of PET degradation on ecosystems and human health.

In the context of PET bottles in use, it is crucial to coat them with special protective coatings and films that can significantly reduce their degradation, enhance their barrier properties, increase their resistance to UV radiation and other external factors, and prevent the migration of PET degradation products and harmful chemicals into drinking water. Although these coatings effectively increase consumer safety and promote the idea of a circular economy, they can simultaneously hinder the subsequent recycling process. Therefore, it is necessary to further improve protective coating technologies to combine the extension of bottle durability with the possibility of their efficient processing. Actions within this strategy also include monitoring the presence of microplastics and chemicals in water and ensuring appropriate storage and transportation conditions for the bottles.

Regarding post-consumer PET bottles, technologies that accelerate their breakdown, such as the use of enzymes and microorganisms, are of key importance. The development of PET biodegradation methods creates an opportunity for more effective elimination of plastic waste from ecosystems, thereby minimizing its long-term impact on the environment and human health. Effective management of post-consumer PET bottles also requires improving recycling and upcycling processes to maximize the use of recovered material and reduce the amount of waste entering the environment. It is crucial to streamline plastic waste sorting systems and increase public awareness about proper waste segregation. An important element of this strategy is also the implementation of more efficient technologies for cleaning and monitoring the presence of microplastics and degradation products in both wastewater and sewage sludge. Strengthening these actions in wastewater treatment plants will allow for more effective protection of the environment and human health.

Despite significant efforts undertaken to understand the degradation processes and factors contributing to microplastic contamination, many fundamental issues remain unresolved. For example, there is a lack of studies and data concerning the relationship between low molecular weight degradation products and the formation of microplastics, as well as analyses of the conditions influencing this relationship. The development of a mathematical model describing the migration paths of microplastics in the environment, particularly one based on well-established principles such as Fick's law, would be beneficial. This task, however, poses a considerable challenge, as it would need to account for highly variable environmental conditions and include a wide range of parameters describing the system. Nevertheless, such a model could support the design of strategies aimed at reducing the risks associated with microplastic pollution. A major unresolved challenge remains the lack of defined dose–response relationships and toxicological limits for micro- and nanoplastics in the human body. Further research is needed to define the physiological limits at which MPs transition from inert to pathologically active substances. Identifying these knowledge gaps is important, as it helps to define future research directions.

Addressing the problem of PET bottle degradation requires a comprehensive approach that includes legal regulations, the development of new technologies, improvement of recycling processes, and actions to increase public awareness about the impact of this phenomenon on health and the environment. Promoting responsible waste management is also an important element. Achieving sustainable development goals and reducing microplastic pollution will only be possible through close cooperation between the scientific community, industry, and institutions responsible for shaping environmental policy.

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Abbreviations

The following abbreviations are used in this manuscript:

AI	artificial intelligence
BHET	bis(hydroxyethyl)terephthalate
CAGR	compound annual growth rate

CVD	chemical vapor deposition
DLC	diamond-like carbon
DMT	dimethyl terephthalate
DRS	deposit return schemes
EG	ethylene glycol
EPR	extended producer responsibility
EU	European Union
LCC	leaf-branch compost cutinase
MBR	membrane bioreactor
MHET	mono(2-hydroxyethyl) terephthalate
MMA-MAc-DAM-MA	methacrylate-methyl acrylate-diallyl maleate-maleic
MP	microplastic
MPF	microplastic fiber
µ-FTIR	FTIR microscopy
µ-Raman	Raman microscopy
O-PTIR	optical photothermal infrared spectroscopy
OVPOSS	octavinyl-polyhedral oligomeric silsesquioxane
PET	polyethylene terephthalate
PICVD	plasma impulse chemical vapor deposition
PPWR Regulation	EU Packaging and Packaging Waste Regulation
PU	polyurethane
PVOH	Poly(vinyl alcohol)
py-GC-MS	pyrolysis gas chromatography-mass spectrometry
QCL-IR	tunable mid-infrared quantum cascade laser technology
rPET	recycled PET
RSF	rapid sand filtration
SERS	Surface-Enhanced Raman Spectroscopy
SMA	stone mastic asphalt
SUP Directive	EU Directive on single-use plastics
TPA	terephthalic acid
UV	ultraviolet
VOC	volatile organic compound
WCCG	quadruple variant of LC-cutinase

References

- Panou, A.; Karabagias, I. Migration and Safety Aspects of Plastic Food Packaging Materials: Need for Reconsideration? *Coatings* 2024, 14, 168. [CrossRef]
- 2. Muhib, M.I.; Uddin, M.K.; Rahman, M.M.; Malafaia, G. Occurrence of microplastics in tap and bottled water, and food packaging: A narrative review on current knowledge. *Sci. Total Environ.* **2023**, *865–876*, 161274. [CrossRef] [PubMed]
- 3. Marfella, R.; Prattichizzo, F.; Sardu, C.; Fulgenzi, G.; Graciotti, L.; Spadoni, T.; D'Onofrio, N.; Scisciola, L.; La Grotta, R.; Frigé, C.; et al. Microplastics and Nanoplastics in Atheromas and Cardiovascular Events. *N. Engl. J. Med.* **2024**, *390*, 900–910. [CrossRef]
- Feng, X.; Zhang, M.; Zhao, T.; Cui, J.; Ye, H.; Zhou, C.; Ye, L.; Zhou, L. Polystyrene microplastics trigger colonic inflammation in rats via the TLR4/NF-κB/COX-2 pathway and modulation of intestinal microbiota. *Toxicology* 2025, *513*, 154090. [CrossRef] [PubMed]
- Brown, D.M.; Wilson, M.R.; MacNee, W.; Stone, V.; Donaldson, K. Size-dependent proinflammatory effects of ultrafine polystyrene particles: A role for surface area and oxidative stress in the enhanced activity of ultrafines. *Toxicol. Appl. Pharmacol.* 2001, 175, 191–199. [CrossRef]
- Amereh, F.; Babaei, M.; Eslami, A.; Fazelipour, S.; Rafiee, M. The emerging risk of exposure to nano(micro)plastics on endocrine disturbance and reproductive toxicity: From a hypothetical scenario to a global public health challenge. *Environ. Pollut.* 2020, 261, 114158. [CrossRef]
- Zhao, J.; Wang, Y.; Li, B.; Zhang, J.; Guan, D.; Hu, Y.; Fan, H.; Sun, Y.; Wang, H.; Guo, L. New insights into microplastics inhibiting kitchen waste dry digestion: Digestion efficiency, microbial communities and microplastic aging mechanisms. *Chem. Eng. J.* 2025, 508, 160907. [CrossRef]

- Wang, Y.; Zhao, J.; Fu, Z.; Guan, D.; Zhang, D.; Zhang, H.; Zhang, Q.; Xie, J.; Sun, Y.; Wang, D. Innovative overview of the occurrence, aging characteristics, and ecological toxicity of microplastics in environmental media. *Environ. Pollut.* 2024, 346, 123623. [CrossRef]
- 9. PET Bottle Market Report by Capacity (High, Medium, Low), Distribution Channel (Business to Business, Retail), Color (Transparent, Coloured), Technology (Stretch Blow Molding, Injection Molding, Extrusion Blow Molding, Thermoforming, and Others), End-Use (Packaged Water, Carbonated Soft Drinks, Food Bottles & Jars, Non-Food Bottles & Jars, Fruit Juice, Beer, and Others), and Region 2025–2033. Available online: https://www.imarcgroup.com/pet-bottle-manufacturing-plant (accessed on 21 April 2025).
- 10. What We Learned About Recycling by Detecting 40 Billion Waste Objects in 2024. Available online: https://www.greyparrot.ai/resource-hub/blog/2024-recycling-data (accessed on 21 April 2025).
- 11. Single-Use Plastics. Available online: https://environment.ec.europa.eu/topics/plastics/single-use-plastics_en (accessed on 5 March 2025).
- 12. Packaging Waste—EU Rules on Packaging and Packaging Waste. Available online: https://environment.ec.europa.eu/topics/ waste-and-recycling/packaging-waste_en (accessed on 23 April 2025).
- 13. Economic Study of Returnable Refillable PET in the EU Soft Drinks Industry. Available online: https://unesda.eu/wp-content/uploads/2024/04/PwC-Economic-study-of-returnable-refillables-PET_2022.pdf (accessed on 5 March 2025).
- 14. AENOR Certifies the 100% rPET Recycled Plastic Content in the Aguas Danone Bottles. Available online: https://www.en.aenor. com/conocenos/sala-de-informacion-aenor/notas-de-prensa/aenor-certifica-el-contenido-plastico-reciclado-rpet-en-las-botellasde-aguas-danone (accessed on 21 April 2025).
- 15. Coca-Cola in Western Europe Transitions to 100% Recycled Plastic (rPET) Bottles in Two More Markets. Available online: https://www.cocacolaep.com/news-and-stories/coca-cola-in-western-europe-transitions-to-100-recycled-plastic-rpetbottles-in-two-more-markets/ (accessed on 21 April 2025).
- 16. TOMRA Is Ready for the Expansion of the Deposit Return System in Germany from January 2022. Available online: https://www.tomra.com/news-and-media/news/2022/expansion-of-deposit-return-systems-in-germany (accessed on 21 April 2025).
- 17. Deposit-Refund System in Poland. Available online: https://www.gov.pl/web/climate/deposit-refund-system-in-poland (accessed on 21 April 2025).
- Recycling Revelations: Study Calls for "Super Cleaning" Investment Amid rPET Bottle Contamination Concerns. Available online: https://www.packaginginsights.com/news/recycling-revelations-study-calls-for-super-cleaning-investment-amidrpet-bottle-contamination-concerns.html (accessed on 21 April 2025).
- 19. Bach, C.; Dauchy, X.; Chagnon, M.C.; Etienne, S. Chemical compounds and toxicological assessments of drinking water stored in polyethylene terephthalate (PET) bottles: A source of controversy reviewed. *Water Res.* **2012**, *46*, 571–583. [CrossRef]
- 20. MacDonald, W.A. New advances in poly(ethylene terephthalate) polymerization and degradation. *Polym. Int.* **2002**, *51*, 923–930. [CrossRef]
- 21. Whelan, T. Polymer Technology Dictionary; Chapman & Hall: London, UK, 1994; p. 327.
- 22. European Commission . Commission Delegated Decision (EU) 2024/1441 of 11 March 2024 Supplementing Directive (EU) 2020/2184 of the European Parliament and of the Council by Laying Down a Methodology to Measure Microplastics in Water Intended for Human Consumption (Notified Under Document C(2024) 1459). Available online: https://eur-lex.europa.eu/eli/dec_del/2024/1441/oj (accessed on 27 March 2025).
- Hartmann, N.B.; Hüffer, T.; Thompson, R.C.; Hassellöv, M.; Verschoor, A.; Daugaard, A.E.; Rist, S.; Karlsson, T.; Brennholt, N.; Cole, M.; et al. Are We Speaking the Same Language? Recommendations for a Definition and Categorization Framework for Plastic Debris. *Environ. Sci. Technol.* 2019, *53*, 1039–1047. [CrossRef] [PubMed]
- 24. Nisticò, R. Polyethylene terephthalate (PET) in the packaging industry. Polym. Test. 2020, 90, 106707. [CrossRef]
- 25. Pinlova, B.; Nowack, B. From cracks to secondary microplastics—surface characterization of polyethylene terephthalate (PET) during weathering. *Chemosphere* 2024, 352, 141305. [CrossRef] [PubMed]
- 26. Horne, F.; Liggat, J.; MacDonald, W.; Sankey, S. Photo-oxidation of poly(ethylene terephthalate) films intended for photovoltaic backsheet. *J. Appl. Polym. Sci.* 2020, *137*, 48623. [CrossRef]
- 27. Fechine, G.; Rabello, M.; Souto Maior, R.M.; Catalani, L.H. Surface characterization of photodegraded poly(ethylene terephthalate). The effect of ultraviolet absorbers. *Polymer* **2004**, *45*, 2303–2308. [CrossRef]
- 28. Rostampour, S.; Cook, R.; Jhang, S.; Li, Y.; Fan, C.; Sung, L. Changes in the Chemical Composition of Polyethylene Terephthalate under UV Radiation in Various Environmental Conditions. *Polymers* **2024**, *16*, 2249. [CrossRef]
- Makhdoumi, P.; Amin, A.A.; Karimi, H.; Pirsaheb, M.; Kim, H.; Hossini, H. Occurrence of microplastic particles in the most popular Iranian bottled mineral water brands and an assessment of human exposure. *J. Water Process Eng.* 2021, 39, 101708. [CrossRef]
- 30. Oßmann, B.E.; Sarau, G.; Holtmannspötter, H.; Pischetsrieder, M.; Christiansen, S.H.; Dicke, W. Small-sized microplastics and pigmented particles in bottled mineral water. *Water Res.* **2018**, *141*, 307–316. [CrossRef]

- 31. Hagelskjær, O.; Hagelskjær, F.; Margenat, H.; Yakovenko, N.; Sonke, J.E.; Le Roux, G. Majority of potable water microplastics are smaller than the 20 μm EU methodology limit for consumable water quality. *PLoS Water* **2025**, *4*, e0000250. [CrossRef]
- 32. Schymanski, D.; Goldbeck, C.; Humpf, H.U.; Fürst, P. Analysis of microplastics in water by micro-Raman spectroscopy: Release of plastic particles from different packaging into mineral water. *Water Res.* **2018**, *129*, 154–162. [CrossRef]
- Taheri, S.; Shoshtari-Yeganeh, B.; Pourzamani, H.; Ebrahimpour, K. Investigating the pollution of bottled water by the microplastics (MPs): The effects of mechanical stress, sunlight exposure, and freezing on MPs release. *Environ. Monit. Assess.* 2023, 195, 62. [CrossRef] [PubMed]
- 34. World Health Organization. *Dietary and Inhalation Exposure to Nano- and Microplastic Particles and Potential Implications for Human Health;* WHO: Geneva, Switzerland, 2022.
- Boutroy, N.; Pernel, Y.; Rius, J.; Auger, F.; Bardeleben, H.; Cantin, J.; Abel, F.; Zeinert, A.; Casiraghi, C.; Ferrari, A.; et al. Hydrogenated amorphous carbon film coating of PET bottles for gas diffusion barriers. *Diam. Relat. Mater.* 2006, 15, 921–927. [CrossRef]
- 36. Welle, F.; Franz, R. SiOx layer as functional barrier in polyethylene terephthalate (PET) bottles against potential contaminants from post-consumer recycled PET. *Food Addit. Contam.* **2008**, *25*, 788–794. [CrossRef] [PubMed]
- 37. Lu, H.; Diaz, D.J.; Czarnecki, N.J.; Zhu, C.; Kim, W.; Shroff, R.; Acosta, D.J.; Alexander, B.R.; Cole, H.O.; Zhang, Y.; et al. Machine learning-aided engineering of hydrolases for PET depolymerization. *Nature* **2022**, *604*, 662–667. [CrossRef]
- Giraldo-Narcizo, S.; Guenani, N.; Sánchez-Pérez, A.M.; Guerrero, A. Accelerated Polyethylene Terephthalate (PET) Enzymatic Degradation by Room Temperature Alkali Pre-treatment for Reduced Polymer Crystallinity. *ChemBioChem* 2023, 24, 1–6. [CrossRef]
- Gerassimidou, S.; Lanska, P.; Hahladakis, J.N.; Lovat, E.; Vanzetto, S.; Geueke, B.; Groh, K.J.; Muncke, J.; Maffini, M.; Martin, O.V.; et al. Unpacking the complexity of the PET drink bottles value chain: A chemicals perspective. *J. Hazard. Mater.* 2022, 430, 128410. [CrossRef]
- 40. Europe Bottled Water Market Report by Product Type (Still, Carbonated, Flavored, Mineral), Distribution Channel (Supermarkets and Hypermarkets, Convenience Stores, Direct Sales, On-Trade, and Others), Packaging Type (PET Bottles, Metal Cans, and Others), and Country 2025–2033. Available online: https://www.imarcgroup.com/europe-bottled-water-market (accessed on 11 March 2025).
- 41. STATISTICS2022. Available online: https://naturalmineralwaterseurope.org/statistics/ (accessed on 11 March 2025).
- 42. Europe Bottled Water Market Size & Outlook, 2024–2030. Available online: https://www.grandviewresearch.com/horizon/ outlook/bottled-water-market/europe (accessed on 2 February 2025).
- 43. UNU-INWEH. Global Bottled Water Industry: A Review of Impacts and Trends. 2023. Available online: https://collections.unu. edu/eserv/UNU:9106/BottledWater_Report_Final_-compressed.pdf (accessed on 29 April 2025).
- 44. Top Bottled Water Consuming Countries. Available online: https://www.worldatlas.com/articles/top-bottled-water-consuming-countries.html (accessed on 5 February 2025).
- 45. Orset, C.; Barret, N.; Lemaire, A. How consumers of plastic water bottles are responding to environmental policies? *Waste Manag.* **2017**, *61*, 13–27. [CrossRef]
- 46. Benyathiar, P.; Kumar, P.; Carpenter, G.; Brace, J.; Mishra, D.K. Polyethylene Terephthalate (PET) Bottle-to-Bottle Recycling for the Beverage Industry: A Review. *Polymers* **2022**, *14*, 2366. [CrossRef]
- 47. Zheng, Y.; Yanful, E.K.; Bassi, A.S. A review of plastic waste biodegradation. Crit. Rev. Biotechnol. 2005, 25, 243–250. [CrossRef]
- 48. SaveMoneyCutCarbon. Available online: https://www.savemoneycutcarbon.com/learn-save/how-long-does-it-take-for-plastic-to-biodegrade/ (accessed on 30 April 2025).
- 49. Dhaka, V.; Singh, S.; Anil, A.G.; Sunil Kumar Naik, T.S.; Garg, S.; Samuel, J.; Kumar, M.; Ramamurthy, P.; Singh, J. Occurrence, toxicity and remediation of polyethylene terephthalate plastics. A review. *Environ. Chem. Lett.* **2022**, *20*, 1777–1800. [CrossRef]
- 50. Elvall, M.T.; Lundqvist, M.; Kelpsiene, E.; Šileikis, E.; Gunnarsson, S.B.; Cedervall, T. Nanoplastics formed during the mechanical breakdown of daily-use polystyrene products. *Nanoscale Adv.* **2019**, *1*, 1055–1061. [CrossRef]
- 51. Ivleva, N.P.; Wiesheu, A.C.; Niessner, R. Microplastic in Aquatic Ecosystems. *Angew. Chem. Int.* 2017, 56, 1720–1739. [CrossRef] [PubMed]
- 52. Zhang, K.; Hamidian, A.H.; Tubić, A.; Zhang, Y.; Fang, J.K.; Wu, C.; Lam, P.K. Understanding plastic degradation and microplastic formation in the environment: A review. *Environ. Pollut.* **2021**, 274, 116554–116568. [CrossRef] [PubMed]
- 53. Arthur, C.; Baker, J.; Bamford, H. Proceedings of the International Research Workshop on the Occurrence, Effects, and Fate of Microplastic Marine Debris; NOAA Technical Memorandum NOS—OR R—30; NOAA: Silver Spring, MD, USA , 2009; pp. 1–49.
- 54. Aini, S.A.; Syafiuddin, A.; Benet, G.A. The presence of microplastics in air environment and their potential impacts on health. *Environ. Toxicol. Manag.* **2022**, *2*, 31–39. [CrossRef]
- 55. Faure, F.; Saini, C.; Potter, G.; Galgani, F.; de Alencastro, L.F.; Hagmann, P. An evaluation of surface micro- and mesoplastic pollution in pelagic ecosystems of the Western Mediterranean Sea. *Environ. Sci. Pollut. Res.* **2015**, *22*, 12190–12197. [CrossRef]
- 56. Sa'adu, I.; Farsang, A. Plastic contamination in agricultural soils: A review. Environ. Sci. Eur. 2023, 35, 13–24. [CrossRef]

- 57. Alfaro-Nunez, A.; Astorga, D.; Caceres-Farias, L.; Bastidas, L.; Villegas, C.S.; Macay, K.; Christensen, J.H. Microplastic pollution in seawater and marine organisms across the Tropical Eastern Pacific and Galapagos. *Sci. Rep.* **2021**, *11*, 6424–6432. [CrossRef]
- Yang, J.; Monnot, M.; Sun, Y.; Asia, L.; Wong-Wah-Chung, P.; Doumenq, P.; Moulin, P. Microplastics in different water samples (seawater, freshwater, and wastewater): Removal efficiency of membrane treatment processes. *Water Res.* 2023, 232, 119673–119696. [CrossRef]
- 59. Roslan, N.; Lee, Y.; Ibrahim, Y.; Tuan Anuar, S.; Yusof, K.; Lai, L.; Brentnall, T. Detection of microplastics in human tissues and organs: A scoping review. *J. Glob. Health* **2024**, *14*, 04179. [CrossRef]
- 60. Zhang, J.; Wang, L.; Trasande, L.; Kannan, K. Occurrence of Polyethylene Terephthalate and Polycarbonate Microplastics in Infant and Adult Feces. *Environ. Sci. Technol. Lett.* **2021**, *8*, 989–994. [CrossRef]
- 61. Sun, J.; Sui, M.; Wang, T.; Teng, X.; Sun, J.; Chen, X. Detection and quantification of various microplastics in human endometrium based on laser direct infrared spectroscopy. *Sci. Total Environ.* **2024**, *906*, 167760–167770. [CrossRef] [PubMed]
- Liu, S.; Guo, J.; Liu, X.; Yang, R.; Wang, H.; Sun, Y.; Chen, B.; Dong, R. Detection of various microplastics in placentas, meconium, infant feces, breastmilk and infant formula: A pilot prospective study. *Sci. Total Environ.* 2023, *854*, 158699–158709. [CrossRef] [PubMed]
- 63. Zhu, L.; Zhu, J.; Zuo, R.; Xu, Q.; Qian, Y.; AN, L. Identification of microplastics in human placenta using laser direct infrared spectroscopy. *Sci. Total Environ.* **2023**, *856*, 159060–159070. [CrossRef] [PubMed]
- 64. Zhang, D.; Wu, C.; Liu, Y.; Li, W.; Li, S.; Peng, L.; Kang, L.; Ullah, S.; Gong, Z.; Li, Z.; et al. Microplastics are detected in human gallstones and have the ability to form large cholesterol-microplastic heteroaggregates. *J. Hazard. Mater.* **2024**, *467*, 133631–133642. [CrossRef]
- 65. Guo, X.; Wang, L.; Wang, X.; Li, D.; Wang, H.; Xu, H.; Liu, Y.; Kang, R.; Chen, Q.; Zheng, L.; et al. Discovery and analysis of microplastics in human bone marrow. *J. Hazard. Mater.* **2024**, 477, 135266–135277. [CrossRef]
- 66. Xu, H.; Dong, C.; Yu, Z.; Ozaki, Y.; Hu, Z.; Zhang, B.; Yao, W.; Yu, J.; Xie, Y. Detection and analysis of microplastics in tissues and blood of human cervical cancer patients. *Environ. Res.* **2024**, *259*, 119498–119509. [CrossRef]
- 67. Yang, Z.; Wang, M.; Feng, Z.; Wang, Z.; Lv, M.; Chang, J.; Chen, L.; Wang, C. Human Microplastics Exposure and Potential Health Risks to Target Organs by Different Routes: A Review. *Curr. Pollut. Rep.* **2023**, *9*, 468–485. [CrossRef]
- 68. Brown, J.; Zeman, K.; Bennett, W. Ultrafine particle deposition and clearance in the healthy and obstructed lung. *Am. J. Respir. Crit. Care Med.* **2002**, *166*, 1240–1247. [CrossRef]
- 69. Henneberger, A.; Zareba, W.; Ibald-Mulli, A.; Rückerl, R.; Cyrys, J.; Couderc, J.; Mykins, B.; Woelke, G.; Wichmann, H.; Peters, A. Repolarization changes induced by air pollution in ischemic heart disease patients. *Environ. Health Perspect.* **2005**, *113*, 440–446. [CrossRef]
- Fournier, S.B.; D'Errico, J.N.; Adler, D.S.; Kollontzi, S.; Goedken, M.; Fabris, C.; Yurkow, E.J.; Stapleton, P. Nanopolystyrene translocation and fetal deposition after acute lung exposure during late-stage pregnancy. *Part. Fibre Toxicol.* 2020, *17*, 55–66. [CrossRef]
- 71. Medley, E.; Spratlen, M.; Yan, B.; Herbstman, J.; Deyssenroth, M. A Systematic Review of the Placental Translocation of Microand Nanoplastics. *Curr. Environ. Health Rep.* **2023**, *10*, 99–111. [CrossRef] [PubMed]
- 72. Atugoda, T.; Piyumali, H.; Wijesekara, H.; Sonne, C.; Lam, S.; Mahatantila, K.; Vithanage, M. Nanoplastic occurrence, transformation and toxicity: A review. *Environ. Chem. Lett.* **2023**, *21*, 363–381. [CrossRef]
- 73. Li, X.; Li, Z. Perspectives on the Toxic Effects of Micro- and Nanoplastics on the Environment: A Bibliometric Analysis of the 2014 to 2023 Period. *Toxics* 2024, 12, 676. [CrossRef] [PubMed]
- 74. Sawicka, D.; Chojnacka-Puchta, L.; Zapór, L.; Miranowicz-Dzierżawska, K.; Skowroń, J. The exposure routes of micro-and nanoplastics and their potential toxic effects on human health. *Med. Pr. Work. Health Saf.* **2024**, *75*, 81–96. [CrossRef]
- 75. Jones, R.; Wilks, E.; Metanomski, W.; Kahovec, J.; Hess, M.; Stepto, R.; Kitayama, T. *Compendium of Polymer Terminology and Nomenclature: IUPAC Recommendations 2008*; Royal Society of Chemistry: Cambridge, UK, 2009; p. 253. [CrossRef]
- 76. Venkatachalam, S.; Nayak, S.G.; Labde, J.V.; Gharal, P.R.; Rao, K.; Kelkar, A.K. *Degradation and Recyclability of Poly (Ethylene Terephthalate)*; IntechOpen: Rijeka, Croatia, 2012; Chapter 4, pp. 75–98. [CrossRef]
- 77. Roslaniec, Z.; Pietkiewicz, D. Synthesis and Characteristics of Polyester-Based Thermoplastic Elastomers: Chemical Aspects: Sections 1–5; John Wiley & Sons, Ltd.: Hoboken, NJ, USA, 2002; Chapter 13, pp. 579–629. [CrossRef]
- 78. Pirzadeh, E.; Zadhoush, A.; Haghighat, M. Hydrolytic and Thermal Degradation of PET Fibers and PET Granule: The Effects of Crystallization, Temperature, and Humidity. *J. Appl. Polym. Sci.* 2007, *90*, 1544–1549. [CrossRef]
- 79. Sang, T.; Wallis, C.J.; Hill, G.; Britovsek, G.J. Polyethylene terephthalate degradation under natural and accelerated weathering conditions. *Eur. Polym. J.* 2020, 136, 109873. [CrossRef]
- Hurley, C.; Leggett, G. Quantitative investigation of the photodegradation of polyethylene terephthalate film by friction force microscopy, contact-angle goniometry, and X-ray photoelectron spectroscopy. ACS Appl. Mater. Interfaces 2009, 1, 1688–1697. [CrossRef]

- 81. Hurley, R.; Lusher, A.L.; Olsen, M.; Nizzetto, L. Validation of a Method for Extracting Microplastics from Complex, Organic-Rich, Environmental Matrices. *Environ. Sci. Technol.* **2018**, *52*, 7409–7417. [CrossRef]
- 82. Pfeiffer, F.; Fischer, E. Various Digestion Protocols Within Microplastic Sample Processing—Evaluating the Resistance of Different Synthetic Polymers and the Efficiency of Biogenic Organic Matter Destruction. *Front. Environ. Sci.* **2020**, *8*, 572424. [CrossRef]
- Shi, Y.; Zheng, L.; Huang, H.; Tian, Y.; Gong, Z.; Liu, P.; Wu, X.; Li, W.; Gao, S. Formation of Nano- and Microplastics and Dissolved Chemicals During Photodegradation of Polyester Base Fabrics with Polyurethane Coating. *Environ. Sci. Technol.* 2023, 57, 1894–1906. [CrossRef]
- 84. Colnik, M.; Pecar, D.; Knez, Z.; Gorsek, A.; Skerget, M. Kinetics Study of Hydrothermal Degradation of PET Waste into Useful Products. *Processes* **2021**, *10*, 24. [CrossRef]
- 85. Grossetête, T.; Rivaton, A.; Gardette, J.; Hoyle, C.; Ziemer, M.; Fagerburg, D.; Clauberg, H. Photochemical degradation of poly(ethylene terephthalate)-modified copolymer. *Polymer* **2000**, *41*, 3541–3554. [CrossRef]
- 86. Palme, A.; Peterson, A.; de la Motte, H.; Theliander, H.; Brelid, H. Development of an efficient route for combined recycling of PET and cotton from mixed fabrics. *Text. Cloth. Sustain.* **2017**, *3*, 4. [CrossRef]
- McMahon, W.; Birdsall, H.A.; Johnson, G.R.; Camilli, C.T. Degradation Studies of Polyethylene Terephthalate. J. Chem. Eng. Data 1959, 4, 57–79. [CrossRef]
- Hosseini, S.S.; Taheri, S.; Zadhoush, A.; Mehrabani-Zeinabad, A. Hydrolytic degradation of poly(ethylene terephthalate). J. Appl. Polym. Sci. 2007, 103, 2304–2309. [CrossRef]
- 89. Lionetto, F.; Lionetto, M.G.; Mele, C.; Corcione, C.E.; Bagheri, S.; Udayan, G.; Maffezzoli, A. Autofluorescence of Model Polyethylene Terephthalate Nanoplastics for Cell Interaction Studies. *Nanomaterials* **2022**, *12*, 1560. [CrossRef]
- 90. Helmberger, S.M.; Frame, K.M.; Grieshop, M. Counterstaining to Separate Nile Red-Stained Microplastic Particles from Terrestrial Invertebrate Biomass. *Environ. Sci. Technol.* **2020**, *54*, 5580–5588. [CrossRef]
- 91. Majcen, A.; Tassoti, S.; Spitzer, P. Lighting Up for Learning—Fluorescence Analysis of Microplastic Particles by Secondary School Students Using Nile Red. *J. Chem. Educ.* **2023**, *100*, 4007–4012. [CrossRef]
- 92. Maes, T.; Jessop, R.; Wellner, N.; Haupt, K.; Mayes, A.G. A rapid-screening approach to detect and quantify microplastics based on fluorescent tagging with Nile Red. *Sci. Rep.* **2017**, *7*, 44501. [CrossRef]
- 93. Prata, J.C.; Reis, V.; Matos, J.T.; da Costa, J.P.; Duarte, A.C.; Rocha-Santos, T. A new approach for routine quantification of microplastics using Nile Red and automated software (MP-VAT). *Sci. Total Environ.* **2019**, *690*, 1277–1283. [CrossRef]
- 94. Erni-Cassola, G.; Gibson, M.I.; Thompson, R.C.; Christie-Oleza, J.A. Lost, but Found with Nile Red: A Novel Method for Detecting and Quantifying Small Microplastics (1 mm to 20 μm) in Environmental Samples. *Environ. Sci. Technol.* 2017, *51*, 13641–13648. [CrossRef] [PubMed]
- 95. Baruah, A.; Sharma, A.; Sharma, S.; Nagraik, R. An insight into different microplastic detection methods. *Int. J. Environ. Sci. Technol.* 2022, 19, 5721–5730. [CrossRef]
- 96. Lin, X.; Gowen, A.A.; Chen, S.; Xu, J.L. Baking releases microplastics from polyethylene terephthalate bakeware as detected by optical photothermal infrared and quantum cascade laser infrared. *Sci. Total Environ.* **2024**, *924*, 171408. [CrossRef] [PubMed]
- Ma, X.; Pavlidis, G.; Dillon, E.; Beltran, V.; Schwartz, J.J.; Thoury, M.; Borondics, F.; Sandt, C.; Kjoller, K.; Berrie, B.H.; et al. Micro to Nano: Multiscale IR Analyses Reveal Zinc Soap Heterogeneity in a 19th-Century Painting by Corot. *Anal. Chem.* 2022, 94, 3103–3110. [CrossRef] [PubMed]
- 98. Marchetti, A.; Beltran, V.; Nuyts, G.; Borondics, F.; De Meyer, S.; Van Bos, M.; Jaroszewicz, J.; Otten, E.; Debulpaep, M.; De Wael, K. Novel optical photothermal infrared (O-PTIR) spectroscopy for the noninvasive characterization of heritage glass-metal objects. *Sci. Adv.* 2022, *8*, eabl6769. [CrossRef] [PubMed]
- 99. Primpke, S.; Godejohann, M.; Gerdts, G. Rapid Identification and Quantification of Microplastics in the Environment by Quantum Cascade Laser-Based Hyperspectral Infrared Chemical Imaging. *Environ. Sci. Technol.* **2020**, *54*, 15893–15903. [CrossRef]
- 100. Xu, G.; Cheng, H.; Jones, R.; Feng, Y.; Gong, K.; Li, K.; Fang, X.; Tahir, M.A.; Valev, V.K.; Zhang, L. Surface-Enhanced Raman Spectroscopy Facilitates the Detection of Microplastics <1 μm in the Environment. *Environ. Sci. Technol.* 2020, 54, 15594–15603. [CrossRef]
- 101. Prata, J.C.; da Costa, J.P.; Duarte, A.C.; Rocha-Santos, T. Methods for sampling and detection of microplastics in water and sediment: A critical review. *TrAC—Trends Anal. Chem.* **2019**, *110*, 150–159. [CrossRef]
- 102. Dümichen, E.; Eisentraut, P.; Bannick, C.G.; Barthel, A.K.; Senz, R.; Braun, U. Fast identification of microplastics in complex environmental samples by a thermal degradation method. *Chemosphere* **2017**, *174*, 572–584. [CrossRef]
- 103. Rivera-Rivera, D.M.; Quintanilla-Villanueva, G.E.; Luna-Moreno, D.; Sánchez-Álvarez, A.; Rodríguez-Delgado, J.M.; Cedillo-González, E.I.; Kaushik, G.; Villarreal-Chiu, J.F.; Rodríguez-Delgado, M.M. Exploring Innovative Approaches for the Analysis of Micro- and Nanoplastics: Breakthroughs in (Bio)Sensing Techniques. *Biosensors* 2025, 15, 44. [CrossRef]
- 104. Behera, A.; Mahapatra, S.R.; Majhi, S.; Misra, N.; Sharma, R.; Singh, J.; Singh, R.P.; Pandey, S.S.; Singh, K.R.; Kerry, R.G. Gold nanoparticle assisted colorimetric biosensors for rapid polyethylene terephthalate (PET) sensing for sustainable environment to monitor microplastics. *Environ. Res.* 2023, 234, 116556. [CrossRef] [PubMed]

- 105. Marcela Melo Cardozo, I.; Pereira dos Anjos, J.; Oliveira Campos da Rocha, F.; de Andrade, J.B. Exploratory analysis of the presence of 14 carbonyl compounds in bottled mineral water in polyethylene terephthalate (PET) containers. *Food Chem.* 2021, 365, 130475. [CrossRef] [PubMed]
- 106. Soufizadeh, M.; Morello, R.; Ferraro, A.; Spasiano, D.; Fratino, U. Technical and Economic Feasibility Investigation for the Treatment of Microplastic-Contaminated Marine Sediments Through an Environmentally Sustainable Separation Process. *Sustainability* 2025, 17, 1258. [CrossRef]
- 107. Bellasi, A.; Binda, G.; Pozzi, A.; Boldrocchi, G.; Bettinetti, R. The extraction of microplastics from sediments: An overview of existing methods and the proposal of a new and green alternative. *Chemosphere* **2021**, *278*, 130357. [CrossRef]
- 108. European Council. Directive (EU) 2018/852 of the European Parliament and of the Council of 30 May 2018 Amending Directive 94/62/EC on Packaging and Packaging Waste. Available online: https://eur-lex.europa.eu/eli/dir/2018/852/oj (accessed on 2 March 2025).
- 109. European Council. Directive (Eu) 2019/904 of the European Parliament and of the Council of 5 June 2019 on the Reduction of the Impact of Certain Plastic Products on the Environment. Available online: https://eur-lex.europa.eu/eli/dir/2019/904/oj (accessed on 2 March 2025).
- Council Decision (EU, Euratom) 2020/2053 of 14 December 2020 on the System of Own Resources of the European Union and Repealing Decision 2014/335/EU, Euratom. Available online: https://eur-lex.europa.eu/eli/dec/2020/2053/oj (accessed on 2 March 2025).
- 111. Bach, C.; Dauchy, X.; Severin, I.; Munoz, J.F.; Etienne, S.; Chagnon, M.C. Effect of temperature on the release of intentionally and non-intentionally added substances from polyethylene terephthalate (PET) bottles into water: Chemical analysis and potential toxicity. *Food Chem.* 2013, 139, 672–680. [CrossRef]
- 112. Liu, R.; Liao, Z.; Zheng, J.; Wu, X.; Zongyi Tan and, H.O. Characterizing the photodegradation-induced release of volatile organic compounds from bottled water containers. *Eco-Environ. Health* **2024**, *3*, 145–153. [CrossRef]
- 113. Idris, A.; Muntean, A.; Mesic, B.; Lestelius, M.; Javed, A. Oxygen Barrier Performance of Poly(vinyl alcohol) Coating Films with Different Induced Crystallinity and Model Predictions. *Coatings* **2021**, *11*, 1253. [CrossRef]
- 114. Zhong, W.; Yang, X.; Sun, J.; Gao, H.; Bai, Y.; Shao, L. Polyacrylate Decorating Poly(ethylene terephthalate) (PET) Film Surface for Boosting Oxygen Barrier Property. *Coatings* 2021, 11, 1451. [CrossRef]
- 115. Kang, T.; Tang, L.; Qu, J. Preparation and Properties of High Hardness Ultraviolet Curable Polyethylene Terephthalates Surface Coatings Modified with Octavinyl-Polyhedral Oligomeric Silsesquioxane. *Coatings* **2018**, *8*, 411. [CrossRef]
- 116. KHS on the Importance of Innovative PET Bottle Coatings. Available online: https://packagingeurope.com/khs-on-theimportance-of-innovative-pet-bottle-coatings/11048.article (accessed on 26 February 2025).
- 117. Nakaya, M.; Kodama, K.; Yasuhara, S.; Hotta, A. Novel G as Barrier SiOC Coating to PET Bottles through a Hot Wire CVD Method. *J. Polym.* **2016**, 2016, 4657193. [CrossRef]
- 118. Shirakura, A.; Nakaya, M.; Koga, Y.; Kodama, H.; Hasebe, T.; Suzuki, T. Diamond-like carbon films for PET bottles and medical applications. *Thin Solid Film.* **2006**, 494, 84–91. [CrossRef]
- 119. Cheng, D.; Cai, G.; Wu, J.; Ran, J.; Wang, X. UV protective PET nanocomposites by a layer-by-layer deposition of TiO2 nanoparticles. *Colloid Polym. Sci.* 2017, 295, 2163–2172. [CrossRef]
- Kocayavuz, O.; Demirel, B.; Yaraş, A.; Akkurt, F.; Daver, F. A way to enhance the mechanical performance and uv visible-light barrier of polyethylene terephthalate packaging material: Synthesis and application of takedaite (ca3b2o6). *Polym. Adv. Technol.* 2022, 33, 3359–3367. [CrossRef]
- Inaner, N.B.; Demirel, B.; Yaras, A.; Akkurt, F.; Daver, F. Improvement of environmental stress cracking performance, load-carrying capacity, and UV light barrier property of polyethylene terephthalate packaging material. *Polym. Adv. Technol.* 2022, 33, 2352–2361. [CrossRef]
- 122. Jung, H.; Shin, G.; Kwak, H.; Hao, L.T.; Jegal, J.; Kim, H.J.; Jeon, H.; Park, J.; Oh, D.X. Review of polymer technologies for improving the recycling and upcycling efficiency of plastic waste. *Chemosphere* **2023**, *320*, 138089. [CrossRef]
- 123. Pinter, E.; Welle, F.; Mayrhofer, E.; Pechhacker, A.; Motloch, L.; Lahme, V.; Grant, A.; Tacker, M. Circularity Study on PET Bottle-To-Bottle Recycling. *Sustainability* **2021**, *13*, 7370. [CrossRef]
- 124. Brouwer, M.T.; Chacon, F.A.; van Velzen, E.U.T. Effect of recycled content and rPET quality on the properties of PET bottles, part III: Modelling of repetitive recycling. *Packag. Technol. Sci.* **2020**, *33*, 373–383. [CrossRef]
- 125. Tanasupawat, S.; Takehana, T.; Yoshida, S.; Hiraga, K.; Oda, K. Ideonella sakaiensis sp. nov., isolated from a microbial consortium that degrades poly(ethylene terephthalate). *Int. J. Syst. Evol. Microbiol.* **2016**, *66*, 2813–2818. [CrossRef]
- 126. Wallace, N.E.; Adams, M.C.; Chafin, A.C.; Jones, D.D.; Tsui, C.L.; Gruber, T.D. The highly crystalline PET found in plastic water bottles does not support the growth of the PETase-producing bacterium Ideonella sakaiensis. *Environ. Microbiol. Rep.* 2020, 12, 578–582. [CrossRef]
- 127. Thomsen, T.B.; Almdal, K.; Meyer, A.S. Signifiance of poly(ethylene terephthalate) (PET) substrate crystallinity on enzymatic degradation. *New Biotechnol.* 2023, 78, 162–172. [CrossRef]

- 128. Son, H.F.; Cho, I.J.; Joo, S.; Seo, H.; Sagong, H.Y.; Choi, S.Y.; Lee, S.Y.; Kim, K.J. Rational Protein Engineering of Thermo-Stable PETase from Ideonella sakaiensis for Highly Efficient PET Degradation. *ACS Catal.* **2019**, *9*, 3519–3526. [CrossRef]
- 129. Tournier, V.; Topham, C.M.; Gilles, A.; David, B.; Folgoas, C.; Moya-Leclair, E.; Kamionka, E.; Desrousseaux, M.-L.; Texier, H.; Gavalda, S.; et al. An engineered PET depolymerase to break down and recycle plastic bottles. *Nature* 2020, 580, 216–219. [CrossRef] [PubMed]
- 130. Gupta, A.; Agrawal, S. Machine learning-based enzyme engineering of PETase for improved efficiency in plastic degradation. *J. Emerg. Investig.* **2023**, *6*, 1–10. [CrossRef]
- 131. Mrigwani, A.; Thakur, B.; Guptasarma, P. Conversion of polyethylene terephthalate into pure terephthalic acid through synergy between a solid-degrading cutinase and a reaction intermediate-hydrolysing carboxylesterase. *Green Chem.* **2022**, *24*, 6707–6719. [CrossRef]
- Li, X.; Liu, X.; Zhang, J.; Chen, F.; Khalid, M.; Ye, J.; Romantschuk, M.; Hui, N. Hydrolase and plastic-degrading microbiota explain degradation of polyethylene terephthalate microplastics during high-temperature composting. *Bioresour. Technol.* 2024, 393, 130108. [CrossRef]
- 133. Guo, B.; Lopez-Lorenzo, X.; Fang, Y.; Bäckström, E.; Capezza, A.J.; Vanga, S.R.; Furó, I.; Hakkarainen, M.; Syrénl, P. Fast Depolymerization of PET Bottle Mediated by Microwave Pre-Treatment and An Engineered PETase**. *ChemSusChem* 2023, 16, e202300742. [CrossRef]
- 134. Joseph, T.M.; Azat, S.; Ahmadi, Z.; Jazani, O.M.; Esmaeili, A.; Kianfar, E.; Haponiuk, J.; Thomas, S. Polyethylene terephthalate (PET) recycling: A review. *Case Stud. Chem. Environ. Eng.* **2024**, *9*, 100673. [CrossRef]
- 135. Cao, F.; Wang, L.; Zheng, R.; Guo, L.; Chena, Y.; Qian, X. Research and progress of chemical depolymerization of waste PET and high-value application of its depolymerization products. *RSC Adv.* **2022**, *12*, 31564. [CrossRef]
- 136. Guo, Z.; Wu, J.; Wang, J. Chemical degradation and recycling of polyethylene terephthalate (PET): A review. *RSC Sustain.* 2025, *3*, 2111–2133. [CrossRef]
- 137. Mahler, A.S.H.; Lemming, M.; Jaime-Azuara, A.; Pedersen, T.H.; Hinge, M. Chemical recycling of polymer contaminated poly(ethylene terephthalate) by neutral hydrolysis. *Waste Manag.* **2025**, *192*, 12–19. [CrossRef]
- Cho, M.H.; Song, Y.J.; Rhu, C.J.; Go, B.R. Pyrolysis Process of Mixed Microplastics Using TG-FTIR and TED-GC-MS. *Polymers* 2023, 15, 241. [CrossRef] [PubMed]
- 139. Wu, J.C.; Chang, J.Y.; Yuan, X.; Khan, E.; Ok, Y.S.; Hou, C.H. Upcycling waste polyethylene terephthalate (PET) bottles into high-performance activated carbon for electrochemical desalination. *Chemosphere* **2024**, *364*, 143029. [CrossRef] [PubMed]
- 140. Chan, K.; Zinchenko, A. Conversion of waste bottles' PET to a hydrogel adsorbent via PET aminolysis. *Chemosphere* **2021**, *9*, 106129. [CrossRef]
- 141. Ahmadinia, E.; Zargar, M.; Karim, M.R.; Abdelaziz, M.; Ahmadinia, E. Performance evaluation of utilization of waste Polyethylene Terephthalate (PET) in stone mastic asphalt. *Constr. Build. Mater.* **2012**, *36*, 984–989. [CrossRef]
- 142. Veropalumbo, R.; Oreto, C.; Viscione, N.; Pirozzi, F.; Pontoni, L.; Trancone, G.; Race, M.; Russo, F. Exploring the effect on the environment of encapsulated micro- and nano-plastics into asphalt mastics for road pavement. *Environ. Res.* 2023, 216, 114466. [CrossRef]
- 143. Guo, Y.; Xia, X.; Ruan, J.; Wang, Y.; Zhang, J.; LeBlanc, G.A.; An, L. Ignored microplastic sources from plastic bottle recycling. *Sci. Total Environ.* **2022**, *838*, 156038. [CrossRef]
- 144. Habib, R.Z.; Thiemann, T.; Kendi, R.A. Microplastics and Wastewater Treatment Plants—A Review. J. Water Resour. Prot. 2020, 12, 1–35. [CrossRef]
- 145. Krishnan, R.Y.; Manikandan, S.; Subbaiya, R.; Karmegam, N.; Kim, W.; Govarthanan, M. Recent approaches and advanced wastewater treatment technologies for mitigating emerging microplastics contamination—A critical review. *Sci. Total Environ.* 2023, 858, 159681. [CrossRef]
- 146. Talvitie, J.; Mikola, A.; Koistinen, A.; Setala, O. Solutions to microplastic pollution—Removal of microplastics from wastewater effluent with advanced wastewater treatment technologies. *Water Res.* **2017**, *123*, 401–407. [CrossRef]
- 147. Cho, S.J.; Choi, J.H.; Yoon, Y.S.; Um, N.I. Microplastic Pollution in Sewage Sludge from Wastewater Treatment Plants and Estimation of Microplastic Release. *Water* 2025, *17*, 387. [CrossRef]
- 148. Iyare, P.U.; Ouki, S.K.; Bond, T. Microplastics removal in wastewater treatment plants: A critical review. *Environ. Sci. Water Res. Technol.* 2020, *6*, 2664–2675. [CrossRef]
- 149. Cao, Z.; Xia, W.; Wu, S.; Ma, J.; Zhou, X.; Qian, X.; Xu, A.; Dong, W.; Jiang, M. Bioengineering Comamonas testosteroni CNB-1: A robust whole-cell biocatalyst for efcient PET microplastic degradation. *Bioresour. Bioprocess.* **2023**, *10*, 94. [CrossRef] [PubMed]
- 150. Wilkes, R.A.; Zhou, N.; Carroll, A.L.; Aryal, O.; Teitel, K.P.; Wilson, R.S.; Zhang, L.; Kapoor, A.; Castaneda, E.; Guss, A.M.; et al. Mechanisms of Polyethylene Terephthalate Pellet Fragmentation into Nanoplastics and Assimilable Carbons by Wastewater Comamonas. *Environ. Sci. Technol.* 2024, *58*, 19338–19352. [CrossRef] [PubMed]
- 151. Sun, J.; Dai, X.; Wang, Q.; van Loosdrecht, M.C.; Ni, B.J. Microplastics in wastewater treatment plants: Detection, occurrence and removal. *Water Res.* 2019, 152, 21–37. [CrossRef]

- 152. Lares, M.; Ncibi, M.C.; Sillanpaa, M.; Sillanpaa, M. Occurrence, identification and removal of microplastic particles and fibers in conventional activated sludge process and advanced MBR technology. *Water Res.* **2018**, *133*, 236–246. [CrossRef]
- 153. European Council. Directive (EU) 2024/3019 of the European Parliament and of the Council of 27 November 2024 Concerning Urban Wastewater Treatment. Available online: https://eur-lex.europa.eu/eli/dir/2024/3019/oj (accessed on 11 March 2025).

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