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Detection, occurrence and removal

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Review

Microplastics in wastewater treatment plants: Detection, occurrence and removal

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ABSTRACT

Microplastics have aroused increasing concern as they pose threats to aquatic species as well as human beings. They do not only contribute to accumulation of plastics in the environment, but due to absorption they can also contribute to spreading of micropollutants in the environment. Studies indicated that wastewater treatment plants (WWTPs) play an important role in releasing microplastics to the environment. Therefore, effective detection of the microplastics and understanding their occurrence and fate in WWTPs are of great importance towards microplastics control. In this review, the up-to-date status on the detection, occurrence and removal of microplastics in WWTPs are comprehensively reviewed. Specifically, the different techniques used for collecting microplastics from both wastewater and sewage sludge, and their pretreatment and characterization methods are reviewed and analyzed. The key aspects regarding microplastics occurrence in WWTPs, such as concentrations, total discharges, materials, shapes and sizes are summarized and compared. Microplastics removal in different treatment stages and their retention in sewage sludge are explored. The development of potential microplastics-targeted treatment technologies is also presented. Although previous researches in microplastics have undoubtedly improved our level of understanding, it is clear that much remains to be learned about microplastics in WWTPs, as many unanswered questions and thereby concerns still remain; some of these important future research areas are outlined. The key challenges appear to be to harmonize detection methods as well as microplastics mitigation from wastewater and sewage sludge.

Contents

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1. Introduction

Microplastics, often defined as plastic particles < 5 mm (Thompson, 2015), have aroused increasing concerns as they pose threat to aquatic species as well as human beings. Microplastics can be directly manufactured, known as primary microplastics, and be used in many personal care and cosmetic products (PPCPs) (Napper et al., 2015; Van Wezel et al., 2016). Also, they can be formed by erosion of large plastic debris via exposure to environmental stressors such as water, wind and sunlight, which are defined as secondary microplastics (Galgani et al., 2013; Singh and Sharma, 2008). The massive usage of plastic products and poor management of plastic waste disposal lead to microplastics being ubiquitously found in aquatic water bodies, including rivers, lakes, estuaries, coastlines and marine ecosystems (Eerkes-Medrano et al., 2015; Li et al., 2018a; Thompson, 2015; Van Cauwenberghe et al., 2015).

Microplastics do not pose acute fatal effects on living organisms, they could however cause chronic toxicity, which is considered as a key issue in long term exposure (Li et al., 2018a). Microplastics induce toxic effects through several mechanisms. Firstly, the toxicity could be directly caused by the polymer materials used for manufacturing plastic products. Polystyrene (PS) is a good example, which is widely used in protective packaging, containers, bottles and lids, but is found to be able to translocate in blood circulation and cause reproductive disruption for marine filter feeders (Chen et al., 2006; Sussarellu et al., 2016). Secondly, microplastics could inflict damage on organisms and cause inflammation due to their small size and sharp ends. It has been observed that ingestion of tiny microplastics could cause malnutrition and alterations in reproduction for some organisms (Besseling et al., 2014). A study also indicated that small microplastics (<10 μm) could be translocated from the gut into the circulatory system of aquatic species (Browne et al., 2008). In addition, additives incorporated into plastics to improve their properties could also be toxicants to organisms. For instance, phthalates and polybrominated diphenyl ethers are two common additives to improve plasticity and fire resistances of the plastics. However, they are also well known as endocrine disrupting compounds (EDCs). Studies have confirmed these chemicals were present in human bodies and were anticipated to be accumulated through bioaccumulation processes (Talsness et al., 2009; Teuten et al., 2009). Furthermore, due to large surface area to volume ratio and hydrophobic nature of the microplastics, they are effective in adsorbing persistent organic pollutants (POPs), such as polychlorinated biphenyles (PCBs) (Bakir et al., 2012). The POPs adsorbed to plastics could reach up to 1 million times higher concentrations than ambient concentrations and these compounds can be further desorbed inside organisms, exacerbating POPs bioaccumulation at higher trophic levels (Browne et al., 2013; Rios et al., 2010).

A recent study indicated that wastewater treatment plants (WWTPs) potentially played an important role in releasing microplastics to the environment (Browne et al., 2011). Microbeads added into facial cleanser, toothpaste can be directly discharged into wastewater through human activities (Cheung and Fok, 2017; Fendall and Sewell, 2009). Also, synthetic clothing, such as polyester (PES) and nylon, might shed thousands of fibers into wastewater during the washing process (Browne, 2015; Napper and Thompson, 2016). The WWTP may remove some of the microplastics depending on the treatment units employed. However, it has been shown that microplastics could bypass the WWTP, entering into the aquatic water bodies and finally accumulated in the environment (Carr et al., 2016; Murphy et al., 2016). The issue of releasing of microplastics from WWTP has drawn attention of a growing number of researchers more recently, with the publication number growing exponentially in last three years according to the search in the database of “Web of Science” (http://apps.webofknowledge.com/) (Fig. S1(A), Supporting Information). A large proportion of these studies worked on the development of approaches suitable for sampling and identifying microplastics in WWTPs (Fig. S1(B), SI). Wastewater and sewage sludge samples were mainly collected using a container or a separate pump and the microplastics were then extracted through various steps. Spectroscopic techniques have been developed and optimized to characterize the extracted microplastics. Another part of the studies investigated the occurrence and removal of microplastics in WWTPs (Fig. S1(B), SI). The reported concentrations of microplastics in wastewater and sludge varied greatly in different WWTPs and the polymer materials covered a wide range. The removal efficiencies of microplastics by different treatment units were also evaluated recently. However, so far, no attempt has been made to provide a comprehensive summary on these findings.

This article aims to provide a comprehensive review to better understand key aspects on WWTPs as an important pathway of microplastics introducing to the environment, which includes: (1) techniques applied to sample and detect microplastics in WWTPs; (2) the occurrence of microplastics in WWTPs and their properties; and (3) the removal of microplastics in WWTPs with different treatment processes. Based on the review, the outlooks on key future research directions are also discussed.

2. Techniques for microplastics detection in WWTPs

The detection of microplastics in WWTPs usually contains three steps, i.e., sample collection, sample pretreatment and microplastics characterization/quantification, as summarized in Fig. 1,

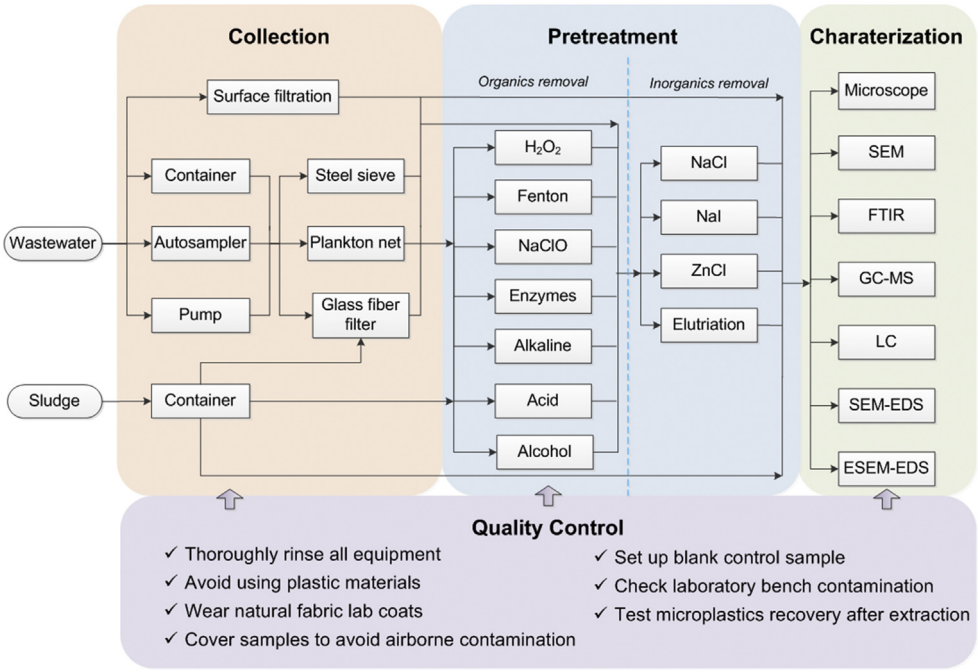

Fig. 1. Flow chart summarizing steps and techniques used for microplastics detection in WWTPs.

though the methods used in each step are not standardized yet. Different techniques may be applied according to sample characteristics, since microplastics can be present in both wastewater and sewage sludge. Also, the techniques for identification of microplastics can result in different dimensions in the final analysis output. In addition, quality control was conducted in most of the studies to avoid potential bias introduced by sample contamination and sample loss.

2.1. Sample collection

The microplastics in wastewater can be collected in different ways, mainly including container collection (Magnusson and Norén, 2014; Murphy et al., 2016; Tagg et al., 2015), autosampler collection (Michielssen et al., 2016; Talvitie et al., 2016), separate pumping and filtration (Mason et al., 2016; Mintenig et al., 2017; Talvitie et al., 2015; Ziajahromi et al., 2017) and surface filtration (Carr et al., 2016). Collecting microplastics with containers or autosamplers is easy for practicing. However, it can only get a limited volume of wastewater sample, usually only a few liters per collection event (Fig. 2A). In this sense, it is more suitable for collecting microplastics in wastewater which contains high content of organic matters and solids, such as the influent of the WWTP, given the ease of the following filtration process. Separate pumping and filtration can effectively increase the sampling volume to hundreds of liters or even hundreds of cubic meters, depending on the wastewater characteristics and mesh sizes of the filtration devices (Fig. 2A). For this sampling process, wastewater was pumped from the water stream into a filtration device using an extraction pump and the microplastics were intercepted (Fig. 3A). The method is frequently used for collecting microplastics in the effluent of WWTPs. Distinctively, Carr et al. (2016) designed a surface filtering assembly for skimming the water surface at the final fall location in WWTPs (Fig. 3B). This method enables the sampling volume to be further increased to thousands of cubic meters. However, the application of this method had a practical limitation, as it can only be applied at the water falls. Also, since the surface filtering assembly was deployed in an open channel, the fugitive airborne contamination can hardly be avoided, which needs to be considered during the microplastics quantification. In addition, the method is likely to underestimate the microplastics count as skimming the water surface may only intercept the microplastics with low density.

Due to relatively low concentrations of microplastics as well as their uneven temporal and spatial distributions in wastewater, the representativeness of the sample should be considered during the collection. The current attempts for taking representative sample include increasing the sampling volume and taking 24-h composite samples (Mason et al., 2016; Talvitie et al., 2016). A microplastics sampling guide is worthy to be developed in future, aimed at offering an efficient and informed choice on the appropriate sampling mode and frequency to minimize sampling errors and maximize data quality. It has been proposed to reduce sampling uncertainty of micropollutants in the wastewater system by adjusting the sampling mode and the sampling frequency according to the goal of the study as well as the flow characteristics (Ort et al. 2010a, 2010b). This sampling guide of micropollutants might be served as a basis for designing the microplastics sampling protocols. However, other aspects such as the particle dynamics related to density and geometry of microplastics should also be taken into consideration.

Collected wastewater samples are usually filtered in order to concentrate the microplastics. Thus, the mesh/pore sizes of sieves and filters will have a profound effect on the amount of microplastics collected (Magnusson and Norén, 2014). However, so far, the mesh/pore sizes applied in the related studies have not been unified. Mesh sizes used vary from around 1 μm to 500 μm (Fig. 2B). Also, in many studies, a stack of sieve pans was used for the filtration (Fig. 3A). This method helps to increase the total volume for the filtration and allows distinguishing size categories of microplastics. However, the mesh-based size categorization can be far beyond accurate. Michielssen et al. (2016) observed that some particles would not pass through the sieves even if sufficiently small due to their irregular shapes. Also, the morphology of fibers

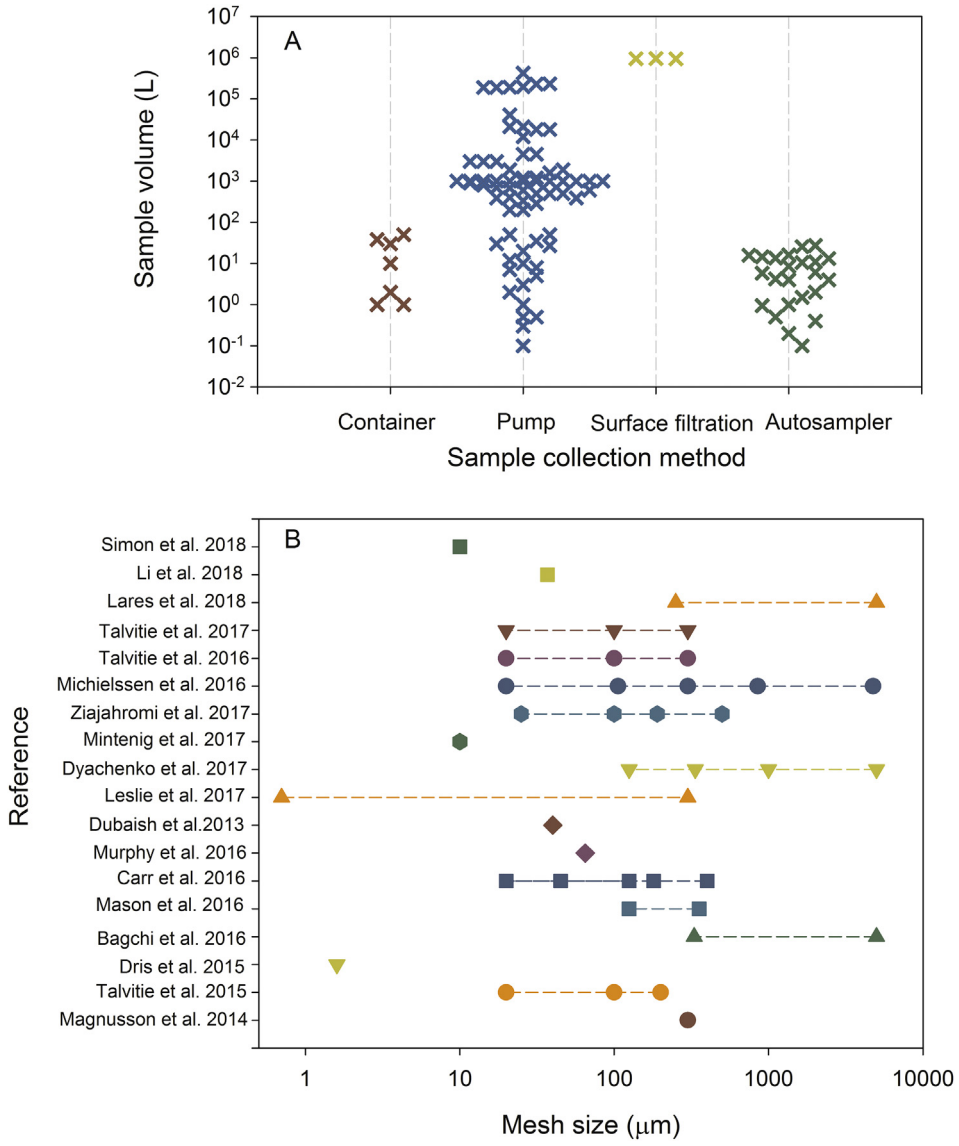

Fig. 2. (A) Volumes of wastewater collected based on different sample collection methods. Based on: Dubaish and Liebezeit (2013), Magnusson and Norén (2014), Dris et al. (2015), Tagg et al. (2015), Bagchia et al. (2016), Mason et al. (2016), Carr et al. (2016), Murphy et al. (2016), Dyachenko et al. (2017), Mintenig et al. (2017), Michielssen et al. (2016), Talvitie et al. (2016), Leslie et al. (2017), Ziajahromi et al. (2017), Talvitie et al. (2017), Lares et al. (2018), Simon et al. (2018). (B) Mesh sizes applied in different studies related to microplastics in WWTPs.

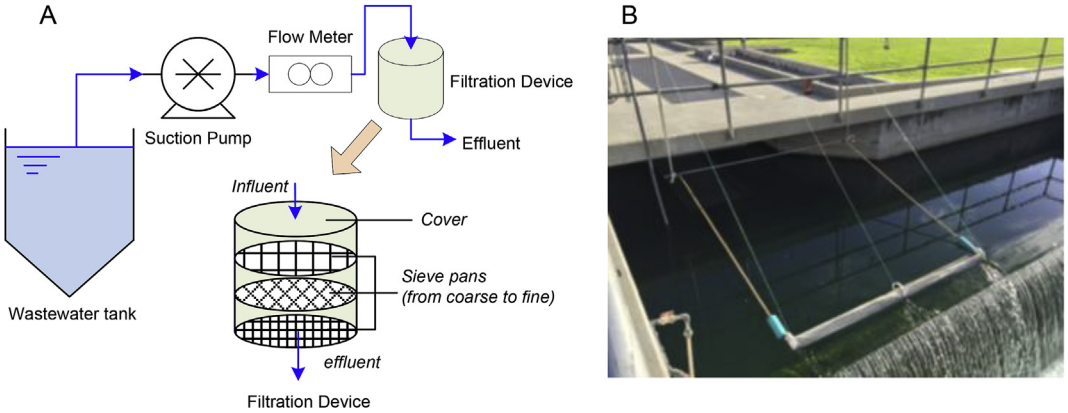

Fig. 3. A. Microplastics collection through separate pumping and filtration, with a filtration device consist of a stack of sieve pans. B. Microplastics collection using a surface filtering assembly (Carr et al., 2016).

enable them to pass longitudinally through smaller filters (Mintenig et al., 2017; Ziajahromi et al., 2017). The microplastics could also be separated *in situ* from surface water by using a neuston plankton net and manta trawl (Li et al., 2018a; Prata et al., 2019). These methods have been commonly used to collect and separate microplastics from freshwater or seawater, but have not been applied to collect microplastics in WWTPs yet.

For microplastics in the sewage sludge, as the sludge has much higher fractions of solids and organic matter than wastewater, separation through direct filtration was seldom applied (only applied to spectroscopic measurement) (Magnusson and Norén, 2014; Murphy et al., 2016). A common practice is to collect the sludge sample (~5–20g) in a glass container and refrigerate it in the dark (~4 °C) before transporting to the laboratory for further sample processing and microplastics extraction using the methods described in Section 2.2.

2.2. Sample pretreatment

Since samples obtained from WWTPs, sludge samples particularly, may contain a high concentration of organic matter or inorganic solids, various methods are applied to purify and extract microplastics from their original matrices. These processes are expected to facilitate the following microplastics quantification and identification. Especially, the removal of organic matter is essential for chemical identification of microplastics.

A commonly employed method to remove organic matter in WWTP samples is (catalytic) wet peroxidation (WPO). Chemicals including H_2O_2, NaClO and Fenton reagents are usually used for oxidizing organic matter. This method is also widely used to pretreat microplastic samples collected from seawater, freshwater, sediments and organisms (Erni-Cassola et al., 2017; Karami et al., 2016; Masura et al., 2015). The majority of plastic debris are considered remain unchanged during the WPO processes, apart from a slight change in the size of polyethylene and polypropylene particles (McCormick et al., 2014; Nuelle et al., 2014). Tagg et al. (2015) demonstrated that the 83% of organic matter could be successfully removed with the Fourier transform infrared spectroscopy (FTIR) spectra of microplastics remaining unchanged after the samples being exposed to H_2O_2 (30%) for 7 days. However, this method might not be efficient for samples with a larger volume (>1L) or with increased organic detritus due to prolonged treatment time. Fenton reagents, on the other hand, are able to rapidly break down organic compounds within a short timeframe, without impacting the microplastics (Tagg et al., 2017). This method was also recommended by the US National Oceanic and Atmospheric Administration (NOAA) to analyze microplastics in the marine environment (Masura et al., 2015).

Another emerging approach to purify microplastics from organic matter is enzymatic degradation. During the degradation process, microplastics samples are submerged in a mixture of technical enzymes such as lipase, amylase, proteinase, chitinase, and cellulase (Cole et al., 2014; Löder et al., 2015). The proteins, lipids and carbohydrates can be specifically removed with the microplastics remaining unaffected. More recently, Mintenig et al. (2017) applied a multi-step, plastic-preserving enzymatic maceration method to treat the wastewater sample. The method combined the usage of enzymes (protease, lipase and cellulase) with sodium dodecyl sulphate (SDS, 5% w/vol) and H_2O_2 (35%, in some of the samples). However, the whole degradation procedure could take longer than 13 days. The effectiveness of the enzymatic degradation for organics removal was further proven by Löder et al. (2017). They also modified the protocol by optimizing the incubation conditions, increasing the SDS concentration, changing buffer composition, adding two optional enzymes for samples with high organics, and so on. The modified protocol improved the efficiency of enzyme purification and enhanced the removal of polysaccharide and lipids, which could be used to purify samples in a broader range of environmental matrices, such as seawater samples, freshwater plankton samples, extracted sediment samples, wastewater samples, and tissue samples of aquatic species.

Alternative methods for organic matters removal in wastewater and sludge samples include alkaline treatment and acid treatment (Ziajahromi et al., 2017). However, the application of these methods required extra concern. Coles et al. (2014) found that the harsh condition (10M of NaOH at 60 °C) would damage the microplastics. Also, the strong oxidizing acid such as sulphuric acid and nitric acid would destroy or damage the low pH intolerant polymers (e.g. polyamide, polystyrene) (Claessens et al., 2013). Acid treatment was usually performed with either a heating block or microwave digestion at 110–120 °C, while some microplastics were even observed to melt at 90 °C (Carr et al., 2016). Only one study used isopropyl alcohol to remove the organic matter impregnated on the microplastics, while the removal efficiency remained untested (Bayo et al., 2016). Other approaches such as ultrasonication combined with deionized water or SDS solution have also been applied to treat seawater samples in the past, but not for wastewater samples, probably due to formation of even smaller microplastics from the brittle plastic samples by applying these methods (Cooper and Corcoran, 2010; Li et al., 2018a).

The inorganic materials in the wastewater and sludge samples are usually removed based on density separation using salt solution. The saturated sodium chloride (NaCl) solution (density: 1.2 kg/L) is commonly used because of its low cost and non-toxicity (Leslie et al., 2017; Li et al., 2018a). However, microplastics with a high density such as polyvinyl chloride (PVC) (density: 1.14–1.56 kg/L) and polyethylene terephthalate (PET) (density: 1.32–1.41 kg/L) can also be settled for removal, resulting in the underestimation of total count (Duis and Coors, 2016). Therefore, to separate all plastic in wastewater and sludge samples, denser salt solutions including NaI solution (density: 1.6–1.8 kg/L) or ZnCl solution (density: 1.5–1.7 kg/L) have been applied (Mintenig et al., 2017; Ziajahromi et al., 2017). Carr et al. (2016) also separated microplastics in the influent of WWTPs using the technique of elutriation, which was originally developed by Claessens et al. (2013) for extracting microplastics from sediments. The microplastics were isolated by exploiting their inherent bouyancies based on combination of water flow and aeration.

2.3. Microplastic characterization

In general, the analysis of microplastics can be classified into physical characterization and chemical characterization. The physical characterization mainly refers to characterizing the size distribution of microplastics as well as assessing other physical parameters such as shape and color. On the other hand, chemical characterization was mainly applied to explore the composition of microplastics.

For the physical characterization, stereomicroscope is the most widely used facility. It can be used straightforwardly to measure the size, characterize the morphology and enumerate the count of the microplastics. However, the visual identification of microplastics is open to bias, as it is size-limited due to relatively low magnification factor of stereomicroscope and the result depends strongly on the operator. It has been estimated that up to 70% error ratio could be observed and this error increased with the decreasing particle size (Hidalgoruz et al., 2012). For example, it is occasionally difficult to distinguish between synthetic and natural fibers, such as textile fibers made of cotton (Magnusson and Norén, 2014). Also, items of similar color to background paper might be overlooked (Murphy

et al., 2016). It is even likely to duplicate or miss counts due to the large microplastics amount. Therefore, different measures have been taken to avoid potentially errors. To facilitate particle counting, gridded petri dishes with sequentially numbered grids were used (Carr et al., 2016). To distinguish synthetic fibres from biological ones, a series of criteria have been applied (Dris et al., 2015; Hidalgoruz et al., 2012): i) synthetic fibres had to be equally thick through their entire length; ii) synthetic fibres should not be entirely straight, which indicates a biological origin; iii) no cellular or organic structures should be visible to consider a fibre as microplastic, and iv) transparent and green fibres were examined with higher magnification to confirm their nature. Magnusson and Norén (2014) also used the alcohol burner for distinguishing, as plastics would melt with the heat while non-synthetic fibers would not. In addition, a staining method was used to minimize overestimation of the suspected microplastics (Ziajahromi et al., 2017). By applying Rose-Bengal solution (4,5,6,7-tetrachloro-20,40,50,70-tetraiodo-fluorescein, Sigma-Aldrich, 95% dye content), the natural micro particles, such as natural fibres, could be stained pink, allowing visual separation of plastic and non-plastic particles (Liebezeit and Liebezeit, 2014). With measures mentioned above, the accuracy of microplastics characterization with stereomicroscopy can be improved. However, the method cannot distinguish polymer types and is time-consuming since no automatization is possible.

Chemical characterization of microplastics can increase the accuracy of microplastics identification and further explore their composition. The current chemical analysis method include destructive technique, such as gas chromatography coupled to mass spectrometry (GC-MS), including pyrolysis-GC-MS and thermal extraction desorption-GC-MS (Dekiff et al., 2014; Dümichen et al., 2017; Fries et al., 2013; Nuelle et al., 2014), and liquid chromatography (LC) (Elert et al., 2017; Hintersteiner et al., 2015), as well as non-destructive spectroscopic techniques, such as FTIR spectroscopy (Browne et al., 2011; Löder et al., 2015; Mintenig et al., 2017) and Raman spectroscopy (Araujo et al., 2018; Erni-Cassola et al., 2017; Lares et al., 2018). Among these methods, the spectroscopic techniques were most widely used to analyze microplasics in environmental samples. However, it is difficult to identify tiny microplastics (ca. <1 μm) with these techniques due to the equipment limitation. The comprehensive comparison of these methods with their advantages and limitations can be found elsewhere (Hidalgoruz et al., 2012; Rocha-Santos and Duarte, 2015).

FTIR is the most frequently reported method used for analyzing microplastics obtained in WWTPs. With this technique, microplastic particle is exposed to infrared radiation and a spectrum is obtained where characteristic peaks correspond to specific chemical bonds between atoms. The obtained spectrum can be used to identify the sample composition by comparing with the reference spectra library. However, these reference spectra always represent very clean and ideal samples, not typically found in the environment (Murphy et al., 2016). Therefore, it is necessary to create a library of non-typical reference plastics taken from various sources, including WWTPs, which allows a comparison to much more environmentally relevant samples. In addition, the traditional FTIR analysis is labor-intensive as microplastics need to be firstly selected under the light microscopy and then be analyzed for the spectra of each particle individually. (Harrison et al., 2012). Thus, FTIR was usually served to assist the visual identification for most of the studies and only ambiguous fragments or sub-samples were examined (Carr et al., 2016; Leslie et al., 2017; Murphy et al., 2016). The recent development of focal plane array (FPA) based micro-FTIR imaging could be more effective to evaluate the spectra of individual particles in a sample, resulting in a high throughput analysis of total microplastics in a sample (Löder et al., 2015). Mintenig et al. (2017) used FPA-based transmission micro-FTIR to identify microplastics in both wastewater and sludge samples, as they considered that IR-transmission showed better images than IR-reflectance. However, the FPA-micro-FTIR technique still has its limitation. Mintenig et al. (2017) found that the technique of imaging was stretched to its limits to identify fibres. In addition, as the lateral resolution of micro-FTIR spectroscopy is always limited to certain diffraction range (e.g. 10 μm at 1000 cm^{-1}) and samples down to 10 μm-20 μm can hardly be analyzed (Li et al., 2018a).

Raman spectroscopy is another frequently used spectroscopic method to identify microplastics. This is a vibrational spectroscopy technique based on the inelastic scattering of light. It provides information on the molecular vibrations of a system in the form of a vibrational spectrum, allowing identification of the components present in the sample (Schymanski et al., 2018). Compared with FTIR, Raman techniques show better spatial resolution (down to 1 μm) (Ribeiro-Claro et al., 2017). Also, it has higher sensitivity to non-polar functional groups and is insensitive towards disturbing signals of water and atmospheric CO_2 (Li et al., 2018a). However, Raman spectroscopy is prone to fluorescence interference, resulting from microbiological, organic or inorganic items in samples. Therefore, the purification of samples should be performed carefully to avoid undesirable sample modification prior to Raman analysis (Elert et al., 2017). Also, fluorescent dyes, such as Nile Red, have been used in some studies to pretreat the microplastics sample for fast and accurate Raman detection (Erni-Cassola et al., 2017; Maes et al., 2017). Currently, only one study used Raman spectroscopy to detect microplastics in the wastewater system (Lares et al., 2018).

Microplastics can also be analyzed by scanning electron microscopy (SEM) based techniques. The traditional SEM produces images of microplastics by scanning the surface with a focused beam of electrons, which has been used for characterizing surface morphology of microplastics in the sewage sludge (Mahon et al., 2017). In addition, SEM-energy dispersive X-ray spectroscopy (SEM-EDS) and environmental scanning electron microscopy-EDS (ESEM-EDS) could be used both for characterizing the surface morphology of microplastics and determining the elemental composition of polymers based on diffraction and reflection of emitted radiation from microplastics surfaces (Dubaish and Liebezeit, 2013; Eriksen et al., 2013; Vianello et al., 2013).

On the other hand, the GC-MS based techniques and LC based techniques can be used for fast identification of plastic in the sample. The GC-MS methods are usually applied with thermoanalytical techniques which identifies microplastics by analyzing the mass spectrometry of their thermal degradation products (Dümichen et al., 2015; Fries et al., 2013). The LC based techniques can be carried out in the form of size exclusion chromatography, which separates dissolved analytes from their hydrodynamic volume as a function of the effective size of the molecules (Elert et al., 2017). Both methods are able to analyze polymer types and quantitative results could be obtained with proper calibration, which facilitate the assessment of the contamination of the studied ecosystem with plastic particles. Unlike spectroscopic techniques, these methods do not have any requirement on microplasics size during the measurement, since they do not deliver direct information regarding the size and number of particles. It is still being discussed how to link the two dimensions of the analysis output, i.e. mass and number, to gain a holistic view on microplastics concentrations. Also, these methods are still at the development stage for environmental sample analysis and have not yet been applied for analyzing microplastics obtained from WWTPs.

2.4. Quality control

During the sample collection and pretreatment, contaminations

of samples might be induced from atmospheric fallout, the equipment and devices used and even clothing of workers (Duis and Coors, 2016; Mintenig et al., 2017; Talvitie et al., 2017). Therefore, different measures should be taken to avoid the potential bias caused by these contaminations. For example, all equipment should be thoroughly rinsed before use and the usage of plastic materials needs to be avoided as much as possible. Laboratory coats made of natural fabric were recommended to wear during all laboratory procedures. The samples were commonly sealed in petri dishes or covered with aluminium foil to minimize airborne plastics contamination. It has also been suggested to setup a blank control sample, processed in parallel with experimental samples with all steps of sample processing, to determine any potential plastic contamination from the laboratory (Ziajahromi et al., 2017). In addition, Murphy et al. (2016) applied the tape lifting technique, frequently used in forensic science laboratories, to check laboratory benches for fiber and particle contamination. With this method, any plastic trace particles appeared on the bench could be adhered to the glue on the tape and could then be examined for identification.

Another issue related to microplastics detection is the potential sample loss during the sample extraction (Bagchia et al., 2016; Löder et al., 2017; Tagg et al., 2017). Thus, the recovery of the microplastics after the extraction processes is recommended to be tested. To determine the recovery rate, a certain amount of microplastics with the specific size and color can be added into the Milli-Q water and undergo the same pretreatment and extraction processes as the wastewater or sludge samples do. Afterwards, number of the added specific microplastics is counted and the recovery rate can be calculated. It has been reported that a high recovery rate of $84.5 \pm 3.3\%$ was achieved after enzymatic purification using polyethylene (PE) as a model polymer (Löder et al., 2017). However, it is preferable that the model microplastics cover a wide range of materials and types.

3. Occurrence of microplastics in WWTPs

3.1. Microplastics concentrations in influent and effluent of WWTPs

Microplastics were readily detected in both influent and effluent of WWTPs, with their typical appearances as shown in Fig. 4 (A and B). Table 1 summarizes the reported microplastics concentrations in the influent and effluent of different WWTPs. The concentrations in each individual WWTP are listed in Table S1 (SI). Microplastics in the influent were only measured in a few WWTPs, with the particle concentrations reported varying from 1 to 10044 particle/L. The measured microplastics concentrations in the effluent of WWTPs were much lower, which were in the range of 0–447 particle/L (Table 1). The large variations in microplastics concentrations in these WWTPs could be partially related to different sample collection, pretreatment and analysis methods applied in these studies. For example, a higher microplastics concentration might be observed when a finer mesh size was applied (Leslie et al., 2017; Simon et al., 2018). Also, studies without chemical characterization were very likely to induce bias in quantification, especially distinguishing natural and synthetic fibers. As a result, the count of natural fibers were included in some studies, (Mason et al., 2016; Talvitie et al., 2015). Therefore, for better comparing microplastics concentrations in different studies, the standardization or harmonization of methods of microplastics sampling and analyzing is urgently needed.

The inter-plant differences of microplastics concentrations could also be related to a complex variety of factors such as catchment sizes, population served, adjacent surrounding land use, combined sewer systems, wastewater sources (residential, commercial or industrial), etc. As a large part of microplastics in the wastewater are originated from household discharges, the human activities in the served catchment, such as preference of residents for wearing synthetic clothes or using plastic products, might directly affect the microplastics concentration in the wastewater. Mason et al. (2016) conducted statistical analysis over 17 WWTPs in the United States and the results showed that the served population was positively associated with the total microplastics particles in the wastewater. However, Mintenig et al. (2017) found that no significant correlation was observed between microplastics ($<500\,\mu m$) counts and population equivalents of 12 evaluated WWTPs in Germany. The different information derived from studies in different countries indicates that it is worthwhile to investigate microplastics in wastewater from areas with different economic levels and different living habits in future to fully evaluate their occurrence.

The combined sewer system was found to be associated to increased number of fragment in the wastewater, which could be related to the adjacent surrounding land use as well as transport-related emissions, such as microplastics released from wear of tires and brakes (Mason et al., 2016; Wagner et al., 2018). The results of the study conducted by Michielssen et al. (2016) also indicated that WWTPs connected with combined sewer systems might result in high concentrations of microplastics and other small anthropogenic litter (SAL) in the influent. Although combined sewer might increase the microplastics load of the WWTP, it also plays an important role for reducing microplastics emission to surface water. It has been reported that if the extra microplastics in combined sewer systems are derived from run-off from roads, the emission to surface waters will be minimized in these areas compared to systems with separate sewer systems (Wanger et al., 2018). In the WWTP, the microplastics can be partially removed from wastewater (See section 4.1).

The microplastics concentration in the effluent of WWTPs will be affected by the wastewater treatment processes applied. In general, the WWTPs with tertiary treatment processes had a lower microplastics concentration (0–51 particle/L) in the effluent than those with primary or secondary treatment processes only (9×10^{-4} - 447 particle/L) (Table 1). However, studies also showed the tertiary treatment in some WWTPs did not further decrease the microplastics concentration in the effluent (Mason et al., 2016; Mintenig et al., 2017). This could be a result of different treatment processes applied, which will be further discussed in Section 4. So far, only one study reported the microplastics concentration of the effluent from WWTP which had primary treatment alone (Ziajahromi et al., 2017). The concentration was about one order of magnitude higher than those from WWTPs with secondary and tertiary treatments as reported in the same study (the same microplastics sampling and analyzing methods were used).

Despite of relatively low concentrations of microplastics in the effluent of WWTPs, the total discharges of the microplastics from WWTPs were still considerably high, as most of these facilities process millions of liters of wastewater every day. As listed in Table 1, the median value of the total daily discharge of microplastics (estimated based on annual efflux and effluent concentration) in the studied WWTPs was 2×10^6 particle/day, corresponding to an average annual efflux of $5 \times 10^7\ m^3$/year. In some WWTPs in the Netherland and United States, the total daily discharge could even be more than 1×10^{10} particle/day. The WWTPs with high microplastics discharges had annual effluxes over $1 \times 10^7\ m^3$ and population equivalents of more than 1×10^6. The high microplastics discharges from WWTPs suggest that microplastics-targeted treatment technology are urgently needed to avoid their massive emission into aquatic system.

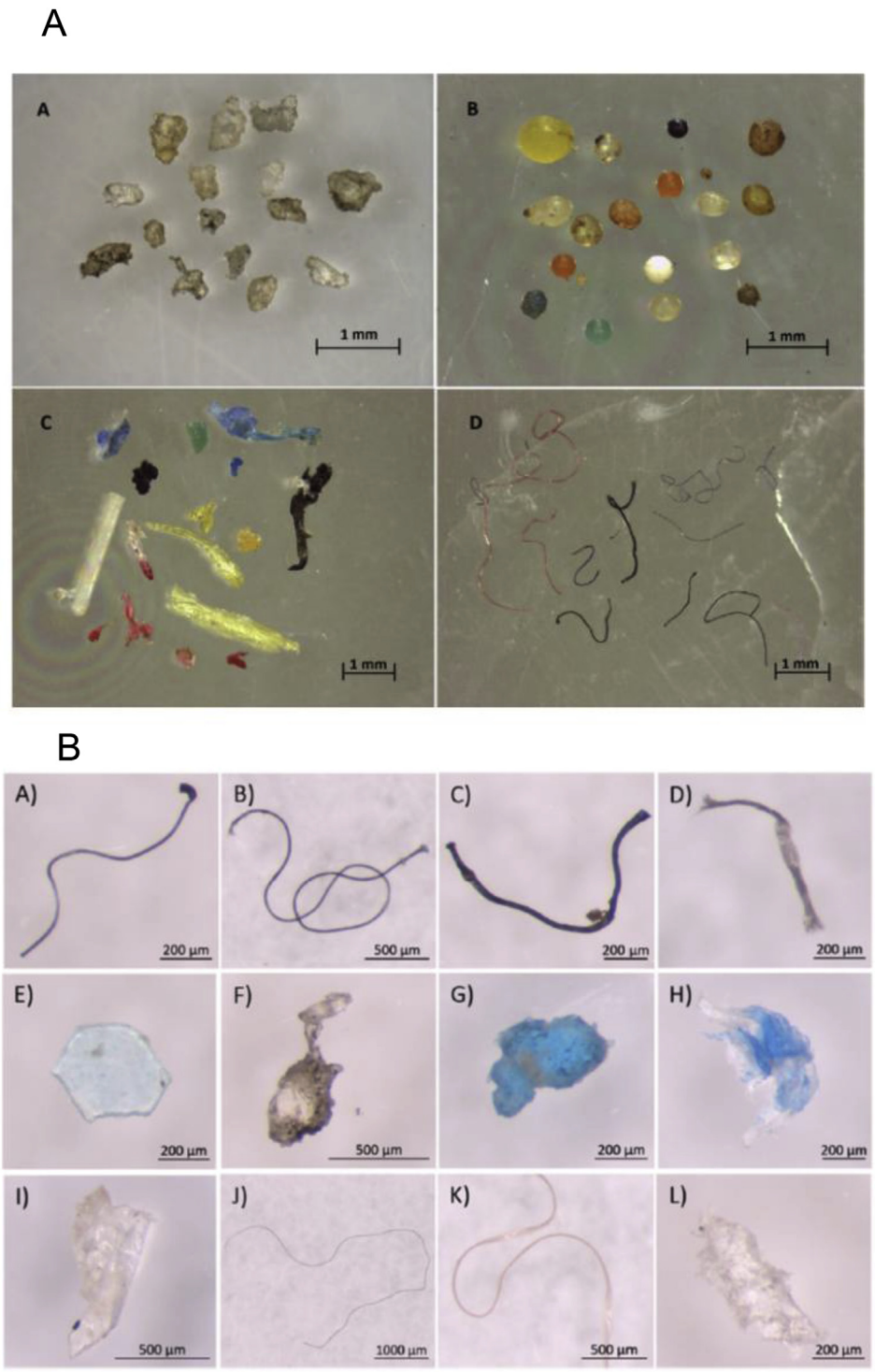

Fig. 4. A. primary microplastics (Microbeads derived from personal care products) and Secondary microplastics (Fragments from break-down of larger plastics and synthetic textile fibers) collected from WWTPs (Talvitie et al., 2017). B. Typical appearance of different polymers detected in different stages of the WWTP and recipient lake and identified by micro-FTIR and/or micro-Raman (Lares et al., 2018).

3.2. Microplastics properties in WWTPs

3.2.1. Materials

So far, over 30 kinds of microplastic polymers have been detected in influent and effluent of WWTPs (Table 2). The most common polymers found in influent and effluent of WWTPs were polyester (PES, ~28%–89%), polyethylene (PE, ~4%–51%), polyethylen terephthalat (PET, ~4%–35%) and polyamide (PA, ~3%–30%). The PES, PET and PA are all widely used in synthetic clothes, while PE are used in personal care products, including body and facial scrubs as well as food packaging films and water bottles (Cheung and Fok, 2017; Lares et al., 2018; Mintenig et al., 2017; Shah et al., 2008; Ziajahromi et al., 2017). Polymers like acrylate, alkyd, polypropylene (PP), polystyrene (PS), polyurethane (PU), PS

Table 1
Influent and effluent concentrations, daily discharges and removal ratios of microplastics in wastewater treatment plants with different treatment processes in different countries.

	Location	Treatment processes	WWTP number	Population equivalent	Efflux (m^3/y)	Sampling method	Detection method	Finest mesh	Influent (P/L)	Effluent (P/L)	Discharge (P/day)	Removal (%)	Ref.
1	Australia	Primary	1	1.2×10^6	1.12×10^8	Pump	Visual/FTIR	25 μm		1.5	4.60×10^8		(1)
2	Sweden	Primary, Secondary	1	1.2×10^4	1.88×10^6	Container/Pump	Visual/FTIR	300 μm	15.1	0.00825	4.25×10^4	99.9	(2)
3	France	Primary, Secondary (Biofilter)	1	8.0×10^5	8.76×10^7	Autosampler	Visual	100 μm	293	35	8.40×10^9	88.1	(3)
4	United States	Primary, Secondary	12	3.5×10^3 -5.6×10^7	8.58×10^5 -1.40×10^8	Pump	Visual	125 μm		0.004 −0.195	5.28×10^4 -1.49×10^7		(4)[a]
5	United States	Primary, Secondary	1		5.51×10^8	Pump	Visual/FTIR	100 μm	1	8.8×10^{-4}	9.30×10^5	99.9	(5)
6	Scotland	Primary, Secondary	1	6.5×10^5	9.52×10^7	Container	Visual	65 μm	15.7	0.25	6.52×10^7	98.4	(6)
7	Netherlands	Primary, Secondary	7	1.3×10^4	3.37×10^6 -2.63×10^8	Container	Visual	0.7 μm	68−910	55−81	7.48×10^8 -4.32×10^{10}	11−94	(7)
8	United States	Primary, Secondary	1	6.8×10^5	7.89×10^7	Container	Visual/FTIR	125 μm		0.023	4.97×10^6		(8)
9	Germany	Primary, Secondary	8	7.0×10^3 -4.6×10^4	1.90×10^5 -1.40×10^6	Pump	FTIR	10 μm		0.08−7.52	4.19×10^4 -1.24×10^7		(9)
10	Australia	Primary, Secondary	1	6.7×10^4	6.21×10^6	Pump	Visual/FTIR	25 μm		0.4	8.16×10^6		(1)
11	United States	Primary, Secondary	1	2.4×10^6	9.13×10^8	Container	Visual	20 μm	133	5.9	1.48×10^{10}	95.6	(10)[b]
12	Damark	Primary, Secondary	9			Autosampler, Container	FTIR	10 μm	2223 −10044	29−447			(11)
13	Finland	Primary, Secondary	1		3.65×10^6	Container	Visual/FTIR/Raman	250 μm	57.6	1	1.00×10^7	98.3	(12)[a]
14	Germany	Primary, Secondary	1		1.13×10^4	Container	Visual	40 μm		80.4	2.47×10^6		(13)
15	Netherlands	Primary/MBR	1		2.03×10^6	Container	Visual	0.7 μm	68	51	2.83×10^8	25.0	(7)
16	United States	Primary/AnMBR	1	pilot		Container	Visual	20 μm	91	0.5		99.4	(10)[b]
17	Finland	Primary/MBR	1	pilot	1.10×10^3	Container	Visual/FTIR/Raman	250 μm	57.6	0.4		99.3	(12)
18	United states	Primary, Secondary, Tertiary (GF,BAF)	5	1.2×10^4 -2.5×10^5	4.75×10^6 -7.77×10^7	Pump	Visual	125 μm		0.009 −0.127	1.01×10^5 -9.63×10^6		(4)[a]
19	United states	Primary, Secondary, Tertiary (Gravity Filter)	3		1.30×10^7 -3.13×10^7	Surface filtration	Visual/FTIR	40 μm/125 μm		0 -2.43×10^{-6}	0-2.08×10^2		(5)
20	Germany	Primary, Secondary, Tertiary (Post-Filtration, microplasticsD)	4	1.1×10^4 -2.1×10^5	1.30×10^7	Pump	FTIR	10 μm		0.01−0.38	2.79×10^5 -2.62×10^6		(13)
21	Australia	Primary, Secondary, Tertiary, RO	2	1.5×10^4	4.75×10^6 -1.75×10^7	Pump	Visual/FTIR	25 μm		0.21−0.28	3.60×10^6 -1.00×10^7		(1)
22	United states	Primary, Secondary, Tertiary (GF)	1	9.9×10^3	6.23×10^5	Container	Visual	20 μm	91	2.6	4.43×10^6	97.2	(10)
23	Finland	Primary, Secondary, Tertiary (BAF)	1	8.0×10^5	9.86×10^7	Pump	Visual	20 μm	610	13.5	3.65×10^9	97.8	(14)[c]
24	Finland	Primary, Secondary, Tertiary (BAF, DF,MBR,DAF,RSF)	6	Pilot-8.0×10^5	8.03×10^2 -8.82×10^7	Pump	Visual/FTIR	20 μm		0.02−0.3	1.26×10^6 -6.59×10^7		(15)
25	Damark	Primary, Secondary, Tertiary (RSF)	1			Autosampler /container	FTIR	10 μm	8149	19			(11)

Treatment processes: Secondary treatment: conventional activated sludge process expected for where specified; MBR: membrane bioreactor; AnMBR: anaerobic membrane bioreactor; GF: granular filter; BAF: biological aerated filter; microplasticsD: maturation pond; RO: reverse osmosis. DF: discfilters; DAF: dissolved air flotation; RSF: rapid sand filter.
Detection Method: Visual: Visual observation; TFIR; Fourier transform infrared spectroscopy; Raman:Raman spectroscopy; P/L: particle/L; P/day: particle/Day.
Reference: (1) Ziajahromi et al. (2017); (2) Magnusson and Norén (2014); (3) Dris et al. (2015); (4) Mason et al. (2016); (5) Carr et al. (2016); (6) Murphy et al. (2016); (7) Leslie et al. (2017); (8) Dyachenko et al. (2017); (9) Mintenig et al. (2017); (10) Michielssen et al. (2016); (11) Simon et al. (2018); (12) Lares et al. (2018); (13) Dubaish and Liebezeit (2013); (14) Talvitie et al. (2016); (15) Talvitie et al. (2017).
[a] Concentration include all textile fiber.
[b] Concentration include microplastics and some other small anthropogenic litter.
[c] Concentration include microplastics and some other microliter.

Table 2
Polymers detected in wastewater treatment plants with their densities and relative abundances.

No	Polymer	Abbreviation	Density g/cm^3[a]	Relative abundance [b]
1	Acrylic	–	1.09–1.20	++
2	alkyd	–	1.24–2.01	++
3	polyethylen terephthalat	PET	0.96–1.45	+++
4	polyamide (nylon)	PA	1.02–1.16	+++
5	polyaryl ether	PAE	1.14	+
6	polyester	PES/PEST	1.24–2.3	+++
7	polyethylene	PE	0.89–0.98	+++
8	polypropylene	PP	0.83–0.92	++
9	polystyrene	PS	1.04–1.1	++
10	polyurethane	PU/PUR	1.2	++
11	Polyvinyl fluoride	PVF	1.7	+
12	polyvinyl acetate	PVAC	1.19	+
13	polyvinyl chloride	PVC	1.16–1.58	+
14	Polytetrafluoroethylene	PTFE	2.1–2.3	+
15	styrene acrylonitrile	SAN	1.08	+
16	ethylene vinyl acetate	EVA	0.92–0.95	+
17	polyvinyl alcohol	PVAL	1.19–1.31	++
18	acrylonitrile butadiene styrene	ABS	1.04–1.06	+
19	polylactide	PLA	1.21–1.43	++
20	Vinyl-acetate-acrylic copolymer	–	1.22	+
21	Polyethylene-Polypropylene copolymer	–	0.94	+
22	Poly(phthalimide)	–	1.10	+
23	polycarbonate	PC	1.2–1.22	+
24	Terpene resin	–	0.98	+
25	Plexar resin	–	0.92	+
26	Poly(oxymethylene)	POM	1.41	+
27	Polysulfone	PSU	1.24	+
28	Silicone	–	1.1–1.2	+
29	polystyrene acrylic	PS acrylic		++
30	polyvinyl acrylate	PV acrylate		+
31	polyvinyl ethelene	PVE		+

[a] Density: Based on Hidalgoruz et al. (2012), Duis and Coors (2016), Simon et al. (2018), Rosato et al. (2010).
[b] Relative abundance: Based on Mintenig et al. (2017), Murphy et al. (2016), Ziajahromi et al. (2017), Lares et al. (2018), Talvitie et al. (2016), Li et al. (2018c), Simon et al. (2018). + refers to Low abundance, ++ refers to medium abundance +++ refers to high abundance.

acrylic, polyvinyl alcohol (PVAL) and polylactide (PLA) were also observed in the wastewater with their highest abundance between about 5%~27%. The other polymers (Table 2) only represented a limited proportion of total microplastics in the wastewater, which were commonly less than 5%, and could even reach below 1% in some samples. Therefore, the research priority could be given to the most common polymers instead of all presented particles. The material information of microplastics detected in WWTPs suggested that a large part of microplastics in wastewater were originating from our daily life. However, the overall sources and routes of microplastics entering the WWTPs have not been fully understood yet. Such knowledge could help with the microplastics source control for preventing related pollution and should be comprehensively investigated in future.

3.2.2. Shapes

Shape is another important indicator used for microplatics classification. The shape of the microplastics can not only affect their removal efficiency in the WWTP, but also have impact on the interaction between microplastics with other contaminants or microorganisms in wastewater (McCormick et al., 2014; Wang et al., 2018). Simply, microplastics could be categorized into fiber (significantly longer than wide) and particles (similar width and length) (Gouveia et al., 2018; Talvitie et al., 2015). Some researchers also divided the category of particle into irregular shapes and spherical bead/pellet. A few studies further included shapes such as flake/flim (very thin particle), foam and chip into the classification (Magnusson K. and F. 2014; Mason et al., 2016; Murphy et al., 2016; Talvitie et al., 2016). The relative abundances of different shapes of microplastics observed in WWTPs are presented in Fig. 5A.

Fibers accounted for the highest proportion of the observed microplastics in the wastewater, with an average percentage of 52.7%. The presence of numerous fibers in the wastewater may be explained by that a large amount of fibers were released through domestic washing machine discharges (Browne et al., 2011; Napper and Thompson, 2016; Pirc et al., 2016). This result is in accordance with high abundance of polymers in the wastewater from manufacturing synthetic clothes (Table 2). Also, the high abundance of fiber in some samples could attribute to the difficulty of distinguish synthetic fibers from natural fibers, and some studies also included the natural fibers during the quantification. Studies showed that the natural fibers such as cotton and linen could account for more than a half of fibers in some wastewater samples (Talvitie et al., 2016). Thus, effectively differentiate and detect synthetic and natural fibers is essential for precisely quantify microplastics in the WWTP.

Irregular fragments form another usually observed shape of microplastics in the wastewater, which accounted for an average percentage of 28.8% (Fig. 5A). The irregular fragments could result from eroded plastic products for the daily use. Or they could be microplastics originating from personal care products, such as toothpaste (Carr et al., 2016). The microplastics in the shape of film, pellet and foam were also found in wastewater with their average abundance around 10% or below. The microplastic film and foam could be mainly sourced from the erosion of plastic bags and packing products, while pellet were mostly primary microplastics added to personal products.

3.2.3. Size and mass

Currently, there are two commonly used methods for the size classification. One is based on the microplastics retention on different sizes of sieves. However, as aforementioned, the accuracy

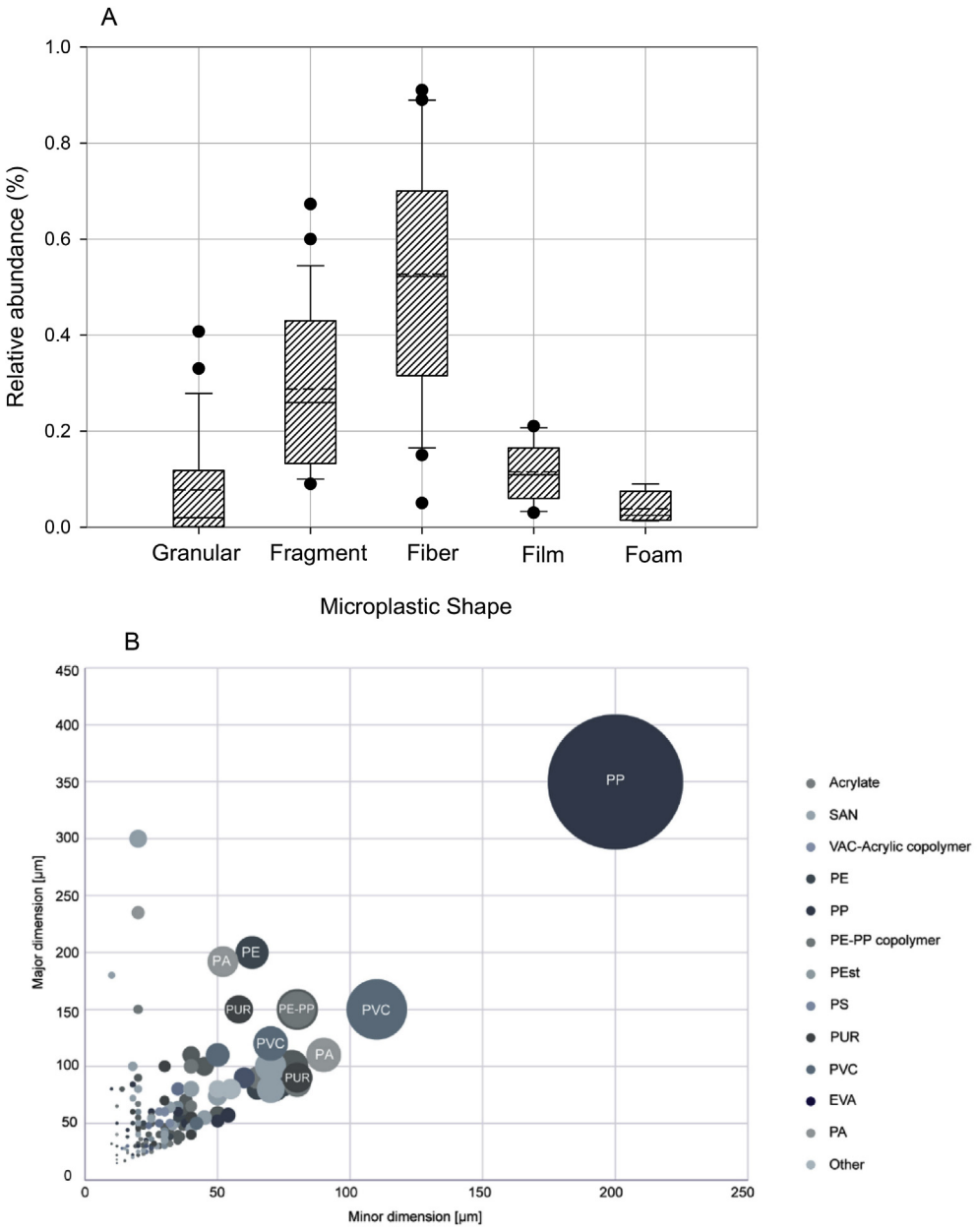

Fig. 5. A. Relative abundance of different shapes of microplastics in the wastewater. The median, 10th 25th 75th and 90th percentiles were ploted as vertical boxes with error bars. The dots indicate outer liners and the dashed lines refer to the average. Based on: Dyachenko et al. (2017), Lares et al. (2018), Mason et al. (2016), Michielssen et al. (2016), Murphy et al. (2016), Talvitie et al. (2016), Ziajahromi et al. (2017). B. The major dimension of particles of each polymer type from raw wastewater plotted against their minor measured dimension. The size of the data points represents the calculated mass of the particles. The polymer type named "Other" includes poly(phthalimide), polycarbonate, terpene resin, Plexar resin and poly(oxymethylene) particles which were sparse throughout the samples (Simon et al., 2018).

of this method is problematic due to irregular shapes of microplastics. The other method is to use microscopic imaging techniques (Lares et al., 2018; Mintenig et al., 2017; Simon et al., 2018). However, describing the size of microplastics with only one number can be insufficient due to the irregular shapes. Therefore, standard parameters applied in colloid science were suggested to be used to obtain reliable and comparable data about the actual size of microplastic particles in a normalized manner (Filella, 2015).

The dimensions of 25 µm, 100 µm and 500 µm were most frequently used for size classification (Dris et al., 2015; Lares et al., 2018; Mintenig et al., 2017; Simon et al., 2018; Talvitie et al., 2016; Ziajahromi et al., 2017). In the influent of the WWTP, the number of microplastics over 500 µm could sometimes reach over 70% (Dris et al., 2015; Lares et al., 2018). While in the effluent, on average, over 90% of microplastics were smaller than 500 µm and in some of the samples, around 60% of microplastics were smaller than 100 µm, (Mintenig et al., 2017; Simon et al., 2018; Ziajahromi et al., 2017). However, the size distribution of the microplastics could be affected by the mesh size used for sample collection, with a large mesh size being likely to miss out most of small particles (Lares et al., 2018). A most recent study showed that microplastics <25 µm had a significant abundance in the wastewater (Simon et al., 2018). This result is in accordance with the observation in the Atlantic Ocean that microplastics under 40 µm in size accounted for 64% of all detected microplastic particles, among which more than half were under 20 µm in size (Enders et al., 2015).

Therefore, the tiny microplastics in WWTPs are worthy to be investigated in future study to fully understand their removal and routes in aquatic environments.

The mass of the microplastics in the wastewater has not been paid much attention in the past. Most recently, Simon et al. (2018) quantified the mass of microplastics in the wastewater based on both the major and minor dimension of the particle. They found that although PP particles were not the most abundant, they contributed the most to the total mass of microplastics (Fig. 5B). This work highlighted the importance of describing microplastics concentrations not only by particle number but by plastic mass as well. However, the method they used just allowed the rough estimation of the microplastics mass only. Future studies could be working on increasing the accuracy of the mass quantification and thermoanalytical techniques might have this potential (Dümichen et al. 2015, 2017; Fischer and Scholz-Böttcher, 2017). Although the thermoanalytical techniques will destroy the microplastic samples, the accurate determination of microplastic mass can be complementary to common, particle-related characterization for better understanding the extent of MP pollution. Especially, together with standardized sample collection and pretreatment methods, applying mass as the conserved base of MP quantification allows consistent comparison with their sources and occurrences, contributing to a differentiated understanding of microplastics behavior in the environment.

4. Removal and retention of microplastics in WWTPs

4.1. Removal of microplastics in WWTPs

The removal of the microplastics by the WWTP was calculated based on their concentrations in both the influent and effluent (Table 1). Except for the study reported by (Leslie et al., 2017), the overall microplastics removal efficiencies of WWTPs without tertiary treatment were above 88% and the number increased to over 97% in the WWTPs with tertiary treatment. The relatively low removal efficiency reported by Leslie et al. (2017) was possibly because they collected only 2 L of wastewater for microplastics detection, while in most of other studies, tens to thousands liters of wastewater samples were collected for analysis. However, it could also be caused by the decreasing performance of some reactors, such as membrane reactors, as suggested in this study. Fig. 6 shows the estimated particle flow of microplastics based on literature reported value ranges, indicating the removal efficiency of microplastics during preliminary, primary, secondary and tertiary treatment, respectively.

4.1.1. Preliminary and primary treatment

The preliminary and primary treatment (pre-treatment) could effectively remove the majority of microplastics in the wastewater. It was reported that approximately 35%~59% of the microplastics could be removed during the preliminary treatment and 50%–98% of the microplastics were removed after primary treatment (Fig. 6). The removal at this stage were mainly achieved via skimming the light floating microplastics during the grease skimming or surface skimming on primary clarifiers as well as settling of the heavy microplastics or microplastics trapped in solid flocs during grit removal and gravity separation in primary clarifiers.

The pre-treatment had the largest impact on microplastics size distribution, as it could effectively remove microplastics of larger size. Dris et al. (2015) found that the fraction of large particles (1000 μm–5000 μm) drastically decreased from 45% to 7% after the primary treatment. In terms of microplastics shapes, studies showed that the pre-treatment could more effectively remove fibers than fragments from the wastewater, with the relative abundance of fibers decreasing after the pre-treatment (Magnusson and Norén, 2014; Talvitie et al., 2015; Ziajahromi et al., 2017). This could probably be due to fibers being more easily entrapped in flocculating particles and separated by sedimentation. Also, Murphy et al. (2016) found that microbeads were effectively removed by skimming, since the majority of these microbeads were made of PE, which were positively buoyant in water and were likely to sit on the surface of the wastewater or fat, grease and oil particles where they could be easily skimmed off. This result was consistent with other studies conducted by Michielssen et al. (2016) and Sutton et al. (2016), both studies found that microbeads were absent in the effluent of the WWTPs. In contrast, a survey conducted in the WWTPs in New York, United States, showed that 4 out of 10 WWTPs still release microbeads (Schneiderman, 2015). This difference might be due to the different amount of fat, grease and oil in the wastewater, since these compounds could be positive for microplastics being skimmed off.

4.1.2. Secondary treatment

The secondary treatment (usually comprise of biological treatment and clarification) managed to further decrease the microplastics in the wastewater to 0.2%–14% (Fig. 6). During this state, sludge flocs or bacterial extracellular polymers in the aeration tank are likely to aid the accumulation of the remained plastic debris, which would then being settled in the secondary clarification tank.

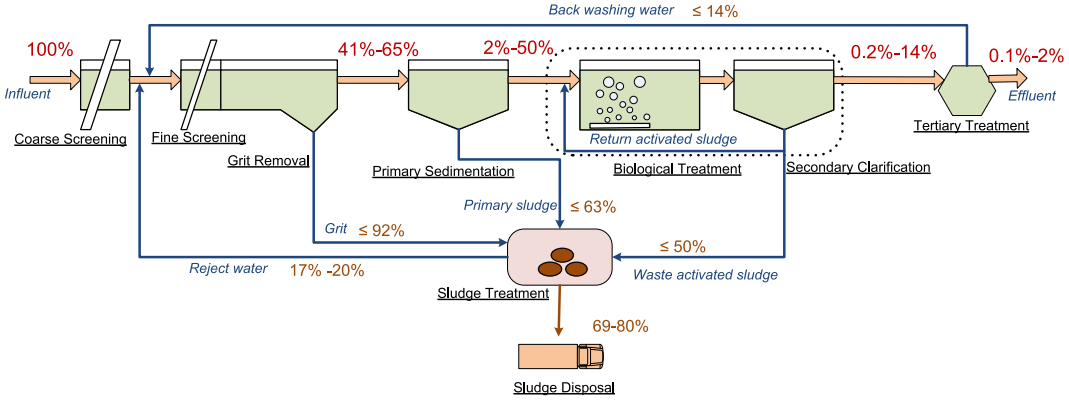

Fig. 6. Estimated microplastics particle flow in wastewater treatment plant with primary, secondary and tertiary treatment processes. The particle flow in liquid phase was summarized based on reported data (Dris et al., 2015; Michielssen et al., 2016; Murphy et al., 2016; Talvitie et al., 2015), (Lares et al., 2018; Talvitie et al., 2016). The particle flow in sludge phase was estimated according to the particle balance.

Also, microplastics might be trapped into sludge flocs due to the ingestion of protozoa or metazoan (Jeong et al., 2016; Scherer et al., 2018). In addition, chemicals such as ferric sulfate or other flocculating agents used during the secondary treatment could pose positive effect on microplastics removal, as they could cause the suspended particulate matters to aggregate together forming a "floc" (Murphy et al., 2016). However, how exactly the microplastics interact with microbial or chemical flocs and to what extent it could help with microplastics removal was unclear yet. It is also likely that some microplastics could be trapped in unstable flocs and may not be settled in an efficient manner, which would lead to a dynamic redistribution of these particles in the aqueous phase and consequently escaping removal during the settling stage (Carr et al., 2016).

Another factor regarded important for the microplastics removal from the secondary discharges is the contact time of microplastics with wastewater in the treatment train. Carr et al. (2016) found that a longer contact time was associated with increased potential for surface biofilm coating on the microplastics. Such bio-coatings may act as wetting agents, modifying the surface properties or (Rummel et al., 2017) relative densities of the microplastics. Such changes could measurably impact removal efficiency of microplastics, as neutrally buoyant particles are more likely to escape from both skimming and settling processes. Therefore, the impact of contact time as well as nutrient level in wastewater on microplastics surface fouling and microplastics removal efficiency may be an area worthy of further investigation. The experimental methods and mathematical models applied to study the biofilm formation on microplastics and the effect on particle transport in freshwater and marine environment offer good references for carrying out such investigation in wastewater system (Besseling et al., 2017; Fazey and Ryan, 2016; Rummel et al., 2017).

Different from the pre-treatment, the secondary treatment removed more fragment particles than fibers. This was supported by studies showing that the relative abundance of microplastics fragments decreased while that of fibers increased after the secondary treatment (Talvitie et al. 2015, 2016; Ziajahromi et al., 2017). One possible reason is that the easily settled or skimmed fibers had already been largely removed during the pre-treatment, whereas the remains might have some nature, such as neutral buoyancy, which was resistant to be further removed.

In terms of sizes, large microplastics particles can be further removed during the secondary treatment, resulting in relatively low abundance in the secondary effluent. Studies showed that microplastics with size larger than 500 μm were almost absent from the secondary effluent (Mintenig et al., 2017; Ziajahromi et al., 2017). Talvitie et al. (2016) found that microparticles >300 μm only account for 8% after secondary treatment. In contrast, Dris et al. (2015) found that microplastics within the size range of 500 μm–1000 μm still accounted for 43% after secondary treatment. The reason for this high proportion was unclear. It might be related to specific microplastic removal efficiency achieved by various secondary treatment processes with different operational conditions, which has to be further investigated in future.

4.1.3. Tertiary treatment

The tertiary treatment may provide substantial additional polishing on microplastics removal. Overall, the microplastics in the wastewater further decreased to 0.2%–2% relative to the influent after the tertiary treatment. The microplastics removal efficiency depends on the treatment processes applied, with the membrane related technologies showing the best performance. Talvitie et al. (2017) compared the removal efficiency of different tertiary treatment processes, i.e., discfilter (DF), rapid sand filtration (RSF) and dissolved air flotation (DAF) treating secondary effluent, as well as membrane bioreactor (MBR) treating primary effluent. They found that MBR obtained the highest removal efficiency (99.9%), which was followed by RSF and DAF, with the removal efficiency of 97% and 95% respectively. The removal efficiency of DF varied from 40% to 98.5%. Similarly, in the aforementioned survey conducted in WWTPs in New York, two plants with membrane filters did not release microbeads while the other four with other advanced filter did (i.e. a rapid sand filter, a continuous backwash filter and two filters with unspecified type) (Schneiderman, 2015). Also, Ziajahromi et al. (2017) observed that microplastics concentration significantly reduced after ultrafiltration and reverse osmosis. On the other hand, it was found that the granular advanced filtration did not reduced microplastics discharges from WWTPs (Mason et al., 2016). Additionally, the biological active filter (BAF) and maturation pond did not significantly impact the microplastics discharge as measured in WWTPs in Netherlands and Germany (Mintenig et al., 2017; Talvitie et al., 2016). It is worthwhile to note that the microplastics concentration in both influent and effluent of the tertiary treatment unit could be very low (<1 particle/L in most of the cases), and as a result, limited sample volume (dozens of liters) might give false zero results. Therefore, larger sampling volumes are required for the reliable assessment of microplastics removal efficiency by the tertiary treatment processes than that require for assessing the pre-treatment and secondary treatment processes.

After the tertiary treatment, the smallest sizes fraction (20–100 μm and 100–190 μm) were found to be the most abundant (Ziajahromi et al., 2017). Also, the relative abundance of fiber might be increased in the final effluent in some cases compared to the secondary effluent. This could probably be due to that fibres could more easily pass filter or membrane through longitudinally. Therefore, this highlights the need for final stage technologies to remove particularly small size and fiber-like microplastics from the effluent. Solids removed through backwashing of filters are typically sent back to the beginning of the WWTP. Hence, the microplastics retained by tertiary treatment may not be removed from the WWTP and could be added to the microplastics loading of the WWTP (Michielssen et al., 2016). With the increasing of the contact time, this part of microplastics might be removed through pre-treatment or secondary treatment. However, the return of microplastics in the wastewater stream might also increase the possibility for escaping from the treatment processes.

4.2. Retention of microplastics in sewage sludge

The relatively high removal efficiency of microplastics by WWTPs (Table 1) indicated that most of microplastics were retained in the sewage sludge. However, so far, studies focusing on microplastics in the sewage sludge including their occurrence, transformation and further mobilization are very limited. Table 3 listed the reported abundance of microplastics in the sludge samples obtained from WWTPs. The microplastics concentration in the sludge varied from ~400 to 7000 particle/kg WW (wet weight). This concentration was substantially higher than that in the liquid phase in WWTPs. If measuring based on the dry weight of the sludge, the concentration could reach ~1500–170000 particle/kg DW. The large variation of microplastics concentration in sludge also emphasizes the importance of representative sampling and harmonized detection method in future studies.

The size of microplastics in the sewage sludge was found significantly different from that in the wastewater as reported by Murphy et al. (2016). Their results showed that the average size of microplastics obtained from sludge was relatively larger than that obtained from wastewater, including influent after coarse

Table 3
Reported microplastics concentrations in sewage sludge.

No.	Location	Sampling point	Smallest mesh size	Concentration particle/kg	Reference
1	Sweden	Slightly dewatered sludge	300 μm	720 ± 112	Magnusson and Norén (2014)
2	United States	Returned activated sludge	N.a.	50	Carr et al. (2016)
		Primary tank skimming		4000–5000	
		Biosolid		1000	
3	Scotland	Grit	65 μm	1440	Murphy et al. (2016)
		Grease skimming		7868	
		Sludge cake		1200	
4	Netherlands	Sludge	0.7 μm	660–760	Leslie et al. (2017)
6	Finland	Activated sludge	250 μm	23000 (DW)	Lares et al. (2018)
		Digested sludge		170900 (DW)	
		MBR sludge		27300 (DW)	
7	China	Sewage sludge	37 μm	1565-56386 (DW)	Li et al. (2018c)
8	Ireland	Sewage sludge	45 μm	4196-15385 (DW)	Mahon et al. (2017)
9	Germany	Sewage sludge	10 μm	1000-24000 (DW)	Mintenig et al. (2017)

Note: Note: DW means that the concentration was presented based on dry weight of the sludge.
N.a: Not applicable.

screening, grits and grease effluent, primary effluent and final effluent. This supports that larger microplastics are easier to be removed by WWTPs than the smaller ones. In terms of the shape, most recently, Li et al. (2018c) examined the microplastics in the sludge from 28 WWTPs in China and found that 63% of microplastics in the sludge were fibers. Also, Lares et al. (2018) found that fibers in the sludge sample in Finland could reached more than 80%. Actually, synthetic fibers have been proposed as an indicator of land application of the sludge (Zubris and Richards, 2005). It was found that fibers were detectable in soil column over 5 years after application and they were detectable in field site soil up to 15 years after application. Thus, the effect of accumulation of microplastics fibers and other particles on the soil applied with sewage sludge should not be ignored. This also accounts for emission to surface waters due to run-off from fields where sludge has been applied.

A few studies also measured microplastics concentrations in the sludge generated at different stages for wastewater treatment. Studies conducted by both Carr. et al. (2016) and Murphy et al. (2016) found that sludge generated by the skimming unit contained the highest microplastics abundances (4000–7000/kg WW) which was about 5–10 times higher than that in the grits and biosolids. This further supports that early stage skimming of floating solids is a very efficient removal mode for microplastics. In addition, the microplastics in the digested sludge was about 5 times higher than that in the activated sludge and MBR sludge as reported by Lares et al. (2018).

So far, only one study has investigated effect of different sludge treatment processes on the microplastics characteristic in the sludge (Mahon et al., 2017). It was found that significantly higher abundance of microplastics in smaller size class was observed in the sludge samples received in lime stabilization. This indicating that lime stabilization may shear the microplastics particles, which could be a result of elevated pH, temperature and mechanical mixing during the treatment. Melting and blistering of the microplastics was observed in thermally dried sludge.

Overall, the majority of the microplastics in WWTPs go into the sewage sludge at different treatment units. Talvitie et al. (2016) calculated that 20% of the microliter (including microplastics) in the sewage sludge was returned to the wastewater flow via reject water while the remained 80% ended up in the dry sludge for disposal. Based on the total sludge production, it was estimated that approximately 4.6×10^8 microplastic particles were discharged daily from the a WWTP with a capacity of 10000 m^3/day in Finland and the average amount of sludge-based microplastics entering into natural environmental was estimated to be 1.56×10^{14} particles per year in China (Lares et al., 2018; Li et al., 2018c). Microplastics in the sludge may be burnt during the sludge incineration. However, these microplastics could also be released into the terrestrial system with the sludge land application, exacerbating the problem of land-spread microplastics pollution. For example, the surface weathering of the microplastics would result in the attachment of organic pollutants and heavy metals, which could be transported with microplastics in the soil and to waterbodies nearby (Peyton et al., 2016; Turner and Holmes, 2015). It is also likely that microplastics would have potential adverse effects on soil living organisms, as they could ingest microplastics, thus leading to accumulation in the soil detrital food web (Rillig, 2012). Therefore, knowledge gaps regarding the mobilization and transport of microplastics which are likely to affect the pathway of land-spread sewage sludge microplastics pollution should be addressed to evaluate and prevent the associated risks.

4.3. Microplastics-targeted treatment technology

Though microplastics in the wastewater could be removed through skimming, sedimentation and tertiary filtration in WWTPs, none of these processes are originally designed for microplastics removal. As a result, still a significant amount of microplastics in WWTPs could escape with the effluent and enter into the receiving water system (Table 1). Also, as most microplastics in WWTPs are retained in sewage sludge, more microplastics could be released to the environment through sludge land application compared with those released through direct wastewater discharge. However, no specific treatment process aimed at microplastics removal has been applied in any full-scale WWTP yet and the microplastics-targeted treatment technology is still at the preliminary research stage.

Beljanski et al. (2016) designed a gravity-powered filtration system aimed at removing microplastics in the secondary effluent in WWTPs. Similar to most rapid filtration units, the system could be operated in two modes, i.e. filtration mode to filter microplastics out of wastewater and backflush mode to wash the microplastics out of the system. The effect of filtration materials, water pressures on the flow rate and microplastics recovery of the system was tested, with a 3D filter and a lower water pressure (1.68 kPa) showing the best result. However, the system was only tested with an artificial microplastics-water solution and its efficiency on real wastewater was not evaluated. More recently, Li et al. (2018b) evaluated the feasibility of micro-particles removal by dynamic membranes and they suggested that this technology could be further developed to remove microplastics in an energy efficient way. However, the construction and operational cost should be

fully evaluated when utilizing an extra unit for microplastics removal.

An alternative economical solution for developing microplatics-targeted treatment technology is to adjust the relevant operational parameters of current wastewater treatment processes to improve microplastics removal efficiency. For example, as current skimming and sedimentation units have proven their ability of microplastics removal to some extent, it is worth investigating the effect of the operational conditions, such as hydraulic retention time (HRT), on microplastics removal in future for improving the removal efficiency. Also, the pore sizes, materials and flux loading of different filtration and membrane systems with their correlation to microplastics removal are worthy to be evaluated. In addition, the enhancement in flocculation/coagulation could also play an important role in microplastics removal. Ma et al. (2019) observed that Al-based coagulant showed better microplastics removal efficiency than Fe-based coagulant and PAM played an important role in increasing microplastics removal efficiency in drinking water systems. These results imply the possibility of improving microplastics removal from wastewater by optimizing flocculation/coagulation processes, which required further investigation.

The treatment for microplasitcs in sewage sludge can be very different from that in wastewater, as it is not easy to separate microplasitcs from sludge. One optimal solution is to improve the microplastics removal through grease removal stage and to treat the grease separately for preventing large number of microplastics entering the waste sludge. On the other hand, pyrolysis techniques, including thermal pyrolysis, catalytic pyrolysis and microwave-assisted pyrolysis have been applied to treat the plastic waste, which could decompose the long chain polymer in to oligomers (Juliastuti et al., 2018; Undri et al., 2014). As the calorific value of the plastics is comparable to that of hydrocarbon fuel, this process could convert plastics into fuels in a relative low cost (Wong et al., 2015). So far, this method has just applied on the plastic waste with relatively large volume. However, the recent development of co-pyrolysis with biomass may provide a solution for treating microplastics-containing sewage sludge (Burra and Gupta, 2018; Jin et al., 2019).

In addition, since a significant part of microplastics are originated from our daily life, future efforts could also be dedicated to developing household-scale microplastics treatment technology, which might help to prevent microplastics pollution at the source. For example, it has been widely recognized that the effluent of washing machines contains a high amount of microplastics in the form of fiber (Hernandez et al., 2017; Napper and Thompson, 2016). If techniques could be developed to reduce fibers releasing from washing machine or fibers in this kind of wastewater could be separated at the household, the overall microplastics in the wastewater are expected to be significantly reduced. Also, implementing regulation of banning plastic microbeads in more regions could serve as an important complementary action for source control (Cheung and Fok, 2017).

5. Conclusions and perspectives

Wastewater treatment plants (WWTPs) are regarded as an important pathway of microplastics entering natural aquatic systems. In this work, the detection, occurrence and removal of microplastics in WWTPs are comprehensive reviewed. The key conclusions are:

(1) Methods for sampling and detection of microplastics in wastewater treatment plants vary greatly among studies, which results in difficulties in comparing results between studies. Sample collection through separate pumping and filtration can effectively increase the sampling volume with less practical limitation. So far, Micro-FTIR or Raman techniques might offer the best options to characterize the microplastics in complex samples, as they provide comprehensive information about the tested particle including number, size and chemical composition.

(2) Microplastics are generally detected in both influent and effluent of WWTPs, with reported influent concentrations varying from 1 to 10044 particle/L and effluent concentrations in the range of 0–447 particle/L. The most common polymers detected in WWTPs are polyester, polyethylene, polyethylen terephthalate and polyamide, with fibers accounting for the largest fraction of the observed microplastics in the classification of different shapes. Despite of relatively low concentration of microplastics in the effluent of WWTPs, the total discharge of the microplastics from the WWTPs still has a reported median value of 2×10^6 particle/day, corresponding to an average annual efflux of 5×10^7 m^3/year.

(3) Microplastics are effectively removed in WWTPs. Especially in the grease removal stage a high number is removed. This might be a target for further optimization of microplastic removal, potentially preventing large numbers of microplastics in the waste sludge if the grease is separately treated. As can be expected, membrane filtration technology is most efficient in reducing the microplastics in the final effluent.

(4) The removed load of microplastics is recovered in the final waste sludge. This might form an important route for environmental emissions when land application is practiced. Sludge burning can be an effective way to fully prevent microplastics from wastewater to enter in the environment.

Based on the current state of the knowledge related to microplastics in WWTPs, the following aspects should be addressed in the future research for better understanding and mitigating microplastics:

(1) Since the microplastics sampling and detection methods can significantly affect the result of its quantification and identification, it is urgent to come to harmonization. The method might focus on the major plastics observed in wastewater or try to consider all plastics.

(2) Current studies related to microplastics in WWTPs mainly investigated the microplastics > 20 μm. However, reports indicate that smaller microplastics have a high abundance in water environment and may have more severe biotoxicity as they could enter into the circulatory system of aquatic species. Therefore, it is worthwhile to include tiny microplastics (<20 μm) in future studies. Raman spectroscopy and thermoanalytical techniques might be options for analyzing these tiny microplastics, which require further exploration.

(3) As most microplastics end up in sewage sludge, future study should be carried out to investigate the potentially environmental impact of land application of sewage sludge.

(4) Microplastics-targeted treatment processes need to be developed for reducing amount of microplastic discharged from WWTPs and released from sewage sludge.

(5) Source control can provide an alternative solution for preventing microplastics pollution. Future efforts could be devoted to separate microplastics from wastewater at the household scales as well as improve plastic regulation.

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Appendix A. Supplementary data

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