Wastewater treatment plant effluent as a source of microplastics: review of the fate, chemical interactions and potential risks to aquatic organisms

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ABSTRACT

Wastewater treatment plant (WWTP) effluent has been identified as a potential source of microplastics in the aquatic environment. Microplastics have recently been detected in wastewater effluent in Western Europe, Russia and the US. As there are only a handful of studies on microplastics in wastewater, it is difficult to accurately determine the contribution of wastewater effluent as a source of microplastics. However, even the small amounts of microplastics detected in wastewater effluent may be a remarkable source given the large volumes of wastewater treatment effluent discharged to the aquatic environment annually. Further, There is strong evidence that microplastics can interact with wastewater-associated contaminants, which has the potential to transport chemicals to aquatic organisms after exposure to contaminated microplastics. In this review we apply lessons learned from the literature on microplastics in the aquatic environment and knowledge on current wastewater treatment technologies with the aim of identifying the research gaps in terms of (i) the fate of microplastics in WWTPs, (ii) the potential interaction of wastewater-based microplastics with trace organic contaminants and metals, and (iii) the risk for aquatic organisms.

Key words | fate, microplastics, removal, risk, sorption capacity, wastewater

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ABBREVIATIONS USED

DDT = Dichlorodiphenyltrichloroethane; FT-IR = Fourier Transform Infrared; LOAEC = lowest observable adverse effect concentration; PAH = Polycyclic Aromatic Hydrocarbon; PBDE = Polybrominated Diphenyl Ether; PCB = Polychlorinated Biphenyl; PVC = Polyvinyl Chloride; TrOC = Trace Organic Contaminants; UPVC = Unplasticized Polyvinyl Chloride; WWTP = Wastewater Treatment Plant; K_{PE-W} = Polyethylene-water partition coefficient; K_{OW} = Octanol-water partition coefficient.

INTRODUCTION

The occurrence of plastic debris as a widespread pollutant has been reported in various aquatic environments, including rivers, lakes, estuaries, coastlines and marine ecosystems, and represents an increasing environmental concern (Eerkes-Medrano *et al.* 2015).

Currently, the polymers that contribute to 90% of plastic products are (low-density and high-density) polyethylene,

polypropylene, polyvinylchloride, polystyrene, and polyethylene terephthalate (or polyester) (Zarfl & Matthies 2010; Ivar do Sul & Costa 2014). Consequently, the majority of released plastic pollutants in the environment are composed of these polymers (Ivar do Sul *et al.* 2014).

Microplastics, which are plastic fragments in the micrometer size range (<1 mm) (Browne *et al.* 2011), are available to a wide range of aquatic organisms as they occupy the same size range as their natural food sources (Zhao *et al.* 2014). Microplastics have the capacity to interact with a variety of contaminants in the surrounding environment (Holmes *et al.* 2012; Bakir *et al.* 2014; Wang *et al.* 2015), which may also pose a risk for aquatic organisms. Further, additives that are included in the polymer structure during manufacturing may also lead to harmful consequences if they are leached to exposed organisms (Browne *et al.* 2013). Nevertheless, the degree to which various microplastics themselves and their associated contaminants may cause a risk to aquatic organisms still remains poorly understood.

Generally the sources of microplastics in the aquatic environment have been classified as (i) aquatic-based sources and (ii) land-based sources and include both, primary or secondary microplastics (Law *et al.* 2010; Hammer *et al.* 2012; Eriksen *et al.* 2013b). Primary microplastics are manufactured as microplastics, while secondary microplastics are produced from the breakdown of larger plastic particles (Zhao *et al.* 2014). Aquatic-based microplastics may originate from the degradation of larger plastic objects discarded through human activities, such as littering, fishing and shipping activities, and are considered as secondary microplastics (Hammer *et al.* 2012). Land-based sources include run-off containing microplastics (solid wastes). After heavy rainfall microplastics can be flushed into the aquatic environment, especially from poorly managed or illegal landfills (Teuten *et al.* 2009; Hammer *et al.* 2012; Wagner *et al.* 2014; Duis & Coors 2016), and these particles can also be classified as secondary microplastics given that they undergo breakdown before entering the aquatic environment (Eerkes-Medrano *et al.* 2015). Wastewater treatment plant (WWTP) effluent is another land-based source of microplastics (Cole *et al.* 2011; Wagner *et al.* 2014; Eerkes-Medrano *et al.* 2015) and can include both primary and secondary microplastics. Depending on the wastewater treatment facilities, effluent may be released directly into the ocean or into the fresh water environment, such as rivers or lakes, which may then be transported to the marine environment (McCormick *et al.* 2014).

While land-based sources are assumed as one of the most important sources of microplastics (da Costa *et al.* 2016), there is a lack of quantitative estimation on the relative contribution of different sources to the total concentration of microplastics in the aquatic environment (Koelmans *et al.* 2014; Duis & Coors 2016).

Amongst the different sources of microplastics to the aquatic environment, wastewater effluent has recently received considerable attention as a potential point source of microplastics (Browne *et al.* 2011; Carr *et al.* 2016). However, the contribution of microplastics released via wastewater effluent compared to other sources is largely unknown (Magnusson & Norén 2014; Eerkes-Medrano *et al.* 2015).

The importance of WWTP effluent as a source of microplastics may increase in the future, given current population growth as well as rapid urbanization and industrialization, which can lead to increasing volumes of wastewater (UNEP 2010; Browne *et al.* 2011). Subsequently, the contribution of wastewater-based microplastics into the receiving environment may continue to increase if its importance is

not fully realised and no further actions are taken to prevent and reduce microplastics in wastewater.

The current review highlights the presence and potential importance of WWTP effluent as one of the sources of microplastics in the aquatic environment by reviewing the current state of knowledge. The review will also provide an in-depth analysis on the fate of microplastics in WWTPs, the potential mechanisms involved in the interaction of wastewater-based microplastics and trace organic contaminants (TrOCs), and the adverse effects of microplastics and their associated TrOCs from a risk-based perspective. The review will conclude by highlighting areas of future research.

SOURCES OF WASTEWATER-BASED MICROPLASTICS INTO THE AQUATIC ENVIRONMENT

Over the last few years, various types of microplastics with a suspected WWTP origin have been reported in aquatic ecosystems (Table 1). One of the common wastewater-based microplastics include microbeads, such as polyethylene and polypropylene, which contribute to over 90% of microbeads associated with cosmetic and personal care products (Zitko & Hanlon 1991; Fendall & Sewell 2009; Leslie 2014; Wagner *et al.* 2014; Dris *et al.* 2015c; Gouin *et al.* 2015). While a number of cosmetic companies have voluntarily started to remove plastic microbeads from their products and replace them with natural compounds (Gouin *et al.* 2015), it was recently reported that 80 body and facial scrub products in the UK market still contain polyethylene microbeads that can be consumed by millions of consumers worldwide (Beat the Microbeads 2015; Napper *et al.* 2015). Moreover, a very recent study of eight municipal WWTPs effluent across Southern California revealed that the majority (over 90%) of detected microplastics had a profile resemble the blue polyethylene microplastics extracted from toothpaste (Carr *et al.* 2016). Thus, microbeads in personal care products are currently an important potential source of microplastics in WWTP effluent.

Recently, the US Government established the 'Microbead-Free Waters Act of 2015', which prohibits the production or sale of rinse-off personal care products (e.g. toothpaste or facial scrubs) containing microbeads from 2017 (Congress.gov. 2015).

Other plastic polymers also used in cosmetic products include polylactic acid, polyethylene terephthalate (Rochman *et al.* 2015), polyethylene isoterephthalate, nylon-12,

Table 1 | Some studies detecting microplastics in the aquatic environment pointing to a suspected wastewater origin

Study	Study location	Sampled environment	Average abundance of sampled microplastics	Mesh size of sampling method	Origin of sampled microplastics
Zhao <i>et al.</i> (2014)	Yangtze Estuary and the coastal waters of the East China Sea	Surface water	4,137 particles/m ³ in Yangtze Estuary, and 0.167 particles/m ³ coastal waters of East China Sea	333 µm mesh neuston net	Approximately 80% were fibres, which were suspected to derive from sewage and/or from degradation of rope material and fishery activities.
Castañeda <i>et al.</i> (2014)	St. Lawrence River, Canada	Sediment	13,832 particles/m ²	500 µm sieve	99.9% of polyethylene microbeads had the similar size, shape and chemical composition of microbeads in cosmetic products
Claessens <i>et al.</i> (2011)	Belgian coast	Sediment	37,000–149,000 particles/m ²	Bulk sample of sediment was taken and passed through 38 µm mesh	Nylon and polypropylene fibres following polyethylene and polypropylene fragments were the dominant type of microplastics that were suggested to be associated with carpet and clothing and cosmetic products, respectively
Browne <i>et al.</i> (2011)*	Australia, Oman, United Arab Emirate, Chile, Philippines, Portugal, USA, South Africa, Mozambique, United Kingdoms	Sediment	2–31 particles /250 ml of sediment	NA	The dominant types of collected microplastics were polyester and acrylic, followed by polypropylene, polyethylene, and polyamide fibres, with detected fibres similar to fibres found in wastewater effluent, most likely from washing synthetic clothes
Dris <i>et al.</i> (2015a)*	River Seine, France	Surface Water	30 and 0.35 particles/m ³ for plankton net and manta trawl, respectively	80 and 330 µm-mesh, plankton net and manta trawl	The dominant type of sampled microplastics were fibres and urban water system discharges, including wastewater, was considered as one of the sources of the detected microplastics.
Eriksen <i>et al.</i> (2013a)	Lakes Superior, Huron and Erie, USA	Surface water	43,000 particles/km ²	333 µm mesh manta trawl	Many microplastics were multi-coloured with a spherical shape and were suspected to be microbeads from cosmetic products associated with wastewater
Gallagher <i>et al.</i> (2015)	Solent Estuarine Complex, UK	Water column	Total of 2,759 particles of four different sampling site	300 µm plankton net trawl	The majority of detected microplastics were fibres (expected to be polyester) and rounded pellets (expected to be polyethylene and polypropylene), which were likely to be associated with wastewater effluent discharge.

(continued)

Table 1 | continued

Study	Study location	Sampled environment	Average abundance of sampled microplastics	Mesh size of sampling method	Origin of sampled microplastics
Talvitie <i>et al.</i> (2015)*	Helsinki archipelago coastal Gulf of Finland.	Water column & sediment	0.01 and 0.65 fibres/L, 0.5 and 9.4 synthetic particles/L	20–200 µm using a filtration device	The fibres and synthetic particles with properties similar to microplastics detected in wastewater effluent were found in the receiving water body of a WWTP.
McCormick <i>et al.</i> (2014)	North Shore Channel, Illinois, USA	Surface water	17.93 particles/m ³ downstream of WWTP effluent input 1.94 particles/m ³ upstream, of WWTP effluent input	333 µm mesh neuston nets	1.21 and 10.57 particles/m ³ were fibres and 0.45 and 0 particle/m ³ were pellet, in downstream and upstream samples, respectively. Microplastics were suspected to be associated with synthetic textiles and cleansing products in wastewater.
Woodall <i>et al.</i> (2014)	Mediterranean Sea, SW Indian Ocean and NE Atlantic Ocean	Sediment	14,000 particles/m ² of Indian Ocean Sediment	Collected sediments passed through 32 µm mesh sieve	The dominant type of detected fibres was polyester (53%) followed by polyamide (34%) and acrylic (12%). Textile was suggested as one of the potential sources.

Note: 70 kg sediment, 0.1 m² of sampling site (Claessens *et al.* 2011).

Note: 'NA': Not available.

*Studies that confirmed wastewater as a source of detected microplastics using Fourier transform infrared spectroscopy.

nylon-6, poly(methyl methacrylate), polytetrafluoroethylene, and polyurethane (Leslie 2014), and these polymers may enter into wastewater effluent following production and use of those products. Additionally, microplastics with physical properties that resemble microbeads, such as polyethylene, polystyrene and thermoset polyester polymer applied in air and sand blasting technologies, are also a potential source of microplastics in wastewater (Eriksen *et al.* 2013a).

Microfibers, such as polyester, acrylic, and nylon associated with clothes, carpets and other textile products (Browne *et al.* 2007; Browne *et al.* 2011; Claessens *et al.* 2011), may unintentionally find their way into the aquatic environment via wastewater during washing and/or manufacturing of synthetic fibres and represent another type of wastewater-based microplastics (Teuten *et al.* 2007; Browne *et al.* 2011; Law & Thompson 2014; Driedger *et al.* 2015; Rocha-Santos & Duarte 2015). More than 100 fibres per litre of laundry wastewater can be released into WWTPs from washing polyester clothes (Browne *et al.* 2011; Dris *et al.* 2015c). Thus, textile and carpet industries as well as commercial laundries are likely to release significant amounts of microplastic fibres to WWTPs and, depending on the level

of treatment, this wastewater effluent may represent a relatively large source of microplastic fibres. However, this has yet to be estimated. McCormick *et al.* (2014) reported high concentrations of microplastics, mostly fiber associated with synthetic fabrics, in the North Shore Channel of the Chicago River, which receives secondary effluent from a domestic treatment plant that currently operates with the secondary clarifiers (Storm Water Solution 2013).

Microplastics with a similar shape and composition as microbeads used in cosmetic products have been detected in surface waters from three of the Great Lakes in North America, with their source suspected to be wastewater effluent discharge (Eriksen *et al.* 2013a). This finding was supported by the detection of microbeads in wastewater effluent discharged into the Great Lakes (Schneiderman 2015). Further, Claessens *et al.* (2011) found high concentrations of microplastics with the dominant shape of fibres and granules in marine sediment along the Belgian coast. Although, this study was not able to determine the source of the detected microplastics, it was speculated that the granules, which included polyethylene and polypropylene, were likely to have originated from cosmetic products, air blasting media and the plastics industry. Additionally, a

proportion of fibres, such as nylon, were suspected to be derived from fibrous material including clothes and carpets (Claessens *et al.* 2011), and thus are likely to be associated with domestic and/or industrial wastewater discharge.

Deep-sea sediments sampled at different locations worldwide, including the Mediterranean Sea, South West Indian Ocean and North East Atlantic Ocean, showed that polyester and acrylic polymers contributed to the majority (86%) of analyzed microplastics and were suspected to derive from sources such as packaging material, electronics and textiles (Woodall *et al.* 2014). wastewater was suggested as one of the potential sources of microplastics in all above studies, the actual contribution of wastewater to the detected microplastics has yet to be established. In contrast, Klein *et al.* (2015) detected microplastics in sediment along the shore of the Rhine and Main Rivers in Germany and found lower concentrations of microplastics near WWTPs compared to other monitored areas, with no relationship between the number of microplastics detected and the proximity to WWTPs found. Moreover, the detected microplastics primarily included polyethylene, polypropylene and polystyrene, which suggests they originate from discarded plastic products, as these polymers comprise 55.7% of European plastic demand (Klein *et al.* 2015).

A number of factors may affect the contribution of wastewater as a source of microplastics and also the dominant type of wastewater-based microplastics in a specific area including (i) population density, (ii) the number and type of industries involved in producing plastics, (iii) the level of wastewater treatment, (iv) the public awareness with respect to microplastics and their subsequent effects and (v) waste management processes (Free *et al.* 2014; Jambeck *et al.* 2015). The correlation between some of the above-mentioned factors and input into aquatic environment has been reported for other pollutants (Abril *et al.* 2002). Thus, it is likely that highly populated and industrialized communities with low levels of wastewater treatment, as well as inefficient waste disposal management, may be major contributors to the release of wastewater-based microplastics to the environment. It is reported that 90% of all wastewater in developing countries is discharging untreated to the aquatic environment such as lakes, rivers and the oceans (UNEP 2010).

FATE OF WASTEWATER-BASED MICROPLASTICS IN WWTP

While the presence of wastewater-based microplastics in the aquatic environment has recently attracted the attention of

scientists (Fendall & Sewell 2009; Cole *et al.* 2011; Eriksen *et al.* 2013a; McCormick *et al.* 2014), less attention has been drawn to the fate of microplastics during wastewater treatment (Duis & Coors 2016).

Before returning to the aquatic system, wastewater may be treated in WWTPs. Depending on the type and scale of the facility, a WWTP can have primary, secondary and tertiary treatment trains to purify the influent using physical, chemical and biological treatment processes. During physical and chemical processes many suspended and floating solids including plastic particles are removed through screening, sedimentation, flotation, coagulation-flocculation and filtration within treatment ponds or settling tanks (Stephenson 2009). These processes generally capture a large amount of solid particles, which may include plastic debris depending on the size and density of particles as well as the size of the screen bars and filters (Stephenson 2009). The degree to which these process can specifically remove microplastics has recently been investigated by Talvitie *et al.* (2015), who found that the number of microfibrils in wastewater influent reduced from 180 fibres per litre to 14.2 and 13.8 fibres per litre after primary and secondary sedimentation processes, respectively.

To investigate the fate of microplastics during treatment processes and the role of wastewater as a source of microplastics, it is necessary to estimate the mass of microplastics entering WWTPs and the amount released with wastewater effluent. Recently, there have been a number of pilot studies assessing the load of microplastic in WWTPs in Europe, Russia and the US (HELCOM 2014; Magnusson & Norén 2014; Schneiderman 2015), with the results summarised in Table 2. One such study demonstrated that mechanical, chemical and biological treatment can remove about 99% of microplastics, from 15,000 particles/L in the influent to 8 particles/L in the effluent (Magnusson & Norén 2014). Likewise, in a Russian WWTP the number of microplastics (textile fibres) reduced from 467 particles/L in the influent to 16 particles/L in the effluent (~96% reduction in microplastics after the secondary treatment) (HELCOM 2014). While this is a promising finding, given the volume of treated wastewater of 2.18 million m³/d from the above Russian WWTP (Vodokanal 2015), over 34 × 10⁹ particle/L are still discharged to the receiving environment every day. A recent study on a WWTP in Finland also reported an average concentration of 4.9 and 8.6 microplastic fibres and particles, respectively, per litre in tertiary treated effluent (~98% removal after tertiary treatment) (Talvitie *et al.* 2015). Therefore, a daily discharge of 1,323 × 10⁶ fibres and 2,322 × 10⁶ particles to the receiving

Table 2 | Some reviews of used techniques to detect microplastics in wastewater and reported results

Study	Study country	Sampling method	Size range of sampled particles (μm)	Identification method	Dominant shape & type of detected microplastics	Number of microplastics influent	Number of microplastics in final effluent	Treatment type	Removal rate
Talvitie <i>et al.</i> (2015)	Finland	Filtration device	20–200	Visual observation & FT-IR spectroscopy	Synthetic particle and textile fibres	430 synthetic particle/L, 180 textile fibres/L	8.6 synthetic particle/L, 4.9 textile fibres/L	Tertiary	98% synthetic particle, 97% for fibres
Dris <i>et al.</i> (2015b)	France	Automatic sampler	ND	Visual observation	Fibres	260–320 particle /L	14–50 particle/L	Primary & biological treatment	84–94%
Murphy <i>et al.</i> (2016)	Scotland	Steel bucket and steel sieve	>65	Visual observation & FT-IR spectroscopy	Fibres (polyester, polyamide, acrylic)	15.7 particle/L	0.3 particle/L	Secondary	98.41%
Carr <i>et al.</i> (2016)	U.S	Sieving	20–400	Visual observation & FT-IR spectroscopy	Irregularly shaped polyethylene	1 particle/L	~0.0007 particle/L	Tertiary	99.9%
Magnusson and Norén (2014)	Sweden	Filtration	>300	Visual observation & FT-IR spectroscopy	Fibres and fragment	15 particle/L	8.3 particle/L	Mechanical, chemical & biological	99.9%
HELCOM (2014)	Russia	A filtration device	20–300	Visual observation	Fibres & synthetic particle	467 fibres/L, 160 synthetic particle/L	16 fibres/L, 7 synthetic particle/L	Mechanical treatment and purification	96%
Browne <i>et al.</i> (2011)	Australia	Glass bottle (750 ml), filtration	ND	Visual observation & FT-IR spectroscopy	Fibres	ND	1 particle/L	Tertiary	ND

Note: 'ND': Not detected; 'FT-IR': Fourier transform infrared spectroscopy.

environment can be expected from that one WWTP, given a flow rate of around 270 million litres of wastewater per day (Talvitie *et al.* 2015). Carr *et al.* (2016), reported the removal efficiency of 99.9% of microplastics in tertiary WWTPs in the U.S, with total daily discharge of 0.93×10^6 microplastics. It should be noted that the lack of a uniform and standardized method for sampling, sample processing and analysis of microplastics may lead to incomparable results. The methods used by some studies that detect microplastics in WWTPs are shown in Table 2.

Thus, while 96 to 99.9% is a significant reduction, the small percentage remaining in the effluent is likely to nevertheless represent an important source of microplastics to the aquatic environment given the large volume of effluent produced annually.

Currently, WWTPs are not specifically designed to remove microplastics from the wastewater effluent (Schneiderman 2015). However, the results of recent studies showed that the majority of microplastics are captured through skimming of floating solids during the primary treatment stage (Carr *et al.* 2016; Murphy *et al.* 2016). In this process microplastics removal is more dependent on density rather than size, as lower density microplastics may float and be removed by surface skimming (Carr *et al.* 2016). Moreover, there is evidence that tertiary treatment showed greater removal of microplastics compare to secondary and primary (Talvitie *et al.* 2015). Analyzing the final effluent of a German WWTP revealed that advanced filtration could only remove 93% of microplastics in the size range of $<500 \mu\text{m}$ (Mintemig *et al.* 2014) as cited in Duis & Coors 2016). More research is needed to assess the capacity of WWTPs to capture microplastics.

The physiochemical properties of microplastics can be altered during treatment processes. A recent study provides evidence regarding the presence of a layer of biofilm on the observed wastewater-based microplastic. These biofilms may influence the surface properties and relative density of microplastics (Carr *et al.* 2016), and consequently the fate of wastewater-based microplastics in the aquatic environment. Further, Lobelle & Cunliffe (2011) showed that biofilm can significantly modify the physiochemical properties of plastic debris, by making them less hydrophobic and more buoyant. Further research is required to study physical and chemical modifications of microplastics during wastewater treatment processes.

When considering the fate of microplastics in WWTPs another important issue is what happens to the retained microplastics. It has been reported that a large proportion of the captured microplastics will be retained in the sludge

phase (Magnusson & Norén 2014), with a number of studies finding high concentrations of microplastics in WWTP sludge samples (Brandsma *et al.* 2014; Magnusson & Norén 2014; Carr *et al.* 2016). Carr *et al.* (2016) reported 5 microplastic particles in 5 g of biosolid samples of a WWTP that based on the daily production of biosolid, 1.9×10^9 particles is estimated to remove daily with the biosolid. Thus, the use of treated sewage sludge as biosolid may be an important source of microplastics to the environment given their wide application in agriculture and forestry for soil amendment (Rillig 2012; Dris *et al.* 2015c; Eerkes-Medrano *et al.* 2015; Rocha-Santos & Duarte 2015). It is also suggested that a proportion of microplastics in soil may eventually enter the aquatic environment via run off after heavy rain, irrigation or by transportation by the wind (Hammer *et al.* 2012; Rochman *et al.* 2015).

INTERACTION OF MICROPLASTICS WITH TROCS AND METAL IONS

Given that wastewater contains a large number of organic and inorganic pollutants, it is likely that microplastics can interact with the variety of contaminants present in wastewater. This can potentially make microplastics carriers of wastewater-based contaminants, including industrial chemicals, pharmaceuticals, hormones, pesticides and metal ions, and thus modify their environmental fate. Using the textile industry as an example, substantial amounts of chemicals are used in bleaching, dyeing and finishing processes, thus auxiliary chemicals and heavy metals such as Cr, As, Cu and Zn can be released into the wastewater (Carmen & Daniela 2012; Ghaly *et al.* 2013) and can potentially interact with the microfibrils associated with textiles.

The interaction of microplastics and metal ions such as Al, Fe, Mn, Cu, Zn, Pb, Ag, Cd, Co, Sr, Mo, Sb, Sn, and Ni and the capacity of plastics to act as a 'transport vector' of toxic metals has been extensively reported in the literature (Ashton *et al.* 2010; Holmes *et al.* 2012; Nakashima *et al.* 2012; Rochman *et al.* 2014). Metallic sorption can occur in both aged/weathered and virgin microplastics. It was assumed that the sorption mechanism in weathered microplastics would occur through the interaction of bivalent cations and oxyanions with the charged regions of modified plastic surfaces (Holmes *et al.* 2012; Holmes *et al.* 2014). However, it was demonstrated that the interaction of virgin microplastics and trace metals (Cd, Co, Ni, Pb) occurs through the rapid adsorption of existing organic matter in water to the surface of the microplastics, leading

to the acquisition of a negative charge from the organic matter function groups, due to the fact that virgin plastics do not exhibit inherent charge or acid-base behaviour (Holmes *et al.* 2014). Therefore, the potential interaction of microplastics with metal ions in wastewater, as well as the common treatment chemicals, such as aluminium and iron used for flocculation-coagulation processes (Ukiwe *et al.* 2014), may be an issue that needs further investigations.

In both the laboratory and the field, microplastics have been shown to sorb a wide range of TrOCs at many orders of magnitude higher than the surrounding water, with the reported concentrations ranging from 1 to 10,000 ng/g (Rios *et al.* 2010; Koelmans *et al.* 2013). Further, microplastics are able to accumulate more contaminants in highly polluted environments, such as industrial areas (Frias *et al.* 2010; Hirai *et al.* 2011; Karapanagioti *et al.* 2011; Antunes *et al.* 2013; Mizukawa *et al.* 2013). Thus, it is expected that microplastics in wastewater will behave similarly and will be able to accumulate high concentrations of TrOCs and metals. The sorption of some organic pollutants including dichlorodiphenyltrichloroethane (DDT) and phenanthrene to polyethylene microbeads extracted from shower gel has recently been showed to be comparable with the sorption capacity of polyethylene originated from other sources (Napper *et al.* 2015). Moreover, there is evidence of the interaction of microplastics and wastewater derived TrOCs, such as perfluorooctanesulfonate (PFOS) and perfluorooctanesulfonamide (PFOSA), with perfluorochemicals found frequently in municipal and industrial wastewater and sewage sludge (Ma & Shih 2010; Wang *et al.* 2015). To date, research on the interaction of microplastics and TrOCs has mainly focused on the marine environment, and more recently fresh water and estuarine environments, but the sorption behaviour and chemical fate of microplastics in wastewater still remains unexplored.

Sorption capacity of microplastic for TrOCs

The sorption of TrOCs to microplastics polymers is likely to occur through (i) absorption and (ii) adsorption mechanisms, which are strictly dependent on the physico-chemical characteristics of both TrOCs and microplastic (Mato *et al.* 2001; Matsuzawa 2010; Rochman *et al.* 2013a). Thus, different microplastics are expected to show different sorption affinities for different types of TrOCs.

Adsorption is assumed to be the general sorption mechanism that would occur not only through the interaction of hydrophobic surfaces of the polymer and TrOCs (Matsuzawa 2010) (non-polar surface interaction/hydrophobic

interaction), but also by specific interactions, such as electrostatic interactions and hydrogen bonding (Pan & Xing 2008). The polymer adsorption capacity to uptake TrOCs is likely to be positively correlated with the polymer surface area, which provides more adsorption sites at the surface of the polymer (Teuten *et al.* 2007; Fotopoulou & Karapanagioti 2012; Rochman *et al.* 2013a; Rochman 2015). Thus, the smaller the microplastic size, the more surface area and consequently the greater the sorption capacity (Rochman 2015). The surface interaction is likely to reach a threshold once the adsorption sites are saturated and the time required to reach the saturation may also vary depending on the polymers and TrOCs (Rochman *et al.* 2013a).

Absorption, on the other hand, refers to the diffusion of contaminants into the matrix of sorbent, which is strongly associated with the polymer molecular structure (Rochman *et al.* 2013a). Polymers have been classified as rubbery and glassy based on their molecular structure (Crawford 1998). The structure of glassy polymers is more condensed, with little void spaces, whilst rubbery polymers exhibit greater flexibility, with large amounts of free volumes between molecules (Teuten *et al.* 2009; Bakir *et al.* 2012). It has been shown that rubbery polymers, such as polyethylene and polypropylene, have a greater sorption capacity for TrOCs, such as phenanthrene, DDT, polychlorinated biphenyl and polycyclic aromatic hydrocarbons compared to glassy polymers, such as polyvinyl chloride (PVC) (Bakir *et al.* 2012; Rochman *et al.* 2013a). Consequently, it is obvious that the sorption capacity of microplastics for TrOCs varies appreciably among different types of polymer depending on their surface area, crystallinity and diffusivity (Rochman *et al.* 2013a; Rochman 2015).

While specific types of polymer may show a high tendency to sorb a specific class of TrOCs, this may not extend to all classes of TrOCs (Rochman *et al.* 2013a; Rochman 2015). The chemical properties of TrOCs such as hydrophobicity (octanol-water partition coefficient, $\log K_{ow}$) and molecular weight (Smedes *et al.* 2009) can also affect the sorption rate and the equilibrium time (Rochman *et al.* 2013a).

Other factors that are likely to affect the partition behaviour and sorption rate of a TrOC to microplastics are (i) the concentration of organic matter and (ii) the presence of other contaminants in the system. The relative importance of organic matter compared to microplastics for sorption of hydrophobic TrOCs has been recently suggested using a multimedia model (Gouin *et al.* 2011). Based on an environmental realistic volume ratio of polyethylene to organic matter ($1:1.26 \times 10^7$) for the marine coastal system, it has been predicted that the majority of TrOCs, even those with

a high polyethylene-water partition coefficient ($\log K_{PE-W} > 7$), will tend to partition more (>99%) to organic matter than polyethylene (Gouin *et al.* 2011). Thus, the organic matter-microplastic volume ratio may also affect the sorption rate of TrOCs to microplastics. This is particularly relevant for wastewater-based microplastics, as wastewater can contain high concentrations of organic matter, microplastics and TrOCs.

The competition behaviours between various TrOCs to sorb to microplastics can also influence the sorption rate of a TrOC to microplastics (Bakir *et al.* 2012). Given that wastewater will contain a complex mixture of TrOCs, it is likely that the sorption capacity of TrOCs by microplastics in wastewater would be lower than expected based on single compound experimental studies (Bakir *et al.* 2012). More research is required to investigate sorption behaviour in real but more complex matrices, such as WWTP effluent.

A RISK-BASED PERSPECTIVE OF BIOLOGICAL EFFECTS OF MICROPLASTICS ON AQUATIC ORGANISMS

To evaluate the risk that microplastics may pose to aquatic organisms, it is important to understand to what extent microplastics and their associated contaminants contribute to adverse effects and subsequent health outcomes in aquatic organisms. Toxicity may be related to acute or chronic effects through short or long-term exposure to microplastics and/or associated contaminants (Hammer 2008).

Biological effects of microplastics themselves

The adverse health effects from exposure to microplastics can result from physical and/or chemical effects (Wright *et al.* 2013b). According to a Marine Strategy Framework Directive Report, the possible direct detrimental effects of ingested microplastics include interference with enzyme production, nutrient dilution, reduced growth rate, decreased steroid hormone levels, delayed ovulation, reproductive failure and death (Galgani *et al.* 2010). Microplastics can also cause physical damage through accumulation in the digestive tract following ingestion (Wright *et al.* 2013b), although the amount of microplastics necessary to produce sub-lethal or lethal effects vary as the process is influenced by the type and sensitivity of the organism (Wright *et al.* 2013a; Wright *et al.* 2013b), exposure duration (Eisler 2000), the daily uptake dose of microplastics through normal

food ingestion, the egestion of microplastics through excrement and the retention time of microplastics in the organism's body, as well as the type, size and shape of ingested microplastics (Huang *et al.* 2010; Wright *et al.* 2013b). To date, there is limited work on assessing the risks after exposure to a range of microplastic concentrations (Syberg *et al.* 2015). Lee *et al.* (2013) reported reduced survival of copepods (*Tigriopus japonicus*) exposed to 0.05 and 0.5 μm microplastic beads at concentrations of 1.25 and 25 mg/L, respectively, and no effect of 6 μm beads at concentrations as high as 25 mg/L.

The negative effects of microplastics on exposed organisms have been reported even after short-term exposure duration. For instance, aggregated polyethylene microplastics were observed in digestive gland lysosomes in blue mussels (*Mytilus edulis*), which caused a considerable decrease in lysosomal membrane stability at the cellular level by increasing granulocytoma formation during 96 h exposure as a cellular self-protection response to contaminants (von Moos *et al.* 2012). The physical impact of unplasticized polyvinyl chloride (UPVC) microplastics within the size range of 20–100 μm was also investigated on lugworms, where 30% and 50% depletion of lipid reserves was observed after exposure to 1% and 5% PVC, respectively, and was directly associated with a reduction in feeding activity due to the long retention time of microplastics in the organism's gut (Wright *et al.* 2013a). Similarly, a recent study with copepods exposed to 20 μm polystyrene microbeads over 24 h showed reduced energy supplies and growth rate due to interference with feeding capacity at concentration of 75 particles mL^{-1} , which were 10% of their available food (Cole *et al.* 2015).

It should be noted that some studies have conversely shown that microplastics can have negligible effects on some organisms. Hamer *et al.* (2014), recently demonstrated limited physical effects on isopods (*Idotea emarginata*) after 6 months of continuous exposure to three different microplastic types (polystyrene microbeads, polystyrene fragments and polyacrylic fibers), under low and high concentrations, with 100% of the ingested microplastics detected in the feces and no accumulation in the organism's gut. Consequently, it is possible that some aquatic organisms may exhibit no negative effects to any concentrations of microplastics. Therefore, improved understanding of the adverse effect of microplastics requires a comprehensive risk assessment procedure that considers all factors mentioned above. Some studies on the effects of microplastics on various organisms, and the factors influencing biological effects, are summarized in Table 3.

Table 3 | Some reviews of the biological effects of microplastics on various organisms

Study	Exposed Organism	Polymer type & size	Concentration	Retention time	Exposure duration	Biological effects
Browne <i>et al.</i> (2008)	Mussel (<i>M. edulis</i>)	Polystyrene (3–9.6 µm)	0.51 g/L	Over 48 days	3 hours	No toxicological effects were observed
Carlos de Sa <i>et al.</i> (2015)	Common goby (<i>P. microps</i>)	Polyethylene (420–500 µm)	30 particles alone & 15 particles in combination with 15 preys	NA	3 min	Reduction of predatory performance and efficiency
Cole <i>et al.</i> (2015)	Copepod (<i>C. helgolandicus</i>)	Polystyrene (20 µm)	75,000 particles/L	NA	24 hours and 9 days	Prolonged exposure to microplastics led to: Reduction of carbon biomass that may cause energetic deficiencies and lower growth; producing smaller eggs; and reduced hatching success
Hamer <i>et al.</i> (2014)	Isopods (<i>I. emarginata</i>)	Polystyrene (1–100 µm) microbeads and fragments, and polyacrylic fibres (20–2,500 µm)	<ul style="list-style-type: none"> • 12 or 120 microbeads/mg of food • 20 or 350 fragments/mg of food • 0.3 µg fibers/mg food 	No retention was observed	6 to 7 weeks	No adverse effects on survival or growth rate were observed
Peda <i>et al.</i> (2016)	European sea bass (<i>D. labrax</i>)	PVC pellets (>300 µm)	0.1% (W/W) of their diet	NA	30, 60, and 90 days	<ul style="list-style-type: none"> • After 30 and 60 days of exposure: the majority of studied organisms showed moderate structural changes such as, shortening and swelling of villi, circulatory changes and worsening of inflammatory changes • After 90 days: half of the individuals showed serious changes such as reduction of perivisceral fat and reduction of intestinal function • No mortality were observed after 90 days exposure

(continued)

Table 3 | continued

Study	Exposed Organism	Polymer type & size	Concentration	Retention time	Exposure duration	Biological effects
Lee <i>et al.</i> (2013)	Copepod (<i>T. japonicas</i>)	Polystyrene (three sizes: 0.05, 0.5 and 6 µm)	Acute toxicity test: 0, 6, 13, 31, 63, 187, 250 and 313 mg/L Chronic toxicity test: 0.125, 1.25, 12.5 and 25 mg/L	NA	96 hours (Acute effects) 14 days (chronic effects)	Acute effects: <ul style="list-style-type: none"> No mortality was observed with any of three different sizes at the highest concentration (acute NOAEC > 313 mg/L) Chronic effects: <ul style="list-style-type: none"> Mortality observed at concentrations > 1.25 mg/L for the 0.05 µm particles (LOAEC = 1.25 mg/L), with no effects on fecundity Decrease fecundity in 0.5 µm particles at the lowest concentrations (LOAEC = 0.125 mg/L), and in survival at the highest concentration tested Decrease in fecundity at the lowest concentration tested (LOAEC = 0.125 mg/L) and no effect on survival with the 6 µm beads
Davarpanah & Guilhermino (2015)	Microalgae (<i>T. chuii</i>)	Polyethylene microspheres (1–5 µm)	0.046 to 1.472 mg/l	NA	96 hours	<ul style="list-style-type: none"> No significant effects of microplastics on the growth rate of <i>T. chuii</i> were found up to a concentration of 1.472 mg/l.
von Moos <i>et al.</i> (2012)	Mussel (<i>M. edulis</i>)	Polyethylene (0–80 µm)	2.5 g/L of sea water	NA	Up to 96 hours	Strong inflammatory response, disruption of lysosomal membrane stability
Wright <i>et al.</i> (2013a)	Lugworm (<i>A. marina</i>)	UPVC (mean 230 µm)	0.5–5% by weight (mixed with sediment)	Prolonged residence time	28 days	Reduced feeding activity, reduced lipid and available energy reserve, inflammation

With an estimated 40.5–215 mg of polyethylene (PE) microbead/person/d in the UK (Napper *et al.* 2015), and assuming an average wastewater production of 150 L/person/d, the expected concentration of PE in UK wastewater is 0.27–1.4 mg/L. This is in range of the lowest observable adverse effect concentration (LOAEC) reported in Lee *et al.* (2013), indicating a potential risk to the aquatic environment from wastewater-based microplastics.

Another potential risk of microplastics for aquatic organisms may be from binding of microorganisms, such as pathogens and bacteria, to the microplastic surface, since wastewater-based microplastics have been shown to be covered with a layer of biofilm (Carr *et al.* 2016). Thus, more research is required to understand the possible interactions of microplastics with existing microorganisms in wastewater and during wastewater treatment (such as biological processes), as these microorganism communities may be transported to the receiving environment.

Biological effects from contaminants associated with microplastics

Considering the evidence of TrOC sorption to microplastics, it is conceivable that the toxicity of microplastics to organisms may be enhanced by the toxic effects of sorbed TrOCs. Indeed, studies have suggested that sorbed TrOCs can migrate from microplastics to exposed aquatic organisms (Browne *et al.* 2013; Rochman *et al.* 2013b; Chua *et al.* 2014).

A recent study by Luis *et al.* (2015) demonstrated that the presence of polyethylene microplastics caused a significant increase in the toxicity of Cr (VI) in marine fish (*Pomatoschistus microps*) compared to chromium alone. Likewise, Avio *et al.* (2015) reported significant transfer of pyrene to marine mussels (*Mytilus galloprovincialis*) through ingestion of contaminated polystyrene and polyethylene microplastics.

Some studies have suggested that the transfer of sorbed TrOCs from contaminated microplastics to the organism is likely to be a less significant source compared to the other exposure pathways (Gouin *et al.* 2011; Koelmans *et al.* 2013). Using a biodynamic model to simulate the uptake of TrOCs on plastics in a sediment-water system, Koelmans *et al.* (2013) found that the role of microplastics for TrOC bioaccumulation in marine lugworm was insignificant compared to other TrOC exposure pathways, such as dermal uptake and uptake from sediment. Moreover, a recent study has also demonstrated much lower uptake of PBDE, and subsequently lower health risks, in amphipods tissue

after 72 h exposure to PBDE sorbed to polyethylene microplastics compared to those exposed to PBDE alone (Chua *et al.* 2014). One proposed mechanism that can explain this result is the strong tendency of PBDEs to remain in/on the microplastics rather than being taken up into amphipod tissue. Consequently, such interactions may also reduce the bioavailability of sorbed TrOCs in organisms (Gouin *et al.* 2011; Hirai *et al.* 2011). While this study demonstrated the reduced contribution of hydrophobic TrOCs associated with microplastics to the organism's health compared to the direct exposure pathway, it is thought that microplastics still act as a carrier of TrOCs to organism's tissue (Chua *et al.* 2014).

Thus, it appears that the influence of microplastics on the bioavailability of contaminants and their potential to act as a vector of contaminants to organisms, as well as the degree to which the sorbed contaminants pose a risk, may vary between different types of microplastics, contaminants and organisms (Koelmans *et al.* 2013). Further, the potential of sorbed contaminants to migrate from microplastics to the organism tissue can be influenced by factors including the kinetics of TrOC desorption between plastic and tissues and the residence time of contaminated microplastics in the organism's body (Andrady 2011; Chua *et al.* 2014). There is evidence that the desorption and incorporation of TrOCs from microplastics to the organism's tissue can be facilitated by the organism's digestive fluid through desorbing sorbed TrOCs on contaminated microplastics, which may be enhanced by increasing the retention time of microplastics in the digestive tract (Bakir *et al.* 2014; Chua *et al.* 2014).

The risks associated with microplastics may also come from the chemicals embedded into the plastic polymer during manufacture, particularly if the components are not stable and can leach into the organism's body after ingestion. The biological effects of some additives, including phthalates, bisphenol A, alkylphenols and PBDEs, on different wildlife species has been widely reported (Foster 2006; Richter *et al.* 2007; Oehlmann *et al.* 2009; Jarmolowicz *et al.* 2013). The concerns regarding leaching of plastic additives in aquatic organisms is associated with microplastics derived from the breakdown of larger plastic objects in the environment, but there is limited experimental data regarding adverse biological effects from additives incorporated in wastewater-based microplastics, such as microbeads and fibres. For example, inclusion of PBDEs as flame retardant in textiles and clothing is reported extensively (Rahman *et al.* 2001; Frederiksen *et al.* 2009). Brominated and chlorinated phthalocyanines are other fire retardant additives

that can be introduced to enhance the thermal stability of fabric (Gladunova *et al.* 2013), and can be toxic to marine organisms (Carmen & Daniela 2012). However, the leaching rate of the above contaminants from wastewater-based microfibers, as well as potential risks for the variety of aquatic organisms, is still unexplored.

CONCLUSIONS AND FUTURE AREA OF RESEARCH

The concerns regarding microplastics in WWTP effluent has rapidly increased in the last few years and preliminary research has confirmed wastewater as a source of microplastics in the receiving environment, with the amount contributed depending on the level of wastewater treatment. Due to the fact that even advanced treatment technologies cannot always remove 100% of microplastics (Schneiderman 2015; Talvitie *et al.* 2015), preventing the release of microplastics to wastewater influent would be the best action to reduce the discharge of wastewater-based microplastics to the aquatic environment. While preventive measures have been taken to remove some microplastic beads from the personal care products, this is more challenging for other sources of microplastics, particularly as synthetic clothing is also expected to be an important source of microfibrils in wastewater. Consequently, further research on developing a sound mitigation strategy to prevent release to the environment would be beneficial.

This review has identified that there are still research gaps in this recently developing field of research, specifically: (i) to investigate the contribution of wastewater discharge from WWTPs to microplastics in the aquatic environments compared to other potential sources, (ii) to understand the behaviour of microplastics in WWTPs, and their physical and chemical modifications through wastewater treatment processes, and (iii) to understand the impact of sorption to microplastics on contaminant fate and toxicity in a more complex mixture that resemble wastewater. This research is necessary to better understand the relative contribution and significance of wastewater-based microplastics.

Since some wastewater-based microplastics from WWTPs can be transported from the discharged area and dispersed widely in the aquatic environment, collaborations at regional and global levels are required to establish management frameworks in terms of the maximum levels of microplastics in the wastewater effluent and to reduce the use of treated sewage sludge or biosolids with high concentrations of microplastics.

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