Removal of microplastics via drinking water treatment: current knowledge and future directions

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Abstract

The occurrence of microplastics in drinking water systems has increasingly become a major environmental challenge. Although the potential impacts of microplastics in drinking water on humans are not yet fully understood, microplastics attract the public health concern when they are consumed by humans through drinking water systems. Current drinking water treatment plants constitute an obstacle to the entry of microplastics from raw water into daily drinking water. Therefore, understanding the behaviors of drinking water treatment process and the fates of microplastics in drinking water treatment plants are very important. We systematically reviewed the available knowledge on the global existence of microplastics in raw water, treated water and tap water in this paper. This will offer a new perspective for the threat of microplastics in drinking water to human health and help to formulate effective strategies for microplastic monitoring. The existing knowledge of microplastic removal by different treatment processes was also thoroughly discussed. Additionally, the potential challenges of microplastic removal from treatment processes and remediation strategies of microplastics in drinking water were also put forward. The relationship between the properties and behavior of microplastics during different treatment processes is suggested to explore in the future.

Keywords: Microplastics; Drinking water treatment; Removal technology; Environmental assessment; Remediation strategies

1. Introduction

Plastic was once praised as "the great invention of the 20th century", but now the harm caused by improper management of plastic products is increasingly recognized by the masses. Global plastic production has reached 359 million tons in 2018 (PlasticsEurope, 2019; Shen et al., 2020; Shen et al., 2019b), and up to 13 million tons of them were discharged into the oceans (Jambeck et al., 2015). It is expected that a total of 250 million tons of plastics will be discharged by 2025 (Jambeck et al., 2015). The use value of plastics may range from one to fifteen years, which depends on how they are used before being treated as plastic waste (PlasticsEurope., 2017). Plastics in the environment will gradually be decomposed to microplastics under various conditions (Auta et al., 2017; Shen et al., 2019a; Shen et al., 2019c). Microplastics usually refer to plastic particles with a size ranging from 0.1 μm – 5 mm (Anderson et al., 2017; Thompson et al., 2004; Wen et al., 2018), while plastic particles with a size less than 0.1 μm are defined as nanoplastics (Da Costa et al., 2016; Enfrin et al., 2019; Shen et al., 2019d). However, this classification of microplastics and nanoplastics has not yet formally recognized (Frias and Nash, 2019; Hartmann et al., 2019a; Hartmann et al., 2019b; Stark, 2019).

The presence of microplastics in water environment, such as rivers, lakes and oceans, has thoroughly been reviewed (Horton et al., 2017; Jiang, 2018; Mendoza et al., 2018). Freshwater (surface water and groundwater) is the main source of human drinking water (Yi et al., 2018). The open and closed freshwater systems can be used as microplastic pipelines and sink tanks. Evidences have shown that microplastics have been detected in freshwaters around the world (Anderson et al., 2017; Di and Wang, 2018; Eriksen et al., 2013; Wang et al., 2018b), even in remote regions (Free et al., 2014). The occurrence of microplastics in freshwater ranged from almost zero to several millions particles per cubic meter.

Drinking water is closely related to human health, therefore, it is suspected that microplastics from drinking water pose a potential risk to humans (Novotna et al., 2019). Despite limited information on human health risk of microplastics (Triebskorn et al., 2019), they should be considered as emerging pollutants in drinking water, at least in some ways. Neither governmental legislative standard restriction on the presence of microplastics nor any direct microplastic removal techniques in drinking water have been done. Recently, several techniques and methods, such as coagulation and membrane separation, have been tried to remove microplastics from drinking water (Ma et al., 2018; Ma et al., 2019). However, due to the limitations of technologies, there are still great challenges this aspect. Each removal process has its own advantages and disadvantages. Evidence showed that the concentrations of microplastics varied from zero to thousands particles per litre in drinking water worldwide (Novotna et al., 2019). As such, the presence of microplastics in drinking water and their removal cannot be overlooked.

Although a growing studies focus on microplastics in drinking water, there is a lack of corresponding connection between them. Understanding the origin of microplastics, the mechanisms of microplastic removal and the potential risks of microplastics in drinking water to human health may help to develop new strategies for monitoring and mitigating microplastic persistence in drinking water systems (Song et al., 2017). In this paper, the available data of microplastic occurrence in drinking water systems (tap water, treated distributed water, and bottled water) and main sources of microplastics in raw water were systematically and critically summarized with the purpose of establishing effective monitoring and mitigating strategies. The removal of microplastics by drinking water treatment processes and impact on subsequent processing were discussed. Remediation strategies of microplastics were examined by reviewing the different aspects including source control, enhancement of microplastic removal efficiency, and development of new methods for plastic removal. Lastly, the future directions are also put forward. This study can provide a guidance to find research needs and knowledge gaps in drinking water treatment and microplastics.

2. Sources, occurrence and impacts of microplastics in drinking water

2.1 Microplastic sources and occurrences in drinking water

Freshwater is the main raw water source for agricultural, industry, energy production and human consumption. Seawater is sometimes used, as only freshwater sources are scarce. However, seawater desalination treatment requires high energy and costs. Surface freshwater, including river, lake and reservoir water, and groundwater are the main raw sources for drinking water. These raw water sources are easily contaminated by agricultural and industrial activities, and animal farming discharges (**Fig. 1**). Microplastic has been detected in different surface waters. The average abundance of microplastics in freshwater environment ranges from several to millions tons (Pivokonsky et al., 2018). These great differences are mainly influenced by the locations, natural conditions, human activities, etc. **Table 1** summarizes the occurence and detection methods of microplastics in some freshwater environments around the world. Microplastics enter the surface water environment via the discharges of wastewater containing microplastics (washing wastewater and bath wastewater) (Chang, 2015; Hartline et al., 2016), decomposition of environmental plastic wastes (Lambert et al., 2014), and abrasions of plastic products (Duis and Coors, 2016), etc. Additionally, atmospheric deposition is also an important source of microplastics for aquatic environment (Free et al., 2014; Shao et al., 2019).

The drinking water treatment processes, coagulation, sedimentation and filtration, directly affect the removal of microplastics from raw water. Currently, few studies on investigating the presence of microplastics in drinking water were reported. A research performed by Kosuth et al. (2018) implied that microplastics were detected in approximately 81% of 159 global drinking water samples (156 tap water samples from 14 countries and 3 bottled water samples from USA). The concentrations of microplastics in water samples ranged from 0 – 62 particles L^{-1}, with an average number of 5.45 particles L^{-1}, and most of these polymers were fibers (about 98.3%), with a length from 0.1 – 5 mm. Pivokonsky et al. (2018) analyzed raw water and drinking water from three drinking water treatment plants in Czech Republic for studying the occurrence of microplastics. The plastic concentrations were in the range of 1648 – 2040 (mean = 1812), 1384 – 1575 (mean = 1473), and 3123 – 4464 (mean = 3605) particles L^{-1} for raw water, and 369 – 485 (mean = 338), 243 – 466 (mean = 443), and 562 – 684 (mean = 628) particles/L for treated water, respectively. By contrast, Mintenig et al. (2019) determined the presence of microplastics in groundwater and drinking water from Germany. The authors reported that only few polymer particles were observed in groundwater and drinking water (only 0 – 7 (mean = 0.7) particles m^{-3}). The difference among aforementioned researches may be because of different water source supplies. The former is surface freshwater and the latter is groundwater.

Consumption of bottled water is another possible way for microplastic exposure. Mason et al. (2018) have analyzed the occurrence of microplastics in bottled water. The 93% samples (259 bottles in total) exhibited sign of microplastic contamination. The concentrations varied from several to thousands, with an average number of 10.4 particles L^{-1}, and fragments were the most common form (66%), followed by fibers. Another research carried out by Oßmann et al. (2018) showed that the number of microplastics was measured to be 2649, 4889, and 6292 particles L^{-1} in the single use PET plastic bottles, reusable PET plastic bottles and glass bottles, respectively, and most particles (90%) were < 5 µm. In water from glass bottles, PE, PP and PET were also found. Possible contamination sources are the washing machinery or other steps during filling process. In addition, water from frequently reused PET bottles showed higher amounts of microplastics than water from newish PET bottles. This was possibly caused by aging of the bottle material. Contrarily, Schymanski et al. (2018) showed that microplastic content was 118 particles L^{-1} in returnable bottles, and only 11 and 14 particles L^{-1} in the beverage cartons and single use plastic bottles, respectively. This difference may be due to different water treatment processes and materials used in water treatment processes. It is worth noting that, actually, the bottles mostly are plastics, which may be a possible origin of microplastics in drinking water.

Concentrations of microplastics in drinking water from different countries or regions are illustrated in **Fig. 2**. According to **Fig. 2**, it can be found that the information on microplastics from different drinking water is rare and results are uneven. Limited information shows that global drinking water has been contaminated by microplastics. At the same time, problematically, we also note that sample collection methods, pretreatment methods and detection techniques used in the literature are not uniform, and individual methods may bring false positive results. There are great differences in the concentration of microplastics from drinking water.

2.2 Potential implications for humans

Drinking water is closely related to human health. Exposure and risk assessment includes the risk characteristics of microplastics to human beings through drinking water. According to the published data (**Fig. 2**), the greatest microplastic average concentrations in researches of bottled water (6292 particles L^{-1} in the single use PET plastic bottles) (Oßmann et al., 2018), treated water (628 particles L^{-1} from a plant) (Pivokonsky et al., 2018) and tap water (9.24 particles L^{-1} from a US water sample) (Kosuth et al., 2018) are considered to assess the potential risks for humans. Generally, women and men need to supplement more than 2.2 and 2.3 L of water daily to maintain their health. As such, at the worst case, women and men obtain a daily particle uptake rate of ~13842, ~1382 and ~21 particles, and ~14472,

~143, and ~22 particles, respectively. Once microplastics enter human body, there are potential risks for the health. Microplastics may induce toxic effects in the human body. The microplastics could produce oxidative stress and induce tissue damage and chronic inflammation (Schirinzi et al., 2017). Recently, a review done by Wright and Kelly (2017) investigated the potential impacts of microplastic uptake on human health through gastrointestinal tract absorption. The uptake and translocation of microplastics depend on many factors, and smaller particles translocate more effectively. It is easy for the larger plastics (> 2 μm) to stay in the intestinal tract. Although low concentration microplastics can enter the blood circulation, it is difficult to enter the deep tissue through the cell membrane due to its size limitation, and it is cleared by the spleen (Bouwmeester et al., 2015). As the size of microplastic decreases, it may enter peripheral tissues and circulatory system though lymphatic aggregation, leading to systemic exposure. However, for patients with intestinal diseases, the translocation efficiency of microplastics will be significantly increased due to the tissue permeability caused by inflammatory infections (Schmidt et al., 2013). In addition, the release of toxic pollutants induced by them is also affected by many factors, so it is not certain that toxic substances will be released in the human body. Despite recent sporadic studies on the interaction of microplastics with human cells, the harm of microplastics to the body has been demonstrated (Schirinzi et al., 2017; Triebskorn et al., 2019; Wang et al., 2018a). The research on the potential impact of microplastics even nanoplastics on human health has just begun, and toxic effects needed to be further investigated and confirmed.

3. Strategies for removing microplastics by drinking water treatment

Drinking water treatment plants are generally designed to ameliorate water quality to meet the standard for social consumption. The purpose of traditional drinking water treatment is to ensure the safety of drinking water to humans through removing physical, chemical and biological contaminants, such as suspended particles, heavy metal and microbes. Therefore, DWTPs play a vital role in preventing microplastics from transferring into the drinking water from raw water. However, at present, neither any direct removal techniques for microplastics nor governmental legislative standard limit for the presence of microplastics in drinking water has been performed. It is because different countries and regions have different standards for drinking water treatment (Kosuth et al., 2018). Several techniques and methods have been tried to remove microplastics from drinking water (Pivokonsky et al., 2018). Assessment of microplastic removal rate is from two aspects: (1) measurement of microplastic content from the influent and effluent of DWTPs or a specific technological step, and (2) research on the impact of different treatment processes on removal rate of microplastics from drinking water under specific conditions.

3.1 Traditional treatment processes

Traditional drinking water treatment mainly includes coagulation, sedimentation, sand filtration, and clarification. As particle matter, microplastics and suspended solid particulates have great similarities in physical properties. After filtration, the abundance of microplastics can be decreased (Pivokonsky et al., 2018). Currently, few studies are available on the removal of microplastics from drinking water by traditional treatment process. Pivokonsky et al. (2018) investigated the microplastic content (size up to less than 1 μm) in raw water and treated water from three DWTPs supplied by surface rivers and reservoirs in Czech Republic. For confidentiality reasons, the names of three DWTPs were not defined in the research, marked as DWTP1 – 3. The process of DWTP1 includes coagulation/flocculation and sand filtration, that of DWTP2 uses coagulation/flocculation, sedimentation, sand filtration and activated carbon filtration, and that of DWTP3 operates coagulation/flocculation, flotation, sand filtration and activated carbon filtration (Pivokonsky et al., 2018). The microplastic contents in treated water were significant decreased compared to the raw water. The removal efficiency of microplastics reached 70% for DWTP1, 81% for DWTP2, and 83% for DWTP3, respectively. However, DWTP1 operates a low microplastic fibre removal rate (only about 25%) compared to DWTP2 and DWTP3 (more than 80%). Furthermore, the removal efficiency by single treatment step, such as coagulation/flocculation, sedimentation, filtration, was not investigated in the paper. Mintenig et al. (2019) determined the presence of microplastics in drinking water from the whole drinking water supply chain. The drinking water from five DWTPs, treated by several filtrations and aerations, and one selected traditional household was analyzed. After treatment, the drinking water was directly supplied and transported to humans or stored in storage tanks. The microplastic content was found at very low level, only 0 – 7 particles/m^3. Problematically, the authors showed that the raw water does not contain microplastics, however, some were found in the treated water from household. Five types of polymers were found in the test, and four of which were identical with the plastics contained in purification and water conveying equipment. Therefore, the drinking water supply chain may also be a potential source of microplastic pollution in drinking water.

In addition, some studies on the removal of microplastics from drinking water by traditional treatment processes under laboratory conditions are also carried out (Ma et al., 2018; Ma et al., 2019). Polyethylene (PE) with different particle size (< 0.5 – 5 mm) was tested in experiments. Generally, coagulation and flocculation in drinking water treatment aim to combine dissolved or colloidal compounds to form larger aggregates suitable for subsequent removal. The needed size approximately ranged from dozens of micrometers (Pivokonsky et al., 2011). Chemicals found in water samples were the same as used for label printing, indicating the bottle cleaning process as possible contamination route

(Oßmann et al., 2018). It is doubtful whether agglomerated particles are needed in the millimeter range. As such, in this paper, the results of PE particle less than 500 μm were discussed. Ma et al. (2018) reported that the best PE removal efficiency (13.27% ± 2.19%) was achieved using $FeCl_3 \cdot 6H_2O$ as coagulant at approximately 112 mg L^{-1} Fe (2.0 mmol L^{-1}) at pH of 8. The dosage of coagulant is higher than that of treatment of other impurities in drinking water (less than 20 mg L^{-1} Fe) (Baresova et al., 2017; Gonzalez-Torres et al., 2014; Pivokonsky et al., 2015). When the doses of coagulant were used at real conditions (0.2 mmol L^{-1} Fe), the PE removal efficiency was only 6.71% ± 1.26%. In addition, Ma et al. (2019) determined the performance of $AlCl_3 \cdot 6H_2O$ and $FeCl_3 \cdot 6H_2O$ in PE removal, and found that aluminum salt coagulant ($AlCl_3 \cdot 6H_2O$) performs better. The PE removal efficiency could reach 36.89 ± 1.06% at 405 mg L^{-1} Al (15 mmol L^{-1}) at pH of 7, nevertheless, the needed doses of aluminum salt were very high in the experiment. At common doses, the PE removal efficiency was only to be 8%. Furthermore, the authors also examined the impact of adding flocculant polyacrylamide (PAM) on the coagulation by coagulants of PE microplastics. The results showed that the PE removal efficiency was determined to be 50 – 60% and 85 – 90%, respectively, at 5 mmol L^{-1} Al and 2.0 mmol L^{-1} Fe combined with 3 – 15 mg L^{-1} PAM at pH of 7 (Ma et al., 2018; Ma et al., 2019). But, the addition of PAM concentrations has far exceeded the maximum allowable level (not more than 1 mg L^{-1}) in drinking water treatment (World Health Organization, 2011). For further ultrafiltration, PE microplastics can be completely rejected by a polyvinylidene fluoride membrane (average pore size of 30 nm), sight membrane fouling was observed. Although coagulation and ultrafiltration applied in these researches have some shortcomings, these technologies have potential application prospects in drinking water treatment so as to eliminate microplastics from the lab scale to the industrial scale.

In general, traditional treatment processes are promising approaches for microplastic removal and are worth attention. For small microplastic particles (size up to less than 1 μm), the traditional treatment methods are more effective for microplastic removal from raw waters. Laboratory conditions should mirror the applicable conditions in drinking water treatment practices. Additionally, the microplastic particle size distribution in raw water and the interaction among other contaminants and microplastics are needed to explore to work out a better scheme for microplastic removal.

3.2 Electrocoagulation

Electrocoagulation provides a cheaper tertiary treatment process that does not rely on chemicals or microbes used in general chemical coagulation and conventional activated sludge processes. Electrocoagulation uses metal electrodes to electrically produce coagulants, thereby making the coagulation process simple and robust (Garcia-Segura et al., 2017; Moussa et al., 2017). Electrocoagulation is a complex process in which cations are produced by metal electrodes under the action of electric field. From the generation of ions and to formation of flocs, there are three successive stages (**Fig. 3A**): (1) Under action of electric field, electrons are generated in the anode to form "micro-coagulants", hydroxides of Fe^{3+} or Al^{3+}; (2) The suspended particles and colloidal pollutants in water lose their stability under the action of coagulants; and (3) After destabilization, pollutant particles and micro-coagulants collide with each other to form micro-flocs. Because the product of electrolytic reaction in the process of electrocoagulation is only ion, no oxidant or reductant is needed, and no or little pollution is produced to the environment, it is called an environmental friendly water treatment technique. Electrocoagulation has the advantages of environmental compatibility, easy automation, sludge minimization, energy efficiency and low capital cost (Zeboudji et al., 2013), and has been used to remove other pollutants in drinking water (Behbahani et al., 2011; Millar et al., 2014).

Recently, Perren et al. (2018) studied the performance of electrocoagulation to the removal of microplastics from wastewater under laboratory conditions. The effects of water characteristics, such as pH, current density and conductivity, and concentrations (0 − 0.1 g L^{-1}) and particle sizes (300 − 355 μm) of microplastics (PE) on removal efficiency were thoroughly studied. It was found that removal of microplastics from water by electrocoagulation is effective and the removal efficiencies of PE microplastics all exceeded 90%. The optimum removal efficiency was to be 99.24% observed under the condition of pH 7.5 (Perren et al., 2018). The results demonstrated that a higher neutral pH of water is more conductive to pollutant removal owing to the higher production of coagulants under this condition, and other researches also have reported this phenomenon (Yavuz and Ögütveren, 2018). At high current density, the reduction of the removal efficiency of PE microplastics is mainly due to the increase of energy consumption, but the improvement of the removal efficiency is not obvious. Water conductivity has no obvious impact on removal efficiency, and the removal efficiency increased with time and reached a steady state after 60 minutes (Perren et al., 2018). Additionally, the authors also showed that the operating cost of the electrocoagulation was measured to be 0.05 £ per m^3, which is lower than the operating cost of other pollutant treatment, such as iron (0.22 $ per m^3) (Hashim et al., 2017), dye (0.26 $ per m^3) (Dalvand et al., 2011), bleaching effluent (1.56 $ per m^3) (Sridhar et al., 2011), etc. It is a viable option to remove microplastics from drinking water. Although some limitations exist in this research, considering the removal efficiency of microplastics and operating costs, this technique is transferable and reproducible from laboratory to industry (Pico and Barcelo, 2019).

3.3 Magnetic extraction

Very recently, Grbic et al. (2019) studied the performance of magnetic extraction for microplastic removal from waters. Magnetic extraction is a separation technology which uses magnetic seeds and acid with external magnetic field to improve the separation speed. Fe nanoparticles were chosen as magnetic seeds in this research due to their low cost, high specific surface area and ferromagnetic properties. Fe nanoparticles were coated with hexadecyltrimethoxysilane to make them hydrophobic, thereby allowing the isolation of microplastics from water by magnetic extraction (Grbic et al., 2019). Three size ranges of microplastics, large (1 – 8 mm), medium (200 μm – 1 mm) and small (< 20 μm), were tested in experiments. **Fig. 3B** illustrates the preparation of coated Fe nanoparticles and the removal of microplastics by magnetic extraction. It was found that the recoveries of medium microplastics (200 μm–1 mm) were measured to be 84% and 78% from freshwater and sediment, respectively. For small microplastics (< 20 μm), 92% of PE and PS microplastics were recovered from seawater. The results showed that magnetic extraction has better removal of small microplastics. For sediments, the recovery was low because soil particles prevent Fe nanoparticles from encountering microplastics. Moreover, if there are lipophilic substances or biota in sediment samples, the nonspecific binding of nanoparticles will significantly reduce the effect. Therefore, the authors reported that this method could be better suitable for drinking water treatment (Grbic et al., 2019).

3.4 Membrane separation

Membrane separation technology is often used for advanced treatment of drinking water, which has the advantages of stable effluent quality and simple operation (Park et al., 2017). Depending on the size of the membrane, membrane separation technology can be divided into ultrafiltration, nanofiltration and reverse osmosis. Membrane has strong selectivity and separation, which can effectively remove organic pollutants, multivalent ions and disinfection by-products and at the same time, reduce the hardness of water. **Fig. 3C** shows the principle of membrane separation technology for water purification. Under the action of pressure difference, the size of membrane pore is used to intercept particles in raw waters. Membrane separation technology has been successful in removing bacteria, suspended solids and irons from drinking water (Wu et al., 2019). Now it provides a practical method to combat microplastic contamination found in drinking water streams. Membrane separation technology works as a physical barrier against microplastics. Unlike other devices, that are not generally designed to remove microplastics, the particle size of microplastics is in the same range as that of membranes (Baker, 2012), which enables them to effectively remove microplastics from waters (Talvitie et al., 2017). Microplastics and other separated impurities are safety retained in a small volume ready for further treatment. Nonetheless, there are few researches on the removal of microplastics from drinking water by membrane separation technology. A research carried out by Ziajahromi et al. (2017) investigated the fate of microplastics in wastewater treatment plants in Sydney, Australia. The wastewater treatment plant operated primary, secondary and tertiary treatment process to treat wastewater, including screening, sedimentation, biological treatment, flocculation, disinfection, ultrafiltration, reverse osmosis and decarbonization. It was found that the concentration of microplastics decreased to 2.2 particles L^{-1} after primary treatment, while 0.28 and 0.21 particles L^{-1} were measured after ultrafiltration and reverse osmosis treatment. The results implied that membrane separation technology could be used for post-density separation or rapid separation in clean samples such as drinking water.

4. Challenges to the performance of microplastic treatment processes

4.1 Coagulation

Coagulations are widely used in DWTPs to remove pollutants in water. Due to the high efficiency and low investment, chemical coagulants such as aluminum salts ($AlCl_3 \cdot 6H_2O$, $Al_2(SO_4)_3 \cdot 18H_2O$ and $KAl(SO_4)_2 \cdot 12H_2O$) and iron salts ($FeCl_3 \cdot 6H_2O$ and $Fe_2(SO_4)_3 \cdot 9H_2O$) are usually used. According to the properties of water, a certain amount of coagulants is added to form flocculent substance to encapsulate suspended particles and deposit at the bottom of the pond (**Fig. 4**). This process is controlled by surface charge and concentration of pollutants and pH of water. Environmental microplastic surface usually shows negative charge (Fotopoulou and Karapanagioti, 2012; Triebskorn et al., 2019), which will increase the interaction with chemical coagulants. Accordingly, microplastics may indirectly increase the amount of coagulants required for coagulation process.

4.2 Membrane fouling

Membrane separation technology has its own separation characteristics and can achieve ideal treatment effect. However, the specific operation process also has shortcomings, and membrane fouling is typical representative problem. Membrane fouling is a phenomenon in which particles and macromolecule substances, similar to microplastics, physically and chemically interact with membrane in the process of the treatment, adsorb and deposit on membrane surface or in membrane pore, and the pore size becomes smaller and smaller or blockage occurs (Enfrin et al., 2019) (**Fig. 4**). Theoretically, water pollutants with particle size of 0.1 – 10 times membrane pore size can cause membrane blockage, and that of > 10 times can cause cake layer pollution (Stoller, 2009). The direct consequences of pollution are the decrease of permeable water flow and the increase of transmembrane pressure, thereby causing increased running time, energy costs and maintenance requirements. Ziajahromi et al. (2017) showed that concentration of microplastics in raw water was as high as 10^6 – 10^7 particles/day, invisibly increasing the interaction between microplastics and membrane surface. Because the average pore size of the membrane is smaller than microplastics, a great amount of microplastics poses a risk of polluting the surface and blocking the pore, thus reducing the membrane filtration performance (Ma et al., 2019). Despite extensive studies on the contamination of suspended solids such as silica (Chen et al., 2018), bacteria and colloids (Guo et al., 2012), no research has been published on the membrane fouling owing to microplastic filtration. Moreover, a recent research suggested that microplastics can be decomposed into nanoplastics by wastewater treatment plant process, which increases the number of micro(nano)plastics in water (Enfrin et al., 2019). Another study showed that about 25% of engineering nanomaterials were released into water treatment plants and rejected in secondary or tertiary treatment (Keller and Lazareva, 2014). These particles will aggregate to form a membrane fouling. In addition, microplastics are considered as unignorable vectors for aquatic microorganisms, which settle on microplastics by forming biofilms on the surface (McCormick et al., 2014; Shen et al., 2019e), which may aggregate the membrane biological fouling during water treatment.

According to the raw water quality, choosing reasonable pretreatment process, optimizing process design and selecting correct and efficient membrane cleaning process can effectively prevent and reduce membrane fouling. Membrane separation does not completely remove pollutants in the process of drinking water treatment, but only through physical interception (Enfrin et al., 2019). Therefore, a large number of concentrated water will be produced during membrane operation. At the same time, there will be also produced contaminated cleaning water in the process of membrane cleaning. If the concentrated water and cleaning water are directly discharged into natural water, it will cause water and soil pollution. This is still a problem worth studying and solving in the development of membrane separation technology.

Membrane separation technology can better meet the drinking water quality standards, making an important part of drinking water treatment, while the membrane fouling caused by micro(nano)plastics will become an important issue (Enfrin et al., 2019). However, the impacts of micro(nano)plastics on membrane filtration process performance are not yet fully clear because of the lack of corresponding removal techniques and analysis methods. Exploring the fouling mechanism of micro(nano)plastics on membrane systems is crucial so as to determine their effects on filtration performance. As such, more efforts are desired to improve the acknowledge on micro(nano)plastic fouling.

4.3 Disinfection

Drinking water environment is threatened by many pollutants and environment pollution will increase all kinds of harmful substance in drinking water. Disinfection is an effective method to kill pathogenic microorganisms in water and prevent the spread of diseases. Disinfection is usually the last treatment step during drinking water treatment, so micro(nano)plastics that achieve this process may be most likely to interact with microorganisms in drinking water (**Fig. 4**). The formation of biofilm on the surface of microplastics may reduce the efficiency of disinfection process (Enfrin et al., 2019). Chlorination, ozonation, and ultraviolet irradiation are three common disinfection techniques (Shin and Sobsey, 2008). Chlorination causes bacterial death by inhibiting the activity of their enzymes. However, the occurrence of micron suspended solids in water hinders the effect of chlorine on microorganisms, because they may be tapped by flocs or suspended particles (Narkis et al., 1995). Therefore, microplastics with similar physical properties can act as protective substrates for bacteria, which can resist the disinfection process (Enfrin et al., 2019). The oxidation potential of ozone is 2.08 eV and ozone oxidation can kill chlorine-tolerant microorganisms by attacking cell membranes (Ding et al., 2019). But, microplastics can interact with ozone, thereby reducing the number of ozone molecules available to react with bacteria and leaving unaffected pathogens in water (Enfrin et al., 2019). Ultraviolet radiation can destroy DNA and inactive pathogenic microorganisms, however, the existence of suspended particles protects microorganisms from ultraviolet radiation damage and disinfection (Carré et al., 2018). Therefore, microplastics may reduce the performance of ultraviolet disinfection process. Despite the limited information on presence and behavior of micro(nano)plastics in drinking water, the impact of micro(nano)plastics on the drinking water disinfection process in predicted to be harmful by comparison of similarities, and further research is required.

5. Remediation strategies of microplastic pollution in freshwater environment

Because plastic products are widely used in daily life, the way of accepting contaminants in water environment is more complex. Further research on microplastics in drinking water should focus on as follow:

1) Establishing policy and regulation to control microplastic pollution sources and enhancing the public understanding of microplastics;

2) Strictly supervising the discharge of microplastic wastewater from relevant enterprises, and upgrading or developing removal equipment to clean up the microplastics in drinking water;

3) Strengthening ecological measures of microplastic pollution in typical areas;

4) Using more biodegradation materials, such as starch, cellulose, and lignin, to produce plastics

5) Assessing potential risks from microplastic exposure to contaminated drinking water by epidemiological methods.

These efforts need a multidisciplinary approach to solve the societal, engineering, epidemiological and technological so as to bridge the knowledge gap, especially environmental factors and anthropogenic activities.

5.1 Source control

Currently, some national laws and regulations already have come in force so as to decrease the release of microplastics. Many countries have imposed restrictions on the use of disposable plastic bags. As an example, since 2008, China already forbade to produce, sell and use plastic shopping bags with thickness less than 0.025 mm nationwide. As the same time, the system of paid use of plastic shopping bags was implemented in supermarkets, shopping malls and pedlars' market. More recently, in a press release, the European Union proposed a Europe-wide plastic strategy as a part of the transition to a circular economy (Pico and Barcelo, 2019). According to the new plan, disposable plastic consumption will be reduced and all plastic packing will be recyclable in the European Union markets by 2030. In addition, plastic microbeads have been banned used in personal care products in many countries because they are important potential source of primary microplastics in aquatic environment. These laws and regulations are to enhance the public understanding the potential risks of the environmental (micro)plastics and reduce the use of plastic products and the discharge of plastic wastes. Moreover, fiber in domestic laundry wastewater is also an important source of microplastics in wastewater. Washing methods of washing machines and different washing liquids would directly affect the emission of microplastics. Therefore, it is necessary to find the best washing scheme according to different types of clothes, washing machines and washing liquids.

5.2 Enhancement of removal efficiency

Other strategy that goes hand in hand with restricting the production and consumption of microplastics is to remove microplastics from the aquatic environment. Wastewater treatment plants (WWTPs) are one of important sources of microplastics to aquatic environment (Murphy et al., 2016). Although WWTPs play a vital role in the removal of microplastics from the liquid fraction, sometimes the removal efficiency can reach up 98% (Lares et al., 2018; Leslie et al., 2017), microplastic particle concentrations in effluent can still be ignored because of the large loads of microplastics in WWTP influents and the large volume of effluent constantly discharged every day. Development of advance wastewater treatment technology to manage microplastic pollution is an effective approach. Electrocoagulation is a well-known technique for environmental pollutant removal from wastewaters. The removal efficiency of microplastics by electrocoagulation can reach up 99.24% at a pH 7 (Perren et al., 2018). This technique has been successfully performed in a laboratory stirred-tank batch reactor, and could be feasible on a large scale. Membrane bioreactor (MBR) is one of the most effective techniques for microplastic removal. As shown in **Fig.5**, MBR processes the greatest removal efficiency compared to other traditional wastewater treatment processes (primary, secondary (activated sludge) and tertiary treatment (microfiltration)). A research done by Lares et al. (2018) showed that the removal efficiency of MBR for microplastics reached up 99.4%, with a 0.4 particles L^{-1} in effluent. Another research also reported that while 6.9 particles L^{-1} in the primary effluent, the concentration significantly decreased to 0.005 particles L^{-1} after MBR treatment, with a removal efficiency of 99.9% (Talvitie et al., 2017). Although MBR is more expensive than other treatment process, the successfully application in WWTPs shows its feasibility.

Additionally, membrane fouling in drinking water treatment can be controlled by effective means. Coagulation is one of the common pretreatment processes (Dixon et al., 2013). Coagulation process can be affected by microplastics, while developing a coagulation step to induce flocculation of microplastics is feasible during drinking water treatment (Enfrin et al., 2019). Although microplastics are easy to agglomerate due to their particle size and chemical properties, the agglomeration is unstable and can be dispersed in turbulence (Sumitomo et al., 2018). The use of coagulants can help to generate stable structure that can be removed by precipitation without being destroyed. Ma et al. (2019) suggested that coagulation and membrane separation technology have certain economic potential for removing microplastics from drinking water. Further research on the causes of membrane fouling and establishment of corresponding models will play a positive guiding role in alleviating and avoiding membrane fouling.

Membrane material is the core of membrane separation technology. To solve the problems of membrane fouling, permeability and selectivity of membranes, new membrane materials can be developed. The use of new membranes to reducing fouling caused by micro(nano)plastics is a big challenge because the preparation and modification of new membranes must be adjusted according to the characteristics of micro(nano)plastics in water, while these characteristics are still unclear (Enfrin et al., 2019). Evidence has reported that microplastics surface shows negative charge when entering the aquatic environment (Fotopoulou and Karapanagioti, 2012), thus the negative charge membrane would reduce fouling via the rejection of micro(nano)plastics under ideal conditions. However, using of coagulants may shift their surface charge. It is necessary to understand the surface chemistry of micro(nano)plastics to match the appropriate surface treatment. Therefore, the preparation and modification of new membranes should be able to prevent particulate matter pollution and also control and limit pollution caused by micro(nano)plastics.

5.3 Exploring new solutions

The main problem related to the effect of micro(nano)plastics on drinking water treatment performance is the lack of knowledge about their properties in water (Ogonowski et al., 2018). Most drinking water treatment techniques are not modified to reduce the effect of micro(nano)plastics. Consequently, new approaches must be developed to explore and inhibit the impact of micro(nano)plastics on their performance. In addition, in the actual treatment process, due to the different water quality conditions, the water purification process and the effect is also different. The removal mechanism of micro(nano)plastics can be described by using appropriate theoretical models and numerical simulation methods so as to predict the change of micro(nano)plastics during water treatment and provide guidance for academic research and practical production. Therefore, increasing knowledge of micro(nano)plastics is vital to control their effect on water treatment processes.

6. Conclusions

At present, microplastics have been found in drinking water including tap water and bottled water. However, evidences have shown that the number of microplastics varies greatly from several to thousands particles per volume water. In addition to the diversity of samples investigated, changes in sampling, sample pretreatment, and analytical methods may also lead to some differences in microplastic concentrations. Despite the potential health risk of microplastics to human are not yet understood, the occurrence in drinking water should not be ignored.

This paper systematically reviews the occurrence of microplastics in drinking water and discusses the removal strategies of microplastics by drinking water treatment process so as to provide the research directions of microplastics in drinking water treatment. Currently, the research on the removal of microplastics in drinking water treatment process is still relatively scarce. Because traditional processing technologies are not designed to remove microplastics, as such, there are questions about whether the appropriate adjustment of these technologies can satisfactorily remove microplastics or whether new technologies need to be developed. Existing pretreatments that limit the amount of microplastics in water should be adapted to protect drinking water treatment process. The current treatment technologies are needed to be optimized to meet the challenges of microplastics and to ensure the proper performance of treatment process. In addition, during the treatment process, the possible enrichment of microplastics and removal efficiency by different treatment steps at DWTPs should be investigated in the future. Environmental-related and feasible water treatment conditions should also be emphasized. Drinking water treatment plants have to face the problem of micro(nano)plastics, at least in some places, because this poses a new threat to human health.

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Declaration of interest

The authors have no conflict of interest to declare regarding this article.

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