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Review

Review of Microplastic Distribution, Toxicity, Analysis Methods, and Removal Technologies

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Abstract: Microplastic contamination has become a problem, as plastic production has increased worldwide. Microplastics are plastics with particles of less than 5 mm and are absorbed through soil, water, atmosphere, and living organisms and finally affect human health. However, information on the distribution, toxicity, analytical methods, and removal techniques for microplastics is insufficient. For clear microplastic analytical methods and removal technologies, this article includes the following: (1) The distribution and contamination pathways of microplastics worldwide are reviewed. (2) The health effects and toxicity of microplastics were researched. (3) The sampling, pretreatment, and analytical methods of microplastics were all reviewed through various related articles. (4) The various removal techniques of microplastics were categorized by wastewater treatment process, physical treatment, chemical treatment, and biological treatment. This paper will be of great help to microplastic analysis and removal techniques.

Keywords: microplastic distribution; toxicity; sampling methods; pretreatment methods; analytical methods; removal technology

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1. Introduction

In 2018, global production of plastic products reached almost 360 million metric tons/year [1]. Out of the 2.5 billion metric tons of solid waste generated by 192 countries in 2010, about 275 Mt consisted of plastics [2]. Once a large plastic item is thrown away in nature, it degrades into smaller plastics and it becomes microplastics (MPs). Microplastics are small plastic particles smaller than 5 mm, and plastic particles smaller than 1 µm are defined as nanoplastics [3–5]. Microplastic particles exist in water, are consumed by living things, and can affect human health. Currently, microplastics are detected in various areas, such as streams, rivers, seas, drinking water, and even food in the world [5]. Microplastics can be divided into two parts: primary microplastics and secondary microplastics [6]. Primary microplastic is defined as plastic made of 5 mm or less according to a specific purpose, and it is often included in toothpaste, face wash, cosmetics, industrial abrasives [7,8], and 3D-printer particles [9]. Secondary microplastics were large at the time of production and manufacturing, but the large plastics were crushed or degraded through physical, chemical, and biological weathering, due to the environment, and became microplastics, such as straw foam and mulching vinyl [8]. As such, pollution from microplastics are becoming a serious problem around the world, and many studies on microplastics such as distribution, toxicity, analysis, and removal are being conducted by many researchers. Among the 192 countries of the world, 44 countries have carried out research regarding microplastics; the studies looking at the impacts on organisms have mostly targeted fish (38%), whereas few studies on other highly affected organisms, such as turtles (1%), have been conducted [10].

Commonly used plastics are PET (polyethylene terephthalate), PU (polyurethane), PS (polystyrene), PVC (polyvinylchloride), PP (polypropylene), polyester, PE (polyethylene), and PA (polyamide, nylon) [11]. Plastics produced in 2019 are PE 27.2%, PP 19.3%, PVC

10%, PA 7.9%, PET 7.7%, PS 6.4%, and others 19% [1]. Among these, the main materials of microplastics are polyethylene (PE), polypropylene (PP), and polystyrene (PS), and polyester, acrylic [11,12]. Polyamide (PA) is often found on the beach as secondary microplastics [12]. Plastics, as well as various additives to improve the functionality in the manufacture of plastics, should be checked. Plastic additives, including plasticizers, flame retardants, antioxidants, acid scavengers, light and heat stabilizers, lubricants, pigments, and antistatic agents, can play a role in improving the functional properties of plastics. Representative plastic additives include bisphenols [13,14], phthalates [15], and adipates, and it is necessary to confirm the toxicity and removal method of the additives.

The main sources of microplastics in flash water are from domestic and industrial sewage, sea littering, and runoff water [16]. Household sewage contains many microplastic particles in cosmetics and detergents used in everyday life, which is mainly introduced into sewage when washing faces or showers [17]. Industrial sewage mainly flows from the plastics industry, such as plastic pellets and abrasives used in the manufacture of plastic products [18]. Many countries around the world treat their sewage using adequate water treatment methods. The sewage treatment plant removes many microplastics contained in sewage, but nano-sized microplastics still remains in the treated sewage water [18–22]. Therefore, microplastics are continuously discharged into the surface water by domestic, industrial, and agricultural processes [16–18]. In Wuhan, China, the concentration of microplastics generated from surface water samples of 20 urban lakes and urban reaches of Hanjiang River and Yangtze River of Wuhan was found to be 1600 ± 639 to 8925 ± 1591 MPs/m^3, and most of them were identified by PET and PP [23]. Microplastics exposed to surface water are mainly introduced into the sea through rivers or streams and accumulate downstream or in sediments, mainly showing a high degree of microplastic pollution [18,24]. In seawater, plastic fragments are worn out by extreme environments, such as waves and salinity, and are absorbed in various ways by marine creatures and salt, etc., and are finally swallowed by humans [24,25].

Microplastics are an environmental problem that only occurs through humans, and it has spread widely until it becomes a threat to our lives. Compared to the microplastic pollution situation, the distribution, toxicity, analysis method, removal method, and policy of microplastic are not clearly established. Accordingly, in this paper, research in various fields related to microplastics was reviewed on distribution, toxicity, analysis method, and removal method.

2. Microplastic Distribution

Although it is not possible to know the exact contamination aspect of microplastics, it has been continuously reported to be detected in seawater, freshwater, food, air, and soil. Moreover, it is predicted that the contamination of microplastics will become more serious in the future by several studies [26–28]. A global estimate of plastic emissions from rivers into the world's oceans is between 1.15 and 2.41 million tons per year, and most of the river plastic input is coming from Asian countries with rapid economic development and poor waste management [29]. The concentration of microplastics is expected to be 250 mg/m^3 in 2016 and 500 mg/m^3 in 2030, and it is expected to exceed 1000 mg/m^3, which is four times the 2016 amount, by 2060 [28]. As shown in Figure 1, microplastics are generated in various forms, such as agricultural wastewater, industrial wastewater, litter, sewage treatment plant, household personal products, road runoff, fishing waste, and atmosphere decomposition [24]. Figure 1 shows the source of microplastic contamination. Microplastics in water can be caused by all areas of human life, including agriculture, industry, landfill, household, and roads. Because microplastics are introduced through various sources and routes, it is difficult to establish a clear removal plan. Figure 2 shows the path of contamination and distribution of microplastics in the sea. Microplastics in the ocean, through rivers, land, and air, are firstly absorbed by marine animals and plants. Human digests various marine organisms (e.g., fish, salt, clam, seaweed, etc.) containing microplastics.

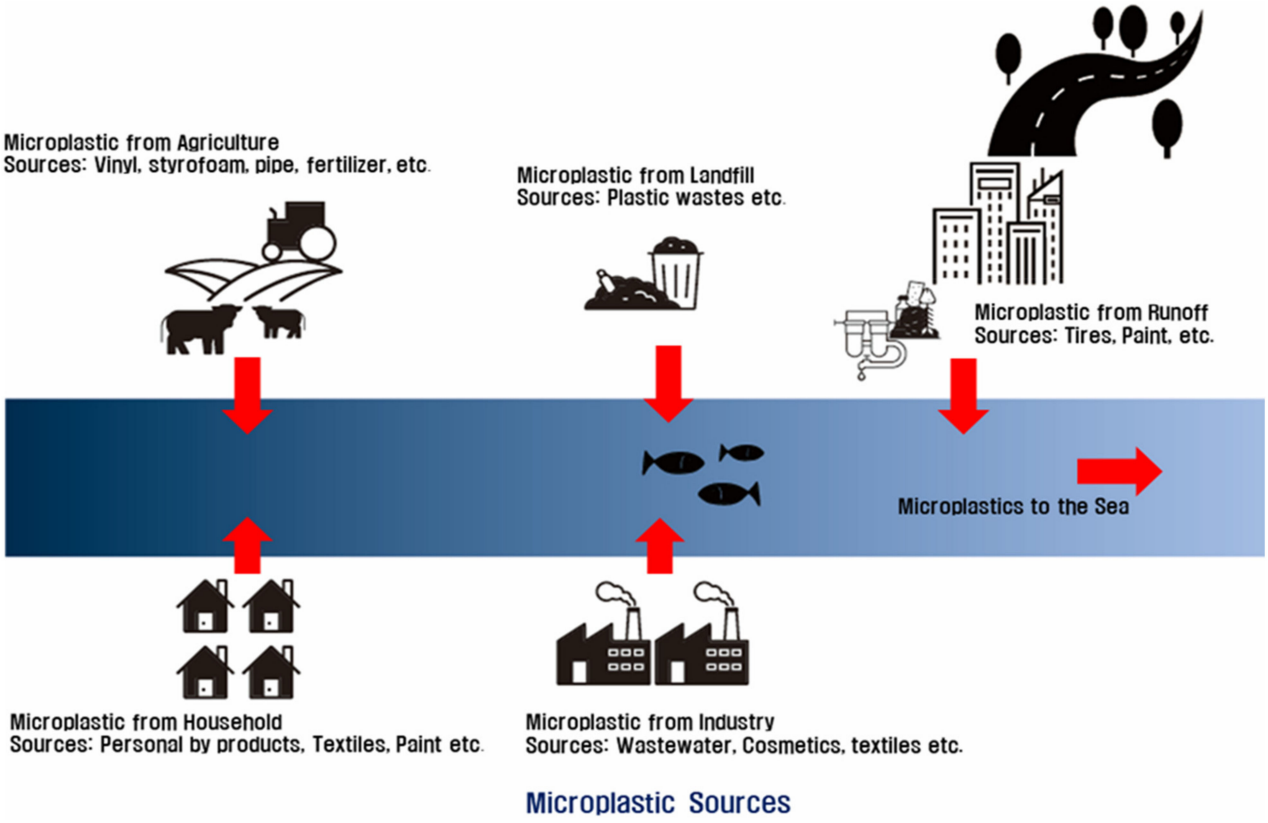

Figure 1. Microplastic-source contamination pathway.

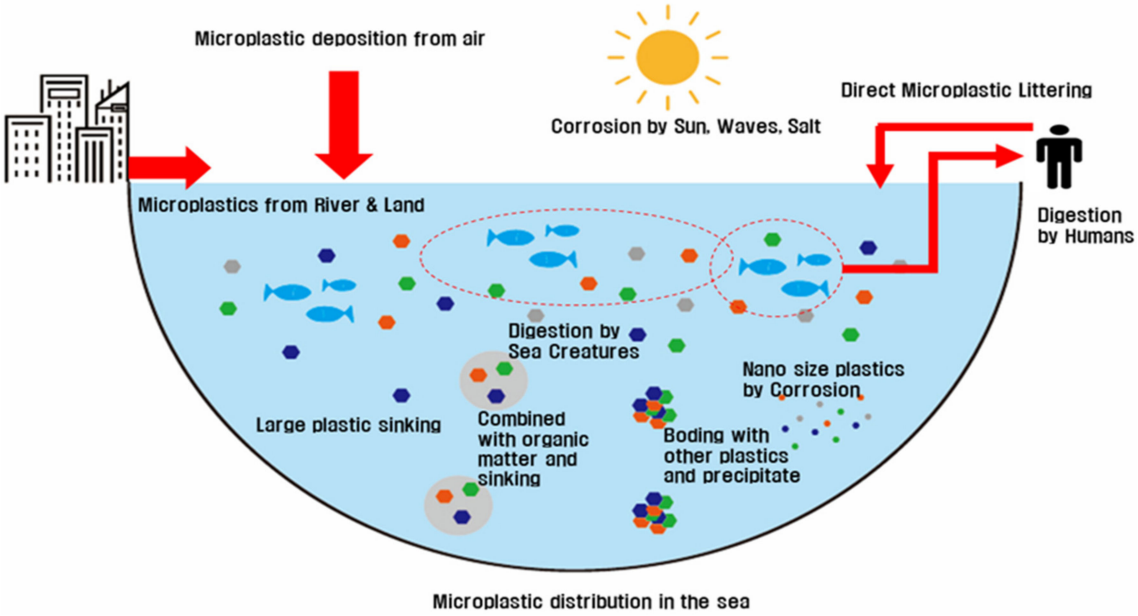

Figure 2. Microplastic contamination distribution pathway in the sea.

Figure 3 is showing the contamination degree of plastics in the world. HYCOM is used by predicting the size of plastics based on the world's oceans from 2007 to 2013 as 0.33–1.00 mm, 1.01–4.75 mm, 4.76–200 mm, and >200 mm [26]. Model prediction of global count density (pieces km^{-2}; see color bar) for each of four size classes (0.33–1.00 mm, 1.01–4.75 mm, 4.76–200 mm, and >200 mm). [30]

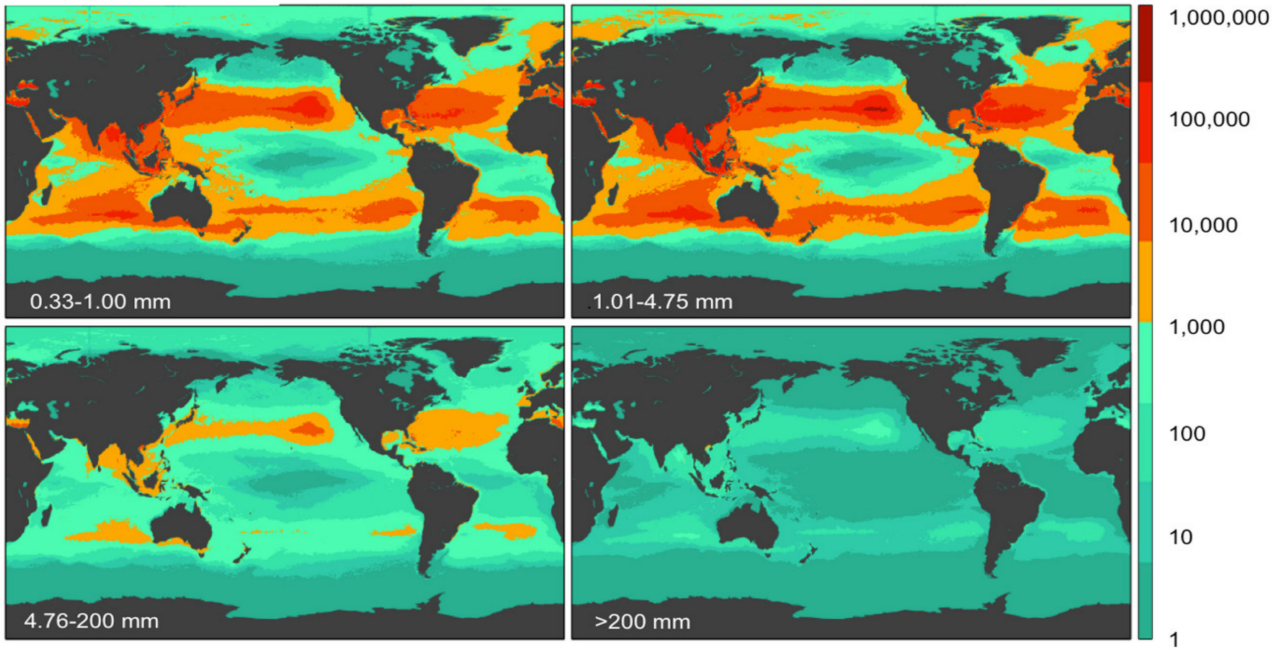

Figure 3. Model results for global count density in four size classes [30].

2.1. Microplastic Distribution in Marine

Research on microplastics started with the ocean contamination in early 2014. Bakir et al. (2014) concluded that circulated contamination zones are easily formed in semi-enclosed seas, such as the Mediterranean coast [31]. Since 2014, many studies on microplastics in the ocean have confirmed the number of microplastic particles in surface water. Research on microplastic pollution in the ocean has mainly focused on the number of microplastic particles in the seawater layer, especially surface water. Iosobe et al. (2015) investigated the concentration of microplastics (<5 mm in size) and mesoplastics (>5 mm in size) in the East Asian sea, around Japan, in 2014, and the total concentration was 1,720,000 MPs/km^2, which is higher than 63,320 MPs/km^2 in the world oceans and 63,320 MPs/km^2 in the North Pacific Ocean [32]. Ressell and Webster (2021) collected surface-sea-level samples from the Scottish seas from 2014 to 2020, using a neuston net, and the concentrations of microplastics ranged from 0 to 91,128 MPs/km^2, and 50 percent of the fragmented plastics were identified as microplastics. Due to the different geographic and temporal ranges of the data, trends in microplastics could not be identified [33]. Pan et al. (2018) collected microplastics from 18 sites in the Western North Pacific Ocean from August to September 2017 and found an average of 1×10^4 MPs/km^2. The MP type was identified as polyethylene (57.8%) > polypropylene (36.0%) > nylon (3.4%) [34]. Among these studies, there are not enough cases in which the total amount of microplastics on the surface of seawater is comparatively analyzed by region. In addition, among the particle sizes in reported microplastic studies, few cases of 0.3 mm or less size have been reported. The reason is that most of the sampling methods for the seawater layer used a mesh size of 333 μm [34,35]. Lindeque et al. (2020) hypothesized that most of the grids used for sampling in the marine sector are 333 μm in size, which may lead to an underestimation of microplastics. They checked the concentration of microplastics in each sample after collecting samples, using 100, 333, and 500 μm nets at the same sampling site. As a result, it was confirmed that, under the statistical significance level, the sample using the grid mesh of 100 μm contained 2.5 and 10 times more microplastics, respectively, than the samples using the grid mesh of 333 and 500 μm [35]. These study results confirmed that there is a limit to the scope of microplastic collection depending on the size of the mesh used for sampling in the microplastic pollution research.

Research on plastic trapping in marine coastal vegetation ecosystems is still in its early stage. Jones et al. (2020) investigated the behavior of microplastics in intertidal and subtidal zones planted with marine plants, such as seagrasses. This was known as

the first study on *Z. marina* to identify microplastics [36]. Piarulli et al. (2020) confirmed the presence of microplastics in salt marshes and 96% of the 330 samples analyzed did not contain microplastics. Suspension and facultative deposit-feeding bivalves (0.5–3%) contains a little compared with omnivores (95%). Although the incidence of MPs was low, the distribution of MP size, shape, and polymer was varied [25]. More studies investigated the presence of microplastics in vegetation, seagrasses and algae in the canopy [37,38]. Cozzolino et al. (2020) divided two intertidal zones and two subtidal zones to determine the degree of microplastic contamination in vegetation developed in Ria Formosa lagoon in Portugal. The microplastic abundance in the sediment was assessed. According to the results of the investigation, the entrapment effect of microplastics in the sediment and crown layer was higher in the subtidal zone than in the intertidal vegetation zone, and the presence or absence of vegetation did not affect the microplastic concentration [39]. As the current research focuses on only one type of vegetation zone, there is a limit to ascertaining the degree of contamination of microplastics according to the vegetation characteristics.

As research on microplastics in the marine sector expands and research cases are accumulated, additional research on the occurrence, distribution, and characteristics of microplastics considering climate and spatial differences in the marine environment is needed.

2.2. Microplastic Distribution in Land

The research on microplastic contamination on land has been relatively insufficient compared to the ocean research. Research studies on freshwater and wastewater treatment plants, using water as a medium among terrestrial environments, are evaluated to be relatively numerous compared to soil and air. Tibbetts et al. (2018) conducted a microplastic survey from a heavily urbanized catchment, the River Tame, and four of its tributaries, which flow through the city of Birmingham, UK [40]. All sediment sampled was found to contain microplastics with an average abundance of 165 MPs/kg [40]. Park (2020) investigated the distribution of microplastics in surface water, fish, and sediment near a sewage treatment plant (STP) in the Tanchon stream, one of the main tributaries flowing into the Han River, Korea, and they found that microplastics concentrations in water varied spatially and temporarily, ranging between 5.3 and 87.3 MPs/m^3 (31.4 $\pm$ 28.5 MPs/m^3). In fish, the concentration in upstream and downstream sites was 7.3 $\pm$ 7.3 MPs/ kg and 12.4 $\pm$ 17.9 MPs/kg. In sediment, microplastic concentration in upstream and downstream sites was 493.1 $\pm$ 136.0 MPs/kg and 380.0 $\pm$ 144.2 MPs/kg [41]. Kosuth et al. (2018) tested the presence of anthropogenic particles in 159 samples of globally sourced tap water, 12 brands of Laurentian Great Lakes beer, and 12 brands of commercial sea salt. A total of 81% were found to contain anthropogenic particles in the tap water. They indicated that the average person ingests over 5800 particles of synthetic debris from these three sources annually, with the largest contribution coming from tap water (88%) [42]. Mason et al. (2018) investigated eleven globally sourced brands of bottled water, purchased in 19 locations in nine different countries, and found that 93% of them contained microplastic synthetic polymer particles among the 259 total bottles [43].

Recent studies have highlighted the discussion that freshwater serves as a major route and source of microplastics between land and ocean [44–46]. Freshwater serves as a major route and source of microplastics transport between land and ocean. Ockelford et al. (2020) pointed out that up to 80% of plastic in the oceans comes from river networks, especially during periods of flooding [44]. Galloway et al. (2017) found that most of microplastics are accumulated in the ocean, and that land is one of the primary sources of microplastics in the ocean [45]. Therefore, microplastic pollution by freshwater is highly dependent on human activity, and factors such as population density and industrialization in the river area can be a major factor affecting marine microplastic pollution [46]. Newly noteworthy research resulted in terrestrial freshwater research that rainfall, such as river overflow or heavy rains due to flooding, affected the movement of microplastics on land [47]. Lebreton et al. (2017) observed that between 1.15 million and 2.41 million tons of plastic waste currently enters the ocean every year from rivers, with over 74% of emissions occurring between

May and October [29]. Hitchcock (2020) emphasized the effect of storms on microplastic pollution in aquatic ecosystems. He measured the abundance of microplastics during the storm and found that the abundance of microplastics was more than 40 times higher during the storm than before the storm. The maximum microplastic abundance in this study was about 17,833 particles/m^3 during the peak of the storm. During the two days of heavy rain, the concentration of microplastics increases from 400 particles/m^3 before the storm to a maximum of 17,383 particles/m^3. The study results may be influenced by microplastics present in the atmosphere, or it may be influenced by the concentration of microplastics in the ocean at the point of occurrence of heavy rain clouds [48]. However, it is difficult to estimate the specific route and source based only on the research. Accordingly, freshwater is more accessible to humans than the ocean, so if it is adjacent to an industrialized or urbanized area, a higher degree of microplastic pollution than the ocean might appear.

The migration of microplastics through the soil has been attributed to the recycling of sewage sludge [49], the use of plastic mulching vinyl [50], the use of organic fertilizers [51], and land application from wastewater treatment plant discharges [41], and other irrigation methods using contaminated river water [52,53] have contributed to the transfer and accumulation of microplastics in agricultural and natural soils. Studies on land contamination of microplastics include a study on open land in an industrial area [54], a study on the concentration of microplastics in soil samples in an open land of flood area in Switzerland [55], and a study on mixed agricultural land [56]. Microplastics, which are confirmed at a high rate, are found more in sludge than in treated wastewater, so reuse of sludge can serve as a source of microplastics in soil. Corradini et al. (2019) confirmed the presence of microplastics in land-use areas which are closely related to human activities. In relation to agricultural soil, research continues to be published that soil is a major storage area for microplastics due to the use of fertilizers recycled from wastewater treatment plant sludge [56]. Regarding the distribution of microplastics in soil, Dioses-Salinas et al. (2020) explained similarities in the morphological type and size of the microplastics in terms of the product purpose. They considered that the characteristics of the source of microplastics, the topography of the surrounding area, hydrological properties, the presence of currents, and the extraction and identification methods used in the survey analysis could affect the difference in microplastic concentrations in individual surveys [52]. Sediment is a particle accumulated in the environment by a medium, and it is difficult to explain it independently as soil [57]. In this regard, sediment should be considered as the main environment in the soil sector, considering the characteristics of the medium of solid particles [57]. Harris (2020) also argues that plastic particles are a solid and transportable material, so it is desirable to look at them as one of the sediments physically [57]. To summarize the above content, microplastic research is being actively conducted in the soil field, especially on agricultural land. Since sludge deposited with microplastics in wastewater treatment facilities can diffuse into the soil, it is necessary to check the use of sludge recycling fertilizers used in agricultural land.

2.3. Microplastic Distribution in Air

As freshwater is considered to be a major part of the migration of microplastics to the ocean, studies on the degree of microplastic pollution in the air field are insufficient compared to the water and sediment fields. However, light microplastics can easily be transported through the air to other environments on land and in the ocean. Yukioka et al. (2020) investigated the abundance of microplastics in road dust in three cities to identify the occurrence and characteristics of microplastics in surface road dust in Kusatsu City, Japan; Danang City, Vietnam; and Kathmandu City, Nepal [58]. The abundance of microplastics among road dust in the three cities surveyed was 0.10 to 39.6 MPs/m^2, and the color, shape, and polymer type of microplastics were all different by region. The proportion of rubber-like microplastics was higher in the dust samples taken from the road than in the samples extracted from the dust present in the general environment of the surveyed areas [58]. It can be inferred that vehicle tires, hoses, shoes, and electrical insulation may be potential

microplastic sources of road dust [59]. These findings are also consistent with the findings of Vogelsang et al. (2018), who estimated that the main source of microplastics from road dust would include the surface wear of automobile tires and paint for pavement road markings [60]. However, detailed information to explain the occurrence and distribution of microplastics in road dust in this study was lacking, and it was difficult to specify the source of microplastics in road dust. In order to explain the occurrence of microplastics distributed in the air and to estimate the distribution and pollution level of microplastics by region or environmental characteristics, more research investigations should be conducted. In the study of Liu et al. (2019), the condition of microplastics among indoor and outdoor dust in 39 cities in China was investigated and the daily human exposure due to indoor dust intake was estimated. They determined that the indoor dust concentration was 1550 to 120,000 mg/kg, the outdoor dust concentration was 212 to 9020 mg/kg, and the daily human exposure by indoor dust intake was 6480 ng/kg-bw/day, estimated to be 17,300 ng/kg-bw/day of PET MPs in children [61]. To summarize, it is only possible to confirm that air is one of the pathways contributing to the movement and diffusion of microplastics. Accordingly, more research should be conducted in order to identify the clear source of origin, migration route, spatial distribution characteristics, and toxicity to the human body.

Microplastic contamination is being studied in all areas of the environment as it includes most parts, such as water, soil, and air. Eriksen et al. (2018) provided sources and potential mitigation measures, as shown in Table 1 [62].

Table 1. Sources, measurements, and strategies for upstream mitigation of microplastics [62].

Category	Source	Potential Mitigation
Production	Microplastics in additives	Removing them from products. Replace with benign alternatives
	Mismanaged preproduction pellets	Regulate pellet handling. Operation clean sweep
Commerce	Industrial abrasives	Improve containment and recovery and require alternatives
	Laundromat exhaust	Improved filtration
	Agriculture-degraded film, pots, and pipes	Improve recovery, biodegradable plastics
Consumer	Tire dust	Technological advances, road surface
	Littering of small plastic items (cigarette filters, torn corners of packaging, small film wrappers, etc.)	Enforcement of fines for littering, Consumer education, EPR on design
	Domestic laundry. Wastewater effluent	Wash with top-load machines, wastewater containment, single-filter woven textiles, textile coatings
Waste management	Fragmentation by vehicles driving over unrecovered waste	Improved waste management
	UV and chemically degraded terrestrial plastic waste	Improved waste management
	Sewage effluent (synthetic fibers)	Laundry filtration, textile industry innovation
	Combined sewage overflow (large items)	Infrastructure improvement
	Mechanical shredding of roadside waste during regular cutting of vegetation (mostly grass)	Better legislation and law enforcement; valorization of waste products

3. Microplastic Toxicity

Microplastics are themselves a problem, but organic contaminants adsorbed on microplastics can also be a problem [23]. The recent research on microplastic toxicity has begun considering the mixed toxicity with other substances. Zhou et al. (2020) studied that microplastics and pharmaceutical and personal care products (PPCPs) could be toxic to organisms when they were exposed in a mixed form [63]. Microplastics could play a role of carrier to mediate PPCPs. Weber et al. (2021), mixed toxicity experiments of microplastics and copper were performed on *Lymnaea stagnalis* (water snails) and showed no change in toxicity [64]. Gao et al. (2019) checked the capability and characteristics of heavy metals on microplastics in marine environment and found that polyvinyl chloride, polypropylene, polyethylene, polyamides and polyformaldehyde could adsorb lead, copper and cadmium and the heavy metals showed higher absorbance on PVC and PP particles compared with PA, PE and POM [65]. Xu et al. (2021) performed a mixed toxicity test with zebrafish (*Danio rerio*) and microplastics, although they did not find any lethal effect, but it was reported that the mixed exposure was correlated with the concentration of microplastics in survival, body length, and heart rate [66]. This may be supported by a study by Lu et al. (2018), who reported that microplastics increase cadmium accumulation in the liver, intestine, and gills of zebrafish and cause oxidative damage and inflammation [67]. Yang et al. (2020) showed that *Chlorella pyrenoidosa* exhibits antagonism when nonylphenol and microplastics are mixed [68]. Although the results of mixed toxicity are diverse, it is worth that the smaller-size plastics could aggravate with prolonged exposure time on algal growth. Stock et al. (2019) performed in vivo experiments in mice and in vitro experiments, using human cells. An oxidative stress response was observed in male mice, suggesting an effect on intestinal immune cells. However, no histological lesions were observed. Experiments using cells confirmed that fine particles can be absorbed into cells. However, the study concluded that oral exposure to microplastics did not cause serious health effects in mammals [69]. Wang et al. (2020) also investigated the effect of microplastics, using human Caco-2 cells, and reported that nanoscale microplastics could increase cytotoxicity [70]. Pop et al. (2021) concluded that bisphenol A (BPA) had a direct or indirect effect on chlorophyll in water microorganisms, and it was confirmed that it was a problem for cell-membrane division, integrity, and survival [14]. Bhatnagar and Anastopoulos (2016) reported that BPA was used as an endocrine disruptor as a major raw material in cosmetics and was found in surface water, groundwater, wastewater, and landfill runoff [15]. Wang and Qian (2021) confirmed that phthalates negatively affect the endocrine system and various organ functions, especially the reproductive organs, such as pregnancy, child growth, and child development [71]. Since there are few experimental studies on microplastic metabolism in the human body, it is judged that caution is needed in the interpretation of the results. In addition, studies related to the toxic effects of microplastics, using organisms and cell lines, are being actively conducted, and many research papers on toxicity have been published recently.

Recently, several studies have been conducted to evaluate the risk of microplastics in ecosystem. Adam et al. (2019) calculated the predicted-no-effect concentration (PNEC) as the fifth percentile of the probabilistic species sensitivity distribution, based on 53 values from 14 freshwater species and found that ecological risks cannot be entirely excluded in Asia, where 0.4% of the RCR values were above 1 [72]. Besseling et al. (2019), preliminary sensitivities for each species of marine water, estuary water, and freshwater were derived and risk assessment was performed. In this study, 168 toxicity data from a total of 66 studies were reviewed, and the HC_5(Hazardous Concentration for 5% of the species) value was determined to be 2.0 ng/L. Based on the species sensitivity distribution, it was found that there is a potential for harm to sensitive species in some hotspot areas [73].

Xu et al. (2018) performed an initial exposure assessment to surface water in the estuary of the Changjiang River in China and the nearby East China Sea. In this study, microplastic sampling was performed at 29 points, and the shape, size, color, and type of microplastics were measured [74]. In addition, they established the risk level criteria for microplastic pollution, using PLI (Pollution Load Index) shown in Table 2.

Table 2. Risk-level criteria for microplastic pollution [74].

Value of polymer index	<10	10–100	100–1000	>1000
Value of pollution load index	<10	10–20	20–30	>30
Risk category	I	II	III	IV

Although risk assessments for microplastics are continuously being conducted on humans and ecology, it is considered that more research is needed for a standard of concentration, and accuracy in microplastic risk.

4. Microplastic Analysis

Scientific methods to quantify and differentiate microplastics are needed to more systematically understand the impact of microplastics on the environment [75]. The measurement and analysis technology for microplastics has not yet been established, and the currently used analysis technologies have limitations in terms of detection limit and speed [76]. Accordingly, many researchers are actively working on developing more accurate microplastic analysis [75]. The analysis process of water microplastics can be roughly categorized into sampling, pretreatment, and analysis. Figure 4 is the procedure for sampling and analyzing microplastics.

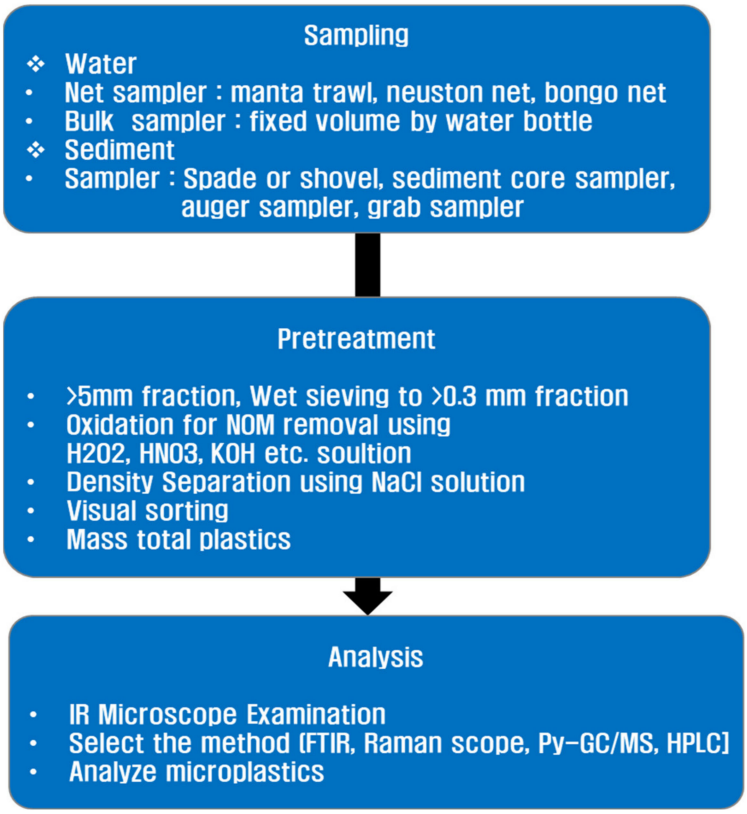

Figure 4. Microplastic analysis steps in water and sediment.

4.1. Sampling

There are various types of microplastic sampling methods, and they are applied differently, depending on water (freshwater and seawater), sediment, soil, cosmetics, and living organisms. However, this paper deals with the most widely used water and sediment sampling.

4.1.1. Water Sampling

Sampling of microplastics in water is highly dependent on the density, shape, properties, water flow, and depth of the microplastics [77]. In particular, the floating state of plastics depends on the salinity density according to the location and depth of sampling, so it is important to understand the density according to the salinity before collection. For microplastic sampling in water, net sampling and bulk sampling are widely used. Nets used for the sampling are manta trawl, neuston net, bongo net, and microplastic traps [77,78]. In bulk sampling, a specific volume is sampled using a water collection container [78]. Figure 5 shows the types of nets for a net sampling.

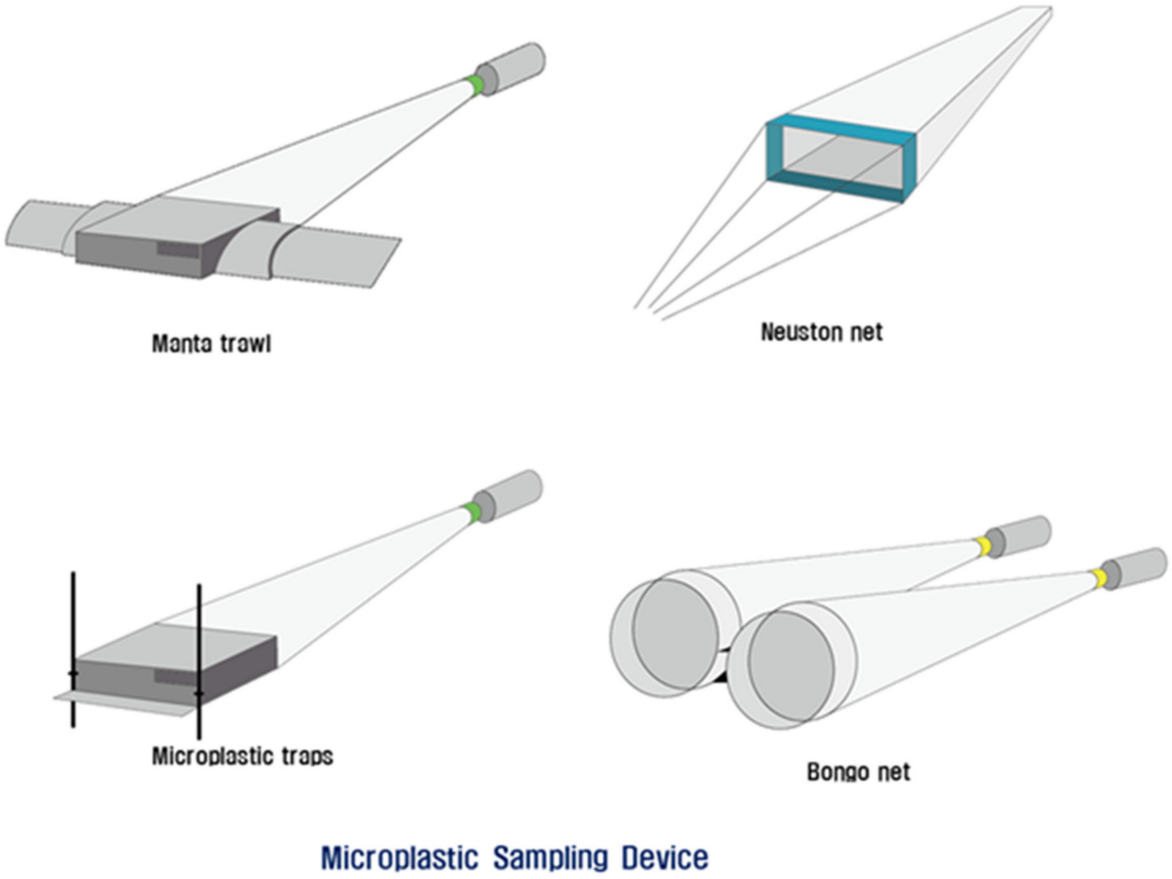

Figure 5. Types of microplastic sampling device for water.

A net sampling method provides the pros of a large area sampling and large volume sampling by a boat [79,80]. At this time, a flow meter should be attached to the net so that the amount of water can be clearly expressed in m^3. However, cons are the size of collected microplastics that limited depending on the size of the mesh [81]. The mesh of manta trawl is generally 330 μm mesh, so it is impossible to collect fine plastics smaller than this [78]. Using a nylon net (100 μm), more microplastics can be collected, but if the size of the mesh is less than 100 μm, it can be quickly clogged by plankton, so it is important to collect it at a low speed to avoid from fast clogging [80]. Vermaire et al. (2017) confirmed the difference between 0.1 MPs/L by nylon net and 0.00135 MPs/L by manta net, in which nylon nets (100 μm) collected microplastics almost 100 times higher than manta nets (330 μm) [82]. Nevertheless, manta net, neuton net, and bongo net mesh sizes (330~350 μm) are widely used, because they can collect a large volume of water. If a plankton net with a small mesh is used, a higher concentration of microplastic can be collected than other nets, but sampling is possible only within 1 min, ensuring a low flow rate [83]. A bulk sampling or grab sampling is collected as much as a fixed volume, and the sampling range is narrower than the net sampling. As an alternative technology, SubCtech

developed a microplastic sampler based on a pump on a ship. It provides sampling at high vessel speed and instantaneous size separation, using low power water pump [84]. These represents that the measured amount of microplastics could vary depending on the sampling methods [77]. To summarize the above, net sampling is mainly used to collect the amount of microplastics in a large area, and a net of 100–350 µm is used depending on the size of the microplastics. When collecting microplastics in a limited space or in a narrow area, grab sampling or bulk sampling is recommended, and a large number of samples is recommended.

4.1.2. Sediment Sampling

The distribution of microplastics in the sediment is uneven and can vary greatly depending on the collection point, number of collections, and depth of extraction [85]. For example, when sediment is collected from a tidal line with high microplastic accumulation, a high concentration of microplastic can be observed [86]. When the depth of sampling is low, the concentration of microplastics can be high, and when the depth of the sample is 15 cm or more (subsoil), it can show low concentration [87]. Accordingly, the standard method used for soil collection is recommended for the sampling method. Soil sampling is limited to the topsoil layer (0~15 cm), and in general, 5 to 10 samples are collected in a zigzag pattern, or 1 representative point of collection and 4 points are selected, and a total of five samples are mixed and tested [88]. Sediment sampling equipment is a general shovel, grab sampler, sediment core sampler, and auger sampler, as shown in Figure 6.

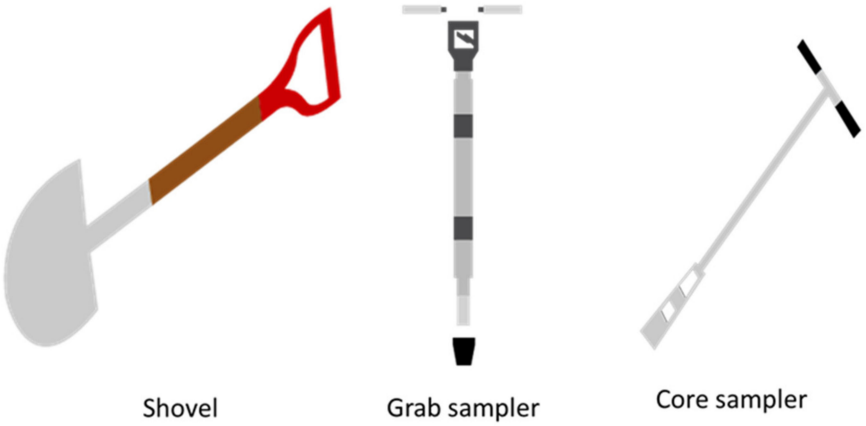

Figure 6. Types of microplastic sampling device for sediment.

In addition, the weight g or volume L of the sample has a great influence on the representativeness of the sample, so 300–500 g is recommended, as with the soil sampling method [89]. The samples collected in this way are sealed in glass bottles of 300 g or more, tested in a refrigerated state at 0 to 4 degrees, and transported to the laboratory [89]. The sample transported to the laboratory is dried at 40 °C for 72 h to correct the moisture content and the moisture content is checked. For soil samples, Tomas et al. suggested sieving to 1, 2, and 5 mm in compliance with standard mesh sizes of commercially available test sieves [90]. Standard sieve sizes vary from 37.5 to 0.075 mm, so 0.5 or 0.15 mm sieve is recommended to collect smaller microplastics.

4.2. Pretreatment

The collected samples contain microplastic particles and various other particulate materials, such as sand, clay, and microorganisms. These samples with other pollutants rather than plastics require pretreatment. The method of pretreatment is largely divided into two parts as density separation and oxidation. The density separation use NaCl or other chemicals to separate microplastics from other materials. The oxidation uses H_2O_2 or other chemicals to remove natural organic matter [91]. The density separation and oxidation can be performed first or later depending on the type of samples and

degree of contamination. For high contaminated microplastic samples, a pretreatment process is required to prevent interference with the measurement of inorganic particles before instrument analysis [92]. Microplastics and inorganic particles are very small in size and are difficult to remove with the naked eye, so separation is mainly used using a difference in density [92]. Many chemicals, such as NaCl, NaBr, NaI, $CaCl_2$, $ZnCl_2$, and $Na_6[H_2W_{12}O_{40}]$, have been used for the density difference separation of microplastics. The solution concentration of the density difference for the separation of various plastics is NaCl solution with a density of 1.2 kg/L, sodium polytungstate with a density of 1.4–1.8 kg/L, $CaCl_2$ with a density of 1.3–1.5 kg/L, NaI with a density of 1.6–1.8 kg/L, $ZnCl_2$ with a density of 1.5–1.7 kg/L, $ZnBr_2$ with a density of 1.7 kg/L [92–94]. Separation by using the difference in density has a problem, in that the recovery rate is as low as 40% when the particle size of the microplastic is less than 1 mm [95]. Recently, with the development of a method to increase the microplastic recovery rate, even small plastic particles can be recovered at the level of 96–99% [92,96]. Density difference separation takes a long time, such as treatment time (2–5 h) and settling time (24–72 h), so some studies are being conducted to save time, such as centrifugation and air bubbling [94,97]. However, a standardized recovery method for microplastic is not established, and so it is necessary to develop a standardized method for microplastic density separation.

In order to remove natural organic matter (NOM) on the surface of microplastics after separating plastics from inorganic particles, 30% hydrogen peroxide, Fenton oxidation, acid decomposition, base decomposition, and enzyme decomposition are used alone or in combination [91,95]. Hydrogen peroxide is excellent in removing organic matter, but it is not suitable for the pretreatment of plastics made of polyethylene and polypropylene; mixed hydrogen peroxide, sulfuric acid, or hydrochloric acid are often used [90,92,95]. The H_2O_2 oxidation results corroborated the findings from previous studies, in that there was polymer degradation despite the effective removal of organic matter from the samples (sludge, 80–86%; soil, 96–108%) [91]. When using acids, low pH can lead to microplastic damage [95]. NaOH alkaline digestion resulted in significant degradation of PET and PC particles but removed 61–67% of the organic matter from the sludge samples and 64–68% from the soil samples. KOH alkaline digestion had minimal impact on the plastic particles; however, it only removed 57% of the organic matter from the sludge samples and 35% from the soil samples [95]. Another method for decomposing organic matter is using enzymes. Cloe M. et al. (2014) used enzymatic digestion techniques, which removed >97% of plankton in seawater, without any damage to the microplastic [98]. However, the method using the enzymatic digestion techniques requires a pretreatment time of about 6 days, and oxidation of NOM removal of microplastics may take 24 h–30 days, depending on the type of sample. Accordingly, it is necessary to standardize a method that can minimize plastic damage and reduce time during pretreatment. Figure 7 shows a simple pretreatment step before analysis.

4.3. Analysis Method

Among the microplastics analysis, the simplest method is to determine the number of microplastics by staining them with 10 ug/mL Nile Red reagent in 10% dimethyl sulfoxide, with a 10 min incubation time and using a counting program [88]. However, it is not easy to consistently dye microplastics of various properties, it is difficult to identify the types of microplastics, and the pretreated samples may contain substances other than microplastics, so the accuracy is low [99]. Kang et al. (2020) compared the Nile Red method to FTIR, with an accuracy of 78% [99]. Shim et al. (2016) found that the recovery rate of polyethylene (100–300 μm) spiked to pretreated natural sand was 98% in the NR stating method, which was not significantly different with Fourier-transform infrared spectroscopy (FTIR) identification [100]. The NR staining method was suitable for discriminating fragmented polypropylene particles from large numbers of sand particles in laboratory experiment [100]. Another easy method is to find the number of moles of solute in a sample by using Beer's law based on the UV–Vis measurement result [101].

This method is capable of quantitative analysis of microplastics to some extent, but qualitative analysis is not possible. In addition, if the microplastic sinks to the floor or jumps on the surface, it can cause various errors. Accordingly, the types of methods that are currently widely used can be broadly divided into two types, namely non-destructive methods and destructive methods [85]. Non-destructive analysis includes microscopy, microscopy with spectroscopy, microscopy with FTIR, and microscopy with Raman spectroscopy [102–105]. Destructive analysis includes liquid chromatography with mass spectrometry (LC/MS/UV) [106,107], gas chromatography connected with mass spectrometry (GC/MS), subsuming pyrolysis/gas chromatography/mass spectrometry (Pyr/GC/MS), and thermal desorption gas chromatography (TD/GC/MS) [108–110]. Among them, the most commonly used methods are a microscopy with FTIR, Raman, and Pyr/GC/MS. The analysis of microplastics is not clearly defined, so many methods are still being used and studied. Tables 3 and 4 review the currently used analysis methods. Recently, for rapid analysis of microplastic, a charge-coupled device (CCD) camera, photodiode [111], dynamic light scattering (DLS) [112], nanoparticle tracking analysis (NTA) [113], fluorescence spectroscopy [114], and atomic force microscopy (AFM) [115] are often used with FTIR, Raman, Pyr/GC/MS, and LC/MS. Asamoah et al. (2019) used photodiode and CCD to measure PET and LDPE among microplastics to determine the type, size, and non-planarity [111]. According to the analysis of microplastic studies, FTIR-microscope and Pyr or TD/GC/MS are the most-used analysis methods. LC/MS/UV and the Nile Red method are often used. The unit used in the result by FTIR-microscope and the Nile Red method is expressed as the number of MP/L or m^3, and the unit used in the result by GC/MS, LC/MS is expressed as weight (ng, g)/L or m^3.

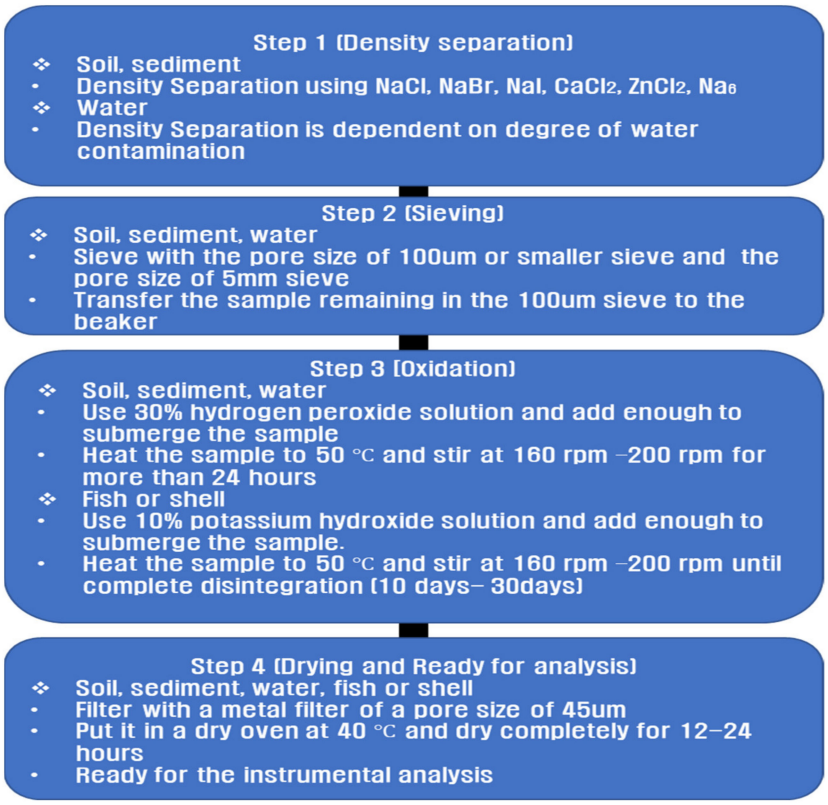

Figure 7. Simple pretreatment steps.

Table 3. Non-destructive analytical method for microplastic analysis.

Methods for Microplastics	Analytical Method	Type of Source	Pretreatment	Particle Size	Types of Polymer	Identification	References
Non-Destructive method	FTIR	Wastewater in Derby, UK	Density separation, oxidation (30% H_2O_2) for 7 days	>5 mm	PE, PP, PVC, PS, nylon-6	Reproducible identification rate: 98.33% 50 MPs/10 L	[116]
	ATR (Attenuated Total Reflection)-FTIR	Agricultural soil in Middle Franconia, Germany	Density separation oxidation (30% H_2O_2)	1 to 5 mm	PE, PS, PVC, PET, PMMA	0.34 ± 0.36 MPs/kg dry soil	[117]
	μFTIR	Sediment of the lagoon in Italy	Density separation	-	-	Up to 2175 MP/kg	[104]
	μFTIR	Air in Denmark	Sonicate, dry	11 μm	PE	9.3 ± 5.8 MPs/m^3	[104]
	FTIR	Sediment, Snow, Ice core in the lake in Finland	Filtration, density separation	>1.2 μm	PA, PE, PP, cellulose, wool	395.5 ± 90.7 MPs/kg, 117.1 ± 18.4 MPs/L, 7.8 ± 1.2 MPs/L	[118]
	Synchrotron-based FTIR	Beach Sediment in Taiwan	Density separation	≥1 mm	PE, PP	4–532 MPs/0.0125 m^3	[119]
	FTIR	25 Beach Sediments in India	Sieving, density separation	<5 mm	PE, PP, PS	178 ± 261 mg/m^2 (low tide) 1323 ± 1228 mg/m^2	[120]
	FTIR	79 fishes in India	Density seperation, oxidation (30% H_2O_2)	-	-	10.1% with 79 fishes	[120]
	FTIR	Treated wastewater, Sediment in Germany	Density separation, sieving, filtratrion	≤500 μm	PE, PP, PET, PS, PVC, PC, PUR, PA	39–37,223 MPs/m^3 (wastewater) 8–20 MPs/m^3 (surface water) 143–1151 MPs/kg (sediment)	[109]
	Raman	Standards from Sigma-Aldrich	-	74 μm PE 37–74 μm PS 27–45 μm PE	PE, PS-DVB	<1 μm visible	[105]
	Raman	Air in London, UK	Density separation,	≥2 μm	PE, PET, PP	PE:2467.9 MPs/m^3, PP:22.4 MPs/m^3, PET:11.2 MPs/m^3	[121]
	Raman	Sea snow in CT, USA	Density separation, 15% H_2O_2	63–600 μm	PP, PET	59 MPs/4 L	[122]

Table 4. Destructive analytical method for microplastic analysis.

Methods for Microplastics	Analytical Method	Type of Source	Pretreatment	Particle Size	Types of Polymer	Identification	References
Non-destructive method	Pyr/GC/MS	Wastewater in Germany	Filtration	Filter (100 µm, 50 µm, 10 µm)	PE, PS	PE: - PS: 0.072 mg/m^3	[123]
	Pyr/GC/MS	Treated wastewater, Sediment in Germany	Density separation, Sieving, Filtration	≤500 µm,	PE, PP, PET, PS, PVC, PC, PUR, PA	6–2525 µg/m^3 (wastewater) 4.2–5.5 µg/m^3 (surface water) 8–144 µg/kg (sediment)	[109]
	TD or Pyr/GC/MS	Standards from BS Partikel GmbH, Sigma-Aldrich	Density separation	PS: 78 nm, 41 µm, PMMA, PEL: 48 µm	PS, PE, PMMA	Sorption of phenanthrene (PMMA << PS 40 µm < 41 µm < PE < PS 78 nm), α-cypermethrin (PS 41 µm < PS 40 µm < PE <PMMA < PS 78 nm)	[110]
	Pyr/GC/MS	Lake water in Western Lake Superior, Canada	Density separation	<5 mm	PP, PS, PVC, PET	Recovery rate: mean 77% PVC (1.38–1.41 g/cm^3) PET (1.38–1.41 g/cm^3) PP (0.85–0.92 g/cm^3)	[124]
	Pyr/GC/MS	Fish in the Texas Gulf Coast in US	Oxidation, Filtration	43 particles	PVC, PET, silicone, nylon, epoxy	PVC, PET 44.1%, nylon 9.3%, silicone 2.3%, epoxy 2.3% 42% samples are not classified	[125]
	LC/UV	Soil, dust, sewage water in Germany	KOH (1 g/100 mL) in 1-pentanol solution	-	PET	3.85–3.99 mg/kg (soil) 12,500–57,000 mg/kg (indoor dust) 1430 mg/kg (sewage water)	[106]
	LC–MS/MS	Sludge, sediments, dust, calm, salt in China	KOH in 1-pentanol solution	-	PC, PET	Indoor dust: 248 mg/kg (PC), 430 mg/kg (PET) Calm:63.7 mg/kg (PC), 127 mg/kg (PET)	[107]
	LC–MS/MS	Indoor dust from 12 countries	KOH in 1-pentanol solution	150 µm	PC, PET	PET: 38–120,000 µg/g PC: <0.11–1700 µg/g	[126]

4.3.1. FTIR Spectroscopy

FTIR spectroscopy is a non-destructive testing method that analyzes a polar functional group such as a carbonyl group by using the degree of scattering of a substance, using infrared rays of a specific wavelength [127,128]. Previously, it was difficult to analyze microplastics under 500 µm and contaminated plastics with FTIR spectroscopy, but recently, microplastics as small as 10 to 5 µm can be analyzed by using a new micro(µ)-FTIR microscopy [104]. FTIR spectroscopy can quickly measure all the frequencies of the infrared source at the same time through pretreated filter paper [117]. However, this method still takes a long time to count small size plastics with irregular shapes and is difficult to apply to samples less than 20 µm [116,117]. Although the condition setting of the FTIR spectroscopy differs depending on the analytical device and microplastic analysis items, the general settings are shown in Table 5.

Table 5. FTIR setting condition for microplastic analysis.

Methods for Microplastics	Pretreatment	Sample Preparation	Settings	Result	References
FTIR	Sieving and flushing with ethanol, Freeze and oxidize with H_2O_2 or other chemicals, Density separation, Sonification	>80 µm use infrared reflective glass slide, <80 µm use CaF_2 infrared transparent window and dried	Focal plane array size: 128 × 128 mm, Objective: 15×, IR Pixel size: 5.5 µm, Number of scans per tile: 30, Number of mosaic tiles: 16 × 16, Total measurement area: 9.8 × 9.8 mm, Spectral resolution 8 cm^{-1}, Spectral range: 3850–850 cm^{-1}, Total scanning time: 3 h, Total number of spectra: 4,200,000	Conform both by mass and by particle count. Use standards to find recovery rate	[116,129–131]

4.3.2. Raman Spectroscopy Method

Raman spectroscopy is a method to determine the presence and type of plastic by scattering light generated when the molecules of microplastic particles are excited in vibrational, rotational, and other states [132]. Micro-Raman spectroscopy can analyze microplastics with a large area and high resolution [133], and it offers high selectivity and reproducibility and requires low sample amounts with a minimal sample preparation and shot data collection time [132,133]. This method is effective, but when the sample is contaminated by organic matter, it acts as an interfering factor for fluorescent materials, so it must be completely removed during pretreatment [96]. Raman spectroscopy is often used with FTIR spectroscopy, because it is difficult to measure a sample of 1 to 50 µm in FTIR. Raman spectroscopy is often used to identify below 10 µm and is capable of resolving particles down to 1 µm or less [132]. Primpke et al. (2020) measured seawater by using FTIR and Raman spectroscopy, and Raman confirmed the number of microplastics by about 23% higher than that of FTIR, and, in particular, it showed a high microplastic discovery rate at ≤500 µm [109]. General Raman spectroscopy settings are offered in Table 6.

Table 6. Raman setting condition for microplastic analysis.

Methods for Microplastics	Pretreatment	Sample Preparation	Settings	Result	References
Raman	Sieving and flushing with ethanol Freeze and oxidize with H_2O_2, Density separation with NaCl, $ZnCl_2$, NaI, salt removal, sonification,	Calcium fluoride slide, samples on disk window placed in a desiccator	Slit: 50 µm Grating blazed: 700 nm with 0.61 nm spectral resolution. Recording spectra: 200–4000 rel./cm (polymer spectral region:2800–3600 rel./cm) Signal to noise ratio: 300:1 Readout rate: 1.8 ms/scan Wavelength: 785 nm, 532 nm	Conform both by mass and by particle count. Use standards to find recovery rate	[102,103,105,121, 132]

Currently, a Laser Direct Infrared (LDIR) chemical imaging system [134] has been developed that improves the analysis time and resolution, which are the disadvantages of FTIR and Raman spectroscopy, and improves the analysis speed.

4.3.3. GC/MS Method

A GC/MS method is a destructive method to determine sample of microplastics by heat-treating the sample and analyzing the gas [104]. There are two types of GC/MS analysis of microplastics: Pyr/GC/MS and TD/GC/MS. In both cases, the amount of microplastics is analyzed by the number of ions released through pyrolysis or thermal desorption [110]. Advantages of GC/MS over FTIR and Raman spectroscopy can analyze both qualitative and quantitative analysis of small microplastics size (<10 µg) with a small sample volume of microplastics, because of its high sensitivity [135]. However, GC/MS is destructive, resulting in the total loss of the particle and subsequently eliminating further particle analysis [125]. Plus, GC/MS allows the analysis of a whole MP particle, in contrast with Raman and FTIR (in reflection mode), which only analyze the surface of the MP particle, being sensitive to interference caused by additives such as pigments [124, 136]. Table 7 shows general setting conditions for microplastic analysis using TD and Pyr/GC/MS

Table 7. TD and Pyr/GC/MS setting condition for microplastic analysis.

Methods for Microplastics	Pretreatment	Sample Preparation	Settings	Result	References
TD/GC/MS, Pyr/GC/MS	Sieving and flushing with ethanol, freeze and oxidize with H_2O_2, freeze-drying of polymers	Application of particles into the pyrolysis tubes, SIM or scan mode (recommended SIM mode operation)	Pyr temperature: 600–800 °C CIS temperature: −50 °C Mode: split or splitless TD temperature: initial, 20 °C, 0.3 min delay time, 1.0 min hold time End: 60 °C/min, hold time 5 min GC/MS: Column: DB-5MS ultra, optima initial: 40–50 °C, hold time 2–4 min, heat 10 °C/min to 300–320 °C maintain for 3 min Split mode <10 µg, splitless mode: >10 µg Mass rage: m/z 10–550 Scan time: 0.2~0.5 s	Conform each m/z result. Use standards to find recovery rate	[123,137–139]

5. Microplastic Removal Technology

5.1. Wastewater Treatment Plants (WWTP)

Industrial wastewater, domestic wastewater, agricultural wastewater, and livestock wastewater contain many microplastics, and wastewater treatment plants cannot remove all microplastics. These wastewaters include micro pellets from cosmetic preparations, especially facial scrubs and textile fibers, the latter due to the shedding of particles during the textile washing process [123]. The wastewater is moved to a municipal wastewater treatment plant, where large plastics are removed; however, microplastic or nanoplastic particles still remain after several water treatment processes, and some of microplastics are still discharged to the final effluent [140]. Accordingly, the efficiency of removing microplastics in wastewater treatment plants has emerged as an important issue, and many studies are being conducted.

Typical WWTP processes are divided into preliminary treatment, primary treatment, secondary treatment, and tertiary treatment, and the process chart is as shown in Figure 8. Preliminary treatment consists of a screen and a sedimentation tank, mainly removing large and large plastics. Primary treatment mainly consists of aeration and sedimentation, and it removes light plastics or heavy plastics by skimming and sedimentation. Secondary treatment is mainly biological treatment to remove organic material. Biological treatment consists of an anaerobic tank, anoxic tank, an aerobic tank, and a settling tank, and most of the microplastics >500 μm are removed [137,141,142]. Tertiary treatment is mainly used as an option in WWTPs and consists of a process of removing phosphorus and nitrogen by using chemicals. At this time, it is possible to increase the removal rate of microplastics by using several coagulants [76,137]. Table 8 confirmed the MPs' removal rate according to the process of WWTPs.

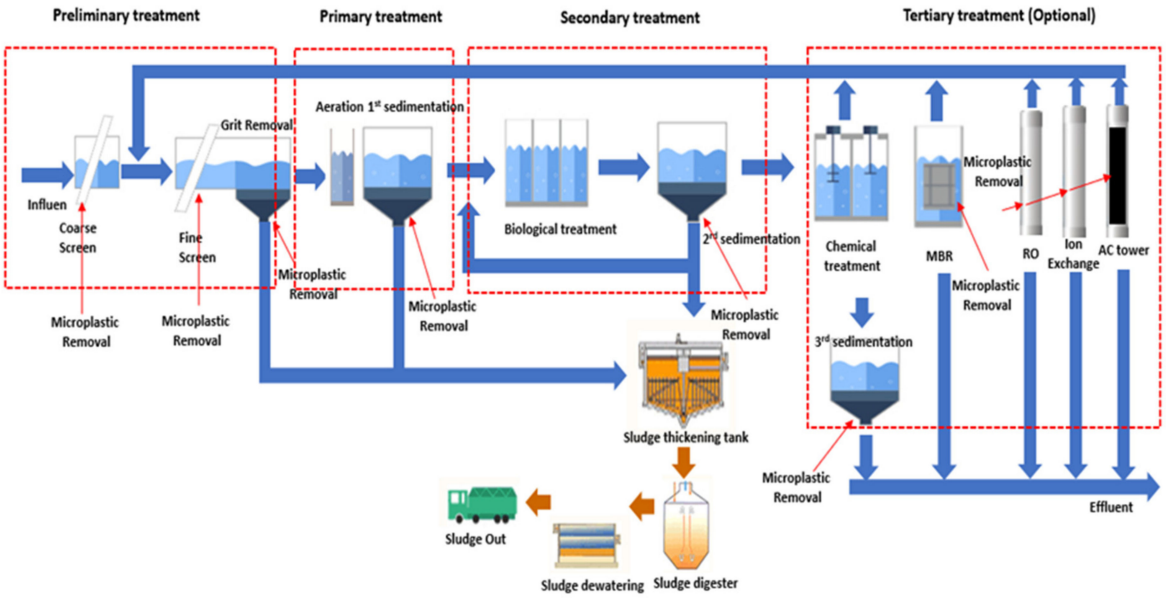

Figure 8. Microplastic-removal process in WWTP.

Table 8. Microplastic-removal reviews in WWTP.

Treatment Process	Removal Technologies	Inlet Concentration	Outlet Concentration	MP Removal Rate	References
Preliminary and primary treatment	Screening, grit removal, pre-aeration, sedimentation	567.8 MPs/L	11.7 MPs/L	82 %	[143]
	Screening, grit removal, primary sedimentation	1737 MPs/L	337 MPs/L	80.6 %	[123]
	Screening, grit removal, physic-chemical lamellar settling	183 MPs/L	43 MPs/L	76.5 %	[123]
	Screening, grit removal, primary sedimentation	35 MPs/L	8 MPs/L	76.9 %	[144]
Secondary treatment	Membrane bioreactor	0.6 MPs/L	0.004 MPs/L	99.3%	[145]
	Biofiltration	43 MPs/L	12 MPs/L	72.1 %	[146]
	A_2O process	128 MPs/L	12.8 MPs/L	90 %	[123]
	A_2O process	1.32 MPs/L	1.1 MPs/L	16.6 %	[140]
Tertiary treatment	Membrane bioreactor (UF)	0.5 MPs/L	0.2 MPs/L	60 %	[147]
	Membrane bioreactor (UF)	0.48 MPs/L	0.28 MPs/L	41.6%	[143]
	Denitrification and UF	12.3 MPs/L	0.59 MPs/L	95 %	[148]

The size and concentration of microplastics in the wastewater entering the wastewater treatment plant require more research depending on the location of the wastewater treatment plant, the type of wastewater, and the purpose of treatment. The concentration of microplastics in the effluent from the wastewater treatment plant was 1 MPs/L or less in many studies when the primary, secondary, and tertiary treatments were completed. In addition, it is difficult to ascertain a clear average removal rate of the reviewed studies because the microplastic treatment rate varies at each treatment stage, but the concentration of the inlet is different. Most of the high concentration of microplastics is removed from the primary treatment and contained in the sludge, so the use of recycled fertilizers by the sludge can cause secondary pollution of microplastics.

5.2. Physical Removal Technology

Physical methods for removing microplastics include flotation, sedimentation, and filtration, but filtration is a representative method. Filtration includes various methods, such as screening, diskfilter, sandfilter, and membrane filtration (microfiltration, MF; ultrafiltration, UF; nanofiltration, NF; dynamic membrane, DF; and reverse osmosis, RO). Screening is used in both general WWTP and drinking-water treatment plants (DWTP). The screening method generally removes large plastic particles by filtering and sedimentation. The microplastic removal rate by the screening method was confirmed to be from about 40% to about 80% [134,141,142]. Diskfilter is often used in WWTP. Simon et al. (2019) showed that the diskfilter removed up to 89.7% of microparticles less than >10 µm [149]. A sandfilter is used in both WWTP and DWTP. Wolff et al. (2021) showed microplastic removal rates of

99.2% ± 0.29% and 99.4% ± 0.15% by a rapid sandfiltration [150]. Most of the microplastic removal using the membrane filtration shows an efficiency of more than 90%, and it appears to be particularly effective in removing microplastics larger than 10 μm [88,99,101–103,119]. However, when microplastics are effectively removed by using the membrane method, membrane contamination can be accelerated by the deposition of microplastics, which can accelerate contamination of other organic matters in the membrane [130]. Accordingly, when using a membrane, a pretreatment process must be installed in order to prevent excessive membrane contamination of organic matters and microplastics. Table 9 provides a positive physical removal technology for microplastic removal

Table 9. Physical removal technology for microplastics.

Removal Methods for Microplastics	Technology Summary	Result	References
Membrane bioreactor (MF)	Source: wastewater MPs: 480 MPs/L	MP removal: 79.01% by MBR MP removal: 75.49% by rapid sandfilter	[151]
Dynamic Membrane (UF)	Source: polycarbonate, cellulose acetate, polytetrafluoroethylene Membrane: 5 μm mesh PMs size: PA, PS (20~300 μm)	MP removal: 94%	[152]
Membrane bioreactor (UF)	Source: wastewater, sludge MPs: 0.1–124.7 MPs/L (wastewater) 8.2–3014 MPs/g (sludge)	MP removal: 99.4% by MBR MP removal: 98.3% by CAS	[147]
Glass membrane	Pore size: 1 μm Plastics: PS, PMMA	MP removal: 90.7%	[153]
RO membrane	Pore size: 0.1->0.005 μm Plastics: PE (0.1 g), PP (0.1 g), PE/PP mixture (0.1 g)	MP removal: >85% Organic removal: >99%	[138]
MF membrane	Source: wastewater Pore size: 0.1 μm Influent MPs: 94–206 MPs/L	MP removal: 98%	[139]
Disk filter	Source: wastewater Plastic size: 10μm Effluent MPs: 3 MP/L	MP removal: 89.7%	[154]

5.3. Chemical Removal Technology

The method of using chemicals for microplastics has been studied in various ways, but in general, the most representatively used method is coagulation/precipitation in water treatment. In particular, there are many differences in the amount of microplastics removed, depending on the type of coagulant, the amount of coagulant, and coagulation retention time. Accordingly, many studies are being conducted to find the optimal coagulant type and conditions, and more clear studies for microplastic removal are needed in the future. In Figure 9, Lapointe et al. (2020) compared the removal rates of polyester (PEST), weathered PE, and pristine PE through a Jar test, using aluminum-based coagulants and polyacrylamide (PAA). When aluminum-based coagulants 2.73 mg Al/L and PAM 0.3 mg PAM/L coagulants were administered to 500 MPs/L water, the optimal removal rates were similar. The removal efficiency of microspheres was PE = 82% of 140 μm, PS = about 80% of 140 μm, PE = about 88% of 15 μm, and PEST fiber = 99% [155]. Table 10 presents a microplastic removal by chemical removal technology.

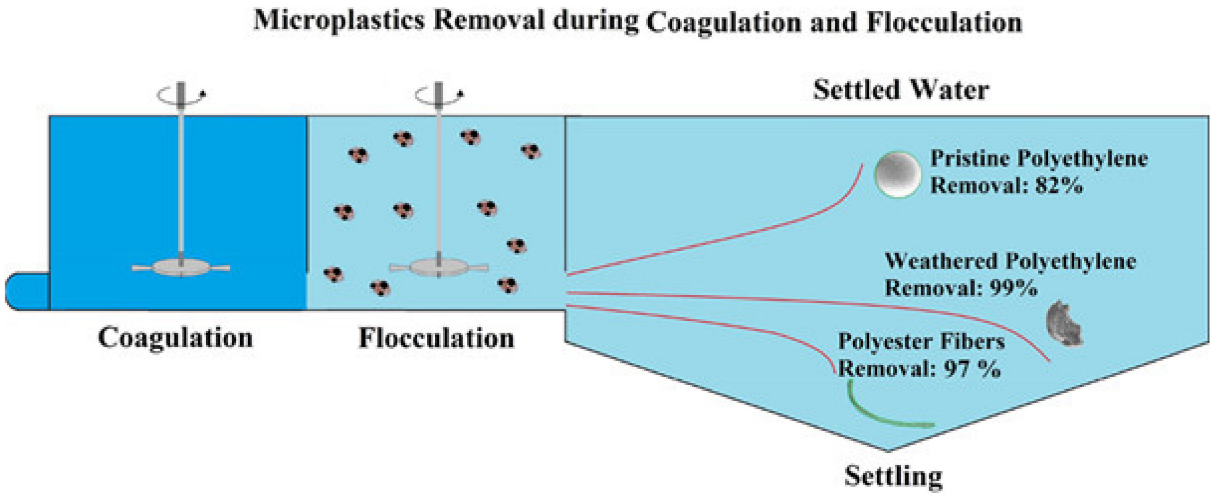

Figure 9. Microplastic removal by coagulation and flocculation [155].

Table 10. Chemical removal technology for microplastics.

Removal Methods for Microplastics	Technology Summary	Result	References
Alum and PAM coagulant	Source: 500 MPs/L Dose: 2.73 mg Al/L, 0.3 mg PAM/L	PE removal: 82% of 140 µm PS removal: 80% of 140 µm PE removal: 88% of 15 µm PEST fiber: 99%	[155]
Electrocoagulation	Source: microbead wastewater Condition: pH 7.5, NaCl concentration: 0–2 g/L, current density: 11 A/m^2	Microbeads: 99.24%	[156]
Fe- and Al-salt coagulation with plant derived tannic acid	Source: PS/PE beads mixed water Dose: 3 mM coagulant (0.5 mL)/1.5 mL (microbead)	PS/PE removal: 95%	[157]
$AlCl_3$ coagulation with and without PAM	Source: PE beads Dose: 5 mM ($AlC_3 \cdot 6H_2O$)	PE removal: about 28%, pH 6.0 PE removal with PAM: about 46%	[158]
$FeCl_3$ with PAM coagulation	Source: wastewater with < 10 µm MPs Dose: 5 mM ($FeCl_3 \cdot 6H_2O$)	MPs removal: up to 99.4%, pH 7.3 to 6.5	[154]

The review results were found to be able to efficiently remove microplastics by combining PAM with Al-based and iron-based coagulants up to 99% dependent on the microplastic size, number, and water conditions, and it was confirmed that electrocoagulation was also effective in removing microplastics.

5.4. Biological Removal Technology

Among the microplastic removal methods, biologic methods include activated sludge treatment, aerobic and anaerobic digestion, lagoons, and septic tanks. In activated sludge systems, bacteria are known to trap microplastics <0.5 mm [158]. However, the activated sludge system only plays an imposing role of trapping microplastics in water, and it is difficult to degrade plastics with a short residence time (7–14 h) in WWTPs [158]. Liu et al. (2019) virgin microplastics do not significantly affect the activities of ammonia oxidizing bacteria, nitrite oxidizing bacteria, and phosphorus accumulating organisms [159]. Cunha et al. (2020) used 10 mg/L of fresh *Cyanothece* sp. and found microplastic removal rate

of up to 47% [160]. Canniff and Hoang (2018) confirmed the growth rate by exposure to PE beads, using *Daphnia magna*, and confirmed that the PE intake rate increased as the particle concentration and exposure time increased. In particular, it was confirmed that *R. subcapitata* exposed to PE beads grew more than those without exposure [161]. As such, the removal of microplastics by using a biologic method is generally low in efficiency, and secondary contamination of microplastics in sludge or sedimentation can be increased. The effect of microplastics on the performance of the bioreactor system should not be overemphasized [162]. Therefore, it can be concluded that high microplastic removal efficiency by biological methods is not very positive.

6. Conclusions

As worldwide plastic production has been increasing, plastic contamination has caused global deterioration of the environment, due to the long plastic-decomposition time [3,4,160]. As a result, microplastic contamination is becoming a global issue, but monitoring, toxicity, analysis, and removal technology are still insufficient for researchers. Therefore, this paper reviewed the distribution, toxicity, analysis method, and removal technologies of microplastics and summarizes the results as follows.

- Microplastics with the high contamination were reported as PET, PU, PS, PVC, PP, PE, and PA.
- Contamination paths of microplastics include agricultural wastewater, industrial wastewater, litter, sewage treatment plant, household personal products, road runoff, fishing waste, and atmosphere decomposition, which finally flow into the sea to pollute sea creatures and are absorbed by humans.
- The pollution of microplastics around the world is sharply increasing, and it is appearing in drinking water, sewage water, rivers, seas, soil, and everywhere. Finally, microplastics will cause a huge problem in the near future.
- Although the toxicity of microplastics has not been studied much, plastics, such as PS, PVC, PP, etc., could still cause problems in human health, and several researchers are conducting research on the risk of microplastics.
- Analysis of microplastics were divided into sampling, pretreatment, and analysis parts. Water sampling and sediment sampling were discussed in the sampling part. In the pretreatment part, how to deal with the density difference separation and how to remove other contaminants rather than microplastics were discussed in detail. In the analysis method part, most of used microplastic analytical methods and methods for a possible application for a microplastic analysis were summarized in detail.
- Non-destructive analytical methods of FTIR and Raman methods were summarized by instrument settings and analytical results from other researchers. Destructive analytical methods of Pyr/GC/MS and LC/MS are summarized by instrument settings and analytical results from other researchers.
- The various techniques for the removal method of microplastics were summarized in WWTP, physical, chemical, and biological technologies. Each technique for microplastic removal rate was summarized in several tables. In particular, the microplastic removal rate in WWTP was found to be more than 70% after secondary treatment, although there was a difference depending on the research papers.

In conclusion, microplastic contamination is scattered around the world and directly affects human life. Accordingly, research on microplastic contamination, analysis, monitoring, and removal technology is insufficient, so deeper interest from researchers and national policies are urgently needed for overcoming microplastic contamination.

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References

1. Plastics Europe. Plastics the Facts 2019: An Analysis of European Plastics Production, Demand and Waste Data. PlasticsEurope, Brussels. 2019. Available online: https://www.plasticseurope.org/application/files/9715/7129/9584/FINAL_web_version_Plastics_the_facts2019_14102019.pdf (accessed on 1 May 2021).
2. Jambeck, J.R.; Geyer, R.; Wilcox, C.; Siegler, T.R.; Perryman, M.; Andrady, A.; Narayan, R.; Law, K.L. Plastic waste inputs from land into the ocean. *Science* **2015**, *347*, 768–771. [CrossRef]
3. Barnes, D.K.A.; Galgani, F.; Thompson, R.C.; Barlaz, M. Accumulation and fragmentation of plastic debris in global environments. *Philos. Trans. R. Soc. B Biol. Sci.* **2009**, *364*, 1985–1998. [CrossRef]
4. Rocha-Santos, T.; Duarte, A.C. A critical overview of the analytical approaches to the occurrence, the fate and the behavior of microplastics in the environment. *Trends Anal. Chem.* **2015**, *65*, 47–53. [CrossRef]
5. Ng, E.L.; Huerta-Lwanga, E.; Eldridge, S.M.; Johnston, P.; Hu, H.-W.; Geissen, V.; Chen, D. An overview of microplastic and nanoplastic pollution in agroecosystems. *Sci. Total Environ.* **2018**, *627*, 1377–1388. [CrossRef] [PubMed]
6. Cole, M.; Lindeque, P.; Halsband, C.; Galloway, T.S. Microplastics as contaminants in the marine environment: A review. *Mar. Pollut. Bull.* **2011**, *62*, 2588–2597. [CrossRef] [PubMed]
7. Da Costa, J.P.; Santos, P.S.M.; Duarte, A.C.; Rocha-Santos, T. (Nano)plastics in the environment-Sources, fates and effects. *Sci. Total Environ.* **2016**, *566*, 15–26. [CrossRef]
8. Andrady, A.L. Microplastics in the marine environment. *Mar. Pollut. Bull.* **2011**, *62*, 1596–1605. [CrossRef] [PubMed]
9. Stephens, B.; Azimi, P.; El Orch, Z.; Ramos, T. Ultrafine particle emissions from desktop 3D printers. *Atmos. Environ.* **2013**, *79*, 334–339. [CrossRef]
10. Ajith, N.; Arumugam, S.; Parthasarathy, S.; Manupoori, S.; Janakiraman, S. Global distribution of microplastics and its impact on marine environment-a review. *Environ. Sci. Pollut. Res. Int.* **2020**, *27*, 25970–25986. [CrossRef]
11. Jones, J.I.; Vdovchenko, A.; Cooling, D.; Murphy, J.F.; Arnold, A.; Pretty, J.L.; Spencer, K.L.; Markus, A.A.; Vethaak, A.D.; Resmini, M. Systematic Analysis of the Relative Abundance of Polymers Occurring as Microplastics in Freshwaters and Estuaries. *Int. J. Environ. Res. Public Health* **2020**, *17*, 9304. [CrossRef]
12. Efimova, I.; Bagaeva, M.; Bagaev, A.; Kileso, A.; Chubarenko, I.P. Secondary Microplastics Generation in the Sea Swash Zone With Coarse Bottom Sediments: Laboratory Experiments. *Front. Mar. Sci.* **2018**, *5*, 313. [CrossRef]
13. Zhang, K.; Hamidian, A.H.; Tubić, A.; Zhang, Y.; Fang, J.K.H.; Wu, C.; Lam, P.K.S. Understanding plastic degradation and microplastic formation in the environment: A review. *Environ. Pollut.* **2021**, *274*, 116554. [CrossRef]
14. Pop, C.-E.; Draga, S.; Măciucă, R.; Niță, R.; Crăciun, N.; Wolff, R. Bisphenol A Effects in Aqueous Environment on Lemna minor. *Processes* **2021**, *9*, 1512. [CrossRef]
15. Bhatnagar, A.; Anastopoulos, I. Adsorptive removal of bisphenol A (BPA) from aqueous solution: A review. *Chemosphere* **2017**, *168*, 855–902. [CrossRef]
16. Boyle, K.; Örmeci, B. Microplastics and Nanoplastics in the Freshwater and Terrestrial Environment: A Review. *Water* **2020**, *12*, 2633. [CrossRef]
17. Sun, Q.; Ren, S.Y.; Ni, H.G. Incidence of microplastics in personal care products: An appreciable part of plastic pollution. *Sci. Total Environ.* **2020**, *742*, 140218. [CrossRef]
18. Carr, S.A.; Liu, J.; Tesoro, A.G. Transport and fate of microplastic particles in wastewater treatment plants. *Water Res.* **2016**, *91*, 174–182. [CrossRef]
19. Talvitie, J.; Mikola, A.; Setälä, O.; Heinonen, M.; Koistinen, A. How well is microlitter purified from wastewater?-A detailed study on the stepwise removal of microlitter in a tertiary level wastewater treatment plant. *Water Res.* **2017**, *109*, 164–172. [CrossRef] [PubMed]
20. Simon, M.; Van Alst, N.; Vollertsen, J. Quantification of microplastic mass and removal rates at wastewater treatment plants applying Focal Plane Array (FPA)-based Fourier Transform Infrared (FT-IR) imaging. *Water Res.* **2018**, *142*, 1–9. [CrossRef]
21. Sun, J.; Dai, X.; Wang, Q.; van Loosdrecht, M.C.M.; Ni, B.J. Microplastics in wastewater treatment plants: Detection, occurrence and removal. *Water Res.* **2019**, *152*, 21–37. [CrossRef]
22. Mason, S.A.; Garneau, D.; Sutto, R.; Chu, Y.; Ehmann, K.; Barnes, J.; Fink, P.; Papazissimos, D.; Rogers, D.L. Microplastic pollution is widely detected in US municipal wastewater treatment plant effluent. *Environ. Pollut.* **2016**, *218*, 1045–1054. [CrossRef]
23. Wang, W.; Ndungu, A.W.; Li, Z.; Wang, J. Microplastics pollution in inland freshwaters of China: A case study in urban surface waters of Wuhan, China. *Sci. Total Environ.* **2017**, *575*, 1369–1374. [CrossRef] [PubMed]
24. Wang, T.; Wang, L.; Chen, Q.; Kalogerakis, N.; Ji, R.; Ma, Y. Interactions between microplastics and organic pollutants: Effects on toxicity, bioaccumulation, degradation, and transport. *Sci. Total Environ.* **2020**, *748*, 142427. [CrossRef] [PubMed]

25. Piarulli, S.; Vanhove, B.; Comandini, P.; Scapinello, S.; Moens, T.; Vrielinck, H.; Sciutto, G.; Prati, S.; Mazzeo, R.; Booth, A.M.; et al. Do different habits affect microplastics contents in organisms? A trait-based analysis on salt marsh species. *Mar. Pollut. Bull.* **2020**, *153*, 110983. [CrossRef]
26. Avio, C.G.; Gorbi, S.; Regoli, F. Plastics and microplastics in the oceans: From emerging pollutants to emerged threat. *Mar. Environ. Res.* **2017**, *128*, 2–11. [CrossRef] [PubMed]
27. Isobe, A.; Iwasaki, S.; Uchida, K.; Tokai, T. Abundance of non-conservative microplastics in the upper ocean from 1957 to 2066. *Nat. Commun.* **2019**, *10*, 417. [CrossRef]
28. Kim, H.J.; Lee, J.Y. Emerging Concerns about Microplastic Pollution on Groundwater in South Korea. *Sustainability* **2020**, *12*, 5275. [CrossRef]
29. Lebreton, L.C.M.; van der Zwet, J.; Damsteeg, J.-W.; Slat, B.; Andrady, A.; Reisser, J. River plastic emissions to the world's oceans. *Nat. Commun.* **2017**, *8*, 15611. [CrossRef]
30. Eriksen, M.; Lebreton, L.C.M.; Carson, H.S.; Thiel, M.; Moore, C.J.; Borerro, J.C.; Galgani, F.; Ryan, P.G.; Reisser, J. Plastic Pollution in the World's Oceans: More than 5 Trillion Plastic Pieces Weighing over 250,000 Tons Afloat at Sea. *PLoS ONE* **2014**, *9*, e111913. [CrossRef]
31. Bakir, A.; Rowland, S.; Thompson, R. Enhanced desorption of persistent organic pollutants from microplastics under simulated physiological conditions. *Environ. Pollut.* **2014**, *185*, 16–23. [CrossRef]
32. Isobe, A.; Uchida, K.; Tokai, T.; Iwasaki, S. East Asian seas: A hot spot of pelagic microplastics. *Mar. Pollut. Bull.* **2015**, *101*, 618–623. [CrossRef]
33. Russell, M.; Webster, L. Microplastics in sea surface waters around Scotland. *Mar. Pollut. Bull.* **2021**, *116*, 112210. [CrossRef]
34. Pan, Z.; Guo, H.; Chen, H.; Wang, S.; Sun, X.; Zou, Q.; Zhang, Y.; Lin, H.; Cai, S.; Huang, J. Microplastics in Northwestern Pacific: Abundance, distribution, and characteristics. *Sci. Total Environ.* **2019**, *650*, 1913–1922. [CrossRef] [PubMed]
35. Lindeque, P.K.; Cole, M.; Coppock, R.L.; Lewis, C.N.; Miller, R.Z.; Watts, A.J.R.; Wilson-McNeal, A.; Wright, S.L.; Galloway, T.S. Are we underestimating microplastic abundance in the marine environment? A comparison of microplastic capture with nets of different mesh-size. *Environ. Pollut.* **2020**, *265*, 114721. [CrossRef] [PubMed]
36. Jones, K.L.; Hartl, M.G.J.; Bell, M.C.; Capper, A. Microplastic accumulation in a *Zostera marina* L. bed at Deerness Sound, Orkney, Scotland. *Mar. Pollut. Bull.* **2020**, *152*, 110883. [CrossRef] [PubMed]
37. Seng, N.; Lai, S.; Fong, J.; Saleh, M.F.; Cheng, C.; Cheok, Z.Y.; Todd, P.A. Early evidence of microplastics on seagrass and macroalgae. *Mar. Freshw. Res.* **2020**, *71*, 922–928. [CrossRef]
38. Goss, H.; Jaskiel, J.; Rotjan, R. Thalassia testudinum as a potential vector for incorporating microplastics into benthic marine food webs. *Mar. Pollut. Bull.* **2018**, *135*, 1085–1089. [CrossRef]
39. Cozzolino, L.; Nicastro, K.R.; Zardi, G.I.; de Los Santos, C. B Species-specific plastic accumulation in the sediment and canopy of coastal vegetated habitats. *Sci. Total Environ.* **2020**, *723*, 138018. [CrossRef]
40. Tibbetts, J.; Krause, S.; Lynch, I.; Smith, G.H.S. Abundance Distribution and Divers of Microplastic Contamination in Urban River Environments. *Water* **2018**, *10*, 1597. [CrossRef]
41. Park, T.-J.; Lee, S.-H.; Lee, M.-S.; Lee, J.-K.; Park, J.-H.; Zoh, K.-D. Distributions of Microplastics in Surface Water, Fish, and Sediment in the Vicinity of a Sewage Treatment Plant. *Water* **2020**, *12*, 3333. [CrossRef]
42. Kosuth, M.; Mason, S.A.; Wattenberg, E.V. Anthropogenic contamination of tap water, beer, and sea salt. *PLoS ONE* **2018**, *13*, e0194970. [CrossRef]
43. Mason, S.A.; Welch, V.G.; Neratko, J. Synthetic Polymer Contamination in Bottled Water. *Front. Chem.* **2018**, *6*, 407. [CrossRef] [PubMed]
44. Ockelford, A.; Cundy, A.; Ebdon, J.E. Storm Response of Fluvial Sedimentary Microplastics. *Sci. Rep.* **2020**, *10*, 1865. [CrossRef]
45. Galloway, T.; Cole, M.; Lewis, C. Interactions of microplastic debris throughout the marine ecosystem. *Nat. Ecol. Evol.* **2017**, *1*, 0116. [CrossRef] [PubMed]
46. Vermeiren, P.; Muñoz, C.C.; Ikejima, K. Sources and sinks of plastic debris in estuaries: A conceptual model integrating biological, physical and chemical distribution mechanisms. *Mar. Pollut. Bull.* **2016**, *113*, 7–16. [CrossRef] [PubMed]
47. Duis, K.; Coors, A. Microplastics in the aquatic and terrestrial environment: Sources (with a specific focus on personal care products), fate and effects. *Environ. Sci. Eur.* **2016**, *28*, 2. [CrossRef]
48. Hitchcock, J.N. Storm events as key moments of microplastic contamination in aquatic ecosystems. *Sci. Total Environ.* **2020**, *734*, 139436. [CrossRef]
49. Van den Berg, P.; Huerta-Lwanga, E.; Corradini, F.; Geissen, V. Sewage sludge application as a vehicle for microplastics in eastern Spanish agricultural soils. *Environ. Poll.* **2020**, *261*, 114198. [CrossRef]
50. Huang, Y.; Liu, Q.; Jia, W.; Yan, C.; Wang, J. Agricultural plastic mulching as a source of microplastics in the terrestrial environment. *Environ. Pollut.* **2020**, *260*, 114096. [CrossRef]
51. Weithmann, N.; Möller, J.N.; Löder, M.G.J.; Piehl, S.; Laforsch, C.; Freitag, R. Organic fertilizer as a vehicle for the entry of microplastic into the environment. *Sci. Adv.* **2018**, *4*, eaap8060. [CrossRef]
52. Dioses-Salinas, D.C.; Pizarro-Ortega, C.I.; De-la-Torre, G.E. A methodological approach of the current literature on microplastic contamination in terrestrial environments: Current knowledge and baseline considerations. *Sci. Total Environ.* **2020**, *730*, 139164. [CrossRef]

53. Wong, G.; Löwemark, L.; Kunz, A. Microplastic pollution of the Tamsui River and its tributaries in northern Taiwan: Spatial heterogeneity and correlation with precipitation. *Environ. Pollut.* **2020**, *260*, 113935. [CrossRef]
54. Fuller, S.; Gautam, A. A Procedure for Measuring Microplastics using Pressurized Fluid Extraction. *Environ. Sci. Technol.* **2016**, *50*, 5774–5780. [CrossRef]
55. Scheurer, M.; Bigalke, M. Microplastics in Swiss Floodplain Soils. *Environ. Sci. Technol.* **2018**, *52*, 3591–3598. [CrossRef]
56. Corradini, F.; Meza, P.; Equiluz, R.; Casado, F.; Huerta-Lwanga, E.; Geissen, V. Evidence of microplastic accumulation in agricultural soils from sewage sludge disposal. *Sci. Total Environ.* **2019**, *671*, 411–420. [CrossRef] [PubMed]
57. Harris, P.T. Review: The fate of microplastic in marine sedimentary environments: A review and synthesis. *Mar. Poll. Bull.* **2020**, *158*, 111398. [CrossRef]
58. Yukioka, S.; Tanaka, S.; Nabetani, Y.; Suzuki, Y.; Ushijima, T.; Fujii, S.; Takada, H.; Van Tran, Q.; Singh, S. Occurrence and characteristics of microplastics in surface road dust in Kusatsu (Japan), Da Nang (Vietnam), and Kathmandu (Nepal). *Environ. Pollut.* **2020**, *256*, 113447. [CrossRef] [PubMed]
59. Rhodes, C.J. Plastic pollution and potential solutions. *Sci. Prog.* **2018**, *101*, 207–260. [CrossRef] [PubMed]
60. Vogelsang, C.; Lusher, A.; Sundvor, I.; Umar, M. *Microplastics in Road Dust-Characteristics, Pathways and Measures*; Technical Report, M-959; Norwegian Institute for Water Research: Oslo, Norway, 2018; Available online: https://www.miljodirektoratet.no/globalassets/publikasjoner/M959/M959.pdf (accessed on 10 July 2021).
61. Liu, C.; Li, J.; Zhang, Y.; Wang, L.; Deng, J.; Gao, Y.; Yu, L.; Zhang, J.; Sun, H. Widespread distribution of PET and PC microplastics in dust in urban China and their estimated human exposure. *Environ. Int.* **2019**, *128*, 116–124. [CrossRef]
62. Eriksen, M.; Thiel, M.; Prindiville, M.; Kiessling, T. Microplastic: What Are the Solution. *Freshw. Microplast.* **2018**, *58*, 273–298.
63. Zhou, R.; Lu, G.; Yan, Z.; Jiang, R.; Bao, X.; Lu, P. A review of the influences of microplastics on toxicity and transgenerational effects of pharmaceutical and personal care products in aquatic environment. *Sci. Total Environ.* **2020**, *732*, 139222. [CrossRef] [PubMed]
64. Webber, A.; van Randow, M.; Voigt, A.-L.; van der Au, M.; Fischer, E.; Meermann, B.; Wagner, M. Ingestion and toxicity of microplastics in the freshwater gastropod Lymnaea stagnalis: No microplastic-induced effects alone or in combination with copper. *Chemosphere* **2021**, *263*, 128040. [CrossRef] [PubMed]
65. Gao, F.; Li, J.; Sun, C.; Zhang, L.; Jiang, F.; Cao, W.; Zheng, L. Study on the capability and characteristics of heavy metals enriched on microplastics in marine environment. *Mar. Poll. Bull.* **2019**, *144*, 61–67. [CrossRef] [PubMed]
66. Xu, K.; Zhang, Y.; Huang, Y.; Wang, J. Toxicological effects of microplastics and phenanthrene to zebrafish (Danio rerio). *Sci. Total Environ.* **2021**, *757*, 143730. [CrossRef] [PubMed]
67. Lu, K.; Qiao, R.; An, H.; Zhang, Y. Influence of microplastics on the accumulation and chronic toxic effects of cadmium in zebrafish (Danio rerio). *Chemosphere* **2018**, *202*, 504–520. [CrossRef] [PubMed]
68. Yang, W.; Gao, X.; Wu, Y.; Wan, L.; Tan, L.; Yuan, S.; Ding, H.; Zhang, W. The combined toxicity influence of microplastics and nonylphenol on microalgae *Chlorella pyrenoidosa*. *Ecotoxicol. Environ. Saf.* **2020**, *195*, 110484. [CrossRef]
69. Stock, V.; Böhmert, L.; Lisicki, E.; Block, R.; Cara-Carmona, J.; Pack, L.K.; Selb, R.; Lichtenstein, D.; Voss, L.; Henderson, C.J.; et al. Uptake and effects of orally ingested polystyrene microplastic particles in vitro and in vivo. *Arch. Toxicol.* **2019**, *93*, 1817–1833. [CrossRef]
70. Wang, Q.; Bai, J.; Ning, B.; Fan, L.; Sun, T.; Fang, Y.; Wu, J.; Li, S.; Duan, C.; Zhang, Y.; et al. Effects of bisphenol A and nanoscale and microscale polystyrene plastic exposure on particle uptake and toxicity in human Caco-2 cells. *Chemosphere* **2020**, *254*, 126788. [CrossRef]
71. Wang, Y.; Qian, H. Phthalates and Their Impacts on Human Health. *Healthcare* **2021**, *9*, 603. [CrossRef]
72. Adam, V.; Yang, T.; Nowack, B. Toward an ecotoxicological risk assessment of microplastics: Comparison of available hazard and exposure data in freshwaters. *Environ. Toxicol. Chem.* **2018**, *38*, 436–447. [CrossRef]
73. Besseling, E.; Redondo-Hasselerharm, P.; Foekema, E.M.; Koelmans, A.A. Quantifying ecological risks of aquatic micro-and nanoplastic. *Crit. Rev. Environ. Sci. Technol.* **2019**, *49*, 32–80. [CrossRef]
74. Xu, P.; Peng, G.; Su, L.; Gao, Y.; Li, D. Microplastic risk assessment in surface waters: A case study in the Changjiang Estuary, China. *Mar. Poll. Bull.* **2018**, *113*, 647–654. [CrossRef] [PubMed]
75. Zhou, Y.; Wang, J.; Zou, M.; Jia, Z.; Zhou, S.; Li, Y. Microplastics in soils: A review of methods, occurrence, fate, transport, ecological and environmental risks. *Sci. Total Environ.* **2020**, *748*, 141368. [CrossRef] [PubMed]
76. Dey, T.K.; Uddin, M.E.; Jamal, M. Detection and removal of microplastics in wastewater: Evolution and impact. *Environ. Sci. Pollut. Res.* **2021**, *28*, 16925–16947. [CrossRef] [PubMed]
77. Campanale, C.; Savino, I.; Pojar, I.; Massarelli, C.; Uricchio, V.F. A Practical Overview of Methodologies for Sampling and Analysis of Microplastics in Riverine Environments. *Sustainability* **2020**, *12*, 6755. [CrossRef]
78. Barrows, A.P.W.; Neumann, C.A.; Berger, M.L.; Shaw, S.D. Grab vs. neuston tow net: A microplastic sampling performance comparison and possible advances in the field. *Anal. Methods* **2017**, *9*, 1446–1453. [CrossRef]
79. Cutroneo, L.; Reboa, A.; Besio, G.; Borgogno, F.; Canesi, L.; Canuto, S.; Dara, M.; Enrile, F.; Forioso, I.; Greco, G.; et al. Microplastics in seawater: Sampling strategies, laboratory methodologies, and identification techniques applied to port environment. *Environ. Sci. Pollut. Res.* **2020**, *27*, 8938–8952. [CrossRef]
80. Abeynayaka, A.; Kojima, F.; Miwa, Y.; Ito, N.; Nihei, Y.; Fukunaga, Y.; Yashima, Y.; Itsubo, N. Rapid Sampling of Suspended and Floating Microplastics in Challenging Riverine and Coastal Water Environments in Japan. *Water* **2020**, *12*, 1903. [CrossRef]

81. Von Ammon, U.; Jeffs, A.; Zaiko, A.; van der Resi, A.; Goodwin, D.; Beckley, L.E.; Malpot, E.; Pochon, X. A Portable Cruising Speed Net: Expanding Global Collection of Sea Surface Plankton Data. *Front. Mar. Sci.* **2020**, *7*, 1109. [CrossRef]
82. Vermaire, J.C.; Pomeroy, C.; Herczegh, S.M.; Haggart, O.; Murphy, M. Microplastic Abundance and Distribution in the Open Water and Sediment of the Ottawa River, Canada, and Its Tributaries. *Facets* **2017**, *2*, 301–314. [CrossRef]
83. Dris, R.; Gasperi, J.; Rocher, V.; Saad, M.; Renault, N.; Tassin, B. Microplastic contamination in an urban area: A case study in Greater Paris. *Environ. Chem.* **2015**, *12*, 592–599. [CrossRef]
84. SubCtech GmbH Subsea Technologies for the Marine Environment (Microplastic Sampler). Available online: https://subctech.com/wp-content/uploads/2020/09/SpecSheet_SubCtech_Microplastic-Sampler_ENG_v1-0.pdf (accessed on 23 June 2021).
85. Yano, K.A.V.; Reyes, N.J.D.G.; Geronimo, F.K.F.; Jeon, M.S.; Kim, Y.; Kim, L.H. A comprehensive review of microplastics: Sources, pathways, and implications. *J. Wetl. Res.* **2020**, *22*, 153–160. [CrossRef]
86. Vereiren, P.; Lercari, D.; Muñoz, C.C.; Ikejima, K.; Celentano, E.; Jorge-Romero, G.; Defeo, O. Sediment grain size determines microplastic exposure landscapes for sandy beach macroinfauna. *Environ. Pollut.* **2021**, *286*, 117308. [CrossRef]
87. Willis, K.A.; Eriksen, R.; Wilcox, C.; Hardesty, B.D. Microplastic Distribution at Different Sediment Depths in an Urban Estuary. *Front. Mar. Sci.* **2017**, *4*, 419. [CrossRef]
88. Kaile, N.; Lindivat, M.; Elio, J.; Thuestad, G.; Crowley, Q.G.; Hoell, I.A. Preliminary Results From Detection of Microplastics in Liquid Samples Using Flow Cytometry. *Front. Mar. Sci.* **2020**, *7*, 856. [CrossRef]
89. Carter, M.R.; Gregorich, E.G. *Soil Sampling and Methods of Analysis*, 2nd ed.; Taylor & Francis Group: Abingdon, UK, 2006; Available online: https://www.niordc.ir/uploads/86_106_Binder1.pdf (accessed on 13 July 2021).
90. Thomas, D.; Schütze, B.; Heinze, M.W.; Steinmetz, Z. Sample Preparation Techniques for the Analysis of Microplastics in Soil-A Review. *Sustainability* **2020**, *12*, 9074. [CrossRef]
91. Hurley, R.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]
92. Lusher, A.L.; Munno, K.; Hermabessiere, L.; Carr, S. Isolation and Extraction of Microplastics from Environmental Samples: An Evaluation of Practical Approaches and Recommendations for Further Harmonization. *Appl. Spectrosc.* **2020**, *74*, 1049–1065. [CrossRef]
93. Nguyen, B.; Claveau-Mallet, D.; Hernandez, L.M.; Xu, E.G.; Farner, J.M.; Tufenkji, N. Separation and Analysis of Microplastics and Nanoplastics in Complex Environmental Samples. *Acc. Chem. Res.* **2019**, *52*, 858–866. [CrossRef]
94. Renner, G.; Nellessen, A.; Schwiers, A.; Wenzel, M.; Schmidt, C.T.; Schram, J. Hydrophobicity–water/air–based enrichment cell for microplastics analysis within environmental samples: A proof of concept. *MethodsX* **2020**, *7*, 100732. [CrossRef]
95. Al-Azzawi, M.S.M.; Kefer, S.; Weißer, J.; Reichel, J.; Schwaller, C.; Glas, K.; Knoop, O.; Drewes, J.E. Validation of Sample Preparation Methods for Microplastic Analysis in Wastewater Matrices-Reproducibility and Standardization. *Water* **2020**, *12*, 2445. [CrossRef]
96. Yang, L.; Zhang, Y.; Kang, S.; Wang, Z.; Wu, C. Microplastics in freshwater sediment: A review on methods, occurrence, and sources. *Sci. Total Environ.* **2021**, *754*, 141948. [CrossRef] [PubMed]
97. Radford, F.; Zapata-Restrepo, L.M.; Horton, A.A.; Hudson, M.D.; Shaw, P.J.; Williams, I.D. Developing a systematic method for extraction of microplastics in soils. *Anal. Methods* **2021**, *13*, 1695–1705. [CrossRef]
98. Cole, M.; Webb, H.; Lindeque, P.K.; Fileman, E.S.; Halsband, C.; Galloway, T.S. Isolation of microplastics in biota-rich seawater samples and marine organisms. *Sci. Rep.* **2014**, *4*, 4528. [CrossRef] [PubMed]
99. Kang, H.; Park, S.; Lee, B.; Ahn, J.; Kim, S. Modification of a Nile Red Staining Method for Microplastics Analysis: A Nile Red Plate Method. *Water* **2020**, *12*, 3251. [CrossRef]
100. Shim, W.J.; Song, Y.K.; Hong, S.H.; Jang, M. Identification and quantification of microplastics using Nile Red staining. *Mar. Pollut. Bull.* **2016**, *113*, 469–476. [CrossRef]
101. Hebner, T.S.; Maurer-Jones, M.A. Characterizing microplastic size and morphology of photodegraded polymers placed in simulated moving water conditions. *Environ. Sci. Process. Impacts* **2020**, *22*, 398–407. [CrossRef] [PubMed]
102. Kumar, B.N.V.; Löschel, L.A.; Imhof, H.K.; Löder, M.G.J.; Laforsch, C. Analysis of microplastics of a broad size range in commercially important mussels by combining FTIR and Raman spectroscopy approaches. *Environ. Pollut.* **2021**, *269*, 116147. [CrossRef]
103. Xu, J.-L.; Thomas, K.V.; Luo, Z.; Gowen, A.A. FTIR and Raman imaging for microplastics analysis: State of the art, challenges and prospects. *TrAC Trends Anal. Chem.* **2019**, *119*, 115629. [CrossRef]
104. Vianello, A. A Journey into Microplastic Analysis Using FTIR Spectroscopy. Ph.D. Thesis, Aalborg University, Aalborg, Denmark, 2020. Available online: https://vbn.aau.dk/en/publications/a-journey-into-microplastic-analysis-using-ftir-spectroscopy (accessed on 6 May 2021).
105. Rocchia, M.; Ruff, I.; Vianello, A. Microplastic Identification and Characterization by Raman Imaging Spectroscopy. In *Thermoscientific White Paper*; Thermoscientific: Milan, Italy, 2017; WP52981_E_10/17M; Available online: https://www.gammadata.se/assets/Uploads/Microplastic-and-Raman.pdf (accessed on 10 May 2021).
106. Müller, A.; Goedecke, C.; Eisentraut, P.; Piechotta, C.; Braun, U. Microplastic analysis using chemical extraction followed by LC-UV analysis: A straightforward approach to determine PET content in environmental samples. *Environ. Sci. Eur.* **2020**, *32*, 85. [CrossRef]

107. Wang, L.; Zhang, J.; Hou, S.; Sun, H. A Simple Method for Quantifying Polycarbonate and Polyethylene Terephthalate Microplastics in Environmental Samples by Liquid Chromatography–Tandem Mass Spectrometry. *Environ. Sci. Technol. Lett.* **2017**, *4*, 530–534. [CrossRef]
108. Funck, M.; Yildirim, A.; Nickel, C.; Schram, J.; Schmidt, T.C.; Tuerka, J. Identification of microplastics in wastewater after cascade filtration using Pyrolysis-GC–MS. *MethodsX* **2020**, *7*, 100778. [CrossRef] [PubMed]
109. Primpke, S.; Fischer, M.; Lorenz, C.; Gerdts, G.; Scholz-Böttcher, B.M. Comparison of pyrolysis gas chromatography/mass spectrometry and hyperspectral FTIR imaging spectroscopy for the analysis of microplastics. *Anal. Bioanal. Chem.* **2020**, *412*, 8283–8298. [CrossRef] [PubMed]
110. Reichel, J.; Graßmann, J.; Letzel, T.; Drewes, J.E. Systematic Development of a Simultaneous Determination of Plastic Particle Identity and Adsorbed Organic Compounds by Thermodesorption—Pyrolysis GC/MS (TD-Pyr-GC/MS). *Molecules* **2020**, *25*, 4985. [CrossRef] [PubMed]
111. Asamoah, B.O.; Kanyathare, B.; Russey, M.; Peiponen, K.-E. A prototype of a portable optical sensor for the detection of transparent and translucent microplastics in freshwater. *Chemosphere* **2019**, *231*, 161–167. [CrossRef]
112. Valsesia, A.; Parot, J.; Ponti, J.; Mehn, D.; Marino, R.; Melillo, D.; Muramoto, S.; Verkouteren, M.; Hackley, V.A.; Colpo, P. Detection, counting and characterization of nanoplastics in marine bioindicators: A proof of principle study. *Microplast. Nanoplast.* **2021**, *1*, 1–13. [CrossRef]
113. Lambert, S.; Wagner, M. Characterisation of nanoplastics during the degradation of polystyrene. *Chemosphere* **2016**, *145*, 265–268. [CrossRef]
114. Karakolis, E.G.; Nguyen, B.; You, J.B.; Rochman, C.M.; Sinton, D. Fluorescent Dyes for Visualizing Microplastic Particles and Fibers in Laboratory-Based Studies. *Environ. Sci. Technol. Lett.* **2019**, *6*, 334–340. [CrossRef]
115. Luo, H.; Xiang, Y.; Zhao, Y.; Li, Y.; Pan, X. Nanoscale infrared, thermal and mechanical properties of aged microplastics revealed by an atomic force microscopy coupled with infrared spectroscopy (AFM-IR) technique. *Sci. Total Environ.* **2020**, *774*, 140944. [CrossRef]
116. Tagg, A.S.; Sapp, M.; Harrison, J.P.; Ojeda, J.J. Identification and Quantification of Microplastics in Wastewater Using Focal Plane Array-Based Reflectance Micro-FT-IR Imaging. *Anal. Chem.* **2015**, *87*, 6032–6040. [CrossRef]
117. Piehl, S.; Leibner, A.; Löder, M.G.J.; Dris, R.; Bogner, C.; Laforsch, C. Identification and quantification of macro- and microplastics on an agricultural farmland. *Sci. Rep.* **2018**, *8*, 17950. [CrossRef]
118. Scopetani, C.; Chelazzi, D.; Cincinelli, A.; Esterhuizen-Londt, M. Assessment of microplastic pollution: Occurrence and characterisation in Vesijärvi lake and Pikku Vesijärvi pond, Finland. *Environ. Monit. Assess.* **2019**, *191*, 652. [CrossRef]
119. Kunz, A.; Walther, B.A.; Lowemark, L.; Lee, Y.C. Distribution and quantity of microplastic on sandy beaches along the northern coast of Taiwan. *Mar. Pollut. Bull.* **2016**, *111*, 126–135. [CrossRef]
120. Karthik, R.; Robin, R.S.; Purvaja, R.; Ganguly, D.; Anandavelu, I.; Raghuraman, R.; Hariharan, G.; Ramakrishna, A.; Ramesh, R. Microplastics along the beaches of southeast coast of India. *Sci. Total Environ.* **2018**, *645*, 1388–1399. [CrossRef]
121. Levermore, J.M.; Smith, T.E.L.; Kelly, F.J.; Wright, S.L. Detection of Microplastics in Ambient Particulate Matter Using Raman Spectral Imaging and Chemometric Analysis. *Anal. Chem.* **2020**, *92*, 8732–8740. [CrossRef] [PubMed]
122. Zhao, S.; Danley, M.; Ward, J.E.; Li, D.; Mincer, T.J. An approach for extraction, characterization and quantitation of microplastic in natural marine snow using Raman microscopy. *Anal. Methods* **2016**, *9*, 1470–1478. [CrossRef]
123. Saur, T. Global Assessment of Microplastic Pollution in Wastewater Treatment Plants. OECD Workshop on Microplastics from Synthetic Textiles in the Environment: Knowledge, Mitigation and Policy. 11 February 2020. Available online: https://www.oecd.org/water/Draft_Agenda_Public_OECD_Workshop_MP_Textile.pdf (accessed on 11 May 2021).
124. Hendrickson, E.; Minor, E.C.; Schreiner, K. Microplastic Abundance and Composition in Western Lake Superior As Determined via Microscopy, Pyr-GC/MS, and FTIR. *Environ. Sci. Technol.* **2018**, *52*, 1787–1796. [CrossRef] [PubMed]
125. Bratton, S.P.; Halbur, J.; Schreiner, K.; Minor, E.C.; Hendrickson, E.; Peters, C.A. Pyr-GC/MS analysis of microplastics extracted from the stomach content of benthivore fish from the Texas Gulf Coast. *Mar. Pollut. Bull.* **2018**, *137*, 91–95. [CrossRef]
126. Zhang, J.; Wang, L.; Kannan, K. Microplastics in house dust from 12 countries and associated human exposure. *Environ. Int.* **2020**, *134*, 105314. [CrossRef] [PubMed]
127. Santos, D.I.; Correia, M.J.N.; Mateus, M.M.; Saraiva, J.A.; Vicente, A.A.; Moldão, M. Fourier Transform Infrared (FT-IR) Spectroscopy as a Possible Rapid Tool to Evaluate Abiotic Stress Effects on Pineapple By-Product. *Appl. Sci.* **2019**, *9*, 4141. [CrossRef]
128. Ahmed, M.B.; Rahman, M.S.; Alom, J.; Hasan, M.S.; Johir, M.A.H.; Mondal, M.I.H.; Lee, D.-Y.; Park, J.; Zhou, J.L.; Yoon, M.H. Microplastic particles in the aquatic environment: A systematic review. *Sci. Total Environ.* **2021**, *775*, 145793. [CrossRef]
129. Wagner, M.; Scherer, C.; Alvarez-Muñnoz, D.; Brennholt, N.; Bourrain, X.; Buchinger, S.; Fries, E.; Grosbois, C.; Klasmeier, J.; Marti, T.; et al. Microplastics in freshwater ecosystems: What we know and what we need to know. *Environ. Sci. Eur.* **2014**, *26*, 12. [CrossRef]
130. Hidalgo-Ruz, V.; Gutow, L.; Thompson, R.C.; Thiel, M. Microplastics in the marine environment: A review of the methods used for identification and quantification. *Environ. Sci. Technol.* **2012**, *46*, 3060–3075. [CrossRef] [PubMed]
131. Olesen, K.B.; van Alst, N.; Simon, M.; Vianello, A.; Liu, F.; Vellertsen, J. Analysis of Microplastics using FTIR Imaging Application Note, Method January 2017. Available online: https://vbn.aau.dk/en/publications/analysis-of-microplastics-using-ftir-imaging-application-note (accessed on 12 May 2021).

132. 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] [PubMed]
133. Löder, M.G.J.; Gerdts, G. Methodology Used for the Detection and Identification of Microplastics—A Critical Appraisal. In *Marine Anthropogenic Litter*; Bergmann, M., Gutow, L., Klages, M., Eds.; Springer: Berlin/Heidelberg, Germany, 2015. [CrossRef]
134. Scircle, A.; Cizdziel, J.V.; Tisinger, L.; Anumol, T.; Robey, D. Occurrence of Microplastic Pollution at Oyster Reefs and Other Coastal Sites in the Mississippi Sound, USA: Impacts of Freshwater Inflows from Flooding. *Toxics* **2020**, *8*, 35. [CrossRef]
135. Ainali, N.M.; Kalaronis, D.; Kontogiannis, A.; Evgenidu, E.; Kyzas, G.Z.; Yang, X.; Bikiaris, D.N.; Lambropoulou, D.A. Microplastics in the environment: Sampling, pretreatment, analysis and occurrence based on current and newly-exploited chromatographic approaches. *Sci. Total Environ.* **2021**, *794*, 148725. [CrossRef] [PubMed]
136. Cowger, W.; Gray, A.; Christiansen, S.H.; DeFrond, H.; Deshpande, A.D.; Hemabessiere, L.; Lee, E.; Mill, L.; Munno, K.; Ossmann, B.E.; et al. Critical Review of Processing and Classification Techniques for Images and Spectra in Microplastic Research. *Appl. Spectrosc.* **2020**, *74*, 989–1010. [CrossRef] [PubMed]
137. Wu, M.; Tang, W.; Wu, S.; Liu, H.; Yang, C. Fate and effects of microplastics in wastewater treatment processes. *Sci. Total Environ.* **2020**, *757*, 143902. [CrossRef]
138. Herbort, A.F.; Sturm, M.T.; Schuhen, K. Technological Approaches for the Reduction of Microplastic Pollution in Seawater Desalination Plants and for Sea Salt Extraction. In *Plastics in the Environment*; IntechOpen: London, UK, 2019. [CrossRef]
139. Yahyanezhad, N.; Bardi, M.J.; Aminirad, H. An evaluation of microplastics fate in the wastewater treatment plants: Frequency and removal of microplastics by microfiltration membrane. *Water Pract. Technol.* **2021**, *16*, 782–792. [CrossRef]
140. Lv, X.; Dong, Q.; Zuo, Z.; Liu, Y.; Huang, X.; Wu, W.-M. Microplastics in a municipal wastewater treatment plant: Fate, dynamic distribution, removal efficiencies, and control strategies. *J. Clean. Prod.* **2019**, *225*, 579–586. [CrossRef]
141. Conley, K.; Clum, A.; Deepe, J.; Lane, H.; Beckingham, B. Wastewater treatment plants as a source of microplastics to an urban estuary: Removal efficiencies and loading per capita over one year. *Water Res. X* **2019**, *3*, 100030. [CrossRef] [PubMed]
142. Edo, C.; González-Pleiter, M.; Leganés, F.; Fernández-Piñas, F.; Rosal, R. Fate of microplastics in wastewater treatment plants and their environmental dispersion with effluent and sludge. *Environ. Pollut.* **2020**, *259*, 113837. [CrossRef] [PubMed]
143. Ziajahromi, S.; Neale, P.A.; Rintoul, L.; Leusch, F.D.L. Wastewater treatment plants as a pathway for microplastics: De-velopment of a new approach to sample wastewater-based microplastics. *Water Res.* **2017**, *112*, 93–99. [CrossRef]
144. Tagg, A.S.; Sapp, M.; Harrion, J.P.; Sinclair, C.J.; Bradley, E.; Nam, Y.J.; Ojeda, J.J. Microplastic Monitoring at Different Stages in a Wastewater Treatment Plant Using Reflectance Micro-FTIR Imaging. *Front. Environ. Sci.* **2020**, *8*, 145. [CrossRef]
145. Talvitie, J.; Mikola, A.; Koistinen, A.; Setälä, O. Solutions to microplastic pollution—Removal of microplastics from wastewater effluent with advanced wastewater treatment technologies. *Water Res.* **2017**, *123*, 401–407. [CrossRef]
146. Ross, P.S. Tackling Microfiber Pollution at Source a Solution-Oriented Partnership Across Public and Private Sectors. OECD Workshop on Microplastics from Synthetic Textiles in the Environment: Knowledge, Mitigation and Policy. 11 February 2020. Available online: https://www.oecd.org/water/Draft_Agenda_Public_OECD_Workshop_MP_Textile.pdf (accessed on 12 May 2021).
147. Lare, M.; Ncibi, M.C.; Sillanpää, 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] [PubMed]
148. Yang, L.; Li, K.; Cui, S.; Kang, Y.; An, L.; Lei, K. Removal of microplastics in municipal sewage from China's largest water reclamation plant. *Water Res.* **2019**, *155*, 175–181. [CrossRef]
149. Simon, M.; Vianello, A.; Vollertsen, J. Removal of >10 μm Microplastic Particles from Treated Wastewater by a Disc Filter. *Water* **2019**, *11*, 1935. [CrossRef]
150. Wolff, S.; Weber, F.; Kerpen, J.; Winklhofer, M.; Engelhart, M.; Backman, L. Elimination of Microplastics by Downstream Sand Filters in Wastewater Treatment. *Water* **2021**, *13*, 33. [CrossRef]
151. Bayo, J.; Lopez-Castellanos, J.; Olmos, S. Abatement of microplastics from Municipal effluents by two different wastewater treatment technologies. *Trans. Ecol. Environ.* **2020**, *242*, 15–26. [CrossRef]
152. Pizzichetti, A.R.P.; Pablos, C.; Álvarez-Fernández, C.; Reynolds, K.; Stanley, S.; Marugán, J. Evaluation of membranes performance for microplastic removal in a simple and low-cost filtration system. *Case Stud. Chem. Environ. Eng.* **2021**, *3*, 100075. [CrossRef]
153. Li, Q.-C.; Lai, Y.-J.; Yu, S.-J.; Li, P.; Zhou, X.-X.; Dong, L.-J.; Liu, X.; Yao, Z.-W.; Liu, J.-F. Sequential Isolation of Microplastics and Nanoplastics in Environmental Waters by Membrane Filtration, Followed by Cloud-Point Extraction. *Anal. Chem.* **2021**, *93*, 4559–4566. [CrossRef]
154. Kim, K.T.; Park, S.H. Enhancing Microplastics Removal from Wastewater Using Electro-Coagulation and Granule-Activated Carbon with Thermal Regeneration. *Processes* **2021**, *9*, 617. [CrossRef]
155. Lapointe, M.; Farner, J.M.; Hernandez, L.M.; Tufenkji, N. Understanding and Improving Microplastic Removal during Water Treatment: Impact of Coagulation and Flocculation. *Environ. Sci. Technol.* **2020**, *54*, 8719–8727. [CrossRef] [PubMed]
156. Perren, W.; Wojtasik, A.; Cai, Q. Removal of Microbeads from Wastewater Using Electrocoagulation. *ACS Omega* **2018**, *3*, 3357–3364. [CrossRef] [PubMed]
157. Park, J.W.; Lee, S.J.; Hwang, D.Y.; Seo, S. Removal of microplastics via tannic acid-mediated coagulation and in vitro impact assessment. *RSC Adv.* **2021**, *11*, 3556–3566. [CrossRef]

158. Ma, B.; Xue, W.; Hu, C.; Liu, H.; Qu, J.; Li, L. Characteristics of microplastic removal via coagulation and ultrafiltration during drinking water treatment. *Chem. Eng. J.* **2019**, *359*, 159–167. [CrossRef]
159. Liu, H.; Zhou, X.; Ding, W.Q.; Nghiem, L.D.; Sun, J.; Wang, Q.L. Do microplastics affect biological wastewater treatment performance? Implications from bacterial activity experiments. *ACS Sustain. Chem. Eng.* **2019**, *7*, 20097–20101. [CrossRef]
160. Cunha, C.; Silva, L.; Paulo, J.; Faria, M.; Nogueira, N.; Cordeiro, N. Microalgal-based biopolymer for nano- and microplastic removal: A possible biosolution for wastewater treatment. *Environ. Pollut.* **2020**, *263*, 114385. [CrossRef]
161. Canniff, P.M.; Hoang, T.C. Microplastic ingestion by Daphina magna and its enhancement on algal growth. *Sci. Total Environ.* **2018**, *633*, 500–507. [CrossRef] [PubMed]
162. Liu, W.; Zhang, J.; Liu, H.; Guo, X.; Zhang, X.; Yao, X.; Cao, Z.; Zhang, T. A review of the removal of microplastics in global wastewater treatment plants: Characteristics and mechanisms. *Environ. Int.* **2021**, *146*, 106277. [CrossRef]