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Microplastic Particles in the Aquatic Environment: A Systematic Review

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Abstract

Microplastics (MPs) pollution has become one of the most severe environmental concerns today. MPs persist in the environment and cause adverse effects in organisms. This review aims to present a state-of-the-art overview of MPs in the aquatic environment. Personal care products, synthetic clothing, air-blasting facilities and drilling fluids from gas-oil industries, raw plastic powders from plastic manufacturing industries, waste plastic products and wastewater treatment plants act as the major sources of MPs. For MPs analysis, pyrolysis-gas chromatography-mass spectrometry (Py-GC-MS), Py-MS methods, Raman spectroscopy, and FT-IR spectroscopy are regarded as the most promising methods for MPs identification and quantification. Due to the large surface area to volume ratio, crystallinity, hydrophobicity and functional groups, MPs can interact with various contaminants such as heavy metals, antibiotics and persistent organic contaminants. Among different physical and biological treatment technologies, the MPs removal performance decreases as membrane bioreactor (> 99%) > activated sludge process (~98%) > rapid sand filtration (~97.1%) > dissolved air floatation (~95%) > electrocoagulation (> 90%) > constructed wetlands (88%). Chemical treatment methods such as coagulation, magnetic separations, Fenton, photo-Fenton and photocatalytic degradation also show moderate to high efficiency of MP removal. Hybrid treatment technologies show the highest removal efficacies of MPs. Finally, future research directions for MPs are elaborated.

***Keywords**:* Microplastics analysis; Contaminant interactions; Physical treatment; Chemical technology; Biological process

1. Introduction

Plastics have found their importance for improving human lives due to their long durability, excellent mechanical properties, resistance to weathering and long service life. Plastic production has started to increase worldwide since 1950, and continues to grow on a massive scale. Its global production was 361 million tonnes in 2019, 299 million tonnes in 2014, and only 1.7 million tonnes in the first year of its journey in 1950 (Auta et al., 2017). Microplastics (MPs) are plastic particles smaller than 5 mm in size (Thompson et al., 2009). MPs are either are derived from large plastic particles in various mechanical and photo-oxidative processes (i.e. secondary MPs) or manufactured in such small sizes deliberately to use for particular purposes (i.e. primary MPs) (Hidalgo-Ruz et al., 2012). The use of primary MPs is increasing tremendously in various fields daily. They are widely used in personal care products (e.g., cosmetics, toothpaste, facial cleansers, deodorant, baby products, peelings, sunscreen, body washes, resin pellets, hair colors, nail polish, bath gels, eye shadows, insects' repellents), air-blasting facilities, synthetic clothing, gas-oil drilling fluids, plastic powders in molding and countless everyday tasks (Alomar et al., 2016). On the other hand, hills of secondary MPs are being formed in the environment every day through the wrecking of various sized plastic debris over time in terrestrial and marine environments (Evangeliou et al., 2020). Biological, physical and chemical processes break down the structural integrity of larger plastic particles which later turn into microscopic fragments. Different environmental factors (e.g. temperature, humidity, salinity), polymer properties (such as morphology, size, shape, density) and biogeographic conditions can influence the formation of secondary MPs (Auta et al., 2017; Min et al., 2020; Chamas et al., 2020; Anjana et al., 2020). Major sources of MPs are shown in the appendix (**Figure A1**).

MPs enter the environment in various ways. MPs from personal care products and other different sources are incorporated directly with municipal wastewater, and due to their microscopic sizes, often are incompletely removed during wastewater treatment plants (WWTPs) and discharged into the aquatic ecosystems. Moreover, atmospheric fallouts, wind advection, storm-water runoff, and improper waste management are the major causes of MP release to the environment (Mason et al., 2016).

To effectively assess the risks of MPs to the aquatic environment and human health requires rigorous analytical methods, which typically follow several steps including sampling, extraction, isolation (or separation), identification and quantification (or classification). The identification of MPs is carried out depending on the physical properties (size, shape and colour) and chemical characteristics (polymer type) of isolated particles in mixtures of inorganic and organic residue particles. Due to the small size of MPs, their quantitative analysis is still a challenge to overcome.

MPs highly persist in the environment due to their slow rates of degradation (Eerkes-Medrano et al., 2015). To date, chronic toxicity of MPs has been reported in various studies due to their extended exposure in human and other organisms, although no evidence has been found of their acute effects (Li et al., 2018b; Sussarellu et al., 2016; Prata et al., 2020; Chen et al., 2020;). They can influence the physiological activities of living communities through leaching of contaminants from plastics (e.g. plasticizers, flame retardants), and by acting as a vector of persistent organic contaminants and heavy metals (Hu et al., 2020; Benson and Fred-Ahmadu, 2020; Luo et al., 2020; Lee et al., 2020; Kalcíková et al., 2020). Some of the abundant types of MPs in the environment, their typical forms of appearance, as well as common organic contaminants and heavy metals for which MPs act as a significant vector to biotic organisms, are listed in the appendix (**Table A1).**

Due to their small size, MPs can be mistakenly ingested by a wide range of organisms which cannot distinguish between their nourishments and MP particles (Kaposi et al., 2014; Lönnstedt and Eklöv, 2016; Dong et al., 2020; Fu et al., 2020). MPs directly impair many species of birds, reptiles, and fishes as the ingested MPs can stay in the gastrointestinal tract for a long time or may clog their stomachs and guts (Carlin et al., 2020; Solleiro-Villavicencio et al., 2020; Roch et al., 2020; Cole et al., 2013).

A number of physical, chemical and biological treatment technologies have been implemented recently to degrade or remove MPs from the environment. Physical methods are most widely used, including adsorption, screening, grit/primary sedimentation, membrane filtration, density separation, dissolve air flotation and magnetic separation. Till now, filtration technology such as ultrafiltration (UF), sand filtration and granular filtration has been found as the most effective, economic, energy-efficient process used in WWTPs of different sizes. In addition, reverse osmosis (RO) has become popular over the past few years for MPs removal. In many cases, UF is coupled with RO process to increase the efficiency of treatment process (Qin et al., 2004). With more than 99% efficiency, membrane bioreactor (MBR) has become the most popular treatment technology among all of the biological treatment methods for MPs removal (Carr et al., 2016;). Other biological methods include activated sludge process, aerobic digestion, anaerobic digestion (AD), biological degradation and constructed wetlands (CWs). Chemical treatment methods include oxidation, photo-oxidation, photo-catalytic degradation, coagulation, Fenton, photo-Fenton and acid-alkali treatment. Electrochemical methods such as electrocoagulation (EC) and electro-Fenton processes have added a new dimension in chemical methods to improve their efficiency. Currently, pyrolysis and co-pyrolysis are being touted as one of the most promising approaches for MPs removal with extra advantages of low-cost fuel production (Ahmed et al., 2011; Burra and Gupta, 2018b; Pinto et al., 2002).

Several review articles are available based on MPs especially on the occurrence of MPs in the marine environment, MPs analysis, and MPs remediation technologies. However, none of these review articles provides a systematic overview of MPs especially from the sources to treatment technology. Hence, the main aim of this review is to provide the state-of-the-art overview of MPs in the aquatic environment by incorporating all relevant aspects and critically analyzing the available information from literature together with advantages and limitations of each technology. Future research directions for MPs are also proposed based on the review.

2. Sources of MPs

2.1. Abundance of MPs

MPs have been found in urban and rural areas (Hirai et al., 2011), seawater, sediment (Hidalgo-Ruz et al., 2012; Van Cauwenberghe et al., 2015;). MPs are also found in the air (Gasperi et al., 2018), soil (Bläsing and Amelung, 2018) and organisms (Zhao et al., 2016). Several reviews have been published on the presence of MPs in rivers, ocean and lakes (Horton et al., 2017; Mendoza et al., 2018). Hidalgo-Ruz et al. (2012) studied the abundance of MPs in the marine environment, and suggested MPs density to be 0.001-1 items/m^2 on the sea surface and 1-100000 items/m^2 in the sediment. Light MPs have been found in the freshwater surface as floating debris ranging from 1×10^3 to 68×10^5 particles/km^2 (Anderson et al., 2017; Free et al., 2014; Su et al., 2016). Furthermore, the denser MPs are usually found in the soil and sediment (Di and Wang, 2018; Su et al., 2016). The abundance of MPs in the aquatic environment is presented in **Table A2.**

The reported MPs concentration in the sediment of St. Lawrence River was 13832 microbeads/m^2, implying high MPs content in association with high population density (Castañeda et al., 2014). Similar suggestion was made in the analysis of MPs distribution in sediment from the Rhine and Main rivers (Klein et al., 2015). However, water column caught a notable quantity of suspended plastics particles (da Costa et al., 2016; Leslie et al., 2017); the measured concentration ranging from 1.6 to 12.6 pieces/L in the three Gorges Reservoir, China (Di and Wang, 2018). During wastewater treatment, MPs are simply removed but remain in sludge as MPs does not degrade. Therefore, wastewater sludge contains relatively high MPs e.g., as high as 56000 particles/kg (Li et al., 2019a). During the measurement of MP in an urbanized river in Chicago, it was found that the MPs concentration exceeded that in oceans, which demonstrated that the WWTP effluent was the primary source of MPs to the coast water (McCormick et al., 2014).

The sources of MPs in the aquatic system included domestic wastewater (Carr et al., 2016; Duis and Coors, 2016), sewage discharge (Eerkes-Medrano et al., 2015; Mintenig et al., 2017), plastic manufacturing industries (Sadri and Thompson, 2014), and decomposition of large plastics (Eerkes-Medrano et al., 2015). Road dust such as tyres, bitumen, and road marking paints are other MPs that transport through the freshwater system to the ocean. It is estimated that an average of 63, 125 and 240 kilo tonnes MPs are discharged through wear and tear of tyres each year in the United Kingdom, Germany and Japan, respectively (Ngo et al., 2019).

Moreover, numerous studies have shown that the abundance of MPs is highly dependent on the population density and type of human activities (Yin et al., 2020). The abundance of MPs has been discussed in ocean (Cole et al., 2011), sediment (Alimi et al., 2021), WWTPs (Murphy et al., 2016), and freshwater including rivers and lakes (Castañeda et al., 2014). The concentrations of MPs from different sources and locations are summarized in **Table A2**.

2.2. MPs in wastewater

Studies show that WWTPs are one of the major sources of MPs in the environment (Carr et al., 2016; Murphy et al., 2016, Alvim et al., 2020). MPs from different personal care products (e.g. hand cleaners, toothpaste, skin cleansers, facial scrubs) have gained considerable public attention during the past few years (Duis and Coors, 2016; Sun et al., 2020;). MPs are incorporated as 'exfoliation tools' in some of these care products and may pass through WWTPs due to their micro size (Chang, 2015). Some plastic pre-products such as plastic resin pellets and plastic powder (flakes and fluffs) are also considered as another source of MP particles, which are used in the fabrication of various plastic products. They may reach the environment by improper handling and accidental losses during transportation. Residues and garbage from plastic recycling and plastic processing factories may also pollute waters as a significant contributor of MPs (Andrady, 2011; Derraik, 2002; Moore, 2008). Protective paints (e.g. ship paints, furniture paints) contain many synthetic polymers such as polyacrylate, polystyrene (PS), poly-urethane, alkyds and epoxy resins which may release MPs during application, removal or abrasion of the paints (Song et al., 2014, Hale et al., 2020, Kwon et al., 2020). Also, abrasion of MPs from some household plastic materials, car tires, and synthetic textiles (e.g. shirts, PS fleeces, blankets) are major sources of MP particles in wastewater (Duis and Coors, 2016). MPs are used in some industrial abrasives, drilling fluids (in gas and oil exploration), cleansing agents of engines as well as some other industrial aids and equipment. Improper disposal and handling of them may cause significant input of MPs in wastewater (Derraik, 2002).

Although significant removal of MPs has been reported in several WTTPs, surprisingly very few of them can remove MPs with 100% efficacy. An observation by the Swedish Environmental Research Institute revealed that release of only 0.009 MPs/L from a WWTP (> 99% MPs removal efficacy) resulted in the total discharge of about 1770 MP particles every hour on average. Murphy et al., (2016) evaluated a significant number of MPs release from a WWTP despite having 98.41% removal efficacy. The release of 0.25 MPs/L of final effluent resulted in almost 65 million MPs release daily with the treated water. In comparison, two tertiary treatment plants in New South Wales, Australia were found to release about 1.0 MP/L of MP particles in the final effluent. Variations in the number of treatment steps, type and operating conditions of treatment technologies, and age of facilities involved in the WWTPs are responsible for the different removal and release of MPs. Some WWTPs from different geographic regions with varying capacities of MPs treatment are compared in **Table A3**, which clearly illustrates WWTPs as a significant secondary source of MPs.

Therefore, the WWTPs are considered as one of the most significant sources of MPs though being a secondary one. Either by implementing more efficient MPs treatment technologies, or by increasing the number of treatment steps, or by treating the contaminated water in two/three repeating cycles instead of the one-time flow system, the release of MPs from this major source can be mitigated significantly.

3. Methods of MPs analysis

To accurately assess the risks of MPs in the aquatic environment requires a standard analysis method for their identification (Koelmans et al., 2019). Analytical method of MPs in environmental samples follows several steps including sampling, extraction, isolation (or separation), identification and quantification (**Figure A2**). First of all, the water and sediment samples are collected from the field which are then filtrated or sieved via density separation techniques. Due to the presence of biological materials in the sample, the digestion step is required to reduce the organic matter without affecting the structural or chemical integrity of MPs (Felsing et al., 2018). In the analysis of MPs, a visual inspection can be used to sort and identify the large-sized MPs (1-5 mm), whereas dissection microscope can be used to sort smaller sized MPs particles (Doyle et al., 2011). The digestion step is mainly recommended before the application of visual inspection for MPs identification, via acid digestion, alkali digestion, oxidizing agent, and enzymatic digestion. However, under high acid concentration and high temperatures, polymers such as nylon, polyethylene terephthalate (PET) and polyamide (PA) have a low resistance to acid and may also be degraded or melted at high temperature, and polymers such as polypropylene (PP) and polyvinyl chloride (PVC) can change their colour (Maes et al., 2017;). In alkali digestion, KOH and NaOH solutions are widely used to recover MPs and remove biological material (Dehaut et al., 2016). In comparison, oxidizing agent such as hydrogen peroxide (30-35% H_2O_2) is more effective to degrade organic matter than NaOH and HCl, with little effect on MPs integrity. On the other hand, enzymatic digestion is less hazardous and causes little impact on MPs integrity when it is used to degrade organic materials (Maes et al., 2017). Enzymatic digestion followed by an incubation of H_2O_2 worked quite well and was able to efficiently destroyed all remaining organic material (Karlsson et al., 2017).

After the extraction and clean-up steps, the identification of MPs is carried out depending on the physical (size, shape and color) and chemical (polymer type) characteristics of isolated particles in mixtures of inorganic and organic residue particles. However, MPs analysis methods are still at development stage, hence there is not yet so-called standard method (Zhang et al., 2020b). The combination of two or more analytical techniques is required to identify MPs of various sizes, shapes and polymer types thoroughly and reliably from complex environmental matrices. Among the available optical analytical methods, Fourier-transform infrared (FTIR) spectroscopy, X-ray photoelectron spectroscopy (XPS) and Raman spectroscopy have been widely used to analyze MPs and nanoplastics (NPs) (Hernandez et al., 2019). XPS along with scanning electron microscopy (SEM) technique is limited to polymer identification but cannot provide elemental information about MPs (Hernandez et al., 2019; Hernandez et al., 2017), whereas Raman can provide elemental information about polymer through fingerprint spectra (Schwaferts et al., 2020). The thermal analysis has also been used to identify MPs.

3.1. Microscopy methods

A microscope is a fundamental instrument which helps to identify MPs particles by measuring their physical property. Stereo microscopy method is broadly-used for MPs identification with size > 100-μm (Kang et al., 2015; Mathalon and Hill, 2014; Nel and Froneman, 2015; Song et al., 2015). Magnified images help to identify ambiguous, plastic-like particles through detailed surface texture and structural information of objects using microscopy. Biological materials from sediment samples make it difficult for microscopic observation to identify MPs because these materials cannot be wholly eliminated by chemical digestion. Several investigations have indicated that false identification of plastic-like particles and transparent particles using microscopy was often over 20% and 70% respectively, which was confirmed with subsequent spectroscopic analysis (Hidalgo-Ruz et al., 2012; Song et al., 2015). Under high magnification, it is difficult to identify transparent or whitish particles using fluorescence microscopy (Löder and Gerdts, 2015).

On the other hand, SEM produces apparent and high-resolution images of the surface texture of MPs particles which are better identified from organic particles (Cooper and Corcoran, 2010). SEM analysis with energy-dispersive X-ray spectroscopy (EDX) provides the fundamental component of these particles which is useful for identifying carbon-dominant plastics from inorganic particles. The combination of SEM and EDX micro-analyzer has also been used in low vacuum mode to determine the chemical and morphological characteristic of MPs particles (Fries et al., 2013). In addition, polarized optical microscopy is an advanced microscopy technique which has been used to identify specific MPs particles such as PE (Von Moos et al., 2012). The main obstacle of the microscopy method is to accurately distinguish between the synthetic and natural fibers (**Table 1**) (Song et al., 2015). Therefore, the microscopy method should be combined with chemical analysis (spectroscopic or thermal analysis) to identify small-sized MPs (<1 mm) (Shim et al., 2017).

3.2. Spectroscopic methods

The spectroscopic technique is more effective for MPs identification, specifically for particles <500 µm. Micro-Raman or micro-FTIR spectroscopy is a fundamental technique which has been applied to evaluating marine samples through detailed information on polymer composition. FTIR and Raman spectroscopy are complementary vibrational techniques which are vital and effective in analysing MPs (Hidalgo-Ruz et al., 2012). These methods can scan the complete sample filters in a short time by combining spectroscopic techniques with fast area-resolved measurements such as FPA detectors, and chemical imaging (Harrison et al., 2012). Fibres of the bigger fraction (> 750 µm) can be spontaneously measured through Raman or FTIR spectroscopy (Käppler et al., 2016).

3.2.1. FTIR spectroscopy

FTIR spectroscopy is the most common method of analysing surface chemical composition or specific chemical bonds of plastic particles (Hidalgo-Ruz et al., 2012). FTIR technique provides unique infrared spectra from the change in the dipole moment of chemical bond and identify MPs by comparison with known reference spectra. The unique spectra of the different bond composition of plastic in FTIR can easily discriminate plastics from other organic or inorganic particles (Löder and Gerdts, 2015), and determine the composition of MPs particle and specific polymer type (Doyle et al., 2011; Harrison et al., 2012). By analysing different band patterns using the FTIR spectroscopy, physicochemical weathering of sampled MPs particles could be identified (Corcoran et al., 2009) (**Table 1**).

Attenuated total reflectance (ATR)-FTIR spectroscopy is employed to detect large particles fast (< 1 min) with high accuracy (Löder and Gerdts, 2015). For example, MPs particles from <500 μm to 5 mm can be determined via ATR-FTIR and transmission measurements (Huppertsberg and Knepper, 2018). The reflectance and ATR mode does not require the sample preparation step for thick and opaque MPs like the transmission mode. Moreover, in the case of irregular MP surfaces, the ATR mode produces stable spectra, compared with unstable spectra in the reflectance mode. MPs as small as the IR beam aperture (e.g., 10 μm) are measurable by the ATR probe.

Micro-FTIR mapping has been successfully applied for MPs identification with sequential measurement of IR spectra at spatially separated and user-defined points on the sample surface (Levin and Bhargava, 2005). But this technique is still very lengthy when targeting the whole sample surface at a high spatial resolution as it uses only a single detector element (Harrison et al., 2012). Only 1.4% of the particles with synthetic polymer origin were detected in sediment with μ-FTIR (Löder and Gerdts, 2015). Surface contact analysis forming ATR-FTR offer a useful method (μ-ATR-FTIR), which has been applied to identify MPs in environmental samples (< 50 μm).

MPs (150–250 μm) with larger surface areas can be detected using single beam mapping focal plane array (FPA)-based reflectance imaging method without compromising spatial resolution (Tagg et al., 2015). The simultaneous recording of several thousand spectra within an area with a single measurement and the generation of chemical images was possible with this technique (Harrison et al., 2012; Levin and Bhargava, 2005). The disadvantages of the FPA method are high instrumental cost and requiring high processing power. The advantage of this method is that no visual pre-selection is needed to automatically analysing the whole filter. This method may facilitate the measurement of large sample areas of filter i.e. whole filter surfaces in one single measurement run. Overall, FPA-micro FTIR method is fast and facilitates fully automated analysis of MPs of small size. Therefore, the FTIR spectroscopy is the most frequently used technique in MPs identification and quantification at now. This technique demonstrates a promising tool for automated MPs analysis. However, more research should be needed to progress automated analysis methodology to increase accuracy and shorten the identification time.

3.2.2. Raman spectroscopy

Raman spectroscopy is a non-destructive scattering method used to identify MPs particles in environmental samples with detailed information about the chemical structure of particles (Cole et al., 2013; Imhof et al., 2012). Molecular vibration of a sample leads to characteristic spectral fingerprint in the Raman spectra which help to identification of MPs by comparison with known reference spectra (Käppler et al., 2016). Once the laser beam (500-800 nm) striking on an object results in different frequencies of back-scattered light based on the molecular structure and atoms present, it produces a unique spectrum for each polymer. This so-called Raman shift can be detected to form substance-specific Raman spectra. This technique can be applied to detect plastic polymers within minutes by comparison with reference spectra as the plastic polymers possess characteristic Raman spectra (Löder and Gerdts, 2015). The smaller diameter of the laser beam in Raman spectroscopy allows for the identification of a broad range of size classes down to tiny plastic particles < 1 μm (Cole et al., 2013). However, chemical additives and pigments of MPs are interfered in this method to identify polymer types (Tagg et al., 2015). In a complex MP identification, the different responses and spectra between FTIR and Raman spectroscopy can compromise each other. Confocal microscopy can be coupled with Raman spectroscopy to detect MPs particles in zooplankton samples (Cole et al., 2013). Raman spectroscopy can also be combined with Raman spectral imaging to generate spatial chemical images of a sample.

Raman spectroscopy has some limitations (**Table 1**) e.g. the laser used to excite the fluorescent samples cannot be analysed since it precludes the formation of interpretable Raman spectra. More research is required to explore the optimum laser wavelength for a compromise between suppressed fluorescence and low signal intensity for assessments of MPs in environmental samples. Usually, before measurements of Raman spectroscopy, a purification step of samples is needed to prevent fluorescence for precise identification of the polymer type of MPs particles (Löder and Gerdts, 2015). However, Raman spectroscopy is a better analytical method to identify small-sized MPs (< 20 μm).

3.2.3. Pyrolysis-gas chromatography-mass spectrometry (Py-GC-MS)

Py-GC-MS can be applied to identify and quantify the types of potential MPs particles with high certainty through their characteristic decomposition products (Dümichen et al., 2015). This method determines the type of polymer through analysing thermally decomposed gases from polymers, by comparing with GC-MS profiles of virgin polymers standards. It also enables to identify the organic plastic additives at a lower temperature in one run (Fries et al., 2013). The Py-

GC-MS technique permits a comparatively good assignment of potential MPs to polymer types, but a drawback is the complete destruction of the sample hence actual chemical composition measurement is difficult to infer. Py-GC-MS analysis method identified PA, PE, PVC, PP, PS, PET and chlorinated or chloro-sulfonated PE MPs particles from sediment samples (Dekiff et al., 2014). Regarding the size limitations of plastic particles, Py-GC-MS method is often used to analyze plastic particles with the maximum size of 1.5 mm (Fries et al., 2013). Its other limitation is long processing time per sample run (**Table 1**).

Py-MS has been used to overcome the limitation of Py-GC-MS method (Zhang et al., 2020b). This process enables the rapid identification of MPs (<1.2 μm) compared with Py-GC-MS and TGA-GC-MS, and can reduce the interference of the environmental matrix in analysing MPs. Another advantage of this method is the ability to analyse mixed polymers in one run without preselection and separation which is difficult in Py-GC-MS method. Py-MS is often used to analyze a large amount of MPs samples. MPs are identified in this process by their chemical fingerprint including characteristic ions and the ratio of characteristic ions from the mass spectra (Zhang et al., 2020b).

3.2.4. Atomic force microscopy (AFM)

The combination of AFM with either IR or Raman spectroscopy can provide an effective method for micro and nanoparticles detection and identification which is the major challenge in MPs research. This combined method can identify polymer types through their chemical composition. Automated FTIR/Raman mapping or particle tracking with simultaneous Raman spectroscopy can resolve the problems such as missing small and transparent MPs particles (Dazzi et al., 2015). AFM-IR can generate IR absorption spectra and absorption images within the spatial resolutions of 50–100 nm (Dazzi et al., 2012). For example, 100-nm PS beads were successfully analyzed by AFM-IR combination.

In addition, Nile Red or NR (9-diethylamino-5H-benzo(α)phenoxazine-5-one) is a strong fluorescent dye which is useful for staining to identify hydrophobic MPs (Andrady, 2011). NR staining can be used as a pre-step of spectroscopic analysis and identify hidden MPs. The NR staining method presents high efficiencies for identifying <100 μm PE, PP and PS particles mixed with high numbers of inorganic particles (Shim et al., 2016). However, co-staining of other natural materials is the main limitations in applying the NR staining method.

3.3. Thermal analysis

Thermal analysis is a destructive method which has been recently used for MPs identification (Castañeda et al., 2014; Majewsky et al., 2016). This method can easily measure the changes in the physical and chemical properties of polymers depending on their thermal stability (Dümichen et al., 2015). Thermo-gravimetric analysis can completely pyrolyze the samples at a temperature up to 600 ºC (Dümichen et al., 2017). The decomposition profile of the TGA is used to measure the homogeneity of the samples. Differential scanning calorimetry (DSC) is a useful method for identification of specific polymer types by comparison with reference polymeric materials. Different plastic has different characteristics in DSC (Kalčíková et al., 2017), which help to identify MPs accurately. Due to the availability of reference materials, this method can identify specific primary MPs such as PE microbeads (Castañeda et al., 2014). The combination of TGA and DSC has been used to identify PE and PP (Majewsky et al., 2016). Due to their overlapping phase transition signal, coupled TGA-DSC method failed to identify PVC, PET, PA, polyester and polyurethane (PU) (Majewsky et al., 2016). TGA can be combined with solid-phase extraction (SPE), coupled with thermal desorption gas chromatography-mass spectrometry (TDS-GC-MS). This combined technique has added advantages of measuring larger sized particles (**Table 1**). TGA-SPE/TDS-GC-MS method is used to identifying PE, PP and PS MPs in different samples (Dümichen et al., 2015). The combination of thermo-extraction and desorption with gas chromatography-mass spectroscopy (TED-GC-MS) has been used for relatively high sample masses and enabling measurement of complex sampling matrix (Dümichen et al., 2017). TED-GC-MS has been used as a fast tool for MPs analysis and the identification of polymers in different solid samples. MPs identification in this process is through pyrolysis and analysis of the decomposition gases (Dümichen et al., 2015).

In summary, spectroscopy analysis should be combined with chemical analysis such as spectroscopic or thermal for MPs <1 mm. Currently, the μ-ATR-FTIR method is recommended for the routine analysis of environmental samples. FPA-FTIR method should be used to analyze samples of known polymer types. TGA-DSC method needs further research to develop the performance for MPs identification. In comparison, spectroscopic (Raman and FTIR) and thermal analysis (Py-GC-MS, TED-GC-MS and Py-MS) methods are the most promising technologies for MPs identification and quantification. However, there is a need for significant improvement in these analytical techniques, especially in the quantification of the MPs. There is also a need to develop tools which can identify and quantify a broad range of MPs.

[Table 1]

4. MPs interactions with other contaminants

Many studies found that MPs interact with contaminants by the adsorption of harmful chemicals, antibiotics and metals on their surface (Li et al., 2019b; Liu et al., 2019a), hence acting as a carrier of other hazardous chemicals. These contaminants can pose a significant impact on microorganisms. Many studies observed the adsorption of organic contaminants or heavy metals on different types of MPs (Bakir et al., 2014; Hüffer and Hofmann, 2016; Wang et al., 2015). Plastics type and environmental factors (e.g. ionic strength and pH) can affect the adsorption of contaminants on MPs (Wang et al., 2015). MPs can easily sorb and concentrate toxic chemicals from the surrounding for their large specific surface area and hydrophobicity which can make them more dangerous for the natural ecosystems (Teuten et al., 2009).

4.1. MPs interaction with toxic organic chemicals

Through adsorption, toxic contaminants can easily interact with MPs, especially those with irregular shapes (Lambert et al., 2017). The shape of MPs could influence the interaction between MPs with other contaminants or microorganisms in water (McCormick et al., 2014; Wang et al., 2018a). Wang et al. (2018a) reviewed the interaction of toxic chemicals with MPs. The large surface area to volume ratio, crystallinity, hydrophobic, physical (e.g. particle size) and chemical (e.g. functional group) properties of MPs promote their adsorption of a wide range of persistent organic contaminants (POPs), such as polychlorinated biphenyls (PCBs) or polycyclic aromatic hydrocarbons (PAHs) (**Figure A3**) (Bakir et al., 2014; Browne et al., 2013; Herbort and Schuhen, 2017). Many additives such as polybrominated diphenyl ethers (PBDE) and phthalate esters are added in the production of plastics (**Table A4**) (Cole et al., 2011). MPs may sorb much more organics than larger plastics particles because of their broad surface area (Lee et al., 2014). For example, PAHs and PCBs were detected in extracts from the plastic fragments in North Pacific Central Gyre, at concentrations of 4-249 and 1-223 ng/g, respectively (Mendoza and Jones, 2015). Similar findings from two beaches of the Portuguese coast showed that the MP pellets contained PAHs, PCBs and dichloro-diphenyl-trichloroethanes (DDTs) at 319.2 ng/g, 15.56 ng/g and 4.05 ng/g, respectively (Frias et al., 2010). MPs can adsorb persistent, bio-accumulative and toxic chemicals from surrounding water and act as a unique medium to transport these chemicals into the food chain, which can lead to deadly effect in ecological habitat (Koelmans et al., 2016). On the other hand, some researcher reported that MPs are not major carriers of toxic chemicals (Gouin et al., 2011; Koelmans et al., 2013). Alimi et al. (2018) reported that PE generally exhibits a higher sorption capacity for contaminants than other MPs types, and discolored MPs adsorb more PCBs than coloured ones. Another study reported that PE particles show higher affinity with organic chemicals than other plastics like PP and PVC pellets.

4.2. MPs interaction with antibiotics

Different antibiotics such as tetracyclines, macrolides, fluoroquinolones and sulfonamides are widely found in the aquatic environment **(Table A5)** (Jiang et al., 2011; Li et al., 2012). Antibiotics can impact the microbial community and generate resistance genes (Yang et al., 2017). In addition, antibiotics are hazardous chemicals which may affect the survival and growth of aquatic organisms (Ahmed et al., 2015). Li et al. (2018c) studied the adsorption of five antibiotics (sulfadiazine, amoxicillin, tetracycline, ciprofloxacin, trimethoprim) on five types of MPs (PE, PS, PP, PA, PVC) in the freshwater and seawater systems. They reported the adsorption capacity decreased as ciprofloxacin > amoxicillin > trimethoprim > sulfadiazine > tetracycline. Due to the pore structure, PS, PP and especially PA showed higher sorption capacity than PE and PVC. Because of the formation of hydrogen bond, amoxicillin, ciprofloxacin and tetracycline are highly adsorbed on PA (Li et al., 2018c). Antibiotics are adsorbed on the MPs because of their large surface area and lipophilic nature (Guo et al., 2018; Guo and Wang, 2019). Further, the adsorption capacities of MPs varied based on their physicochemical properties such as specific surface area, the abundance of rubbery, polarity, degree of crystallinity and chemical properties such as Van der Waals interaction, π-π interaction and hydrogen bonding between antibiotics and organic matters (**Figure-A4**) (Li et al., 2018c; Wang et al., 2015). Solution pH, ionic strength and salinity have a significant effect on antibiotics sorption in MPs (Guo et al., 2018). For this reason, MPs more highly adsorb antibiotic in freshwater than in seawater (Li et al., 2018c). Due to the combined pollution of antibiotics and MPs, a more toxic effect was shown in the freshwater and marine ecosystems (Li et al., 2018c). However, more research is needed on the interactions between MPs and antibiotics, in order to further evaluate their environmental risks.

4.3. MPs interactions with heavy metals

Many studies reported that heavy metals were associated with MPs in the aquatic environment (Holmes et al., 2012; Wang et al., 2019a; Yu et al., 2019). Heavy metals (Ag, Cd, Co, Cr, Cu, Hg, Ni, Pb and Zn) can be adsorbed into MPs in freshwater at pH 6.5 (Turner and Holmes, 2015). Metals highly accumulated on a few specific MPs types such as PET, PVC, PP, low-density PE and high-density PE (Rochman et al., 2013). Cr and Cu were strongly adsorbed with an adsorption capacity of 0.297 and 0.261 μg/g, respectively, while Ni, Co, Pb and Cd showed no obvious adsorption. It was found that adsorption capability of Ag, Cd, Co, Cu, Hg, Ni, and Pb was 0.0128 μg/g, 0.0101 μg/g, 0.0692 μg/g, 0.100 μg/g, 0.170 μg/g, 0.0166 μg/g, and 0.191 μg/g, respectively in freshwater (**Table A6**) (Turner and Holmes, 2015).

The large surface area, polarity, and organic polymer composition are the major causes of heavy metal adsorption on MPs (Ashton et al., 2010). Cations or complexed metals were directly adsorbed on the charged or neutral regions of MPs surfaces (Ashton et al., 2010). The adsorption of Ag, Cd, Co, Ni, Pb and Zn increased with increasing pH of river water (Godoy et al., 2019; Turner and Holmes, 2015). Dissolved organic matter may play a major role in the trace metal adsorption on MPs through interacting with metals or polymers (Godoy et al., 2019). Other factors such as specific surface area, porosity and morphology also play an important role in metal adsorption on MPs (Godoy et al., 2019), who revealed that the adsorption mechanism of metals in MPs might be chemical adsorption due to the adsorption isotherm following the Langmuir model. Intermolecular interaction such as van der Waals interactions, hydrogen bonding interactions, and cavity formation between contaminants and polymers may influence the sorption capacity (Godoy et al., 2019). Many studies indicated that the metal adsorption kinetic was relatively fast (Godoy et al., 2019; Holmes et al., 2012). Metals are specific to adsorption on MPs, due to their different chemical structure, as demonstrated by that Pb, Cr and Zn are significantly adsorbed on the surface of PE and PVC MPs with little adsorption on PET (Godoy et al., 2019). Similarly, Pb, Zn and Co are adsorbed significantly on the surface of PE, PP and PET polymers (Rochman et al., 2013). Zn, Cu, Cr and Pb seem to have a higher affinity for plastics than Ni and Co (Godoy et al., 2019; Holmes, 2013).

5. Toxicity of MP contaminants

5.1. Biological toxicity

Studies have been conducted to evaluate the toxicity of MPs on biotic communities, and revealed higher toxic effects of MPs on the non-selective filter feeders, which mistakenly ingest MPs instead of other nourishing substances. MPs may cause various internal injuries, blockage in alimentary canals, reduction of dietary intake, or translocating into the internal circulatory system (Murray and Cowie, 2011; Shen et al., 2020). MPs ingestion may cause chronic effects, e.g. MPs from high-density polyethylene (HDPE) (particle size < 80 μm) were tracked in the digestive system and epithelial cells of *Mytilus edulis* with adverse impacts on the tissues and intestinal tract (Von Moos et al., 2012). Adverse effects on the fecundity of *Calanus helgolandicus* (pelagic copepods) were reported while exposed to 20-μm MPs beads (Cole et al., 2015). The laboratory-based observations for the toxicity of MPs on biotic communities and organisms are described in **Table A7.**

Different types of chemical additives are incorporated with plastic compounds to obtain and improve the desired properties of the products according to consumer demands. For example, PBDEs are used to increase heat resistance, and phthalates to increase malleability, colorants, plasticizers, and flame retardants. Leachates from the waste plastic debris and their fragments may impose toxicity to a wide range of species (Lithner et al., 2009; Lithner et al., 2011). Batel et al.

(2016) reported that benzo[α]pyrene-loaded MPs would release contaminants retaining in gut and liver of zebrafish. Browne et al., (2013) evaluated the desorption of nonylphenol and phenanthrene from PVC. A mathematical model using equilibrium partitioning and experimental data demonstrated the transfer of contaminants from plastic to organisms (Teuten et al., 2009).

A positive relation was observed between MP concentration in the sediment and both uptake of plastic particles and weight loss by *A. marina* (Besseling et al., 2013). Lungworms exposed to micro-PS and PCBs showed reduced feeding capability and lost weight. In terms of immune function and oxidative status, PVC alone made lungworms $>$30% more susceptible to oxidative stress (Browne et al., 2013). Adverse effects of phenanthrene loaded MPs from low-density polyethylene (LDPE) and PS beads (incorporated with fluoranthene) were observed in African catfish and *Mytilus* spp., respectively (Paul-Pont et al., 2016). Plastic-sorbed chemicals were found to induce liver toxicity in Japanese medaka (*Oryzias latipes*) (Rochman et al., 2013). Luís et al., (2015) investigated the potential influence of MPs on the short-term toxicity of chromium to juveniles of *Pomatoschistus microps.* They discovered a significant decrease of the predatory performance ($\leq$ 67%) and an inhibition of acetylcholinesterase (AChE) activity ($\leq$ 31%).

Furthermore, MPs possess high surface area that supports the particles to act as a significant vector of various contaminants such as POPs and heavy metals during the transports in the environment (Browne et al., 2013). Some of the contaminants can be retained in the tissues of the organisms, e.g. PBDE-47 and triclosan were found in the tissues of lugworm at 330 ng/g and 1250 μg/g, respectively (Browne et al., 2013). Some of the sorbed contaminants such as DDT, phenylalanine and di(2-ethylhexyl) phthalate (DEHP) can transfer from microscopic particles of PVC and PE to the benthic invertebrates (Bakir et al., 2016; Chua et al., 2014). **Table A8** listed the concentrations of contaminants in the tissues of the organisms due to the transfer of chemicals from MPs. Here it is worth mentioning that, the concentration gradient of such adhering pollutants on the MPs surfaces are often too much low to induce potential toxicities alone to the organisms. However, there are several lines of evidences that disclose the joint adverse impacts of MPs and their adhering toxic pollutants on various living organisms. In **Table A8**, some of them have been listed in detail. Overall, MPs can pose a significant risk of contaminating aquatic food chains, and potentially increase human exposure via such sources. Rather than being a vector of various POPs, MP itself pose a huge threat as a persistent pollutant to the environment, to the health of individual organisms and potentially the overall ecosystems as well.

5.2. Impact on the aquatic ecosystems

MPs in the open ocean support various microbial biofilms which are different in taxonomic composition from the microbial assemblages of the surrounding water, in heterotrophs, autotrophs, predators and symbionts. Sediments with *O. edulis* showed significantly different assemblage structures when dosed with MPs, at any density or type of plastic than controls (Green et al., 2017). Negative impacts on the producers of aquatic ecosystems (e.g. algae) have been observed, e.g. reduction of chlorophyll, growth rate and photosynthesis, with implications for all of the producer-dependent primary, secondary and tertiary components of the total ecosystem. Furthermore, individual organisms (e.g. fish) may also ingest MPs mistakenly and undergo various disorders through the direct action of MPs. The influence pathways of MPs in an ecosystem have been illustrated in **Figure 1.**

MPs represent a wide range of stressors to interfere in the ecosystem jointly. For instance, a mixture of additives, sorbed contaminants and copolymers may cause additive effects (Jang et al., 2020; Sait et al., 2020). In the aquatic ecosystem, MPs affect the water translucency (Sjollema et al., 2016), sedimentation (Cole et al., 2016), and thermal conductivity (Carson et al., 2011) as well as the ‘aquatic biocoenosis’. Moreover, they may act as a vector for a large number of pathogens and some other invasive species. MPs may also influence the ‘predator-prey interactions’ and the community structure by interfering with the inter- and intra-species signaling in the ecosystem (Besseling et al., 2014).

[Figure 1]

6. MPs treatment technologies

6.1. Physical treatment

6.1.1. Adsorption through biochar and activated carbon

Biochar (BC) and activated carbon (AC) are extensively used as an adsorbent to treat stormwater containing MPs and NPs (Ahmed et al., 2016; Mohan et al., 2014; Mohanty et al., 2018; Sommer et al., 2018). Adsorbent surface area and porosity are two major properties for effective removal of MPs (Siipola et al., 2020). Siipola et al. (2020) reported that steam activated BC (at 800 ^{o}C) was most suitable adsorbent for MPs removal, even with relatively low surface area (200–600 m^2/g). Despite the small surface area (187 m^2/g) with macro-scale porosity, spruce bark AC resulted in better performance for MPs retention than pine bark ACs with surface area

556-603 m^2/g. Activated BC effectively retained large size MPs particles, whereas 10 μm spherical microbeads did not adsorb as efficiently. Hence, meso- and macro-porosity can be very beneficial for the removal of MPs. The BC surface roughness may influence the retention of large MPs particles most likely through physical attachment. They also found that PE particles and fleece fibers were 100% retained, although the mechanism of MPs adsorption is yet to be identified (Siipola et al., 2020). Both BC and AC may act as a filter when packed in a column for MPs removal (Zhang et al., 2020a). Therefore, adsorption with AC or BC via a filtration setup is an economical process to remove MPs.

6.1.2. Membrane processes

Membrane technologies such as UF, microfiltration (MF), RO and MBR have increased the removal efficiency of MPs and NPs. In the last five years (2015-2020), the application of membrane technology in MPs removal was only reported in 13% studies, with other treatment processes accounting for 87% of studies. The combination of a porous membrane with a biological process (i.e. MBR) could enhance the rate of MPs removal from primary effluent up to 99.9% (Talvitie et al., 2017a). Membrane material, pore size, thickness and surface characteristics affect the performance of the membrane process. In membrane filtration, the major drawback is the fouling phenomena which occur by adsorption of particles on the membrane surface. Following fouling, membrane filtration performance will be decreased which resulted in higher energy cost, operation time and maintenance (Malankowska et al., 2021). Enfrin et al., (2019) briefly reviewed and revealed that MPs could interact with the membrane surface because of their intrinsic physicochemical properties such as hydrophobicity, surface charge and roughness. Nevertheless, membrane technology is highly efficient in the removal of low-molecular weight contaminants such as small MPs (<100 μm) and NPs.

6.1.2.1. Ultrafiltration

UF is represented as the most effective and economical process. UF is a low functioning pressure (1-10 bar) driven membrane process, which can separate dissolved macromolecules and tiny suspended particles in the colloid size range whose molecular weight is higher than a few thousand Dalton from feed fluid. It permits to move solvent and low molecular weight solutes (microsolutes) through the membrane in the range of pore size 0.001-0.1 μm (Basri et al., 2011). Sun et al., (2019) reported that polyethylene (PE, ~4%-51%) is the most abundant types of polymer. Owing to the large particle size, PE particles can be fully removed through the UF membrane (Ma et al., 2019). UF with a nominal size of 0.2 μm in MBR offered 100% removal of MPs. A detailed schematic diagram of MPs behavior during the coagulation and UF processes is shown in **Figure A6**. Overall, UF membrane treatment can be applied to eradicate PE particles, especially with UF and coagulation combination. Pilot-scale UF-MBR hybrid treatment also showed an adequate performance to remove MPs. The pore size of the UF membrane is similar to MPs (Enfrin et al., 2019), but MPs also interact with UF membrane surface and may cause abrasion due to their irregular shape which reduces the filtration performance.

6.1.2.2. Reverse osmosis (RO)

RO is the most common and promising process to treat wastewater (Ibrar et al., 2020). The high pressure applied in the RO membrane process enhances treatment efficiency, but may produce nano plastic particles from MPs by fragmentation (Enfrin et al., 2019). Ziajahromi et al. (2017) studied the performance of RO process to removal MPs from a WWTP in Sydney, Australia. They reported that screening and sedimentation, biological treatment, flocculation, de-chlorination processes and tertiary UF treated effluent reduced plastic particles from 2.2 MPs/L to 0.28 MPs/L, which was finally treated by RO to reach 0.21 MPs/L. Overall, the RO process is not particularly efficient for MPs removal but the application of combined UF-RO/MBR-RO processes may enhance the removal efficacy.

6.1.2.3. Dynamic membrane (DM) technology

The development of DM technology has attracted interest. DMs can be classified into self-formed and pre-deposited (Hu et al., 2018). DM acts as a secondary membrane when particles and other foulants in wastewater are filtered by forming a cake layer in supporting membrane surface (Ersahin et al., 2012). Instead of the traditional membrane, larger pore-sized mesh or other low-cost porous materials such as non-woven fabric or woven filter cloth and stainless steel are used as a supporting layer. DM technology is more effective to removed low density (poor settling) and non-degradable MPs because of the swiftly formed secondary membrane (DM layer) with micro-particles (Li et al., 2018e). In recent years, DM technology is an effective option to filtrate MPs from synthetic wastewater under a gravity-driven operation. Low TMP, low filtrate resistance and easy cleaning are the main benefits of DM (Li et al., 2018e). Li et al., (2018e) designed and operated a lab-scale DM filtration using 90 μm supporting mesh, and reported about 90% of MPs removal from synthetic wastewater. The DM formation is essential in DM technology to resist the higher number of microparticles in wastewater filtration. After 20 min of MPs filtration, the effluent turbidity has been reduced to <1 NTU (Nephelometric Turbidity Unit) which verified the rapid formation of DM inducing better MPs removal performance.

In contrast to MF and UF, DM filtration process has shown lower TMP value which indicates the reduction of energy consumption (Li et al., 2018d). The most substantial TMP value observed is 16 times lesser than conventional MF at 30 kPa (Huang et al., 2017). Overall, DM technology showed excellent performance to remove micro-contaminants including MPs during wastewater treatment, and mitigated the disadvantages of membrane fouling in MBR. The combination of DM technology with coagulation or activated sludge process can be highly effective to remove micro-contaminants and MPs in wastewater treatment (Li et al., 2018d). Further research is needed to unravel the mechanism of DM layer formation.

Overall, membrane treatment technology is not specially designed to remove MPs efficiently, due to common issues of membrane fouling and decreasing water flux. More research should be devoted to minimize the membrane abrasion and fouling in membrane-based treatment technology. However, membrane treatment technology can be attractive if it is combined with biological process such as MBR or chemical process such as coagulation.

6.1.3. MPs removal in primary treatment

Many studies showed that conventional WWTP with primary and secondary treatment could remove MPs up to 99.9% from wastewater (**Table A9**). Most of the MPs were removed during pre-treatment stages i.e. preliminary (35-59%) and primary treatment (50-98%) (Sun et al., 2019). Sun et al., (2019) reported that higher MPs concentration (9×10^{-4} - 447 particles/L) of effluent present after primary and secondary treatment process than from a tertiary treatment process (0-51 particles/L). They also reported that large particles (1-5 mm) were clearly reduced from 45% to 7% after the primary treatment process.

6.1.3.1. Screen

Zhang et al. (2020a) used fine screen (mesh size 2.5-10 mm) to remove larger size MPs (> 2.5 mm) during primary wastewater treatment, as coarse screen (mesh size 50-100 mm) and medium screen (mesh size 10-40 mm) were not capable of removing MPs. Ultrafine screen (mesh size 0.2 -2 mm) was used before MBR as an alternative for primary sedimentation to prevent membrane fouling, which is the main obstacle of MBR. Therefore, the larger size of MPs (> 0.2 mm) could be removed from the wastewater, when the ultrafine screen was employed. Due to MPs' irregular shape, some larger MPs could still pass through the ultrafine screen.

6.1.3.2. Density separation

Density separation is the most reliable and commonly method for the separation of MPs/NPs from sediment or water samples by the density difference between materials of interest and other unwanted materials (Schwaferts et al., 2019; Wang and Wang, 2018). Mainly it is used to isolate the MPs from the environmental samples, particularly sediment samples (Hidalgo-Ruz et al., 2012). Most plastics have a density of 0.8-1.70 g/cm^3, compared with 2.65 g/cm^3 for sand and sediment, which sink to the bottom (Wang and Wang, 2018). It is difficult to remove MPs from water samples because of their density being close to water (**Table A10**). Flotation may be used after primary sedimentation in wastewater treatment plants to improve MPs separation. The low-density MPs (e.g. PP, PE) and moderate-density MPs (e.g. PS) will float at the surface of the wastewater. In density flotation method, saturated NaCl and NaI solutions was widely used to increase the density of water from 1 g/cm^3 to 1.2 g/cm^3 and 1.8 g/cm^3, respectively (Enfrin et al., 2019), so that low densities MPs float more easily even the denser PET. Few studies reported that the majority of MPs was removed in the primary treatment stage of WWTP, mainly in skimming and settling processes (Carr et al., 2016; Lares et al., 2018). Murphy et al., (2016) observed that the majority of microbeads (PE) were effectively removed via skimming. A great amount of MPs was picked up through skimming of floating debris in primary treatment since lower density MPs can float on wastewater (Carr et al., 2016; Murphy et al., 2016). Saturated NaCl solution was widely used due to low cost, widely available and environmentally benign (Li et al., 2018b; Van Cauwenberghe et al., 2015) (**Table A10**). It can be seen that fragments (PP, PE, PET) and fibers (PES, PET, PA) are the most abundant MPs in wastewater. Small amount of granular (PE), film (PP, PE, PA), and foams (PU, PVC, PA) are also found in wastewater (Enfrin et al., 2019; Lares et al., 2018; Sun et al., 2019; Talvitie et al., 2017a). PE, PP and PS were dominated plastic types in some wastewater samples (Murphy et al., 2016; Ziajahromi et al., 2017). Other studies found that polyamide (PA) was the abundant (54.8%) type of MPs in wastewater followed by PE (9.0%), PP (9.6%) and PVC (2.5%) (Liu et al., 2019c). PE is the most abundant MPs in effluents of different WWTPs (Ziajahromi et al., 2017). PES was highly detected in the final effluent of a Scottish (28%) (Murphy et al., 2016) and an Australian WWTP (67%). However, the saturated NaCl solution is less effective to separate denser MPs including PVC (1.3-1.7 g/cm^3) and PET (1.4-1.6 g/cm^3) (Wang and Wang, 2018). To overcome this problem, some high-density salt solution including sodium iodide (1.8 g cm^{-3}) (Wang and Wang, 2018; Ziajahromi et al., 2017), calcium chloride (1.30-1.35

g/cm^3) (Stolte et al., 2015), sodium polytungstate (1.4 g/cm^3) (Corcoran et al., 2009; Zhao et al., 2015) and zinc chloride (1.5-1.7 g/cm^3) (Maes et al., 2017; Mintenig et al., 2017) were used to increase the extraction efficiency of denser MPs. The density of brine solution has been recommended to be >1.45 g/cm^3 for the separation of all plastic polymers from sediments (Van Cauwenberghe et al., 2015). Majewsky et al., (2016) reported 85% and 91% recovery rates of PE and PVC respectively, when ZnCl solution was used for primary density flotation separation. Quinn et al. (2017) recommended 25% $ZnBr_2$ (1.71 g/cm^3) solution for density separation which will allow the flotation of PP, LDPE, HDPE, PE, PS, PVC, PET and PA. By using saturated NaCl, NaI and $ZnBr_2$, Quinn et al. (2017) observed higher recovery rates of 200–400 μm MPs. Moreover, $ZnCl_2$ (1.7 g/cm^3) brine solution recovered 100% of large size (1-5 mm) and 95% of smaller size (< 1mm) MPs from marine sediments. They also recovered 91-99% large size (1 mm) and 40% smaller size (40-309 μm) MPs particles using density separation method. However, heavy salts or higher density solutions are often used which are costly and even hazardous (NaI) for the environment.

6.1.3.3. Grit chamber/primary sedimentation

High-density contaminants (> 1.5 g/cm^3) such as sand and cinder (density around 2.65 g/mL) are separated by grit chamber. Some MPs may be attached on the sand and then settle together in grit, and can be easily removed from the stream. The specific densities range of most of the MPs are 0.8-1.7 g/mL, while PET, PVC and polytetrafluoroethylene (PTFE) are exceptional. PET and PVC have density varied from 1.38 to 1.41 or 2 g/mL, and PTFE has a density 2.10-2.30 g/mL. Density, travelling distance (D) and water flow (V) are the main parameters of grit performance. The MPs will be removed when the MPs settling time is smaller than the D/V. Blair et al. (2019) reported the lower removal efficiency (~6%) of MPs during grit. Generally, two types of grit are applied in wastewater treatment, horizontal and aerated grit chamber. In aerated grit, the lighter MPs were also eliminated along with sediment. For this reason, the aerated grit chamber showed better performance than horizontal grit chamber. Around 60% of 50 μm sized MPs in the wastewater can be removed in aerated grit (Yang et al., 2019). Liu et al. (2019c) demonstrated that large size particles could be efficiently retained from wastewater by the grit and aerated grit chamber (with 6 mm sized screen). In contrast, the smaller MPs are easily absorbed by sludge and settled during the secondary treatment process. They found that after the primary treatment, the reduction rate of MPs was 40.7% when treated with the coarse and fine grit, aerated grit chamber, and primary settlement tank. The combination of preliminary (screening, Grit and grease process) and primary settle treatment could improve the efficiency of removing MPs. Few studies reported that Grit chamber and primary settling tank more efficiently decreased the concentration of MPs from wastewater influent through skimming and settling process (Carr et al., 2016; Lares et al., 2018). This process is effectively used as a first stage of the WWTPs. Murphy et al., (2016) observed that preliminary (grit and grease with screen) and primary settle treatment combinedly removed 78.3% MPs from the wastewater. A very recent study showed a higher MPs removal efficiency (69–79%) by screening and grit removal process (Ziajahromi et al., 2021). Moreover, Sun et al., (2019) reported that preliminary treatment including coarse screening, fine screening and grit removal phases also shown better performance with 35-59% removal efficiency of MPs from wastewater. Consequently, preliminary treatment may play an important role in MPs removal. The grit removal and primary sedimentation process can be used as an effective pre-process in wastewater treatment, but not sufficient for small sized large scale MPs removal. In summary, primary treatment is able to remove large sized MPs in WWTP by 60-98%.

6.1.4. Filtration using granular media

Filtration is an effective process applied after density separation process to retain the MPs particles from bulk water samples or supernatant solutions. Various types of filter media are used in filtration process, among which glass fibers are the most frequently used, followed by nitrocellulose and polycarbonate (Wang and Wang, 2018). The most frequent used pore size of filters is 0.45-1 μm. Because of the microscopic particulates or debris in liquid, performing filter media was lowered by clog formation. To overcome this issue, ferrous salt is added to flocculate solid fraction and set a pre-filtration step with large pore size filter (Crawford and Quinn, 2017).

6.1.4.1. Granular activated carbon (GAC) filtration

GAC is widely used to remove various micro-contaminants from wastewater through a combination of biodegradation and physical adsorption. However, GAC is mostly combined with coagulation and sedimentation processes in drinking water treatment plant to remove MPs (Wang et al., 2020b). In an advanced drinking water treatment plant in China, the overall removal efficiency of MPs reached up to 82.1-88.6% (from 6614 ± 1132 to 930 ± 71 MPs/L) with sand filtration, sedimentation, ozonation and GAC filtration. GAC filtration only showed 56.8–60.9% of MPs removal efficiency mainly for small size MPs (Wang et al., 2020b). Granular filtration showed high ability range to filtering out MPs from 86.9% ± 4.9% (removal for middle sized particles,10-20 μm) to 99.9% ± 0.1% (removal for large sized particles,106-125 μm) (Zhang et al., 2020c). By additional GAC filtration during drinking water treatment, Pivokonsky et al., (2018) reported the average 81% and 83% removal efficiency of MPs (≤ 1 μm) in two plants (**Table A11**). Wang et al., (2020b) showed that the particles size between 1-5 μm was efficiently removed (73.7–98.5%) by GAC filtration process. They also revealed that GAC filtration efficiently removed different types of MPs, including fibers (38-52.1%), spheres (76.8–86.3%) and fragments (60.3–69.1%). Therefore, GAC filtration process would be an effective technology for MPs removal at low MPs concentration ranges. However, the mechanism to remove MPs in GAC is yet unclear.

6.1.4.2. Rapid sand filtration (RSF)

Due to the low operational and maintenance cost, RSF is the most widely applied water treatment worldwide. In the presence of three layers (anthracite grains, silica sand, and gravel), RSF is accelerating to grab suspended solids either by physical adsorption or mechanical straining (**Figure A7**). RSF very efficiently removed solid particles such as MPs from wastewater in the WWTPs (Simon et al., 2018). By the hydrophilic interaction, MPs are adhered to the surface of the sand grains or adsorbed with silica grains and clogged the second layer, which reduces the performance (Enfrin et al., 2019). Previously used coagulation process can increase the production of RSF to reduce the concentration of MPs from wastewater effectively. The addition of coagulant could further improve the adherence (Talvitie et al., 2017a). Enfrin et al. (2019) reported that NPs/MPs adsorption was hard to reverse due to the presence of functional groups such as hydroxyl groups on the surface of NPs/MPs resulting in the stronger interaction (Cai et al., 2018).

In a pilot-scale RSF tertiary treatment in Kakolanmaki WWTP, Turku City, Finland (Talvitie et al., 2017a), RSF process removed 97% MPs (0.7-0.02 MPs/L). of all shapes and size, even the smallest size fractions of 20-100 μm. Hidayaturrahman and Lee (2019) revealed that the removal efficiency of MPs in RSF stage was 73.8%, and the overall removal rate was 98.9% when using the RSF in tertiary stage with coagulation process. Therefore, RSF was regarded as a suitable technology for MPs removal at low MPs concentration range. These studies showed that MPs removal efficiency in RSF process is smaller than the MBR process. Thus, RSF process can be very attractive if this process is combined with other treatment process such as coagulation. Moreover, the efficacy of RSF process is more significant than dissolved air flotation, GAC and membrane filtration, demonstrating its potentials as an effective process for MPs removal in WWTP.

6.1.4.3. Disc filter

Disc filter (DF) is a promising technology to decrease the concentration of MPs in wastewater treatment effluent. Many studies reported that micro screen filtration with DF was applied in the final polishing stage of different WWTPs in many countries (Hidayaturrahman and Lee, 2019; Talvitie et al., 2017a). DF as a full-scale tertiary treatment was used in Viikinmaki WWTP at Helsinki (Talvitie et al., 2017a). Particles were removed with sludge cake formation inside the stacks of round filter based on the physical retention in filters. To remove the sludge cake, high-pressure backwash was executed. The pore size of filter mesh which is a woven material, typically PP, polyester or polyamide, is generally 10-40 μm. Coagulation step before this process enhanced the removal efficiency. Hydraulic retention time, water flow, membrane fouling and pore size are the main factors of DF process. Membrane fouling also occurred in disc-filters and led to more frequent high-pressure backwash (Talvitie et al., 2017a). Due to this high-pressure backwash to clean DF, the secondary membrane layer (biofilm layer on DF) can be lost and MPs easily pass through membrane. Hidayaturrahman and Lee (2019) reported an overall MPs removal efficiency of 99.1% in a WWTP with membrane disc-filter (10 μm pore size), while the MPs removal efficiency of 79.4% at DF stage. In another study, Talvaite et al. (2017a) observed the removal performance of various types of MPs by DF in a WWTP effluent was 40-98.5%. With DF, the MPs concentration was reduced from 0.5 to 0.3 MPs/L with 10 μm filter and from 2.0 to 0.03 MPs/L with 20 μm filter. Generally smaller sized filters shown higher removal efficiency for MPs. However, the opposite result in this investigation was owing to the interruption of preceding treatment stages. After that, DF can effectively remove all shapes and size fractions, however, a portion of smallest size fraction (20–100 μm) passes through the filter entering the final effluent (Talvitie et al., 2017a). From the literature review the DF offered a comparatively low efficiency in MPs removal.

6.1.5. Flotation processes

Wang et al. (2019b) reported that selective flotation could easily remove fine plastics. Flotation with elutriation, combined with a most commonly used hypersaline solution (density flotation) or a surfactant such as sodium dodecyl sulfate, is widely applied for the isolation of MPs from seawater samples. Carr et al. (2016) separated MPs from WWTP influent by using the elutriation method, which was developed by Claessens et al. (2013) to isolate MPs from sediments. The MPs were isolated by exploiting their inherent buoyancies based on a combination of water flow and aeration. Thanks to many MPs' low density and buoyant properties, flotation is a suitable process for those MPs removal from wastewater (da Costa et al., 2016; Di and Wang, 2018). One of the popular flotation processes is dissolved air flotation (DAF), where water is aerated to forming dispersed water at high pressure after processing to promote flocs formation by coagulation, to remove the flocculent materials and remaining suspended compounds including total suspended solids, oil and grease based on their buoyancy. To augment flocculation, flocculation chemical or coagulant is added to wastewater before the flotation. DAF was examined as a full-scale tertiary treatment plant at Paroinen WWTP in the city of Hameenlinna, Southern Finland (Talvitie et al., 2017a). During flotation, low-density MPs were efficiently removed by transported to the surface of the flocs due to their buoyancy and floated to the water which were removed by skimming (Talvitie et al., 2017a). Enfrin et al., (2019) reported that this process led to 20-70 μm sized air bubbles dispersed in water, which adhered to the suspended matter (flocs) and resulted in an air-solid complex to float to the water surface, from where it is removed by skimming. Talvitie et al., (2017a) reported 95% removal performance of various types of MPs by DAF process from the WWTP effluent. They also indicated that dissolved air floating removed any sized of MPs with the smallest size of 20–100 μm. However, there have been insufficient investigations to assess the performance of DAF in removing MPs under various conditions such as density, size, shape, and composition of MPs. Thus, there is a striking research gap for future study.

6.1.6. Magnetic separation process

Among all other extraction methods such as flotation (density separation), chemical digestion and sieving or filtration, magnetic extraction is used to isolate the small-sized (< 20 μm) MPs effectively. Magnetic separation is the most reliable for the separation of MPs/NPs from sediment or water samples under magnetic force, although is not suitable for MPs removal in WWTP. This method is particularly effective for small-sized MPs, because of their large surface area to volume ratio which enhances the binding affinity of MPs with Fe nanoparticles. Grbic et al. (2019) recently observed the performance of magnetic extraction for MPs isolation from seawater, freshwater and sediment. Fe nanoparticles were coated with hexadecyltrimethoxysilane (HDTMS) to create the hydrophobic characteristics for allowing binding with MPs, which helps to isolate the MPs from water under a magnetic field (**Figure A8**). The hydrophobization mechanism of Fe nanoparticles is that the methoxysilane group (-Si-OCH_3) of HDTMS reacts with hydroxyl groups in the native oxide layer of Fe and form siloxane bonds (−Si−O−Si−) which are covalently bonded. As a result, the hydrophobic hydrocarbon tail (alkyl chain) of HDTMS in Fe nanoparticles' surface makes hydrophobic, simplifying to sorption MPs. They reported 92% and 93% removal of small-sized (< 20 µm) PE and PS and large-sized (> 1 mm) MPs from seawater. The recovery rate of medium-sized (200 µm-1 mm) MPs was 84% and 78% from freshwater and sediment. Additionally, lipophilic substances (e.g. fat in fish tissues) in the sediment samples can negatively affect the removal efficiency. Therefore, MPs recovery by magnetic extraction process is particularly suitable for drinking water treatment (Grbic et al., 2019).

6.1.7. Comparison among physical processes

Physical treatment methods can be applied to remove a wide range of MPs from water, with their average removal efficiencies represented in **Figure 2a** and their comparison summarized in **Table 2**. A wide range of MPs can be removed through some filtration process such as GAC filtration, rapid sand filtration and disc filter. Dissolved air flotation process also attractive to remove MPs efficiently with the flocculation process. Membrane treatment such as UF, RO and DM technology can be more effective if this process is combined with MBR. UF process is effective only for PE MPs removal. Among all other membrane treatment technology, DM technology is a desirable, cost-effective and highly efficient technology to remove MPs from synthetic wastewater but still insufficient to remove large scale MPs from wastewater. On the other hand, density separation and magnetic separation are more efficient to remove MPs from sample water. The adsorption process is suitable to adsorb MPs from water but this process was not studied sufficiently. Other physical process which applied in WWTP has not been studied extensively for MPs removal. Moreover, among the physical treatment technologies, the quantitative analysis revealed that filter-based methods showed the better MPs removal efficiency than others. Therefore, filter-based treatment technologies (UF, RSF, DF, GAC filtration) achieved the best performance in eliminating MPs. Among them, RSF treatment process provides rapid and efficient removal of MPs. MPs removal though physical methods followed the order: filtration process > flotation process > adsorption process > membrane process > magnetic and density separation process. Furthermore, a more detailed characterization of MPs in different treatment technologies is needed to select the most suitable methodologies for the efficient removal of MPs from the WWTP effluents.

[Figure 2]

[Table 2]

6.2. Biological treatment technologies

Secondary treatment in a WWTP assembles the (i) biological treatment processes and (ii) clarification process, following the preliminary treatment (35-59% MPs removal) and the primary treatment, removes 50-98% MPs (Sun et al., 2019). Different results have been obtained while evaluating the efficacy of MPs removal from biological wastewater treatment. Murphy et al., (2016) reported < 20% MPs removal efficacy while Sun et al., (2019) reported 0.2-14% decrease in MPs from wastewater in the secondary treatment. Other researchers reported 2-55% removal of MPs in biological treatment processes (Lv et al., 2019; Yang et al., 2019). These variations in MPs removal are due to (i) the change of microbes, (ii) nature of the MPs in the wastewater (size, shape, taste, surface structure), and (iii) abiotic factors (e.g. temperature, pH). Of biological treatment processes, microbial treatments (activated sludge method, biofilm-related process), MBR technology, aerobic digestion, AD and CWs have been identified as the most widely used and efficient method for MPs removal.

6.2.1. Microbial treatment

6.2.1.1. Activated sludge process

In the activated sludge process, the sludge is first completely mixed with oxygen in a reactor, which incites the microorganisms to use the sludge as their food. MPs removal in this process occurs via adsorption, degradation or aggregation. Microorganisms secret EPS to absorb the accessible contaminants as well as MPs and then degrade them to produce desirable products. Sometimes microbes take MPs mistakenly because of the visual similarity with their nourishments and then egest them after agglomerating into flocs due to their inability to degrade or transform the MPs into harmless substances**.** Activated sludge process along with modified processes such as anaerobic-anoxic-oxic process, sequencing batch reactor process and oxidation ditch, achieved 3.6-42.9% removal of MPs from wastewater (Carr et al., 2016; Lares et al., 2018; Mason et al., 2016). Generally, MPs are not degraded or mineralized in the activated sludge processes, but mainly removed from wastewater by the aggregation with sludge flocs. This process exhibited discrimination for different sizes and shapes of MPs. For example, Liu et al., (2019a) found that most of the MPs removed in the activated sludge process were < 300 μm in size, whereas other researchers obtained the most removal efficacy for 1-5mm sized particles (Lares et al., 2018). Zhang et al., (2020a) suggested that MPs removal variations were due to the difference in the MPs shapes, with high MPs removal efficacy for fibre shaped MPs. Since the MP particles are not completely degraded or mineralized in this process and often end up in sludge flocs, this process has been followed in only some rare studies to date. Due to the incomplete disposal of the pollutants, the non-degraded MPs portion present in the sludge further easily incorporate with terrestrial ecosystems and spread again throughout the entire environment. The fate and treatment of these non-degraded MPs in sludge phase have been rarely discussed in literature. Hence, more research is urgently required on the topic.

6.2.1.2. Biofilm process

A tire of micro-organisms growing on the surface of MPs or other carriers is defined as biofilms. In this process, biofilms undergo periodically three repeating steps of (i) growing phase, (ii) stationary phase, and (iii) peeling period. After peeling one tire of biofilm, and another new film starts to form and acts to remove more MPs and other contaminants in wastewater (Zhang et al., 2020a). Biofilm related processes remove contaminants via adsorption and fixation. Like the activated sludge process, firstly MPs are adsorbed by the microorganisms with the help of EPS, secreted by the microorganisms. MPs become an attachable carrier and support the microbes in their augmentation. After a stationary phase, biofilms start to collapse from the surface of the carrier, accumulate the contaminants and emerge with the treated water (**Figure 2d**). Biofilms also play an important role in the biodegradation of MPs.

6.2.1.3. Biodegradation

Biodegradation is an eco-friendly process compared to other treatment methods, by converting organic substances into their fragments and eventually CO_2 (Ahmed et al., 2018; Zheng et al., 2005; Gu et al., 2000). Biodegradation involves depolymerisation and mineralization. The process in which complex polymers impair into their monomers, dimers, or short chains of their oligomers, which can transgress through 'bacterial membranes' and act as a source of energy and carbon, is referred to as depolymerisation. Mineralization refers to the process in which the final products are water, carbon dioxide and methane. MPs undergo microbial breakdown using the activity of exoenzymes which promote depolymerisation or assimilation by the microbial species and result in mineralization (Shah et al., 2008; Yoshida et al., 2016). MPs are usually non-biodegradable but their biodegradability can be increased in two ways, *i.e.* by reducing their polymeric chain-length to such an extent that is accessible for microbial growth, and by increasing their hydrophilicity.

Biodegradation initiates with the formation of biofilms, which are shoals of microorganisms attached on various biotic or abiotic surfaces and deflect the surface characteristics and entity of MPs as well as to improve the hydrophilicity of the surface (O'Toole et al., 2000). Biofilms can be made of a single microbial species but often by multiple species. The formation and development process of biofilms involves four stages (i) initial attachment, (ii) irreversible attachment, (iii) maturation, and (iv) dispersion. The formation, mode of action and MPs removal by biofilms are presented in **Figure 3**.

[Figure 3]

Such type of bio-coatings contributes to MPs removal by acting as (i) a wetting agent for MPs which modifies MPs surface properties (e.g. density) to help the particles for skimming up or settling down in wastewater (Rummel et al., 2017), and (ii) the initial step for the postero biodegradation process.

Various mechanisms for biodegradation of plastics and MPs have been proposed. Lucas et al. (2008) suggested that the biodegradation of plastics and their fragments occurred first by cleaving enzymatically the polymers into their oligomers and monomers which were then assimilating by microbes. Similarly, Gu and Gu (2005) proposed that the biodegradation started with the cleavage of polymer backbone or its side chains by the act of extracellular enzymes, which resulted in the formation of smaller polymer units (i.e. monomers, oligomers). In most cases, it involves the hydrolysis of amides (in polyamides), esters (in polyesters), or urethane (in PU) bonds where the extracellular enzymes act as catalysts. In addition, abiotic hydrolysis can facilitate the polymers for cleavage (Müller et al., 2001). The cleaved and simpler molecules are then absorbed and metabolized by the microorganisms. These alterations further promote biodegradation (depolymerisation and mineralization). Plastics and MPs can undergo biodegradation both in the aerobic and anaerobic environments (Klein et al., 2018), but three main conditions should be maintained:

i. Presence of appropriate microbes capable of depolymerizing polymer substrate and mineralizing monomeric fractions by enzymes through metabolic pathways;

ii. Proper environmental conditions for the biodegradation (e.g. temperature, pH, nutrients);

iii. Morphology of the polymer substrate should be favourable for the microbial attachments and formation of biofilms.

In addition, a number of biotic and abiotic factors (**Table A15**) can affect the uptake and biodegradation of MPs even though the appropriate microbes are present in the environment. For the uptake of MPs (as a food of the microbes), physiological characteristics of MPs, their size and feeding type act as the major biotic factors; while temperature and pH are abiotic factors. The polymeric substrate's molecular weight, chemical composition, hydrophobicity, size of the invaded molecules, and other environmental conditions (e.g. temperature) were reported as essential factors. Obtaining an optimum environmental condition helps to improve the effectiveness of microbial treatments for MPs removal. A combination of field-based assessments and laboratory-based trials is required to facilitate the application of microbes for the degradation of different types of plastics and MPs under different environmental conditions.

6.2.1.4. Microorganisms for microbial treatment

Several studies have reported the ability of some microbes for the degradation and ingestion of MPs from the environment. These microorganisms were isolated from different ecosystems (e.g., terrestrial to marine environment) to evaluate their capability in plastics and MPs removal processes. A large number of marine organisms including amphipods, decapod Crustaceans (Murray and Cowie, 2011), lungworms and barnacles (Thompson et al., 2004) were reported to have the capability of ingesting MPs in several studies. Some studies demonstrated the uptake of MPs by planktons as well as the lungworm *Arenicola marina* (Besseling et al., 2013) and the blue mussel *Mytilus edulis* (Wegner et al., 2012), while others showed the MPs ingestion by gastropods and zooplanktons (e.g. *Daphnia magna*) (Eerkes-Medrano et al., 2015). Planktonic organisms like chaetognatha, copepods, larval fish, salps, zooplanktons have been detected to uptake MPs (Moore et al., 2001). Cole et al., (2013) conducted experiments on 13 zooplankton taxa which included micro-zooplanktons, holoplankton, and meroplankton. However, some negative impacts on the zooplanktons, such as reduced health function of the individuals, transfer of contaminants in the food web, were also observed. Along with zooplanktons, primary consumers such as herbivorous, bacterivorous, detritivorous, and some deposits feeders can also ingest MPs (Scherer et al., 2018). Even some proofs of MP ingestion were found for protozoa or metazoans (Scherer et al., 2018). Most importantly, a diversity of invertebrates such as crustaceans, bryozoans, polychaetes, echinoderms, and bivalves have shown ability to ingest MPs (Murray and Cowie, 2011;).

Suspension feeding organisms including cladocerans, rotifers, mussels, protozoans are enthusiastic to MPs because they swallow suspended particulate matter (SPM) indiscriminately.

Herbivores and bacterivores ciliates (e.g. *Halteria* sp.), rotifers (e.g. *Anuraeopsis fissa),* cladocerans (e.g. *Daphnia* sp.) and flagellates (e.g. *Vorticella* sp.) were reported to feed plastic beads readily. Some aquatic larvae (*Chironomus riparius),* as well as blackworms *(*e.g. *Lumbriculus variegatus)* showed non-selective feeding on a wide range of sediment MPs. Bivalves swallow SPM effectively including MPs, where bivalves such as marine mussels (*Mytilus edulis)*, and freshwater (e.g. *Sphaerium corneum, Anodonta cygnea)* exhibit size selectivity on particles (Scherer et al., 2018). Surface-grazing gastropods (e.g. *Potamopyrgus antipodarum, Physella acuta)* also demonstrated the ingestion of MPs through both foods associated route *(*e.g. *P. antipodarum)* and water sediment-borne route (e.g. *P. acuta*). *Mytilus edulis* showed ingestion of MPs in laboratory experiments. Von Moos et al., (2012) observed the intake of high-density polyethylene, i.e. HDPE in laboratory experiments by the cells and tissues of *Mytilus edulis* although they only ingested particles larger than 80 μm. Microbes of different genera were found to contain such species that have the potential for plastic degradation, including *Aeromonas, Arcobacter, Aquabacterium, Zymophilus* and *Pseudomonas* (McCormick et al., 2014). Various copepods (e.g. *Calanus pacificus, Acartia tonsa*, copepodits, crustaceans (e.g. nauplii), echinoderm larvae, ciliates, *Oxyrrhis marina* (dinoflagellates), and salps showed their ability to uptake MPs, although the size of the MPs was limited to <100 μm range (Cole et al., 2013).

Overall, a large number of microbes have demonstrated their capabilities for MPs uptake, either as their comestibles or mistakenly for various factors of similarity with their nourishments. Therefore, the uptake of MPs does not necessarily mean biodegradation of those particles. Sometimes MPs may have been preserved in the microbial body, affecting the microbes by interrupting their normal biological activities, or may use the microbes as media to diffuse in their ecosystem. Another significant limitation of microbial treatment is that no such microorganism has been found that can interact with all types of MPs under any environmental conditions. Microbes show their selectivity according to the size of MPs, shape of the feeds, taste discrimination, mode of feeding, variability in locus, and individual selectivity of feeds for MPs uptake. For example, *Tenebrio molitor* Linnaeus can chew and swallow styrofoam with the help of their gut bacteria (Yang et al., 2015) while no evidence has been found for their ability or interest to interact with other types of synthetic polymers such as PU and PP. Some bacterial strains (e.g. *Bacillus cereus,* *Pseudomonas otitidis*) were found to degrade only PVC plastic beads (Ali et al., 2014). *Enterobacter* sp., *Alcaligens* sp., *Citrobacter sedlakii*, *Brevundimonas diminuta* exhibited their ability for degradation of high impact PS beads only (Braun, 2004). *Ideonella sakaiensis* 201-F6, a bacterial strain has been found to degrade PET into terephthalic acid and ethylene glycol, and eventually CO_2 and water in aerobic condition with the help of an extracellular and an intracellular hydrolase enzyme (Avérous and Pollet, 2012; Joo et al., 2018; Yoshida et al., 2016). *Bacillus cereus*, *Pseudomonas otitidis* and *Acanthopleurobacter pedis* showed their capability of degrading PVC while some fungi species (e.g. *Lentinus tigrinus, As. fumigatus, As. niger, Phanerochaete chrysosporium, Aspergillus sydowii*) were found to degrade only the plasticized ones (Ali et al., 2014). A significant extent of degradation in PE shopping bags was observed during the simple contact of about 100 *Galleria mellonella* within 12 hours (Yang et al., 2015). *Comamonas acidovorans* was found to degrade only PU as a sole nitrogen and carbon source (Zheng et al., 2005). All of these microorganisms exhibit individual selectivity in MPs uptake. Moreover, the effect of size and shape of MPs have also been noticed in some investigations. For instance, in filter-feeding taxa, the maximum particle size for ingestion is determined by the morphology of mouthparts of the microbes, while in cladocerans, it is determined by the opening width of the carapace. In laboratory experiments with different sized MPs, *Daphnia cucullata* exhibited higher filtering rates for 3 and 6 µm MPs in comparison with the 0.5 µm sized beads where *Conochilus unicornis* (rotifer) filtered 3 µm MPs more frequently than 0.5 µm MPs (Agasild and Nõges, 2005). Therefore, microbial treatment process of MPs requires the presence of suitable microorganisms, appropriate environmental conditions, and specific types of MPs in order to obtain higher efficacy for MPs removal.

6.2.2. Membrane bioreactor

MBR has gained recognition in the last few decades due to its high effluent quality, small footprint, diversity and scalability. Sun et al. (2019) reported that MPs concentration in wastewater was reduced from 10044 particles/L to < 450 particles/L by conventional wastewater treatment. Some researchers showed that WWTP could remove up to 99% MPs (Carr et al., 2016;). On the other hand, some researchers showed that wastewater treatment was not efficient for the removal of MPs (Leslie et al., 2017). Despite the high efficiency to removal MPs by conventional wastewater treatment processes, advanced treatment is required to decrease the number of MPs from the final effluents (Bayo et al., 2020; Carr et al., 2016; Mintenig et al., 2017; Talvitie et al., 2017b; Yang et al., 2019). Lares et al. (2018) observed that MBR treatment technology reduced the concentration of MPs in influent from 57600 MP/m^3 to 400 MP/m^3, and showed a better removal (99.4%) of MPs compared to the activated sludge treatment (98.3%). Mintenig et al. (2017) reported 95% removal of the 20-500 μm MPs. Further, Carr et al. (2016) reported 99.9% removal of MP from the secondary treatment processes. Michielssen et al. (2016) suggested that MBR is more efficient than conventional activated sludge process, and removed 99.4% of MPs from wastewater. Talvaite et al. (2017a) reported 99.9% MPs removal from wastewater by MBR. Blair et al. (2019) demonstrated that advanced WWTP could efficiently remove MPs in the size range 60-2800 μm. The MP particles in the range of 100-200 μm in various WWTPs are widely reported (Carr et al., 2016; Kalčíková et al., 2017; Ziajahromi et al., 2017). Talvitie et al. (2017b) revealed 70% and > 95% removal for MP in size range 20-100 μm and 20-300 μm in wastewater, respectively. Among all shapes of MPs, fibers have been seen to dominate the effluent (average 75%) of WWTPs (Dris et al., 2016; Michielssen et al., 2016). Ruan et al. (2019) observed that the most abundant shape of MPs was fibers in influent (55-71%) which is consistent with previous studies (Lares et al., 2018). Murphy et al., (2016) detected no PE microbeads in the effluent of secondary WWTP in Scotland. A few studies have shown that fibers were more effectively removed than fragments in pre-treatment (Talvitie et al., 2017b), at the same time fragments were more effectively removed than fibers in the secondary treatment process (Sun et al., 2019; Wang et al., 2018a). MBR process showed a high removal capacity for all size fractions (especially the smallest size, 20-100 μm) and all shapes of MPs from wastewater compared to other advanced treatment (Talvitie et al., 2017a). Lares et al., (2018) reported that MBR permeate contained an average of 0.2 fiber MP/L.However, compared with other treatment technologies, the performance of MBR seems to be not influenced by the shape, size and composition of MPs. It can be concluded that MBR is the most promising technology to address MP problem. Further, the effect of MPs on membrane fouling and the degradation and/or transformation of MPs in MBR should be studied in the future research.

6.2.3. Anaerobic and aerobic digestion

Aerobic digestion is the natural attenuation process of heterogeneous organic matters or contaminants by the acts of mixed microbial communities in the presence of oxygen, and occurs in a warm and moist environment (Mohee et al., 2008). It is also designated as an aerobic composting process where the organic matter is decomposed via microorganisms that can sustain in the presence of oxygen. The common end products are water vapour, CO_2 and dark brown to black coloured organic effluents, defined as compost. On the contrary, anaerobic digestion (AD) process is referred to the attenuation process through which the complex organic materials are disintegrated in the absence of oxygen as a consequence of metabolic interactions in microorganisms to yield CO_2, methane, ammonia, hydrogen sulphide, water, hydrogen, and other compost products (Mohee et al., 2008). AD can also be defined as biogasification process that mainly converts the biodegradable organic wastes to biogases, which can be further combusted to produce electricity and heat or further processed to become a source of renewable energies (Van Doren et al., 2017). AD process has drawn attention in recent years because of its capability of destroying pathogenic microbes, reducing sludge amount, recovering sustainable energy as well as the production of biogases at the same time. Mahon et al., (2017) analysed the effluents of seven WWTPs after treatment by AD, lime stabilization and thermal drying, and detected comparatively lower MPs abundances, hence in favour of accepting AD as an effective strategy for MPs treatment. Moreover, while treating sludge without and with plastic wastes in AD process, comparatively high production of biogases in the plastic containing batch proved the feasibility of plastic waste removal (Mahon et al., 2017).

6.2.3.1. Comparison between aerobic and aerobic digestion for MPs treatment

The rate of digestion for some biodegradable plastic polymers has been found to be higher in aerobic conditions than in anaerobic conditions. In experimenting with polycaprolactone, polylactic acid, polybutadiene adipate-co-terephthalate, and starch/polycaprolactone blend in both aerobic and anaerobic conditions, Massardier-Nageotte et al. (2006) demonstrated a higher biodegradation in aerobic conditions than under anaerobic condition. Moreover, polycaprolactone exhibited a significant biodegradability (35%) under aerobic condition while no biodegradation was observed in anaerobic condition. The biodegradation of polybutylene adipate terephthalate (Kijchavengkul et al., 2010) and ecoflex® (Witt et al., 2001) has been reported at aerobic conditions. Therefore, aerobic digestion may be a significant method for the destruction of biodegradable MPs. Some abiotic and biotic factors (e.g. temperature, pH, enzymes, biosurfactants) as well some internal and external characteristics of the substrate (e.g. crystallinity, functional groups, chain flexibility, molecular weight) can influence the rate and extent of biodegradation in aerobic or composting conditions (Dřímal et al., 2007; Kale et al., 2007; Kijchavengkul et al., 2010).

Biodegradation of MPs has gained attention due to its performance of degrading petroleum-based polymers (e.g. PE, PP) (Gómez and Michel, 2013). In addition, methane production from AD has made the process more attractive as a contributor to renewable energies, and therefore offering both environmental and economic rewards. Some of the most commonly used biodegradable plastics reported are poly(lactic acid), poly(ε-caprolactone), poly(β-hydroxybutyrate), poly(β-hydroxybutyrate-co-11.6%-β-hydroxyvalerate) (Gómez and Michel, 2013; Kolstad et al., 2012).

Abou-Zeid et al. (2001) tested the biodegradability of natural and synthetic polyesters, based on the percentage weight loss. Natural polyesters such as poly-β-hydroxybutyrate (PHB) and poly β-hydroxybutyrate-co-11.6%-β-hydroxyvalerate (PHBV) exhibited complete and 60% conversion in aerobic condition respectively, in just 9 days. Poly(o-caprolactone) or PCL, a synthetic polyester showed 30% weight loss at the same time. PHBV polymers were found to be biodegraded at about 85% extent within 45 days in the dry aerobic condition in another research (Mohee et al., 2008). *Clostridium*, an anaerobic species was found to be capable of hydrolysing PCL and PHB polyesters in anaerobic conditions (Abou-Zeid et al., 2001; Perz et al., 2016). Other than virgin biodegradable polymers, some materials have come into the markets with various applications as "biological products", "degradable", "green", "compostable", "oxo-biodegradable", although very few studies have been performed to determine their biodegradation extent in AD and/or composting process (Gómez and Michel, 2013).

Gomez and Michel (2013) observed 20-25% conversion of bio-based materials within 50 days in anaerobic conditions while the additive containing conventional plastics was converted into biogases only at 2%. They tasted with (i) additives containing conventional plastics (e.g. PET incorporated with 1% additives, PP with 2% additives), (ii) conventional plastics incorporated with plastarch and other additives, (iii) co-polyesters with corn-based plastics, (iv) paper pulp and soy wax mixture, and (v) plastarch, to compare their relative biodegradability during composting and anaerobic condition. Plastarch (without additives) were found as the most biodegradable material while conventional plastics with additives exhibited the smallest extent of biodegradation.

Moreover, some synthetic non-biodegradable MPs interrupted the normal digestion of biodegradable materials. Wang et al., (2013) investigated the influence of PE-based MPs on the AD process, and reported a comparatively low rate of methane production from the toxicity of polyethylene MPs (100-200 particles/g) on the normal activity of the relevant microorganisms.

The abundances of hydrolytic microbes (e.g. *Rhodobacter* sp.), certain bacterial genera (relevant to acidification) and protein utilizing *Proteiniclasticum* sp. (relevant to acetogenesis) significantly decreased due to the exposure to PE MPs during AD (Luo et al., 2016; Wang et al., 2018b). MPs of PET polymers inhibited hydrolysis, acetogenesis and acidogenesis in anaerobic fermentation of biodegradable polymers via (i) shifting the microbes towards the opposite direction of hydrolysis and acidification, (ii) raising the ROS (reactive oxygen species) level that causes the death of more microbial cells (Wei et al., 2019b). The negative impact of PVC MPs also has been reported in AD system (Wei et al., 2019a). Therefore, aerobic or AD processes have limited performance in degrading non-biodegradable polymers or MPs. Biodegradable plastics that have predetermined durability, are strongly recommended to be used as alternatives to non-biodegradable plastics. Aerobic and AD with appropriate conditions can be applied as an effective way for their degradation, although the maintenance cost would be higher than that of CWs.

6.2.4. Constructed wetlands

CWs are familiar and natural technology for wastewater treatment with a comparatively lower cost than other biological treatment methods. Studies have been conducted recently to investigate the feasibility of MPs removal from wastewater using CWs (Liu et al., 2019b; Ziajahromi et al., 2020). Vegetated wetlands are the prime locus for detaching, storing, transforming, and finally releasing MP particles (Helcoski et al., 2020). A few studies have been conducted on the contribution or performance of vegetated wetlands, including natural and CWs in MPs removal from polluted water (Helcoski et al., 2020; Wang et al., 2020a). Plenty of macro-invertebrates (e.g. snails, bristle worms, beetle) have been found in the wetlands capable of playing a significant role in controlling the accumulation of sludges (Ouattara et al., 2009). Wang et al. (2020a) showed the effective role of macro-invertebrates in MPs distribution throughout the wetlands. They claimed that macro-invertebrates of the wetlands ingest a non-negligible amount of MPs. Over 90% removal efficacy was achieved in both horizontal and vertical flow type CWs. Wang et al. (2020a) obtained 88% average efficacy of MPs removal in CWs which is comparable with other conventional tertiary treatment methods of WWTPs, such as biological filtration (84%), dissolved air floatation (95%), DF (40-98.5%), MBR (99.9%), and sand filters (97.1%). 98% MPs removal efficacy was obtained through the whole WWTP when CWs were used in its tertiary treatment steps. Therefore, CWs can be an efficient, environmentally friendly and cost-effective tertiary treatment process to reduce MPs significantly from wastewater. Moreover, the efficacy can be enhanced through integrating different features of different types of CWs (*e.g.* surface flow CWs, subsurface flow vertical type, subsurface flow horizontal type CWs). Therefore, further combined applications of such different CWs are strongly recommended for MPs removal from wastewater.

6.2.5. Comparison among different biological processes

Biological treatment methods can be applied to remove MPs at a significant extent from different environmental conditions. A comparison between the efficiencies of different biological treatments is represented in **Figure 2b**. MBR process and CWs showed the best efficacy among all of them. Conventional activated sludge process also reached similar removal percentage, but only in limited works. It is difficult to declare any exact removal percentage for microbial treatment processes because they always fluctuate according to the microorganisms involved. On the other hand, aerobic digestion and AD can be applied efficiently only for biodegradable MP particles. A comparative overview of the biological methods, their advantages and drawbacks are summarized in **Table 3**. The removal of MPs via biological methods decreased in the order: MBR > CWs > activated sludge > microbe processes. The MBR process and CWs have potential in leading biological methods of MPs removal.

[Table 3]

6.3. Efficacy of chemical treatment technologies of MPs

In addition to physical and biological methods, chemical methods are used in MPs treatment purposes, either on their own e.g. chemical oxidation, advanced oxidation process, coagulation, and EC, or used as an aid to improve the efficiencies of physical processes (e.g. density separation process, elutriation). Several approaches e.g. catalytic, thermal, photo-oxidative and chemical (ozonation, advanced oxidation) degradation of waste plastic materials have been discussed in multiple studies.

6.3.1. Oxidation

Several studies have reported MPs removal by oxidizing agents (e.g. ozone, hydrogen peroxides, oxidizing acids) as well as some advanced oxidation methods (e.g. electro-Fenton process, photo-Fenton process). Eventually, chemical oxidation aims to mineralize the polymeric substances and convert them into CO_2, water and other minerals. In some cases, radiation from different sources (e.g. UV-vis radiation, solar energy), electric current, and ultrasound are used to improve the efficiencies of these oxidation process (Miao et al., 2020). Advanced oxidation processes (AOPs) have gained much attention in recent years, by mineralizing the targeted substances by producing highly reactive oxidizing species (*e.g.* $^{\cdot}OH^{-}$ radicals) under moderate conditions (Klavarioti et al., 2009). Although many AOPs have been developed and implemented for wastewater treatment purposes (Feng et al., 2011), very few of them are used for MPs treatment. Ozonation, photo-Fenton, electro-Fenton method, photocatalytic oxidations are the most widely used and efficient methods.

6.3.1.1. Ozonation

Ozone is known as one of the most potent ancient oxidants that can react with various polymeric substances, with the unsaturated bonds as well as the aromatic rings of the polymers (Ahmed et al., 2017). Although very few studies have been reported on the influence of ozone in MPs treatments, several studies have corroborated its effective influences on polymer degradations (Chen et al., 2018), via highly reactive secondary oxidant species (e.g. hydroxyl radicals). This process is applied either as a direct treatment method for MPs removal or used to improve the efficacy of some conventional biological methods by facilitating biodegradation of the suitable polymers. There is some evidence of significant changes in PE, PP, PET polymers exposed to ozone. For instance, Chen et al., (2018) reported a high polymer degradation rate (> 90%) at 35-45 ^{o}C by exposure to ozone. Ozonation may facilitate polymer degradation by increasing polymer surface tension, boosting the polymer surface's adhesion properties, reducing hydrophobicity and increasing solubility, reducing intrinsic viscosity, and decreasing melting points of the polymers and modifying mechanical properties. Hidayaturrahman and Lee (2019) obtained the highest MPs (particle size 1-5 μm) removal efficiency in ozonation method (89.9%) compared to other advanced treatment methods such as membrane disc-filter (79.4%) and rapid sand filtration (73.8%). These methods followed the other primary and secondary treatment methods and coagulation **(Figure A9).** Moreover, enhancement of 17.2-22.2% removal efficacy was obtained by integrating the GAC-filtration method with ozonation. There is evidence of enhancing microbial mineralization and removal efficacy of MPs with ozonation. In a laboratory-based ozone investigation, mineralization of β-^{14}C PS films by *Penicillium variabile* was found to be increased significantly from 0.01±0.003% to 0.15±0.03% (Tian et al., 2017). These cases proved that ozonation could be used as a useful tertiary treatment step in wastewater treatment. The main challenge with ozonation is the high production cost of ozone and environmental issues (Ahmed et al., 2017).

6.3.1.2. Fenton process

Fenton process has been one of the most widely used AOPs for wastewater treatment. In this process, highly reactive hydroxyl radicals are generated from the reaction of hydrogen peroxides (H_2O_2) and Fe^{2+}-containing heterogeneous catalysts, which further drastically oxidize the targeted organic impurities and other contaminants to CO_2, water and mineral products. This process enhances the oxidation power of hydrogen peroxides with the help of an iron-catalyst. As iron is a non-toxic and abundant element of the environment, this method has become popular. MPs are rarely affected by Fenton process. Tagg et al. (2017) examined the influence of Fenton's reagent on PE, PP and PVC MPs, and observed no significant changes in any of the MPs even at three different doses of H_2O_2 and Fe^{2+}-containing catalysts (*i.e.* $FeSO_4.H_2O$). An average of 25.49% removal efficacy was obtained within 24 hours through this modified Fenton process. A significant effect of pH was observed, as an enhancement of 1.69%-3.89% removal efficacy was attained by adding sodium pyrophosphate as a chelating agent (at pH 7.95). Overall, more investigation is required for the effective implementation of the Fenton process in MPs treatment.

6.3.1.3. Electro-Fenton (EF) process

To overcome the limitations of classical Fenton process, EF method has been newly developed in which the primary oxidant (H_2O_2) is generated electrochemically under moderate conditions (de Luna et al., 2012), with the rest of the process being the same as the classical Fenton process (Ganiyu et al., 2018). Despite being a popular AOP for wastewater treatment, very few studies have been reported on MPs treatment with this process. Miao et al. (2020) applied EF method to degrade PVC MPs and obtained 75% dechlorination and 56% weight loss efficacy within 6 hours of the experiment at 100 ºC. TiO_2/C cathode was used to gather such higher efficiency, compared with only 29% dechlorination efficacy using graphite cathode. As this method is effective for PVC MPs, this can be a potential method for other chlorinated species such as 2,4-dichlorophenol, PS, PP and PE also (Miao et al., 2020). More modification in cathodic materials and other environmental conditions may make this method more efficient in future.

6.3.1.4. Photo-Fenton process

The photo-Fenton process is another efficient AOP approach for MPs treatment, which utilizes UV radiations to produce $\cdot OH^-$ radicals from hydrogen peroxide molecules in the presence of iron catalysts and to destruct the targeted contaminants effectively (Ahmed et al., 2017). Enhanced production of the reactive hydroxyl radicals and a higher rate of propagation is induced in acidic or near-neutral pH conditions. In such pH range, Fe^{3+} ions from the catalyst substances form different light absorptive hydroxyl complexes (e.g. $[Fe(OH)]^{2+}$, $[Fe(OH)_2]^{4+}$), which further generate Fe^{2+} ions and ·OH radicals by utilizing the absorbed UV/visible light energy. The produced oxidized reactive ligands then transform or mineralize the target microcontaminants through different reactions (De la Cruz et al., 2012). This process is more rapid than the traditional Fenton process, and recycling of Fe^{2+} ions can occur at a higher rate. Instead of hydroxyl radicals, sometimes alkyl radicals can also be generated in the same way. Fe^{3+} ions precipitate easily by forming amorphous ferric oxyhydroxides at higher pH range. Consequently, it becomes hard to recycle the Fe^{2+} ions (Ahmed et al., 2017). Therefore, the entire process should be conducted at an optimum low pH condition. While traditional Fenton process failed to induce significant changes on MPs with the single effort of its produced ·OH radicals, UV irradiation was reported to enhance the rate of this oxidative degradation. Feng et al., (2011) reported over 99% mineralization of cross-linked sulfonated PS foams within only 250 minutes in photoassisted Fenton process. Despite being one of the widely used AOPs in wastewater treatment, research on this method for MPs treatment purpose is limited and should be strengthened.

6.3.2. Photolysis and photocatalytic degradation

Photolysis has been used in wastewater treatment processes to remove various contaminants (Ahmed et al., 2017). There is evidence that MPs can also be decomposed into the ultimate end products (water and CO_2) if they are irradiated to UV-radiations for a particular length of time (Tao et al., 2019). The major problem is the lower rate of degradation in the natural weathering process. Brandon et al. (2016) experimented with two types of MPs (PE and PP) under natural weathering conditions, and observed only a slight change with the help of FTIR analysis after 3 years. The prolonged degradation of MP particles resulted in hydroxyl, carbonyl groups, C=O bonds, and other minor products. Among the very few studies on the degradation mechanism, luminance and salinity of the corresponding media have been found to affect the degradation of MPs in aquatic environments (Padervand et al., 2020). Enhanced degradation of MPs was observed in the artificial seawater system where no significant deterioration was sighted at the same periods when the MPs were exposed to UV-radiations alone. Both SEM images and FTIR analysis of the initial and end materials confirmed that the salinity of media could facilitate the photodegradation of MPs (Karlsson et al., 2018). Additional chemicals are being used to enhance the photo-degradation rate of MPs, as photocatalysis has gained popularity during the past few years for MPs treatment.

Photocatalysis process initiates with the excitation of the corresponding photocatalyst through the absorption of an appropriate amount of energy from a definite light source. This photo-excitation results in the generation of 'exciton pair's which further propagate other reactive species (e.g. hydroxyl radicals, superoxides) by interacting with moisture or water molecules from the surrounding environment (figure 4b). The highly reactive radicals then oxidize various organic contaminants, including polymers effectively (Ali et al., 2016; Padervand et al., 2020; Qi et al., 2017). It has been proposed to be an energy-efficient, durable, and cost-effective process for polymer degradation (Tofa et al., 2019).

Different mechanisms of MPs photocatalysis have been proposed, among which hydroxyl radicals promoting degradation process (Liang et al., 2013) have been mostly granted, and confirmed via FTIR analysis of the generated intermediated compounds during the degradation process. Various nanostructured semiconductors are used as photocatalysts to generate the desired reactive species, of which metal oxide nano-materials having semiconducting properties with a particular bandgap (ZnO, TiO_2) are most appropriate. As photocatalysis is a type of surface phenomenon, nano-sized materials are especially used because of their high surface to volume ratio. ZnO nanoparticles are considered as one of the most promising photocatalysts due to their appropriate bandgap for catalysis (3.37 eV), high redox potential, non-toxicity, excellent electron mobility and optical properties, ease of synthesis, and flexibility in sizes and shapes to be formed (Qi et al., 2017). Photocatalytic degradation of LDPE based MPs was investigated through heterogeneous rod-like zinc oxide nano-catalysts (Tofa et al., 2019).

Photo-active micromotors have gained massive attention during recent years due to their extensive capability for environmental contaminants remediation and water purification (Eskandarloo et al., 2017; Zhang et al., 2018). Recently, several studies have been conducted on the degradation capability and mechanism of TiO_2-based nano-devices and micromotors in photocatalysis of MPs. MPs have been treated in the photocatalytic process by using Au-decorated TiO_2-micromotors to make this process more efficient (Wang et al., 2019c). Protein-based N-TiO_2 photocatalysts were also reported to hold the potential of degrading MPs in both aqueous and solid phases. Ariza-Tarazona et al. (2019) obtained 6.40% mass loss of high-density polyethylene MPs within 18 hours, while irradiated it with visible light radiations in the presence of N-TiO_2 photocatalysts. The catalyst surface area, as well as the extent and nature of interactions between the MPs and catalyst surface, influenced the removal efficacy significantly (Ariza-Tarazona et al., 2019). As this method is very new in MPs treatment compared to the other conventional and advanced treatment methods, future research is strongly required to obtain more effective and efficient advanced photocatalysts, so that the method can be successfully applied for MPs treatment in real wastewater..

6.3.3. Coagulation

As MP particles are tiny in size (diameter < 5 mm), it is highly challenging to separate them through filtration processes continuously. Difficulties like membrane fouling make them often inefficient and discontinuous, hence, pre-treatment by coagulation or flocculation before filtration process will improve filtration efficiency. Coagulation and agglomeration processes are widely used in WWTPs worldwide to improve MPs removal efficacy through the formation of enlarged contaminant particles that are more easily separated than their discrete tiny particles (Lee et al., 2012). Of various coagulating agents used to coagulate and agglomerate the MP particles, iron-based and aluminium-based salts are used widely. A variety of flocculating agents (e.g. $Fe_2(SO_4)_3$) have been found to be capable of aggregating suspended particulate matters and forming flocs that can impact positively on MPs removal in WWTPs during secondary treatments (Murphy et al., 2016). Ariza-Tarazona et al., (2019) investigated the role of Fe and Al-based salts as coagulating agents in removal of polyethene-based MPs, and found that Al^{3+} ions showed better performances in comparison with Fe^{3+} ions. Ma et al., (2019) also noticed a comparatively better performance of Al-based salts than the Fe-based coagulants, with 36.89±1.06% PE MPs being removed effectively by using $AlCl_3{\cdot}6H_2O$ salts as coagulating agents. A higher dosage of Al^{3+} was implemented (15 mmol/L at pH 7.0) to attain such higher removal efficacy than 8.0% removal with normal doses. They also tested another flocculating agent, polyacrylamide (PAM) for the same polyethylene MPs removal, and interestingly, the removal efficacy reached 85-90% and 50-60%, respectively when PAM (3-15 mg/L dosage) was combined with Fe-based (2 mmol/L dosage) and Al-based coagulants (5 mmol/L dosage). Ariza-Tarazona et al., (2019) also noticed the effect of PAM in MPs treatment, obtaining enhanced MPs removal efficacy (MPs diameter < 0.5 mm) from 25.83% to 61.19% by using 15 mg/L PAM.

The size of the MPs is an important factor in the formation and growth of flocs. PAM showed better performance in the removal of small MPs in comparison with that of the larger particles. Ariza-Tarazona et al., (2019) reported the removal of MPs was raised as the concentration of the coagulant ($FeCl_3{\cdot}6H_2O$) was increased, especially for the smaller MPs (diameter < 0.5 mm). Ma et al., (2019) also determined the effect of coagulant concentration on the removal of MPs, and found that 13.27±2.19% PE MPs was removed using 2 mmol/L $FeCl_3 \cdot H_2O$ coagulant which was decreased to 6.71±1.26% with 0.02 mmol/L of the same coagulant. Therefore, selecting appropriate coagulants and dosage is vital to facilitate the MPs treatment more effectively.

Another new sustainable approach has been proposed recently for the agglomeration-based removal of MPs (Herbort et al., 2018). This process initiates with the synthesis of active molecular precursors at an inert atmosphere which are further applied to form 'bio-inspired' alkoxy-silyl bonds through sol-gel reactions. MPs can become adhered to one another and form comparatively large-sized 3D agglomerates (666% larger particles) through the sol-gel process, which are quickly removed via cost-efficient filtration methods (Herbort et al., 2018) (Figure A10).

Bio-inspired materials are deliberately prepared having a good extraction ability for a diverse range of macro- and micro-sized substances. Their extraction capability may vary according to their molecular structure, the morphology of the precursors and the corresponding hybrid silica-gels. Herbort et al. (2018) used four different types of hybrid silica-gel from four separately synthesized precursors to remove a commercially available polyethylene in laboratory experiments, and obtained > 70% removal efficacy. Among them, N^1,N^4-bis((3-(trimethoxysilyl) propyl carbamoyl) terephthalamide was found as the most efficient precursor (> 95% extraction capacity) to bind the MPs.

Overall, coagulation or agglomeration of MPs can be an effective step in MPs treatment process to enhance the efficacy of WWTPs. Moreover, it can play a significant role to overcome the fouling problems of the membrane-based treatments. The efficacy of MPs removal is accelerated at high concentration of the coagulant, high pH, and the small size of MPs. Anionic PAM exhibited better performance compared to that of the cationic PAM in some studies due to the denser and facile formation of metal-based MPs trapping flocs (*e.g.* Fe-based flocs). Al-based salts showed better performance in comparison with the Fe-based coagulants Efficacy of MPs removal varies with pH, chemical compositions and concentration of the media, and types of the coagulation agents. In addition, alkoxy-silyl induced agglomeration method can be potentially effective for MPs treatment.

6.3.4. Electrocoagulation (EC)

EC, electroflotation, and electro-decantation are cost-effective methods compared to the microbial related methods to remove contaminants from wastewater (Garcia-Segura et al., 2017).

EC is the advanced technology of chemical coagulation process, which is comparatively cost-effective, energy-efficient, and amenable to automation, with the help of electrodes. EC process initiates with the liberation of metal ions from the sacrificial electrodes via the electrolysis process. Primary electrodes are connected to the external electric source and are monopolar (anode and cathode). For convenience, the sacrificial electrodes are made bipolar which break down to form metal ions during the electrolysis. Often aluminium and iron is used as sacrificial electrodes due to the capability of Al^{3+} and Fe^{3+} ions to act as a coagulating agent effectively. The liberated metals ions in the water stream react with hydroxyl ions of the media to form metal hydroxide coagulants. These coagulants break up the emulsion or colloids and make changes in stabilization of the surface charges of suspended MPs which permits them to become close enough to each other and thus attached via Van der Waals forces (Akbal and Camcı, 2011). The coagulation agents trap the suspended solid particles (e.g. MPs, particulates) by forming a sludge blanket. The liberated hydrogen gas (produced from the electrolysis process) helps to elevate the resultant sludge flocs above the water surface, which is referred to as stable flocs (Moussa et al., 2017) (**Figure 4a**). The following anodic and cathodic reactions co-occur throughout the process:

$$M_{(Solid)} \rightarrow M^{n+}_{(aqueous)} + ne^- \qquad \text{(i)}$$

$$2\,H_2O_{(l)} \rightarrow 4\,H^+_{(aqueous)} + O_{2\,(gas)} + 4\,e^- \qquad \text{(ii)}$$

$$2\,H_2O_{(l)} + 2\,e^- \rightarrow H_{2\,(gas)}\uparrow + 2\,OH^-_{(aqueous)} \qquad \text{(iii)}$$

$$M^{n+}_{(aqueous)} + n\,OH^-_{(aqueous)} \rightarrow M(OH)_{n\,(solid)} \qquad \text{(iv)}$$

EC has been reported to be effective over pH 3-10 (Padervand et al., 2020), which makes it more attractive to be an effective pathway for MPs removal from many types of wastewater and their effluents without adding other chemicals. Also, alteration in current density rarely affects the efficacy of MPs removal, which is above 90% (Perren et al., 2018). In the presence of 0.2 g/L of NaCl and pH 7.5, 99.24% microbeads were removed using this method (Padervand et al., 2020). Recently, Akarsu and Deniz (2020) obtained up to 98% MPs removal from laundry wastewater using Fe-Al electrode in this process within only 60 minutes. Despite having such cost-effective and efficient performance in MPs treatments, EC process is constrained with some operational drawbacks such as requirements for continuous replacement of sacrificial anodes, cathodic passivation and high cost of power supply. The development of more viable anodes and future research on operational modifications to avoid the cathodic passivation are required to overcome these limitations.

[Figure 4]

6.3.5. Comparison of chemical treatment technologies

Overall, chemical treatment methods are applied to enhance the MPs removal efficacy of WWTPs significantly. An overview of each of the methods, their advantages, obtained efficiencies and drawbacks is represented in **Table 4**. MPs were better removed via EC and ozonation process. Efficacy of sol-gel agglomeration also reached similar performance, but which varied with the implemented bio-inspired materials. Comparatively very limited research has been conducted on electro-Fenton and photo-Fenton processes for MPs removal. Photocatalytic degradation is a potential strategy but very few WWTPs have implemented this method so far due to their miscellaneous drawbacks **(Table 4)**. Acid-alkali pre-treatment method is still required to be developed to be more cost-efficient. The average efficiencies of MPs removal obtained with chemical methods followed the order: photo-Fenton process > electro-coagulation > ozonation > electro-Fenton process > sol-gel agglomeration > coagulation > modified Fenton process.

To date, the highest removal efficiencies obtained from different chemical methods are presented in **Figure 2c** for a comparative overview (**Table A16**). Unfortunately, none of these treatment strategies can remove MPs from contaminated sludge and wastewater when implemented alone without any other physical or biological treatment strategies. Moreover, by-products, as well as some secondary sludge produced in some methods such as coagulation, EC, sol-gel agglomeration require further treatment.

[Table 4]

6.4. Pyrolysis and co-pyrolysis of MPs

Pyrolysis and gasification processes are designated as thermo-chemical treatment technologies which allow the thermal decomposition of plastic wastes and other biomass particles as well as fuel production from them. In this process, the solid carbonaceous materials (e.g. plastic trash, solid organic contaminants, biomass) are converted into different gaseous products (having different heating values), which are also referred to as syngas. Composition of the produced gases may vary due to the applied temperature range and characteristics of original materials. Some common fractions of the end gaseous products are H_2, CO_2, CO, CH_4, and some hydrocarbons (Burra and Gupta, 2018a). Plastic wastes with long polymeric chains can be degraded into oligomers during various types of pyrolysis techniques such as catalytic pyrolysis, thermal pyrolysis, and microwave aided pyrolysis (Sun et al., 2019). A requirement of high temperature to break down the polymeric chains was one of the most significant challenges for this process. This limitation of higher temperature requirements can be solved through co-pyrolysis of plastic wastes along with other biomass or biogenic materials. Also, it may provide a feasible pathway to treat the MPs of sewage-sludge where they are mixed up with other contaminants (Burra and Gupta, 2018a; Jin et al., 2019). Different mass fractions of different type plastic wastes (e.g. polyethene terephthalate, polypropylene, polycarbonate) were treated in co-pyrolysis process with pinewood and enhanced production of syngas and hydrogen fuel were observed in several studies (Ahmed et al., 2011; Burra and Gupta, 2018b; Pinto et al., 2002). Furthermore, co-pyrolysis of polypropylene with cellulose exhibited a reduction of the pyrolysis activation energy (Burra and Gupta, 2018a).

Although some studies have been conducted on thermal treatment of plastic wastes, there has been a negligible amount of work on MPs. One of the most probable reasons is the requirements of isolation of MPs from the environment first with excess pre-treatment steps before the main thermal treatments. As there is evidence that co-pyrolysis of plastic wastes with biogenic materials requires even comparatively lower heat extent, therefore, co-pyrolysis may become a cost-effective process for MPs removal in future along with another beneficial side of fuel production at the same time without requirements for excessive accessories and costs. More investigations on thermal treatment are still required to establish this process entirely for efficient MPs treatments.

6.5. Efficacy of hybrid treatment technologies of MPs

MPs-targeted wastewater treatment technology is not fully developed, and no specific treatment process aimed at MPs removal has been applied in full-scale WWTP yet. In wastewater treatment, significant improvements have been achieved during the last few years in the application of a variety of hybrid treatment technologies, which consist of the combination of different treatment technologies to obtain the maximum MPs removal efficacies (Ahmed et al., 2017). Recently, hybrid systems have been widely used for the removal of MPs from water and wastewater. Talvitie et al., (2017a) observed the removal efficiency of various types of MPs from WWTP effluents by advanced treatment technologies including DF, RSF, DAF and MBR. They concluded that MBR-UF hybrid process treating primary effluent could remove 99.9% of MPs of almost all sized (> 20 μm) and all shapes, and showed that DF-coagulation and flocculation-DAF hybrid processes removed 98.5% and 95% MPs respectively.

Porous membranes and biological process combination could enhance the MPs removal efficiency up to 99.9% (Talvitie et al., 2017a). Lares et al., (2018) observed the performance of a municipal WWTP operating based on a combination of primary treatment and pilot MBR technology for the removal of MPs. This hybrid system was found to achieve high retention capacity of MPs over 98.3%. Similarly, the combination of MBR-RO is an effective advanced technology for the wastewater treatment to produce high-quality water (Dolar et al., 2012; Qin et al., 2006). RO influences the performances of MBR in wastewater treatment, and RO performance is commonly impacted by membrane fouling from inorganic, organic and biological fouling (Farias et al., 2014;). MBR-based anaerobic/anoxic/aerobic (A/A/O-MBR) systems effectively removed MPs from influent by trapped in sludge and block into permeate (effluent) through micro-membrane (pore size < 0.1 μm) filtration (Lv et al., 2019). This structure could eliminate necessarily all MPs from wastewater. A combination of sorption and filtration methodologies with biological and sedimentation processed showed an excellent efficiency for the treatment of MPs containing wastewater. MBR with other advanced physical and chemical treatment showed higher MPs removal efficiency in the WWTP. While MBR coupled with sorption and filtration process exhibited high removal percentage of MPs from the influent of water treatment plant (Padervand et al., 2020). MBR based hybrid systems are more effective for the removal of high MPs concentrate influent.

EC and agglomeration, coupled with additional filtration stage, showed effective separation of MPs from water. Ma et al., (2019) demonstrated that the coagulation process could remove more than 36.89% of MPs from water. Coagulation with sedimentation could enhance the removal efficiency up to 81.6% MPs of the secondary sediment effluent (Hidayaturrahman and Lee, 2019). Chemical coagulation treatment is the most widely used process to combine with the physical process including UF, DF, RSF, GAC and RO process to reduce the fouling problem or enhance the removal performance. Coagulation is often coupled with rapid sand filtration, membrane filtration and ozone oxidation in tertiary treatment of wastewater treatment plant (Hidayaturrahman and Lee, 2019). This hybrid treatment, including coagulation-RSF, coagulation-ozonation and coagulation-DF achieved the removal efficiency of MPs by 84.8%, 95.2% and 96.2%, respectively. Compared with coagulation (47.1-81.6% removal) and rapid sand filtration

(73% MPs removal), membrane filtration (79.4% MPs removal) and ozone oxidation (89.9% MPs removal) showed better performance to remove MPs. Small MPs are trapped with flocs which formed in coagulation and stopped during filtration. Primary and secondary treated three different WWTP in Daegu, South Korea was finally treated through different tertiary treatment process combined with coagulation (Hidayaturrahman and Lee, 2019). They showed that MPs' overall removal rate in different WWTPs is 99.2%, 99.1% and 98.9%, when using the ozonation, membrane disc filter, and RSF in the tertiary stage. UF process coupled with coagulation as a pre-treatment which is one of the main water treatment technology to remove organic contaminants from wastewater and produce high-quality effluent in current water treatment plant (Park et al., 2017;). UF-Coagulation hybrid system is not perfectly designed for MPs removal from wastewater (Mason et al., 2016). Recently Ma et al., (2019) observed the performance of UF-coagulation to remove PE MPs for potential application in drinking water treatment. After coagulation slight membrane fouling was induced due to the formation of loose cake layer by flocs, although PE particles were completely eliminated during drinking water treatment. However, after coagulation with PE particles (especially small size), membrane fouling was gently eased to increase the action of UF membrane (Ma et al., 2019).

Coagulation/flocculation combined with sedimentation (CFS) and granular filtration is applied to MPs (180 nm – 125 μm) removal in drinking water treatment (Zhang et al., 2020c), but with unsatisfactory removal. MBRs combined the biological activated sludge process with membrane separation provided MPs free effluent. Many studies have shown MBR hybrid systems to be more effective in the removal MPs from water up to 99.9% (Lares et al., 2018; Talvitie et al., 2017a). GAC filtration could effectively remove contaminants such as MPs through a synergistic combination of physical adsorption and biodegradation from the effluent of ozonation (Wang et al., 2020b). It is commonly coupled with ozonation process to remove some emerging contaminants (Fu et al., 2019; Li et al., 2018a) and larger molecular weight matter is converted into a small fraction to enhancing the biodegradability of the influent of GAC filter during drinking water treatment (Ross et al., 2019). When ozonation combined with GAC filtration, it increases the removal efficiency of MPs by approximately 17.2-22.2% (Wang et al., 2020b). So ozonation-GAC filtration couple process reduced 74-83.1% MPs from the final effluent of drinking water treatment plant. GAC filtration also coupled with sand filtration and sedimentation/flotation process for treated drinking water treatment plant (Pivokonsky et al., 2018).

Coagulation/flocculation, sedimentation, sand and GAC filtration hybrid process removed 81% MPs and coagulation/flocculation, flotation, sand filtration and GAC filtration hybrid treatment reduce the MPs 83% in drinking water treatment.

In an advanced drinking water treatment plant (coagulation+sedimentation+sand filtration+ozonation+GAC), the overall MPs removal efficiency was 82.1-88.6%, of which 82.9-87.5% 73.1–88.9% fibers fragments and 89.1–92.7% spheres were removed (Wang et al., 2020b). With GAC filtration combined with coagulation and sedimentation only, the MPs removal was reduced to 56.8-60.9% where 1-5 μm MPs removal was 73.7-98.5%. On the other hand, the combination of coagulation and sedimentation removed only fiber types MPs at 40.5-54.5%. In the coagulation/sedimentation process, it was found that the larger size MPs had a higher removal efficiency. MPs > 10 μm were almost completely removed, followed by the removal efficiency of 44.9–75.0% for 5–10 μm in this process. Despite the high removal efficiency of MPs about 99%, the conventional WWTP with primary and secondary treatment is not specially designed to improve the quality of final effluent. Different technologies could be combined before application to remove MPs in the WWTPs. Among them, a few physical and chemical treatment showed better performance when they use as combined with another process. The effective removal rates of MPs in tertiary stage of different WWTPs can be followed as hybrid MBR with RO or UF > coagulation-membrane disc-filter > coagulation- ozonation > Flocculation-DAF > constructed wetland > coagulation-RSF > ozonation-GAC filtration > coagulation-sedimentation. In summary, the combination of MBR with physical treatment such as RO/UF/NF has been found highly efficient in the removal of a wide range of MPs. CWs based hybrid treatment was found highly efficient, environmentally friendly and cost-effective. Moreover, coagulation with ozonation/GAC/DAF/RSF/ filtration processes are also more cost-effective than MBR based hybrid treatment.

7. Recommendations for future work

Although significant progress has been made in MPs research in terms of their analysis, interactions with other contaminants, toxicological effects, and removal by different treatment technologies, there are still many gaps. Future research directions on MPs are suggested as follows:

- ❖ More reliable methods for the detection and quantitative analysis of MPs are urgently needed to cover a wide range of MPs in the aquatic environment.

- ❖ More studies are needed to determine the toxicological effects of MPs.
- ❖ In membrane-based treatment, more research is needed to minimize the membrane abrasion and fouling to increase the performance in MPs removal.
- ❖ Application of dynamic membrane technology in MBR treatment should be promoted to improve MPs removal performance.
- ❖ The degradation and/or transformation of MPs in MBR should be investigated.
- ❖ More research is needed to isolate and amplify the number of MPs-degrading microbes for their targeted applications.
- ❖ Research on MPs removal from the sludge phase produced from biological treatment methods is urgently needed.
- ❖ CWs should be further developed for application in MPs removal.
- ❖ New materials and cathodic materials should be synthesized for efficient removal of MPs in Fenton and electro-Fenton processes.
- ❖ For commercial-scale photocatalysis treatment plants, the utilization of solar energy should be actively explored instead of UV irradiations.
- ❖ More effective and efficient photo-catalysts are to be synthesized for the photocatalytic degradation for MPs removal.
- ❖ More bio-inspired materials and their cost-efficient synthesis routes for sol-gel agglomeration method should be sought.
- ❖ Development of more viable anodes for EC method is needed.
- ❖ Hybrid treatments are needed to be specially designed to remove MPs.

8. Conclusions

A detailed review of MPs abundance and sources in the aquatic environment, their effective identification and analytical methods, interactions with major contaminants, potential toxicological effects on living organisms, and removal technologies from wastewater have been provided. MPs are widely detected in freshwater and marine environments such as water, sediments and organisms. A comparative evaluation of different MPs identification and analysis methods in environmental samples is presented by describing their applications, advantages, and drawbacks. Py-GC-MS, Py-MS methods, Raman spectroscopy, and FT-IR spectroscopy have been found as the most promising methods for MPs identification and quantification. WWTPs act as a significant source of MPs besides the domestic and industrial sources. MPs are found to act as a significant vector of different contaminants such as heavy metals, additive mixtures, surfactants, antibiotics, pesticides, and pharmaceuticals. Various adverse effects of MPs such as reduced fecundity, increased genotoxicity, growth inhibition, the elevation of reactive oxygen species, neurotoxic effects, reduction of photosynthesis of producers, and interferences with food intake, enzyme activities, plasma activities, lysosome activities and energy balance, are detected in different aquatic organisms. Different MPs treatment methods are discussed on their performance, together with their advantages and limitations. Filtration methods are reported as the most efficient physical treatment method although more developments are still required to implement them in large scale MPs treatment. CWs and MBR technologies are the most efficient among the biological treatment methods. In chemical treatment, EC, coagulation, sol-gel agglomeration, photo-Fenton and electro-Fenton processes show promising results in MPs removal. Hybrid treatment such as MBR-UF/RO system; coagulation followed by ozonation, GAC, DAF, RS, filtration; and CWs based hybrid technologies have shown highly promising results for effective MPs removal.

Conflict of interest

There are no conflicts to declare.

References

Abou-Zeid, D.M., Müller, R.J., Deckwer, W.D., 2001. Degradation of natural and synthetic polyesters under anaerobic conditions. J. Biotechnol. 86, 113-126.

Agasild, H., Nõges, T., 2005. Cladoceran and rotifer grazing on bacteria and phytoplankton in two shallow eutrophic lakes: in situ measurement with fluorescent microspheres. J. Plankton Res. 27, 1155-1174.

Ahmed, I., Nipattummakul, N., Gupta, A., 2011. Characteristics of syngas from co-gasification of polyethylene and woodchips. Appl. Energy. 88, 165-174.

Ahmed, M.B., Zhou, J.L., Ngo, H.H., Guo, W., 2015. Adsorptive removal of antibiotics from water and wastewater: progress and challenges. Sci. Total Environ. 532, 112-126.

Ahmed, M.B, Zhou J.L, Ngo, H.H., Guo, W., Chen M., 2016. Progress in the preparation and application of modified biochar for improved contaminant removal from water and wastewater. Bioresour. Technol. 214, 836-851.

Ahmed, M.B., Zhou J.L., Ngo, H.H., Guo, W., Thomaidis, N.S., Xu, J., 2017. Progress in the biological and chemical treatment technologies for emerging contaminant removal from wastewater: a critical review. J. Hazard. Mater. 323, 274-298.

Ahmed, T., Shahid, M., Azeem, F., Rasul, I., Shah, A.A., Noman, M., Hammed A., Manzoor N., Manzoor I., Muhammad S., 2018. Biodegradation of plastics: current scenario and future prospects for environmental safety. Environ. Sci. Pollut. Res. 25, 7287-7298.

Akarsu, C., Deniz, F., 2020. Electrocoagulation/Electroflotation Process for Removal of Organics and Microplastics in Laundry Wastewater. CLEAN–Soil, Air, Water, 2000146.

Akbal, F, Camcı, S., 2011. Copper, chromium and nickel removal from metal plating wastewater by electrocoagulation. Desalination. 269, 214-222.

Ali, S.S., Qazi, I.A., Arshad, M., Khan, Z., Voice, T.C., Mehmood, C.T., 2016. Photocatalytic degradation of low density polyethylene (LDPE) films using titania nanotubes. Environ. Nanotechnol. Monit. Manag. 5, 44-53.

Alimi, O.S., Fadare, O.O., Okoffo, E.D., 2021. Microplastics in African ecosystems: Current knowledge, abundance, associated contaminants, techniques, and research needs. Sci. Total Environ. 755, 142422.

Alimi, O.S., Farner Budarz, J., Hernandez, L.M., Tufenkji, N., 2018. Microplastics and nanoplastics in aquatic environments: aggregation, deposition, and enhanced contaminant transport. Environ. Sci. Technol. 52, 1704-1724.

Alomar, C., Estarellas, F., Deudero, S., 2016. Microplastics in the Mediterranean Sea: deposition in coastal shallow sediments, spatial variation and preferential grain size. Mar. Environ. Res. 115, 1-10.

Alvim, C. B., Mendoza-Roca, J. A., Bes-Piá, A., 2020. Wastewater treatment plant as microplastics release source–Quantification and identification techniques. J. Environ. Manage. 255, 109739.

Anderson, P.J., Warrack, S., Langen, V., Challis, J.K., Hanson, M.L., Rennie, MD., 2017. Microplastic contamination in lake Winnipeg, Canada. Environ. Pollut. 225, 223-231.

Andrady, A.L., 2011. Microplastics in the marine environment. Mar. Pollut. Bull. 62, 1596-1605.

Anjana, K., Hinduja, M., Sujitha, K., Dharani, G., 2020. Review on plastic wastes in marine environment–Biodegradation and biotechnological solutions. Mar. Pollut. Bull. 150, 110733.

Ariza-Tarazona, M.C., Villarreal-Chiu, J.F., Barbieri, V., Siligardi, C., Cedillo-González, E.I., 2019. New strategy for microplastic degradation: Green photocatalysis using a protein-based porous N-TiO_2 semiconductor. Ceram. Int. 45, 9618-9624.

Arossa, S., Martin, C., Rossbach, S., Duarte, C.M., 2019. Microplastic removal by red sea giant clam (*Tridacna maxima*). Environ. Pollut. 252, 1257-1266.

Ashton. K., Holmes, L., Turner, A., 2010. Association of metals with plastic production pellets in the marine environment. Mar. Pollut. Bullet. 60, 2050-2055.

Auta, H., Emenike, C., Fauziah, S., 2017. Distribution and importance of microplastics in the marine environment: a review of the sources, fate, effects, and potential solutions. Environ. Int. 102, 165-176.

Avérous, L., Pollet, E., 2012. Biodegradable polymers. Environmental silicate nano-biocomposites. Springer, pp 13-39.

Bakir, A., O'Connor, I.A., Rowland, S.J., Hendriks, A.J., Thompson, R.C., 2016. Relative importance of microplastics as a pathway for the transfer of hydrophobic organic chemicals to marine life. Environ. Pollut. 219, 56-65.

Bakir, A., Rowland, S.J., Thompson, R.C., 2014. Enhanced desorption of persistent organic contaminants from microplastics under simulated physiological conditions. Environ. Pollut. 185, 16-23.

Basri, H., Ismail, A.F., Aziz, M., 2011. Polyethersulfone (PES)–silver composite UF membrane: effect of silver loading and PVP molecular weight on membrane morphology and antibacterial activity. Desalination. 273, 72-80.

Batel, A., Linti, F., Scherer, M., Erdinger, L., Braunbeck, T., 2016. Transfer of benzo[*a*]pyrene from microplastics to Artemia nauplii and further to zebrafish via a trophic food web experiment: CYP1A induction and visual tracking of persistent organic contaminants. Environ. Toxicol. Chem. 35, 1656-1666.

Bayo, J., Olmos, S., López-Castellanos, J., 2020. Microplastics in an urban wastewater treatment plant: The influence of physicochemical parameters and environmental factors. Chemosphere. 238, 124593.

Benson, N. U., Fred-Ahmadu, O. H., 2020. Occurrence and distribution of microplastics-sorbed phthalic acid esters (PAEs) in coastal psammitic sediments of tropical Atlantic Ocean, Gulf of Guinea. Sci. Total Environ. 139013.

Besseling, E., Wang, B., Lürling, M., Koelmans, A.A., 2014. Nanoplastic affects growth of S. obliquus and reproduction of D. magna. Environ. Sci. Technol. 48, 12336-12343.

Besseling, E., Wegner. A., Foekema. E.M., Van Den Heuvel-Greve, M.J., Koelmans, A.A., 2013. Effects of microplastic on fitness and PCB bioaccumulation by the lugworm *Arenicola marina* (L.). Environ. Sci. Technol. 47, 593-600.

Blair, R.M., Waldron, S., Gauchotte-Lindsay, C., 2019. Average daily flow of microplastics through a tertiary wastewater treatment plant over a ten-month period. Water Res. 163, 114909.

Bläsing, M., Amelung, W., 2018. Plastics in soil: Anal. Methods and possible sources. Sci. Total Environ. 612, 422-435.

Brandon, J., Goldstein, M., Ohman, M.D., 2016. Long-term aging and degradation of microplastic particles: Comparing in situ oceanic and experimental weathering patterns. Mar. Pollut. Bull. 110, 299-308.

Braun, D., 2004. Poly (vinyl chloride) on the way from the 19th century to the 21st century. J. Polym. Sci. A Polym. Chem. 42, 578-586.

Browne, M.A., Niven, S.J., Galloway, T.S., Rowland, S.J., Thompson, R.C., 2013. Microplastic moves contaminants and additives to worms, reducing functions linked to health and biodiversity. Curr. Biol. 23, 2388-2392.

Burra, K., Gupta, A., 2018a. Kinetics of synergistic effects in co-pyrolysis of biomass with plastic wastes. Appl. Energy. 220, 408-418.

Burra, K., Gupta, A., 2018b. Synergistic effects in steam gasification of combined biomass and plastic waste mixtures. Appl. Energy. 211, 230-236.

Cai, L., Wang, J., Peng, J., Wu, Z., Tan X., 1018. Observation of the degradation of three types of plastic pellets exposed to UV irradiation in three different environments. Sci. Total Environ. 628, 740-747.

Carlin, J., Craig, C., Little, S., Donnelly, M., Fox, D., Zhai, L., Walters, L., 2020. Microplastic accumulation in the gastrointestinal tracts in birds of prey in central Florida, USA. Environ. Pollut. 114633.

Carr, S.A., Liu, J., Tesoro, A.G., 2016. Transport and fate of microplastic particles in wastewater treatment plants. Water Res. 91, 174-182.

Carson, H.S., Colbert, S.L., Kaylor, M.J., McDermid, K.J., 2011. Small plastic debris changes water movement and heat transfer through beach sediments. Mar. Pollut. Bull. 62, 1708-1713.

Castañeda, R.A., Avlijas, S., Simard, M.A., Ricciardi, A., 2014. Microplastic pollution in St. Lawrence river sediments. Can. J. Fish. Aquat. 71, 1767-1771.

Chamas, A., Moon, H., Zheng, J., Qiu, Y., Tabassum, T., Jang, M., J. H., Abu-Omar, Scott, S. L., Suh, S., 2020. Degradation Rates of Plastics in the Environment. ACS Sustain. Chem. Eng. 8, 3494-3511.

Chang, M., 2015. Reducing microplastics from facial exfoliating cleansers in wastewater through treatment versus consumer product decisions. Mar. Pollut. Bull. 101, 330-333.

Chen, R., Qi, M., Zhang, G., Yi, C., 2018. Comparative experiments on polymer degradation technique of produced water of polymer flooding oilfield. IOP Conference Series: Earth and Environmental Science. 113, pp. 012208.

Chen, G., Feng, Q., Wang, J., 2020. Mini-review of microplastics in the atmosphere and their risks to humans. Sci. Total Environ. 703, 135504.

Chua, E.M., Shimeta, J., Nugegoda. D., Morrison, P.D., Clarke, B.O., 2014. Assimilation of polybrominated diphenyl ethers from microplastics by the marine amphipod, Allorchestes compressa. Environ. Sci. Technol. 48, 8127-8134.

Claessens, M., Van Cauwenberghe, L., Vandegehuchte, M.B., Janssen, C.R., 2013. New techniques for the detection of microplastics in sediments and field collected organisms. Mar. Pollut. Bull. 70, 227-233.

Cole, M., Lindeque, P., Fileman, E., Halsband, C., Galloway, T.S., 2015. The impact of polystyrene microplastics on feeding, function and fecundity in the marine copepod Calanus helgolandicus. Environ. Sci. Technol. 49, 1130-1137.

Cole, M., Lindeque, P., Fileman, E., Halsband, C., Goodhead, R., Moger, J., Galloway T.S., 2013. Microplastic ingestion by zooplankton. Environ. Sci. Technol. 47, 6646-6655.

Cole, M., Lindeque, P., Halsband, C., Galloway, T.S., 2011. Microplastics as contaminants in the marine environment: a review. Mar. Pollut. Bull. 62, 2588-2597.

Cole, M., Lindeque, P.K., Fileman, E., Clark, J., Lewis, C., Halsband, C., Galloway T.S., 2016. Microplastics alter the properties and sinking rates of zooplankton faecal pellets. Environ. Sci. Technol. 50, 3239-3246.

Cooper, D.A., Corcoran, P.L., 2010. Effects of mechanical and chemical processes on the degradation of plastic beach debris on the island of Kauai, Hawaii. Mar. Pollut. Bull. 60, 650-654.

Corcoran, P.L., Biesinger, M.C., Grifi, M., 2009. Plastics and beaches: a degrading relationship. Mar. Pollut. Bull. 58, 80-84.

Courtene-Jones, W., Quinn, B., Murphy, F., Gary, S.F., Narayanaswamy, B.E., 2017. Optimisation of enzymatic digestion and validation of specimen preservation methods for the analysis of ingested microplastics. Anal. Methods. 9, 1437-1445.

Crawford, C., Quinn, B., 2017. Microplastic separation techniques. Microplastic Contaminants. Elsevier, pp. 203-218.

da Costa, J.P., Santos, P.S., Duarte, A.C., Rocha-Santos, T., 2016. (Nano) plastics in the environment–sources, fates and effects. Sci. Total Environ. 566, 15-26.

Dazzi, A., Prater, C.B., Hu, Q., Chase, D.B., Rabolt, J.F., Marcott, C., 2012. AFM–IR: combining atomic force microscopy and infrared spectroscopy for nanoscale chemical characterization. Appl. Spectrosc. 66, 1365-1384.

Dazzi, A., Saunier. J., Kjoller, K., Yagoubi, N., 2015. Resonance enhanced AFM-IR: A new powerful way to characterize blooming on polymers used in medical devices. Int. J. Pharm. 484, 109-114.

De la Cruz, N., Giménez, J., Esplugas, S., Grandjean, D., De Alencastro, L., Pulgarin, C., 2012. Degradation of 32 emergent contaminants by UV and neutral photo-fenton in domestic wastewater effluent previously treated by activated sludge. Water Res. 46, 1947-1957.

de Luna, M.D.G, Veciana, M.L., Su, C.C., Lu, M.C., 2012. Acetaminophen degradation by electro-Fenton and photoelectro-Fenton using a double cathode electrochemical cell. J. Hazard. Mater. 217, 200-207.

Dehaut, A., Cassone, A-L., Frère, L., Hermabessiere, L., Himber, C., Rinnert. E., Riviere G., Lambert C., Soudant P., Huvet A., Duflos G., 2016. Microplastics in seafood: Benchmark protocol for their extraction and characterization. Environ. Pollut. 215, 223-233.

Dekiff, J.H., Remy, D., Klasmeier, J., Fries, E., 2014. Occurrence and spatial distribution of microplastics in sediments from Norderney. Environ. Pollut. 186, 248-256.

Derraik, J.G., 2002. The pollution of the marine environment by plastic debris: a review. Mar. Pollut. Bull. 44, 842-852.

Di, M., Wang, J., 2018. Microplastics in surface waters and sediments of the Three Gorges Reservoir, China. Sci. Total Environ. 616, 1620-1627.

Dolar, D., Gros, M., Rodriguez-Mozaz, S., Moreno, J., Comas, J., Rodriguez-Roda, I., Barcelo D., 2012. Removal of emerging contaminants from municipal wastewater with an integrated membrane system, MBR–RO. J. Hazard. Mater. 239, 64-69.

Dong, H., Chen, Y., Wang, J., Zhang, Y., Zhang, P., Li, X., Zou, J., Zhou, A., 2020. Interactions of microplastics and antibiotic resistance genes and their effects on the aquaculture environments. J. Hazard. Mater. 123961.

Doyle, M.J., Watson, W., Bowlin, N.M., Sheavly, S.B., 2011. Plastic particles in coastal pelagic ecosystems of the Northeast Pacific ocean. Mar. Environ. Res. 71, 41-52.

Dřímal, P., Hoffmann, J., Družbík, M., 2007. Evaluating the aerobic biodegradability of plastics in soil environments through GC and IR analysis of gaseous phase. Polym. Test. 26, 729-741.

Dris, R., Gasperi, J., Saad, M., Mirande, C., Tassin, B., 2016. Synthetic fibers in atmospheric fallout: a source of microplastics in the environment? Mar. Pollut. Bull. 104, 290-293.

Duis, K., Coors, A., 2016. Microplastics in the aquatic and terrestrial environment: sources (with a specific focus on personal care products), fate and effects. Environ. Sci. Eur. 28, 2.

Dümichen, E., Barthel, A-K., Braun, U., Bannick, C.G., Brand, K., Jekel, M., Senz R., 2015. Analysis of polyethylene microplastics in environmental samples, using a thermal decomposition method. Water Res. 85, 451-457.

Dümichen, E., Eisentraut, P., Bannick, C.G., Barthel, A.K., Senz, R., Braun, U., 2017. Fast identification of microplastics in complex environmental samples by a thermal degradation method. Chemosphere. 174, 572-584.

Eerkes-Medrano, D., Thompson, R.C., Aldridge, D.C., 2015. Microplastics in freshwater systems: a review of the emerging threats, identification of knowledge gaps and prioritisation of research needs. Water Res. 75, 63-82.

Enfrin, M., Dumée, L.F., Lee, J., 2019. Nano/microplastics in water and wastewater treatment processes – Origin, impact and potential solutions. Water Res. 161, 621-638.

Ersahin, M.E., Ozgun, H., Dereli, R.K., Ozturk, I., Roest, K., van Lier, J.B., 2012. A review on dynamic membrane filtration: materials, applications and future perspectives. Bioresour. Technol. 122, 196-206.

Eskandarloo, H., Kierulf, A., Abbaspourrad, A., 2017. Light-harvesting synthetic nano-and micromotors: a review. Nanoscale 9, 12218-12230.

Evangeliou, N., Grythe, H., Klimont, Z., Heyes, C., Eckhardt, S., Lopez-Aparicio, S., Stohl, A., 2020. Atmospheric transport is a major pathway of microplastics to remote regions. Nat. Commun. 11, 1-11.

Farias, E.L., Howe, K.J., Thomson, B.M., 2014. Effect of membrane bioreactor solids retention time on reverse osmosis membrane fouling for wastewater reuse. Water Res. 49, 53-61.

Felsing, S., Kochleus, C., Buchinger, S., Brennholt, N., Stock, F., Reifferscheid, G., 2018. A new approach in separating microplastics from environmental samples based on their electrostatic behavior. Environ. Pollut. 234, 20-28.

Feng, H-M., Zheng, J-C., Lei, N-Y., Yu, L., Kong, K.H-K., Yu, H-Q., et al., 2011. Photoassisted Fenton degradation of polystyrene. Environ. Sci. Technol. 45, 744-750.

Free, C.M., Jensen, O.P., Mason, S.A., Eriksen, M., Williamson, N.J., Boldgiv, B., 2014. High-levels of microplastic pollution in a large, remote, mountain lake. Mar. Pollut. Bull. 85, 156-163.

Frias, J., Sobral, P., Ferreira, A., 2010. Organic contaminants in microplastics from two beaches of the Portuguese coast. Mar. Pollut. Bull. 60, 1988-1992.

Fries, E., Dekiff, J.H., Willmeyer, J., Nuelle, M.T., Ebert, M., Remy, D., 2013. Identification of polymer types and additives in marine microplastic particles using pyrolysis-GC/MS and scanning electron microscopy. Environ. Sci. Process Impacts. 15, 1949-1956.

Fu. J., Lee, W-N., Coleman, C., Nowack, K., Carter, J., Huang, C.H., 2019. Removal of pharmaceuticals and personal care products by two-stage biofiltration for drinking water treatment. Sci. Total Environ. 664, 240-248.

Fu, Z., Chen, G., Wang, W., Wang, J., 2020. Microplastic pollution research methodologies, abundance, characteristics and risk assessments for aquatic biota in China. Environ. Pollut. 115098.

Ganiyu, S.O., Zhou, M., Martínez-Huitle, C.A., 2018. Heterogeneous electro-Fenton and photoelectro-Fenton processes: a critical review of fundamental principles and application for water/wastewater treatment. Appl. Catal. B. 235, 103-129.

Ganzenko, O., Huguenot, D., Van Hullebusch, E.D., Esposito, G., Oturan, M.A., 2014. Electrochemical advanced oxidation and biological processes for wastewater treatment: a review of the combined approaches. Environ. Sci. Pollut. Res. 21, 8493-8524.

Garcia-Segura, S., Eiband, M.M.S, de Melo, J.V., Martínez-Huitle, C.A., 2017. Electrocoagulation and advanced electrocoagulation processes: A general review about the fundamentals, emerging applications and its association with other technologies. J. Electroanal. Chem. 801, 267-299.

Gasperi, J., Wright, S.L., Dris, R., Collard, F., Mandin, C., Guerrouache, M., Langlois V., Kellly F.J., Tassin B., 2018. Microplastics in air: are we breathing it in? Curr. Opin. Environ. Sci. Health. 1, 1-5.

Godoy, V., Blázquez, G., Calero, M., Quesada, L., Martín-Lara, M., 2019. The potential of microplastics as carriers of metals. Environ. Pollut. 255, 113363.

Gómez, E.F., Michel Jr., F.C., 2013. Biodegradability of conventional and bio-based plastics and natural fiber composites during composting, anaerobic digestion and long-term soil incubation. Polym. Degrad. Stab. 98, 2583-2591.

Gouin, T., Roche, N., Lohmann, R., Hodges, G., 2011. A thermodynamic approach for assessing the environmental exposure of chemicals absorbed to microplastic. Environ. Sci. Technol. 45, 1466-1472.

Grbic, J., Nguyen, B., Guo, E., You, J.B., Sinton, D., Rochman, C.M., 2019. Magnetic extraction of microplastics from environmental samples. Environ. Sci. Technol. Letters 6, 68-72.

Green, D.S., Boots, B., O'Connor, N.E., Thompson, R., 2017. Microplastics affect the ecological functioning of an important biogenic habitat. Environ. Sci. Technol. 51, 68-77.

Gu, J.G., Gu, J.D., 2005. Methods currently used in testing microbiological degradation and deterioration of a wide range of polymeric materials with various degree of degradability: a review. J. Polym. Environ. 13, 65-74.

Guo, X., Pang, J., Chen, S., Jia, H., 2018. Sorption properties of tylosin on four different microplastics. Chemosphere. 209, 240-245.

Guo, X, Wang, J., 2019. Sorption of antibiotics onto aged microplastics in freshwater and seawater. Mar. Pollut. Bull. 149, 110511.

Hale, R. C., Seeley, M. E., La Guardia, M. J., Mai, L., Zeng, E. Y., 2020. A global perspective on microplastics. J. Geophys. Res. Oceans 125, e2018JC014719.

Harrison, J.P, Ojeda, J.J., Romero-González, M.E., 2012. The applicability of reflectance micro-Fourier-transform infrared spectroscopy for the detection of synthetic microplastics in marine sediments. Sci. Total Environ. 416, 455-463.

Helcoski, R., Yonkos, L.T., Sanchez, A., Baldwin, A.H., 2020. Wetland soil microplastics are negatively related to vegetation cover and stem density. Environ. Pollut. 256, 113391.

Herbort, A.F., Schuhen K., 2017. A concept for the removal of microplastics from the marine environment with innovative host-guest relationships. Environ. Sci. Pollut. Res. 24, 11061-11065.

Herbort, A.F., Sturm, M.T., Fiedler, S., Abkai, G., Schuhen, K., 2018. Alkoxy-silyl induced agglomeration: a new approach for the sustainable removal of microplastic from aquatic systems. J. Polym. Environ. 26, 4258-4270.

Hernandez, L.M., Xu, E.G., Larsson, H.C., Tahara, R., Maisuria, V.B., Tufenkji, N., 2019. Plastic teabags release billions of microparticles and nanoparticles into tea. Environ. Sci. Technol. 53, 12300-12310.

Hernandez, L.M., Yousefi, N., Tufenkji, N., 2017. Are there nanoplastics in your personal care products? Environ. Sci. Technol. Letters 4, 280-285.

Hidalgo-Ruz, V., Gutow, L., Thompson, R.C., Thiel. M., 2012. Microplastics in the marine environment: a review of the methods used for identification and quantification. Environ. Sci. Technol. 46, 3060-3075.

Hidayaturrahman, H., Lee, T.G., 2019. A study on characteristics of microplastic in wastewater of South Korea: Identification, quantification, and fate of microplastics during treatment process. Mar. Pollut. Bull. 146, 696-702.

Hirai, H., Takada, H., Ogata, Y., Yamashita, R., Mizukawa, K., Saha, M., Kwan C, Moore C, Gray H, Laursen D, Zettler E.R., 2011. Organic microcontaminants in marine plastics debris from the open ocean and remote and urban beaches. Mar. Pollut. Bull. 62, 1683-1692.

Holmes. L.A., Turner, A., Thompson, R.C., 2012. Adsorption of trace metals to plastic resin pellets in the marine environment. Environ. Pollut. 160, 42-48.

Horton, A.A., Walton, A., Spurgeon, D.J., Lahive, E., Svendsen, C., 2017. Microplastics in freshwater and terrestrial environments: evaluating the current understanding to identify the knowledge gaps and future research priorities. Sci. Total Environ. 586, 127-141.

Hu, L., Fu, J., Wang, S., Xiang, Y., Pan, X., 2020. Microplastics generated under simulated fire scenarios: Characteristics, antimony leaching, and toxicity. Environ. Pollut. 269, 115905.

Hu, Y., Wang, X.C., Ngo, H.H., Sun, Q., Yang. Y., 2018. Anaerobic dynamic membrane bioreactor (AnDMBR) for wastewater treatment: A review. Bioresour. Technol. 247, 1107-1118.

Huang, B.C., Guan, Y.F., Chen, W., Yu, H.Q., 2017. Membrane fouling characteristics and mitigation in a coagulation-assisted microfiltration process for municipal wastewater pretreatment. Water Res. 123, 216-223.

Hüffer, T., Hofmann, T., 2016. Sorption of non-polar organic compounds by micro-sized plastic particles in aqueous solution. Environ. Pollut. 214, 194-201.

Huppertsberg, S., Knepper, T.P., 2018. Instrumental analysis of microplastics—benefits and challenges. Anal. Bioannal. Chem. 410, 6343-6352.

Ibrar, I., Altaee, A., Zhou, J.L., Naji, O., Khanafer, D., 2020. Challenges and potentials of forward osmosis process in the treatment of wastewater. Crit. Rev. Environ. Sci. Technol. 50, 1339-1383.

Imhof, H.K., Schmid, J., Niessner, R., Ivleva, N.P., Laforsch, C., 2012. A novel, highly efficient method for the separation and quantification of plastic particles in sediments of aquatic environments. Limnol. Oceanogr.: Methods. 10, 524-537.

Ivleva, N.P., Wiesheu, A.C., Niessner, R., 2017. Microplastic in aquatic ecosystems. Angew. Chem. Int. Ed. 56, 1720-1739.

Jang, M., Shim, W. J., Han, G. M., Cho, Y., Moon, Y., Hong, S. H., 2020. Relative importance of aqueous leachate versus particle ingestion as uptake routes for microplastic additives (hexabromocyclododecane) to mussels. Environ. Pollut. 116272.

Jiang, L., Hu, X., Yin, D., Zhang, H., Yu, Z., 2011. Occurrence, distribution and seasonal variation of antibiotics in the Huangpu River, Shanghai, China. Chemosphere. 82, 822-828.

Jin, Q., Wang, X., Li, S., Mikulčić, H., Bešenić, T., Deng, S., Vujanović M, Tan H, Kumfer B.M., 2019. Synergistic effects during co-pyrolysis of biomass and plastic: Gas, tar, soot, char products and thermogravimetric study. J. Energy Inst. 92, 108-117.

Joo, S., Cho, I.J., Seo, H., Son, H.F., Sagong, H.Y., Shin, T.J., Choi S.Y., Lee S.Y., Kim K.J., 2018. Structural insight into molecular mechanism of poly (ethylene terephthalate) degradation. Nat. Commun. 9, 1-12.

Kalčíková, G., Alič, B., Skalar, T., Bundschuh, M., Gotvajn, A.Ž., 2017. Wastewater treatment plant effluents as source of cosmetic polyethylene microbeads to freshwater. Chemosphere. 188, 25-31.

Kalcíková, G., Skalar, T., Marolt, G., Kokalj, A. J., 2020. An environmental concentration of aged microplastics with adsorbed silver significantly affects aquatic organisms. Water Res. 115644.

Kale, G., Kijchavengkul, T., Auras, R., Rubino, M., Selke, S.E., Singh, S.P., 2007. Compostability of bioplastic packaging materials: an overview. Macromol. Biosci. 7, 255-277.

Kang, J.H., Kwon, O.Y., Shim, W.J., 2015. Potential threat of microplastics to zooplanktivores in the surface waters of the Southern Sea of Korea. Arch. Environ. Contam. Toxicol. 69, 340-351.

Kaposi, K.L., Mos, B., Kelaher, B.P., Dworjanyn, S.A., 2014. Ingestion of microplastic has limited impact on a marine larva. Environ. Sci. Technol. 48, 1638-1645.

Käppler, A., Fischer, D., Oberbeckmann, S., Schernewski, G., Labrenz, M., Eichhorn, K.J., Voit B., 2016. Analysis of environmental microplastics by vibrational microspectroscopy: FTIR, Raman or both? Anal. Bioannal. Chem. 408, 8377-8391.

Karami, A., Golieskardi, A., Choo, C.K., Romano, N., Ho, Y.B., Salamatinia, B., 2017. A high-performance protocol for extraction of microplastics in fish. Sci. Total Environ. 578, 485-494.

Karlsson, T.M., Hassellöv, M., Jakubowicz, I., 2018. Influence of thermooxidative degradation on the in situ fate of polyethylene in temperate coastal waters. Mar. Pollut. Bull. 135, 187-194.

Karlsson, T.M., Vethaak, A.D., Almroth, B.C., Ariese, F., van Velzen, M., Hassellöv, M., Leslie H.A., 2017. Screening for microplastics in sediment, water, marine invertebrates and fish: method development and microplastic accumulation. Mar. Pollut. Bull. 122, 403-408.

Kijchavengkul, T., Auras, R., Rubino, M., Selke, S., Ngouajio, M., Fernandez, R.T., 2010. Biodegradation and hydrolysis rate of aliphatic aromatic polyester. Polym. Degrad. Stab. 95, 2641-2647.

Klavarioti, M., Mantzavinos, D., Kassinos, D., 2009. Removal of residual pharmaceuticals from aqueous systems by advanced oxidation processes. Environ. Int. 35, 402-417.

Klein, S., Dimzon, I.K., Eubeler, J., Knepper, T.P., 2018. Analysis, occurrence, and degradation of microplastics in the aqueous environment. Freshwater Microplastics. Springer, Cham, pp. 51-67.

Klein, S., Worch, E., Knepper, T.P., 2015. Occurrence and spatial distribution of microplastics in river shore sediments of the Rhine-Main area in Germany. Environ. Sci. Technol. 49, 6070-6076.

Koelmans, A.A., Bakir, A., Burton, G.A., Janssen, C.R., 2016. Microplastic as a vector for chemicals in the aquatic environment: critical review and model-supported reinterpretation of empirical studies. Environ. Sci. Technol. 50, 3315-3326.

Koelmans, A.A., Besseling, E., Wegner, A., Foekema, E.M., 2013. Plastic as a carrier of POPs to aquatic organisms: a model analysis. Environ. Sci. Technol. 47, 7812-7820.

Koelmans, A.A., Nor, N.H.M., Hermsen, E., Kooi, M., Mintenig, S.M., De France, J., 2019. Microplastics in freshwaters and drinking water: critical review and assessment of data quality. Water Res. 155, 410-422.

Kolstad, J.J., Vink, E.T., De Wilde, B., Debeer, L., 2012. Assessment of anaerobic degradation of Ingeo™ polylactides under accelerated landfill conditions. Polym. Degrad. Stab. 97, 1131-1141.

Kwon, O. Y., Kang, J. H., Hong, S. H.,Shim, W. J., 2020. Spatial distribution of microplastic in the surface waters along the coast of Korea. Mar. Pollut. Bull. 110729.

Lambert, S., Scherer, C., Wagner, M., 2017. Ecotoxicity testing of microplastics: Considering the heterogeneity of physicochemical properties. Integr. Environ. Assess. Manag. 13, 470-475.

Lares, M., Ncibi, M.C., Sillanpää, M., Sillanpää, M., 2018. Occurrence, identification and removal of microplastic particles and fibers in conventional activated sludge process and advanced MBR technology. Water Res. 133, 236-246.

Lee, H., Shim, W.J., Kwon, J.H., 2014. Sorption capacity of plastic debris for hydrophobic organic chemicals. Sci. Total Environ. 470, 1545-1552.

Lee, K.E., Morad, N., Teng. T.T., Poh, B.T., 2012. Development, characterization and the application of hybrid materials in coagulation/flocculation of wastewater: A review. Chem. Eng. J. 203, 370-386.

Lee, Y. K., Murphy, K. R., Hur, J., 2020. Fluorescence signatures of dissolved organic matter leached from microplastics: Polymers and additives. Environ. Sci. Technol. 54, 11905-11914.

Leslie, H., Brandsma, S., Van Velzen, M., Vethaak, A., 2017. Microplastics en route: Field measurements in the Dutch river delta and Amsterdam canals, wastewater treatment plants, North Sea sediments and biota. Environ. Int. 101, 133-142.

Levin, I.W., Bhargava, R., 2005. Fourier transform infrared vibrational spectroscopic imaging: integrating microscopy and molecular recognition. Annu. Rev. Phys. Chem. 56, 429-474.

Li, G., Ben, W., Ye, H., Zhang, D., Qiang, Z., 2018a. Performance of ozonation and biological activated carbon in eliminating sulfonamides and sulfonamide-resistant bacteria: A pilot-scale study. Chem. Eng. J. 341, 327-334.

Li. J., Liu, H., Chen, J.P., 2018b. Microplastics in freshwater systems: A review on occurrence, environmental effects, and methods for microplastics detection. Water Res. 137, 362-374.

Li, J., Zhang, K., Zhang, H., 2018c. Adsorption of antibiotics on microplastics. Environ. Pollut. 237, 460-467.

Li, L., Xu, G., Yu, H., 2018d. Dynamic membrane filtration: formation, filtration, cleaning, and applications. Chem. Eng. Technol. 41, 7-18.

Li, L., Xu, G., Yu, H., Xing, J., 2018e. Dynamic membrane for micro-particle removal in wastewater treatment: Performance and influencing factors. Sci. Total Environ. 627, 332-340.

Li, Q., Wu, J., Zhao, X., Gu, X., Ji, R., 2019a. Separation and identification of microplastics from soil and sewage sludge. Environ. Pollut. 254, 113076.

Li, Q., Xu, Z., Pinnau, I., 2007. Fouling of reverse osmosis membranes by biopolymers in wastewater secondary effluent: Role of membrane surface properties and initial permeate flux. J. Membr. Sci. 290, 173-181.

Li, W., Shi, Y., Gao, L., Liu, J., Cai, Y., 2012. Occurrence of antibiotics in water, sediments, aquatic plants, and animals from Baiyangdian Lake in North China. Chemosphere. 89, 1307-1315.

Li, X., Mei, Q., Chen, L., Zhang, H., Dong, B., Dai, X., et al., 2019b. Enhancement in adsorption potential of microplastics in sewage sludge for metal contaminants after the wastewater treatment process. Water Res. 157, 228-237.

Liang, W., Luo, Y., Song, S., Dong, X., Yu, X., 2013. High photocatalytic degradation activity of polyethylene containing polyacrylamide grafted TiO_2. Polym. Degrad. Stab. 98, 1754-1761.

Lin, H., Zhang, M., Wang, F., Meng, F., Liao, B.Q., Hong, H., Chen J., Gao W., 2014. A critical review of extracellular polymeric substances (EPSs) in membrane bioreactors: characteristics, roles in membrane fouling and control strategies. J. Membr. Sci. 460, 110-125.

Lithner, D., Damberg, J., Dave, G., Larsson, Å., 2009. Leachates from plastic consumer products–screening for toxicity with Daphnia magna. Chemosphere. 74, 1195-1200.

Lithner, D., Larsson, Å., Dave, G., 2011. Environmental and health hazard ranking and assessment of plastic polymers based on chemical composition. Sci. Total Environ. 409, 3309-3324.

Liu, F.F., Liu, GZ., Zhu, Z.L., Wang, S.C., Zhao, F.F., 2019a. Interactions between microplastics and phthalate esters as affected by microplastics characteristics and solution chemistry. Chemosphere. 214, 688-694.

Liu, F., Vianello, A., Vollertsen, J., 2019b. Retention of microplastics in sediments of urban and highway stormwater retention ponds. Environ. Pollut. 255, 113335.

Liu, X., Yuan, W., Di, M., Li, Z., Wang, J., 2019c. Transfer and fate of microplastics during the conventional activated sludge process in one wastewater treatment plant of China. Chem. Eng. J. 362, 176-182.

Löder. M.G., Gerdts, G., 2015. Methodology used for the detection and identification of microplastics—A critical appraisal. Marine Anthropogenic Litter. Springer, pp. 201-227.

Löder, M.G.J., Kuczera, M., Mintenig, S., Lorenz, C., Gerdts, G., 2015. Focal plane array detector-based micro-Fourier-transform infrared imaging for the analysis of microplastics in environmental samples. Environ. Chem. 12, 563-581.

Lönnstedt, O.M., Eklöv, P., 2016. Environmentally relevant concentrations of microplastic particles influence larval fish ecology. Science. 352, 1213-1216.

Lucas, N., Bienaime, C., Belloy, C., Queneudec, M., Silvestre, F., Nava-Saucedo, J-E., 2008. Polymer biodegradation: Mechanisms and estimation techniques–A review. Chemosphere. 73, 429-442.

Luís, L.G., Ferreira, P., Fonte, E., Oliveira, M., Guilhermino, L., 2015. Does the presence of microplastics influence the acute toxicity of chromium (VI) to early juveniles of the common goby (*Pomatoschistus microps*)? A study with juveniles from two wild estuarine populations. Aquat. Toxicol. 164, 163-174.

Luo, H., Zhao, Y., Li, Y., Xiang, Y., He, D., Pan, X., 2020. Aging of microplastics affects their surface properties, thermal decomposition, additives leaching and interactions in simulated fluids. Sci. Total Environ. 714, 136862.

Luo, J., Chen, Y., Feng, L., 2016. Polycyclic aromatic hydrocarbon affects acetic acid production during anaerobic fermentation of waste activated sludge by altering activity and viability of acetogen. Environ. Sci. Technol. 50, 6921-6929.

Lv, X., Dong, Q., Zuo, Z., Liu, Y., Huang, X., Wu, W-M., 2019. Microplastics in a municipal wastewater treatment plant: Fate, dynamic distribution, removal efficiencies, and control strategies. J. Clean. Prod. 225, 579-586.

Ma, B., Xue, W., Ding, Y., Hu, C., Liu, H., Qu, J., 2019. Removal characteristics of microplastics by Fe-based coagulants during drinking water treatment. J. Environ. Sci. 78, 267-275.

Macwan, D., Dave, P.N., Chaturvedi, S., 2011. A review on nano-TiO_2 sol–gel type syntheses and its applications. J. Mater. Sci. 46, 3669-3686.

Maes, T., Jessop, R., Wellner, N., Haupt, K., Mayes, A.G., 2017a. A rapid-screening approach to detect and quantify microplastics based on fluorescent tagging with Nile Red. Sci. Rep. 7, 1-10.

Mahon, A.M., O'Connell, B., Healy, M.G., O'Connor, I., Officer, R., Nash, R., Morrison L., 2017. Microplastics in sewage sludge: effects of treatment. Environ. Sci. Technol. 51, 810-818.

Majewsky, M., Bitter, H., Eiche, E., Horn, H., 2016. Determination of microplastic polyethylene (PE) and polypropylene (PP) in environmental samples using thermal analysis (TGA-DSC). Sci. Total Environ. 568, 507-511.

Malankowska, M., Echaide-Gorriz, C., Coronas, J., 2021. Microplastics in marine environment: a review on sources, classification, and potential remediation by membrane technology. Environ. Sci.: Water Res. Technol.

Mason, S.A., Garneau, D., Sutton, R., Chu, Y., Ehmann, K., Barnes, J., Fink P., Papazissimos D., Rogers D.L., 2016. Microplastic pollution is widely detected in US municipal wastewater treatment plant effluent. Environ. Pollut. 218, 1045-1054.

Massardier-Nageotte, V., Pestre, C., Cruard-Pradet, T., Bayard, R., 2006. Aerobic and anaerobic biodegradability of polymer films and physico-chemical characterization. Polym. Degrad. Stab. 91, 620-627.

Mathalon, A., Hill, P., 2014. Microplastic fibers in the intertidal ecosystem surrounding Halifax Harbor, Nova Scotia. Mar. Pollut. Bull. 81, 69-79.

McCormick, A., Hoellein, T.J., Mason, S.A., Schluep, J., Kelly, J.J., 2014. Microplastic is an abundant and distinct microbial habitat in an urban river. Environ. Sci. Technol. 48, 11863-11871.

Mendoza, L.M.R., Jones P.R., 2015. Characterisation of microplastics and toxic chemicals extracted from microplastic samples from the North Pacific Gyre. Environ. Chem. 12, 611-617.

Mendoza, L.M.R., Karapanagioti, H., Álvarez, N.R., 2018. Micro (nanoplastics) in the marine environment: current knowledge and gaps. Curr. Opin. Environ. Sci. Health. 1, 47-51.

Miao, F., Liu, Y., Gao, M., Yu, X., Xiao, P., Wang, M., et al., 2020. Degradation of polyvinyl chloride microplastics via an electro-Fenton-like system with a TiO2/graphite cathode. J. Hazard. Mater. 123023.

Michielssen, M.R., Michielssen, E.R., Ni, J., Duhaime, M.B., 2016. Fate of microplastics and other small anthropogenic litter (SAL) in wastewater treatment plants depends on unit processes employed. Environ. Sci. Water Res. Technol. 2, 1064-1073.

Min, K., Cuiffi, J. D., Mathers, R. T., 2020. Ranking environmental degradation trends of plastic marine debris based on physical properties and molecular structure. Nat. Commun. 11, 1-11.

Mintenig, S., Int-Veen, I., Löder, M.G., Primpke, S., Gerdts, G., 2017. Identification of microplastic in effluents of waste water treatment plants using focal plane array-based micro-Fourier-transform infrared imaging. Water Res. 108, 365-372.

Mohan, D., Sarswat, A., Ok, Y.S., Pittman Jr., C.U., 2014. Organic and inorganic contaminants removal from water with biochar, a renewable, low cost and sustainable adsorbent–a critical review. Bioresour. Technol. 160, 191-202.

Mohanty, S.K., Valenca, R., Berger, A.W., Iris, K., Xiong, X., Saunders, T.M., Tsang D.C., 2018. Plenty of room for carbon on the ground: potential applications of biochar for stormwater treatment. Sci. Total Environ. 625, 1644-1658.

Mohee, R., Unmar, G., Mudhoo, A., Khadoo, P., 2008. Biodegradability of biodegradable/degradable plastic materials under aerobic and anaerobic conditions. Waste Manage. 28, 1624-1629.

Moore, C.J., 2008. Synthetic polymers in the marine environment: a rapidly increasing, long-term threat. Environ. Res. 108, 131-139.

Moore, C.J,, Moore, S.L., Leecaster, M.K., Weisberg, S.B., 2001. A comparison of plastic and plankton in the North Pacific central gyre. Mar. Pollut. Bull. 42, 1297-1300.

Moussa, D.T., El-Naas, M.H., Nasser, M., Al-Marri, M.J., 2017. A comprehensive review of electrocoagulation for water treatment: Potentials and challenges. J. Environ. Manage. 186, 24-41.

Müller, R.J., Kleeberg, I., Deckwer, W.D., 2001. Biodegradation of polyesters containing aromatic constituents. J. Biotechnol. 86, 87-95.

Murphy, F., Ewins, C., Carbonnier, F., Quinn, B., 2016. Wastewater treatment works (WWTW) as a source of microplastics in the aquatic environment. Environ. Sci. Technol. 50, 5800-5808.

Murray, F., Cowie, P.R., 2011. Plastic contamination in the decapod crustacean Nephrops norvegicus (Linnaeus, 1758). Mar. Pollut. Bull. 62, 1207-1217.

Nel, H., Froneman, P., 2015. A quantitative analysis of microplastic pollution along the south-eastern coastline of South Africa. Mar. Pollut. Bull. 101, 274-279.

Ngo, P.L., Pramanik, B.K., Shah, K., Roychand, R., Pathway., 2019. classification and removal efficiency efficienyof microplastics in wastewater treatment plants. Environ. Pollut. 113326.

O'Toole, G., Kaplan, H.B., Kolter, R., 2000. Biofilm formation as microbial development. Annu. Rev. Microbiol. 54, 49-79.

Ouattara, J.M.P., Coulibaly, L., Tiho, S., Gourène, G., 2009. Comparison of macrofauna communities in sediments of the beds of vertical flow constructed wetlands planted with Panicum maximum (Jacq.) treating domestic wastewater. Ecol. Eng. 2009; 35: 1237-1242.

Paço, A., Duarte, K., da Costa, J.P., Santos, P.S., Pereira, R., Pereira, M.E, Freitas A.C., Duarte A.C., Rocha-Santos T.A., 2017. Biodegradation of polyethylene microplastics by the marine fungus Zalerion maritimum. Sci. Total Environ. 586: 10-15.

Padervand, M., Lichtfouse, E., Robert, D., Wang, C., 2020. Removal of microplastics from the environment. A review. Environ. Chem. Lett. 1-22.

Park, H.B., Kamcev, J., Robeson, L.M., Elimelech, M., Freeman, B.D., 2017. Maximizing the right stuff: The trade-off between membrane permeability and selectivity. Science. 356, eaab0530.

.

Paul-Pont, I., Lacroix, C., Fernández, C.G., Hégaret, H., Lambert, C., Le Goïc, N., Frère L, Cassone AL, Sussarellu R, Fabioux C, Guyomarch J., 2016. Exposure of marine mussels Mytilus spp. to polystyrene microplastics: toxicity and influence on fluoranthene bioaccumulation. Environ. Pollut. 216, 724-737.

Perren, W., Wojtasik, A., Cai, Q., 2018. Removal of microbeads from wastewater using electrocoagulation. ACS Omega. 3, 3357-3364.

Perz, V., Hromic, A., Baumschlager, A., Steinkellner, G., Pavkov-Keller, T., Gruber, K., Bleymaier K, Zitzenbacher S, Zankel A, Mayrhofer C, Sinkel C., 2016. An esterase from anaerobic Clostridium hathewayi can hydrolyze aliphatic–aromatic polyesters. Environ. Sci. Technol. 50, 2899-2907.

Pinto, F., Franco, C., André, R., Miranda, M., Gulyurtlu, I., Cabrita, I., 2002. Co-gasification study of biomass mixed with plastic wastes. Fuel. 81, 291-297.

Pivokonsky, M., Cermakova, L., Novotna, K., Peer, P., Cajthaml, T., Janda, V., 2018. Occurrence of microplastics in raw and treated drinking water. Sci. Total Environ. 643, 1644-1651.

Prata, J. C., da Costa, J. P., Lopes, I., Duarte, A. C., Rocha-Santos, T., 2020. Environmental exposure to microplastics: An overview on possible human health effects. Sci. Total Environ. 702, 134455.

Qi, K., Cheng, B., Yu, J., Ho, W., 2017. Review on the improvement of the photocatalytic and antibacterial activities of ZnO. J. Alloys Compd. 727, 792-820.

Qin, J.J., Kekre, K.A., Tao, G., Oo, M.H., Wai, M.N., Lee, T.C., et al.. 2006. New option of MBR-RO process for production of NEWater from domestic sewage. J. Membr. Sci. 272, 70-77.

Qin, J-J., Oo, M.H., Lee, H., Kolkman, R., 2004. Dead-end ultrafiltration for pretreatment of RO in reclamation of municipal wastewater effluent. J. Membr. Sci. 243, 107-113.

Quinn, B., Murphy, F., Ewins, C., 2017. Validation of density separation for the rapid recovery of microplastics from sediment. Anal. Methods. 9, 1491-1498.

Raju, S., Carbery, M., Kuttykattil, A., Senthirajah, K., Lundmark, A., Rogers, Z., et al. 2020. Improved methodology to determine the fate and transport of microplastics in a secondary wastewater treatment plant. Water Res. 173, 115549.

Roch, S., Friedrich, C., Brinker, A., 2020. Uptake routes of microplastics in fishes: practical and theoretical approaches to test existing theories. Sci. Rep. 10, 1-12.

Rochman, C.M., Hoh, E., Kurobe, T., Teh, S.J., 2013. Ingested plastic transfers hazardous chemicals to fish and induces hepatic stress. Sci. Rep. 3, 3263.

Ross, P., van der Aa, L., van Dijk, T., Rietveld, L., 2019. Effects of water quality changes on performance of biological activated carbon (BAC) filtration. Sep. Purif. Technol. 212, 676-683.

Ruan, Y., Zhang, K., Wu, C., Wu, R., Lam, P.K., 2019. A preliminary screening of HBCD enantiomers transported by microplastics in wastewater treatment plants. Sci. Total Environ. 674, 171-178.

Rummel, C.D., Jahnke, A., Gorokhova, E., Kühnel, D., Schmitt-Jansen, M., 2017. Impacts of biofilm formation on the fate and potential effects of microplastic in the aquatic environment. Environ. Sci. Technol. Letters. 4, 258-267.

Sadri, S.S., Thompson, R.C., 2014. On the quantity and composition of floating plastic debris entering and leaving the Tamar Estuary, Southwest England. Mar. Pollut. Bull. 81, 55-60.

Sait, S. T., Sørensen, L., Kubowicz, S., Vike-Jonas, K., Gonzalez, S. V., Asimakopoulos, A. G., Booth, A. M., 2020. Microplastic fibres from synthetic textiles: Environmental degradation and additive chemical content. Environ. Pollut. 268, 115745.

Scherer, C., Weber, A., Lambert, S., Wagner, M., 2018. Interactions of microplastics with freshwater biota. Freshwater Microplastics. Springer, Cham, pp. 153-180.

Schwaferts, C., Niessner, R., Elsner, M., Ivleva, N.P., 2019. Methods for the analysis of submicrometer-and nanoplastic particles in the environment. Trends Anal. Chem. 112, 52-65.

Schwaferts, C., Sogne, V., Welz, R., Meier, F., Klein, T., Niessner, R., Elsner M, Ivleva N.P., 2020. Nanoplastic Analysis by Online Coupling of Raman Microscopy and Field-Flow Fractionation Enabled by Optical Tweezers. Anal. Chem. 92, 5813-5820.

Shah, A.A., Hasan, F., Hameed, A., Ahmed, S., 2008. Biological degradation of plastics: a comprehensive review. Biotechnol. Adv. 26, 246-265.

Shen, M., Ye, S., Zeng, G., Zhang, Y., Xing, L., Tang, W., Wen, X., Liu, S., 2020. Can microplastics pose a threat to ocean carbon sequestration?. Mar. Pollut. Bull. 150, 110712.

Shi, Y., Huang, J., Zeng, G., Gu, Y., Chen, Y., Hu, Y., Tang B, Zhou J, Yang Y, Shi L., 2017. Exploiting extracellular polymeric substances (EPS) controlling strategies for performance enhancement of biological wastewater treatments: an overview. Chemosphere. 180, 396-411.

Shim, W.J., Hong, S.H., Eo, S.E., 2017. Identification methods in microplastic analysis: a review. Anal. Methods. 9, 1384-1391.

Shim, W.J., Song, Y.K., Hong, S.H., Jang, M., 2016. Identification and quantification of microplastics using Nile Red staining. Mar. Pollut. Bull. 113, 469-476.

Siipola, V., Pflugmacher, S., Romar, H., Wendling, L., Koukkari, P., 2020. Low-Cost Biochar Adsorbents for Water Purification Including Microplastics Removal. Appl. Sci. 10, 788.

Simon, M., van Alst, N., Vollertsen, J., 2018. 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. 142, 1-9.

Sjollema, S.B., Redondo-Hasselerharm, P., Leslie, H.A., Kraak, M.H., Vethaak, A.D., 2016. Do plastic particles affect microalgal photosynthesis and growth? Aquat. Toxicol. 170, 259-261.

Solleiro-Villavicencio, H., Gomez-De León, C. T., Del Río-Araiza, V. H., Morales-Montor, J., 2020. The detrimental effect of microplastics on critical periods of development in the neuroendocrine system. Birth Defects Res. 112, 1326-1340.

Sol, D., Laca, A., Laca, A., Díaz, M., 2020. Approaching the environmental problem of microplastics: Importance of WWTP treatments. Sci. Total Environ. 740, 140016.

Sommer, F., Dietze, V., Baum, A., Sauer, J., Gilge, S., Maschowski, C., et al., 2018. Tire abrasion as a major source of microplastics in the environment. Aerosol Air Qual. Res. 18, 2014-2028.

Song, Y.K., Hong, S.H., Jang, M., Han, G.M., Rani, M., Lee, J., et al., 2015. A comparison of microscopic and spectroscopic identification methods for analysis of microplastics in environmental samples. Mar. Pollut. Bull. 93, 202-209.

Song, Y.K., Hong, S.H., Jang, M., Kang, J-H., Kwon, O.Y., Han, G.M., Shim W.J., 2014. Large accumulation of micro-sized synthetic polymer particles in the sea surface microlayer. Environ. Sci. Technol. 48, 9014-9021.

Sornalingam, K., McDonagh, A., Zhou, J.L., 2016. Photodegradation of estrogenic endocrine disrupting steroidal hormones in aqueous systems: progress and future challenges. Sci. Total Environ. 550, 209-224.

Stolte, A., Forster, S., Gerdts, G., Schubert, H., 2015. Microplastic concentrations in beach sediments along the German Baltic coast. Mar. Pollut. Bull. 99, 216-229.

Su, L., Xue, Y., Li, L., Yang, D., Kolandhasamy, P., Li. D., Shi H., 2016. Microplastics in taihu lake, China. Environ. Pollut. 216, 711-719.

Sun, Q., Ren, S. Y., Ni, H. G., 2020. Incidence of microplastics in personal care products: An appreciable part of plastic pollution. Sci. Total Environ. 140218.

Sussarellu, R., Suquet, M., Thomas, Y., Lambert, C., Fabioux, C., Pernet, M.E., Le Goïc N, Quillien V, Mingant C, Epelboin Y, Corporeau C., 2016. Oyster reproduction is affected by exposure to polystyrene microplastics. Proc. Natl. Acad. Sci. U.S.A. 113, 2430-2435.

Tagg, A., Harrison,. J.P., Ju-Nam, Y., Sapp, M., Bradley, E.L., Sinclair, C.J., Ojeda J.J., 2017. Fenton's reagent for the rapid and efficient isolation of microplastics from wastewater. Chem. Commun. 53, 372-375.

Tagg, A.S., Sapp, M., Harrison, J.P., Ojeda. Js.J., 2015, Identification and quantification of microplastics in wastewater using focal plane array-based reflectance micro-FT-IR imaging. Anal. Chem. 87, 6032-6040.

Talvitie, J., Mikola, A., Koistinen, A., Setälä, O., 2017a. Solutions to microplastic pollution–Removal of microplastics from wastewater effluent with advanced wastewater treatment technologies. Water Res. 123, 401-407.

Talvitie, J., Mikola, A., Setälä, O., Heinonen, M., Koistinen, A., 2017b. 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. 109, 164-172.

Tao, L., Wang, Y., Zou, Y., Zhang, N., Zhang, Y., Wu, Y., Wang Y, Chen R, Wang S., 2019. Charge Transfer Modulated Activity of Carbon-Based Electrocatalysts. Adv. Energy Mater. 1901227.

Teuten, E.L., Saquing, J.M., Knappe, D.R., Barlaz, M.A., Jonsson, S., Björn, A., Rowland S.J., Thompson R.C., Galloway T.S., Yamashita R, Ochi D., 2009. Transport and release of chemicals from plastics to the environment and to wildlife. Philos. Trans. R. Soc. Lond., B, Biol. Sci. 364, 2027-2045.

Thompson, R.C., Moore, C.J., Vom Saal, F.S., Swan, S.H., 2009. Plastics, the environment and human health: current consensus and future trends. Philos. Trans. R. Soc. Lond., B, Biol. Sci. 364, 2153-2166.

Thompson, R.C., Olsen, Y., Mitchell, R.P., Davis, A., Rowland, S.J., John, A.W., McGonigle D, Russell A.E., 2004. Lost at sea: where is all the plastic? Science. 304, 838.

Tian, L., Kolvenbach, B., Corvini, N., Wang, S., Tavanaie, N., Wang, L., Ma Y, Scheu S, Corvini PF, Ji R., 2017. Mineralisation of 14C-labelled polystyrene plastics by Penicillium variabile after ozonation pre-treatment. New. Biotechnol. 38, 101-105.

Tofa, T.S., Kunjali, K.L., Paul, S., Dutta, J., 2019. Visible light photocatalytic degradation of microplastic residues with zinc oxide nanorods. Environ. Chem. Lett. 17, 1341-1346.

Turner, A., Holmes, L.A., 2015. Adsorption of trace metals by microplastic pellets in fresh water. Environ. Chem. 12, 600-610.

Van Cauwenberghe, L., Devriese, L., Galgani, F., Robbens, J., Janssen, C.R., 2015. Microplastics in sediments: a review of techniques, occurrence and effects. Mar. Environ. Res. 111, 5-17.

Van Doren, L.G., Posmanik, R., Bicalho, F.A., Tester, J.W., Sills, D.L., 2017. Prospects for energy recovery during hydrothermal and biological processing of waste biomass. Bioresour. Technol. 225, 67-74.

Von Moos, N., Burkhardt-Holm, P., Köhler, A., 2012. Uptake and effects of microplastics on cells and tissue of the blue mussel Mytilus edulis L. after an experimental exposure. Environ. Sci. Technol. 46, 11327-11335.

Wang, F., Shih, K.M., Li, X.Y., 2015. The partition behavior of perfluorooctanesulfonate (PFOS) and perfluorooctanesulfonamide (FOSA) on microplastics. Chemosphere. 119, 841-847.

Wang, F., Wong, C.S., Chen, D., Lu, X., Wang, F., Zeng, E.Y., 2018a. Interaction of toxic chemicals with microplastics: a critical review. Water Res. 139, 208-219.

Wang, F., Yang, W., Cheng, P., Zhang, S., Zhang, S., Jiao, W., Sun Y., 2019a. Adsorption characteristics of cadmium onto microplastics from aqueous solutions. Chemosphere. 235, 1073-1080.

Wang, H., Zhang, Y., Wang, C., 2019b. Surface Modification and Selective Flotation of Waste Plastics for Effective Recycling-A Review. Sep. Purif. Technol. 226, 75-94.

Wang, L., Kaeppler, A., Fischer, D., Simmchen, J., 2019c. Photocatalytic TiO_2 micromotors for removal of microplastics and suspended matter. ACS Appl. Mater. Interfaces. 11, 32937-32944.

Wang, Q., Hernández-Crespo, C., Santoni, M., Van Hulle, S., Rousseau, D.P., 2020a. Horizontal subsurface flow constructed wetlands as tertiary treatment: Can they be an efficient barrier for microplastics pollution? Sci. Total Environ. 137785.

Wang, Q., Ye, L., Jiang, G., Jensen, P.D., Batstone, D.J., Yuan, Z., 2013. Free nitrous acid (FNA)-based pretreatment enhances methane production from waste activated sludge. Environ. Sci. Technol. 47, 11897-11904.

Wang, W., Wang, J., 2018. Investigation of microplastics in aquatic environments: An overview of the methods used, from field sampling to laboratory analysis. Trends Anal. Chem. 108, 195-202.

Wang, Y., Zhao, J., Wang, D., Liu, Y., Wang. Q., Ni, B.J., Chen F, Yang Q, Li X, Zeng G, Yuan Z., 2018b. Free nitrous acid promotes hydrogen production from dark fermentation of waste activated sludge. Water Res. 145, 113-124.

Wang, Z., Lin, T., Chen, W., 2020b. Occurrence and removal of microplastics in an advanced drinking water treatment plant (ADWTP). Sci. Total Environ. 700, 134520.

Wegner, A., Besseling, E., Foekema, E.M., Kamermans, P., Koelmans, A.A., 2012. Effects of nanopolystyrene on the feeding behavior of the blue mussel (*Mytilus edulis* L.). Environ. Toxicol. Chem. 31, 2490-2497.

Wei, W., Huang, Q.S., Sun, J., Wang, J.Y., Wu, S.L., Ni, B.J., 2019a. Polyvinyl chloride microplastics affect methane production from the anaerobic digestion of waste activated sludge through leaching toxic bisphenol-A. Environ. Sci. Technol. 53, 2509-2517.

Wei, W., Zhang, Y.T., Huang, Q.S., Ni, B.J., 2019b. Polyethylene terephthalate microplastics affect hydrogen production from alkaline anaerobic fermentation of waste activated sludge through altering viability and activity of anaerobic microorganisms. Water Res. 163, 114881.

Witt, U., Einig, T., Yamamoto, M., Kleeberg, I., Deckwer, W.D., Müller, R.J., 2001. Biodegradation of aliphatic–aromatic copolyesters: evaluation of the final biodegradability and ecotoxicological impact of degradation intermediates. Chemosphere. 44, 289-299.

Yang, L., Li, K., Cui, S., Kang, Y., An, L., Lei, K., 2019. Removal of microplastics in municipal sewage from China's largest water reclamation plant. Water Res. 155, 175-181.

Yang, L., Zhang, Y., Kang, S., Wang, Z., Wu, C., 2021. Microplastics in freshwater sediment: A review on methods, occurrence, and sources. Sci. Total Environ. 754, 141948.

Yang, Y., Liu, W., Xu, C., Wei, B., Wang, J., 2017. Antibiotic resistance genes in lakes from middle and lower reaches of the Yangtze River, China: effect of land use and sediment characteristics. Chemosphere. 178, 19-25.

Yang, Y., Yang, J., Wu, W.M., Zhao, J., Song, Y., Gao, L., Yang R., Jian L., 2015. Biodegradation and mineralization of polystyrene by plastic-eating mealworms: Part 1. chemical and physical characterization and isotopic tests. Environ. Sci. Technol. 49, 12080-12086.

Yin, L., Wen, X., Du, C., Jiang, J., Wu, L., Zhang, Y., et al., 2020. Comparison of the abundance of microplastics between rural and urban areas: A case study from East Dongting Lake. Chemosphere 244: 125486.

Yoshida, S., Hiraga, K., Takehana, T., Taniguchi, I., Yamaji, H., Maeda, Y., Toyohara K, Miyamoto K, Kimura Y, Oda K., 2016. A bacterium that degrades and assimilates poly (ethylene terephthalate). Science. 351, 1196-1199.

Yu, F., Yang, C., Zhu, Z., Bai, X., Ma, J., 2019. Adsorption behavior of organic contaminants and metals on micro/nanoplastics in the aquatic environment. Sci. Total Environ. 694, 133643.

Zhang, K., Shi, H., Peng, J., Wang, Y., Xiong, X., Wu, C., Lam P.K., 2018. Microplastic pollution in China's inland water systems: A review of findings, methods, characteristics, effects, and management. Sci. Total Environ. 630, 1641-1653.

Zhang, X., Chen, J., Li, J., 2020a. The removal of microplastics in the wastewater treatment process and their potential impact on anaerobic digestion due to contaminants association. Chemosphere. 126360.

Zhang, X., Zhang, H., Yu, K., Li, N., Liu, Y., Liu, X., Zhang H, Yang B, Wu W, Gao J, Jiang J., 2020b. Rapid monitoring approach for microplastics using portable pyrolysis-mass spectrometry. Anal. Chem. 92, 4656-4662.

Zhang, Y., Diehl, A., Lewandowski, A., Gopalakrishnan, K., Baker, T., 2020c. Removal efficiency of micro-and nanoplastics (180 nm–125 μm) during drinking water treatment. Sci. Total Environ. 720, 137383.

Zhao, S., Zhu, L., Li, D., 2015. Microplastic in three urban estuaries, China. Environ. Pollut. 206, 597-604.

Zhao, S., Zhu, L., Li, D., 2016. Microscopic anthropogenic litter in terrestrial birds from Shanghai, China: Not only plastics but also natural fibers. Sci. Total Environ. 550, 1110-1115.

Zheng, Y., Yanful, E.K., Bassi, A.S., 2005. A review of plastic waste biodegradation. Crit. Rev. Biotechnol. 25, 243-250.

Ziajahromi, S., Drapper, D., Hornbuckle, A., Rintoul, L., Leusch, F.D., 2020. Microplastic pollution in a stormwater floating treatment wetland: Detection of tyre particles in sediment. Sci. Total Environ. 713, 136356.

Ziajahromi, S., Neale, P.A., Rintoul, L., Leusch, F.D., 2017. Wastewater treatment plants as a pathway for microplastics: development of a new approach to sample wastewater-based microplastics. Water Res. 112, 93-99.

Ziajahromi, S., Neale, P.A., Telles Silveira, I., Chua, A., Leusch, F.D.L., 2021. An audit of microplastic abundance throughout three Australian wastewater treatment plants. Chemosphere 263, 128294.