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Microplastics in freshwater and terrestrial environments: evaluating the current understanding to identify the knowledge gaps and future research priorities

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Abstract

Plastic debris is an environmentally persistent and complex contaminant of increasing concern. Understanding the sources, abundance and composition of microplastics present in the environment is a huge challenge due to the fact that hundreds of millions of tonnes of plastic material is manufactured for societal use annually, some of which is released to the environment. The majority of microplastics research to date has focussed on the marine environment. Although freshwater and terrestrial environments are recognised as origins and transport pathways of plastics to the oceans, there is still a comparative lack of knowledge about these environmental compartments. It is highly likely that microplastics will accumulate within continental environments, especially in areas of high anthropogenic influence such as agricultural or urban areas. This review critically evaluates the current literature on the presence, behaviour and fate of microplastics in freshwater and terrestrial environments and, where appropriate, also draws on relevant studies from other fields including nanotechnology, agriculture and waste management. Furthermore, we evaluate the relevant biological and chemical information from the substantial body of marine microplastic literature, determining the applicability and comparability of this data to freshwater and terrestrial systems. With the evidence presented, the authors have set out the current state of the knowledge, and identified the key gaps. These include the volume and composition of microplastics entering the environment, behaviour and fate of microplastics under a variety of environmental conditions and how characteristics of microplastics influence their toxicity. Given the technical challenges surrounding microplastics research, it is especially important that future studies develop standardised techniques to allow for comparability of data. The identification of these research needs will help inform the design of future studies, to determine both the extent and potential ecological impacts of microplastic pollution in freshwater and terrestrial environments.

Introduction

Research on microplastics as an environmental contaminant is rapidly advancing. Although marine microplastics research remains at the forefront, in recent years researchers recognising the comparative lack of studies on microplastics in freshwater environments have begun to address this field as a matter of priority, quantifying microplastics in lake and river systems and assessing exposure to, and uptake by, organisms (Dris et al., 2015b; Wagner et al., 2014). Despite the knowledge that microplastics (and indeed plastics of all sizes) are also widespread within terrestrial environments as a result of human activities, there is a dearth of studies that have quantified microplastics in terrestrial environments. In fact, much of the existing information about the environmental presence of microplastics considers terrestrial and freshwater environments only as sources and transport pathways of microplastics to the oceans. However, given that the majority of all plastics will be used and disposed of on land, both terrestrial and adjacent freshwater environments will themselves be subject to extensive pollution by plastics of all sizes, based on large amounts of anthropogenic litter from both point (e.g. wastewater treatment discharge, sewage sludge application) and diffuse (e.g. general littering) sources. As such it is highly likely that soils will act as long term sinks for microplastic debris (Rillig, 2012; Zubris and Richards, 2005). Hence it is important to understand release rates, fate and transport of microplastics entering terrestrial systems as well as freshwater systems in order to allow for the assessment of hazards and risks posed by microplastics, and indeed plastics in general, to ecosystems.

The aim of this review is to synthesise available information relevant to understanding microplastics behaviour, fate and ecological effects within freshwater environments and soils. The review draws primarily on the published literature available from freshwater and the relatively few terrestrial microplastic studies published to date, setting out the key factors that will influence microplastic distribution, fate and exposure. One important consideration is that the processes governing distribution and exposure to plastics are not necessarily exclusive to a specific environmental ‘compartment’ (e.g. plastics within a shallow freshwater system may be exposed to

similar levels of UV radiation as a particle in coastal marine systems) and plastics can be transported between compartments (e.g. from land to rivers and the sea, and from rivers and sea to land during flooding, storm events or tidal surges). Therefore it is not realistic to consider such studies in isolation from the body of marine work. Thus, where appropriate, we also include key studies from the extensive body of marine literature that will inform knowledge of the processes likely to occur in freshwaters and soils.

Microplastics as a term has quite a broad definition and can refer to a wide range of polymers, particle sizes and densities (see section 2). In this review we will predominantly focus on microplastics defined as being any polymer within the size range 1 µm to 5 mm as this is the size range which has been the major focus of reported microplastics research to date. Where information is available, we have in places included relevant information from reported studies for nanoplastics (< 100nm) as contaminants that are also likely to occur in soils and water. For the purposes of this review, microplastics and nanoplastics have been defined as per the study in which they were used/discussed and parallels drawn between the two where appropriate. However, we do not intend to carry out a complete review of nanoplastics or compare them with other nanomaterials as this topic has been has been previously addressed (Syberg et al., 2015). Finally in places throughout the text, we also use the term "plastics" to refer to plastics as a whole class (macro-, micro- and nano-sized plastics). This is in order to capture the relevant influence of processes such as wind or water flow, exposure to UV, temperature fluctuations and associations with organic matter that can, alone or together, commonly affect the fate and behaviour with different sized plastic materials. The reality is that there are likely to be significant similarities between the effects and behaviours of plastics of different size classifications, for example when comparing 'large nanoplastics' to 'small microplastics'. As the size and state of plastics within the environment can change with time, we believe it is necessary to include information that extends beyond plastics in the micron size range to fully understand the drivers of microplastic and indeed all plastic transport, fate and resulting bioavailability.

Available information on plastic usage and presence on land is used in order to make informed estimations about the likely presence and effects of microplastics within terrestrial environments. This includes considering relevant data on plastic sources and transport through different environmental compartments, and therefore the organisms that may encounter and be affected by these plastics. We evaluate the available literature on ecological effects of microplastics to freshwater species (using both studies with freshwater species and any studies in comparable marine species) that can be directly related to organisms occupying the same ecological niche within aquatic and terrestrial environments. Finally, we review chemical associations and plasticiser leaching, including examples from microplastics and also large plastic products ('macroplastics') that may have implications for the toxicity of microplastics within freshwater and terrestrial environments. If we are to fully understand or predict the effects of microplastic pollution within the environment as a whole, a multidisciplinary approach will be needed to integrate knowledge on presence and behaviour of plastic waste, particles and associated chemical pollution in the environment. Our review sets out to reflect this by drawing together knowledge from all relevant fields including waste management, nanotechnology, agriculture and toxicology. By using all available knowledge we are able to establish how previous studies can inform our knowledge of presence and effects of microplastics in terrestrial and freshwater environments and, thus, make recommendations for further research.

2. Plastic as an environmental contaminant

2.1. Plastic pollution in the environment

In 2014, annual plastic production exceeded 311 million tonnes, an increase of nearly 84 million tonnes since 2004 (PlasticsEurope, 2015; Thompson et al., 2005). By 2050 it is estimated that this may increase to a colossal 33 billion tonnes (Rochman et al., 2013a). Of anthropogenic waste materials released to the environment, plastic can constitute up to 54% by mass (Hoellein et al., 2014). Established widespread uses of plastic include packaging materials (39.5% total plastic production),

120 building materials (20.1%), automotive components (8.6%), electronic appliances (5.7%) and
121 agricultural materials (3.4%), with the remainder including products such as household appliances and
122 sporting equipment (PlasticsEurope, 2015). There are approximately 30,000 different polymer
123 materials registered for use in the European Union. A 'polymer' is difficult to characterise as definitions
124 will vary between manufacturers, with much information commercially confidential. However the
125 European Commission report states that 84% of this 30,000 are represented by thermoplastics (Postle
126 et al., 2012). Although they share similar characteristics, each polymer has different physical
127 properties with respect to their plasticity and density. The density of the material in particular will be
128 important for determining environmental fate. For example, density will influence how particles
129 partition in the aquatic environment including whether they float on water surfaces or settle to
130 sediment and the ease with which they will be transported by wind action across land (Zylstra, 2013).
131 However, even when properties are known, it can be difficult to predict the fate of polymers. For
132 example, it has been observed that supposedly buoyant particles such as polyethylene and
133 polypropylene can be retained within sediments (Horton et al., 2016). This could be due to biofouling
134 or agglomeration with organic materials. These differences highlight polymers to be complex
135 environmental pollutants.

136 For many plastic products their useful lifetimes are often relatively short. This is especially the
137 case for single-use packaging materials. However, the qualities which make plastic a good material for
138 consumer products: waterproof, durable and resistant to wear and biodegradation, can also make
139 plastic extremely persistent (Barnes et al., 2009; Imhof et al., 2012). Many commonly-used polymers
140 are extremely resistant to biodegradation, for example polyethylene and polystyrene (Gautam et al.,
141 2007). Common characteristics of plastics that can impede biodegradation are high molecular weight,
142 hydrophobicity and cross-linked chemical structure (Gautam et al., 2007; Shah et al., 2008). There is
143 evidence that biodegradation of polymers by some organisms can occur, for example bacteria, fungi
144 and mealworms (due to gut bacteria) (Gu, 2003; Yang et al., 2015a, b). However, when biodegradation
145 does occur, it is reliant on exposure of polymers to these and other specific degrading organisms that

146 have the ability to degrade these specific polymers – conditions that may not necessarily be
147 encountered in the environment. Indeed it has been proposed that no polymers can be efficiently
148 biodegraded in landfill sites (Shah et al., 2008). Therefore, apart from incineration, it is understood
149 that the vast majority of plastic ever made is still present in the environment in some form (Barnes et
150 al., 2009; Thompson et al., 2005). It is this persistence that makes plastic pervasive as an
151 environmental pollutant and is a main driver underpinning current concerns about the possible
152 ecological impacts of the growing burden of plastic materials present in ecosystems. Plastic litter is
153 present in terrestrial, freshwater, estuarine, coastal and marine environments, particularly in
154 urbanised regions (Cole et al., 2011; Free et al., 2014; Zylstra, 2013). Plastics have been observed even
155 in remote areas of the world including deep-sea sediments (Van Cauwenberghe et al., 2013; Woodall
156 et al., 2014), submarine canyons (Pham et al., 2014) and encapsulated in Arctic sea ice (Obbard et al.,
157 2014), far from any potential land-based source. It has even been observed in some locations that
158 plastic debris can fuse together, becoming associated with volcanic rocks, sediment and organic
159 materials forming 'plastiglomerates', solid rock-like substances, that have the potential to become
160 preserved in the fossil record. As human influence begins to dominate even the most fundamental
161 processes on earth, the potential for this evidence of human impact to last far into geological records
162 has prompted the suggestion that we are moving into a new geological epoch from the Holocene to
163 the 'Anthropocene' (Corcoran et al., 2014).

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165

166 **2.2. Microplastics: a brief background**

167 Plastic debris is broadly classified by size: mega-debris (> 100 mm), macro-debris (> 20 mm),
168 meso-debris (20-5 mm) and micro-debris (< 5 mm) (Barnes et al., 2009). Although microscale plastic
169 particles were first observed in the marine environment in the early 1970s (Buchanan, 1971; Carpenter
170 and Smith, 1972), it was not until 2004 that the term "microplastic" became commonly used as the

result of a study by Thompson et al. (2004). Microplastics are now commonly defined as particles with the largest dimension smaller than 5 mm, although no lower size limit has been specifically defined (Arthur and Baker, 2009; Duis and Coors, 2016; Faure et al., 2012). It is understood that plastic particles in the environment will continue to degrade and become steadily smaller, eventually forming 'nanoplastics' (Koelmans et al., 2015; Mattsson et al., 2015). Microplastics in environmental samples can currently be detected down to a size of 1 µm, however few environmental studies identify particles <50 µm due to methodological limitations (Hidalgo-Ruz et al., 2012; Imhof et al., 2016).

Microplastics fall within two categories: primary and secondary. Primary microplastics are specifically manufactured in the micrometre size range, for example those used in industrial abrasives for sandblasting, either acrylic or polyester beads (von Moos et al., 2012; Zitko and Hanlon, 1991), plastic pre-production pellets ('nurdles') or in personal care products such as exfoliating agents in creams and cleansers containing polyethylene 'microbeads' (Napper et al., 2015). Primary microplastic particles are likely to be washed down industrial or domestic drainage systems and into wastewater treatment streams (Fendall and Sewell, 2009; Lechner and Ramler, 2015). Despite the capability of some sewage treatment works to remove up to 99.9% microplastic particles from wastewater (dependent on the processes employed by the treatment plant), the sheer number of particles entering the system may still allow a significant number to bypass filtration systems and be released into the freshwater environment with effluent (Carr et al., 2016; Murphy et al., 2016).

Secondary microplastics are formed as a result of meso and macroplastic litter fragmentation. Plastics are susceptible to the effects of UV radiation and high temperatures which can cause chemical changes making plastics brittle and thus more susceptible to fragmentation (Andrady, 2011; Barnes et al., 2009; Hidalgo-Ruz et al., 2012; Ivar do Sul and Costa, 2014; Rillig, 2012; Shah et al., 2008). Fragmentation increases surface area and number of particles per unit of mass. Both exposure to sunlight and wave action are primary causes of fragmentation in marine waters. On land, especially at the soil surface, fragmentation of plastics is thought to occur readily as a result of direct exposure to UV radiation from sunlight, aided also by temperature fluctuations which will generally be greater

than those in sea water (Andrady, 2011). Similarly, exposure to UV may be higher in small shallow aquatic systems such as ponds and rivers than in large lakes or the open ocean. However many freshwater environments may lack the fragmentation potential that is offered by turbulence and wave action in coastal waters, especially in rocky tidal areas (Barnes et al., 2009). An additional source of secondary microplastics is derived from synthetic fabrics, which can shed up to 1900 fibres per garment during washing (Browne et al., 2011). Although microfibres are secondary particles they will be released to the environment along with primary microplastics through wastewater effluents and sludge application. Hence in this respect the fate and transport of these fibres may be more closely aligned with that of primary microplastics, based on similar release routes.

3. Sources, environmental presence and transport of microplastics

3.1. Sources of microplastics to freshwater and terrestrial environments

A significant direct input of primary microplastics to terrestrial environments has been identified as being through the application of sewage sludge containing synthetic fibres or sedimented microplastics from personal care or household products to land (Habib et al., 1996; Zubris and Richards, 2005). Polymers used in synthetic textiles include polyester and nylon, while polyethylene or polypropylene are commonly used as microbeads or glitter in cosmetics. As sewage treatment works are efficient in removing the majority of microplastic particles from wastewater, many of the particles that are removed will be retained within the sludge (Magnusson and Norén, 2014; Mintenig et al., 2017). This suggests that the major routes of release for secondary microfibres and primary microplastics are the same. In Europe it is common practice to compost and pasteurise sewage sludge for use as agricultural fertiliser as well as dispose of large quantities of sludge produced by wastewater treatment to land (DEFRA, 2012). Between four and five million tons dry weight of sewage sludge are applied to arable land every year in the European Union (Cieślik et al., 2015; Willén et al., 2016), although application rates are highly variable between countries (Nizzetto et al., 2016b). Despite

222 regulations on harmful substances within sludge applied to land, microplastics are not yet considered
223 by these and thus the mass of microplastics inadvertently applied to land annually may exceed
224 400,000 tonnes – higher than the mass currently estimated to be present in oceanic surface waters
225 worldwide (Nizzetto et al., 2016b). Zubris & Richards (2005) found that soils with a known history of
226 sewage sludge application contained significantly higher concentrations of synthetic microfibres than
227 soils which had not received sewage sludge. In some field sites, synthetic microfibres were found 15
228 years after the last sludge application (Zubris and Richards, 2005). This suggests that microplastics and
229 synthetic fibres are likely to accumulate in soils after repeated sludge applications.

230 Those particles that are not retained within the sewage sludge, or removed by skimming
231 during the treatment process, will enter the environment via effluent input to rivers. For primary
232 microplastics and secondary microfibres, effluent from sewage treatment is thought to be a major
233 source of microplastics to freshwater bodies. Synthetic microfibres have been identified by many
234 studies as the most abundant microplastic particle type found throughout freshwater, terrestrial and
235 marine environments (Browne et al., 2011; Dubaish and Liebezeit, 2013; Free et al., 2014; Zubris and
236 Richards, 2005), with primary microbeads from personal care products also likely to be a significant
237 contributor to microplastic pollution (Castañeda et al., 2014; Murphy et al., 2016; Napper et al., 2015).
238 However, it must be noted that the sampling equipment and methodology will influence the size of
239 particles observed, and therefore may determine the dominant particle type observed. For example,
240 because fibres have at least one very small dimension, they may not always be retained on a mesh
241 even if the length of the fibre exceeds the mesh size. This variation in sampling methodology could
242 lead to fragments or pellets being erroneously identified as the most abundant particle type and may
243 make comparison of particle types and abundances between studies difficult (Dris et al., 2015b; Ivleva
244 et al., 2016).

245 Due to the small size of primary microplastics they are unlikely to be removed by existing
246 screening of debris, with coarse screens retaining particles >10 mm and even the finest screens
247 retaining particles >1.5 mm (Fendall and Sewell, 2009). An important predictor of microplastic

248 partitioning in sewage treatment will be particle density, with dense particles settling to sludge and
249 buoyant particles floating in effluents (Fig. 1). The extent to which this occurs will also depend on a
250 number of relevant processes that may affect the characteristics of the microplastics. For example,
251 the aggregation of microplastic particles, either with themselves or more likely with other (organic)
252 particulate materials can increase size and density leading to an increase in sedimentation rate (Long
253 et al., 2015). The growth of bacterial biofilms on microplastic surface may again increase particle
254 weight and density, resulting in settling (Cozar et al., 2014; Kowalski et al., 2016; Moret-Ferguson et
255 al., 2010).

256 Figure 1 shows a schematic diagram of waste water treatment processes and how particle
257 partitioning is likely to occur through processing. Removal of coarse debris with physical screens,
258 primary settling lagoons and aerobic oxidation are common across many treatment plants, additional
259 settling lagoons and tertiary treatments may also be present. Plastic materials will generally not be
260 degraded at any point throughout the process and as a consequence, any plastic not removed for
261 disposal during the initial filtering steps will remain in the solids or the effluent after processing. Many
262 microplastics from sewage treatment works will therefore ultimately be directly released to the
263 environment in effluents or through sludge application to land. Other methods of sludge disposal
264 include landfilling, incineration and even in production of cement for use in construction. In these
265 cases, plastic particles are likely to be well-contained and so unlikely to leach into the surrounding
266 environment (Browne et al., 2011; Cieślik et al., 2015; Dubaish and Liebezeit, 2013; Rillig, 2012; Zubris
267 and Richards, 2005).

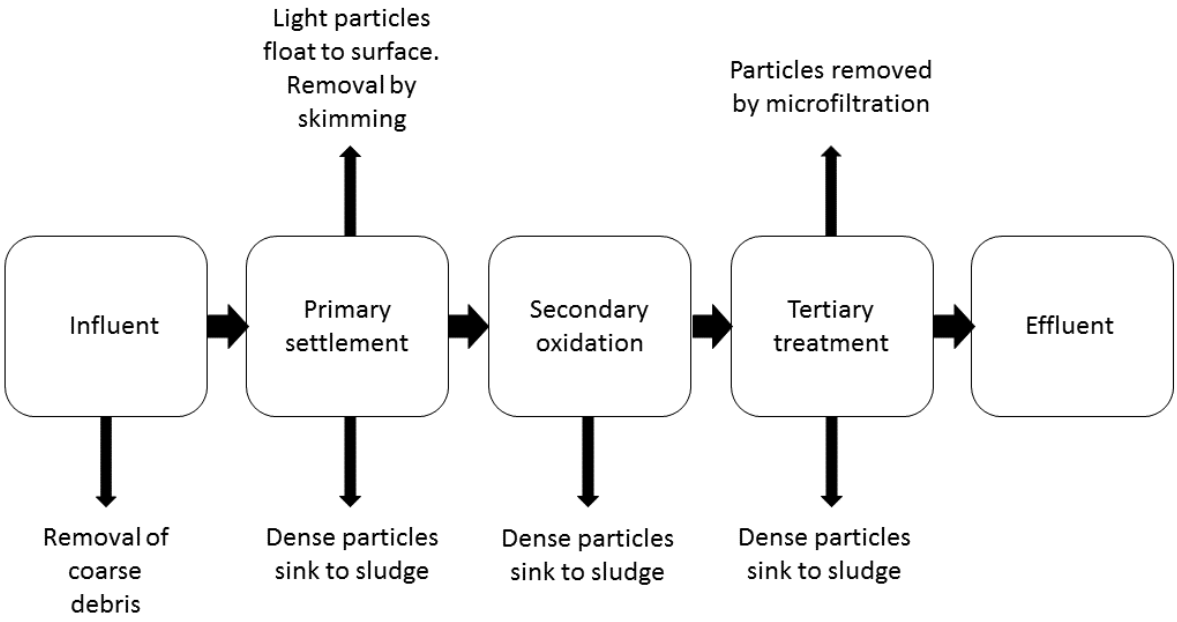

Figure 1. Schematic diagram of standard wastewater treatment processes and particle behaviour influenced by density at each stage of treatment. Adapted from Baird and Cann (2012).

A recent study observed microbeads originating from cosmetic products in wastewater treatment influents and effluents at seven wastewater reclamation plants in California, in which waste waters were treated for reuse with tertiary treatment. The treatment processes at these plants resulted in the complete removal of microparticles (45–400 µm) from water outputs, as a result of tertiary treatment including surface skimming, sludge settling and microfiltration processes (Carr et al., 2016). After secondary treatment only (elimination microfiltration), effluents contained on average one plastic particle per 1140 litres of effluent, compared to an estimated one particle per litre in the influent (Carr et al., 2016). No fibres were found despite these being the most frequently reported kind of microplastics found in environmental samples, however as previously highlighted, this may be a result of the sampling technique used. Murphy et al. (2016) similarly found that microplastics were significantly reduced in effluent following a secondary treatment process. In this study, plastic flakes and fibres were the two most abundant microplastic types (67.3% and 18.5%

285 respectively), with microbeads only contributing to 3% of total particles. For this mixture of materials,
286 average microplastic concentrations reduced from 15.7 particles litre^{-1} (± 5.23) in sewage treatment
287 influents to 0.25 particles litre^{-1} (± 0.04) in final effluents, which represents a 98% reduction in
288 microplastic concentrations (Murphy et al., 2016). Other recent studies have reported similar high
289 removal rates: 95% (Talvitie et al., 2017), 97% (Mintenig et al., 2017) and 99% (Magnusson and Norén,
290 2014). Notably, these proportions of partitioning between solid waste and effluent are similar to
291 estimates that have been provided for nanomaterials: 90% removal of titanium (Ti) associated with
292 titanium dioxide (TiO_2) nanoparticles (Johnson et al., 2011), 96% removal of Ti (Westerhoff et al.,
293 2011), 94% removal of surfactant-coated silicon dioxide (SiO_2) nanoparticles (Jarvie et al., 2009). This
294 suggests that similar processes may affect the fate of microplastics as they do poorly soluble and
295 potentially inert nanomaterials such as gold and titanium dioxide during waste water treatment (e.g.
296 heteroaggregation), and highlights the importance of interdisciplinary research for understanding the
297 fates and behaviours of microplastics and nanoparticles and the parallels that can be drawn between
298 them (Bouwmeester et al., 2015; Syberg et al., 2015). Despite the significant removal of particles from
299 treated wastewater, given the large volumes passing through wastewater treatment plants the
300 remaining 5%, or less, of the microplastics that are not filtered out will likely represent a large number
301 and mass entering the freshwater environment in effluent (Murphy et al., 2016; Ziajahromi et al.,
302 2016). It is also important to note that these results are based on efficient current-generation
303 wastewater treatment processes that may not be widely available or utilised worldwide. In many
304 countries, untreated sewage is input directly to watercourses without treatment (Duis and Coors,
305 2016; Hammer et al., 2012). Where the most modern facilities are not available, these estimates could
306 fall short by up to 100-fold in places.

307 Sources of secondary microplastics derived from plastic litter are both numerous and diverse,
308 ranging from releases during municipal solid waste collection, processing and land-filling, release from
309 transportation and disposal systems to individuals creating litter either accidentally or intentionally
310 (Fig. 2). This includes large plastic items and sanitary waste input to rivers via combined sewage

311 overflows (CSOs). Runoff via drainage ditches from agricultural land, or storm drains from roads
312 containing plastics such as tyre wear particles, vehicle-derived debris or fragments of road-marking
313 paints is another significant source of riverine microplastic loads (Browne et al., 2010; Eriksen et al.,
314 2013; Galgani et al., 2015; Horton et al., 2016; Tibbetts, 2015). Additionally, wind action may also
315 transport lighter plastic items into water bodies or across land (Zylstra, 2013) and there is evidence to
316 suggest that anthropogenic fibres can be transported and deposited by atmospheric fallout. This
317 appears to be especially significant in urban areas, with deposition increasing during periods of rain
318 (Dris et al., 2016). Although the fibres found in atmospheric studies were not exclusively synthetic
319 (<33% fibres were pure polymers), with an estimated deposition of between 3-10 tonnes of fibres
320 deposited annually in an area approximately 2500 km^2 (based on the Paris region), this may therefore
321 still represent a significant pathway of microplastics from consumer products to the environment (Dris
322 et al., 2017; Dris et al., 2016). Airborne particles are determined to originate from a variety of sources
323 including construction materials, artificial turf and household dust (Magnusson et al., 2016).

324 Another direct source of secondary microplastics to land is the use and fragmentation of
325 agricultural plastics. For example, plastic mulches and polytunnels are used to control temperature
326 and moisture, and retard weed growth in agricultural and horticultural applications (Kasirajan and
327 Ngouajio, 2012; Kyrikou and Briassoulis, 2007; Rillig, 2012; Steinmetz et al., 2016). Polymer seed
328 coatings can also be used to control germination (Clayton et al., 2004). These may consist of various
329 polymers and often contain incorporated pesticides and fertilisers. Commonly used polymers for seed
330 coatings are non-biodegradable and therefore following germination, will remain in the soil (Schultz
331 et al., 2014; Turnblad and Chen, 1998). Additional products used in agriculture include bale twines and
332 wraps, containers, packaging and netting, all of which have the potential for dispersal within the
333 environment (Scarascia-Mugnozza et al., 2012). Exposure of these materials to sunlight and high
334 temperatures may lead to their relatively rapid fragmentation after which they are difficult to
335 completely remove from soils. Dense polymers are more likely to remain in soil and ultimately to be
336 transported into deeper soil layers, whereas lighter polymers will be more likely to be transported by

337 wind and water action either to other terrestrial locations or to surface waters. To our knowledge, to
338 date there are no studies which quantify microplastic presence at terrestrial field sites. Based on the
339 above evidence, however, it is highly likely that microplastics will be present within terrestrial
340 environments and, if investigated in detail, may be found to be as equally pervasive as they are in
341 freshwater and marine environments (Nizzetto et al., 2016a).

342

343 **3.2. Presence of microplastics in the freshwater environment**

344 Studies of microplastics in freshwater environments are rapidly advancing, with microplastic
345 particles found across a range of freshwater environments worldwide, including lakes and rivers. Area
346 of water surface, depth, wind, currents and density of particles are all factors determining transport
347 and fate of particles within these aquatic systems (Eriksen et al., 2014; Eriksen et al., 2013; Fischer et
348 al., 2016; Free et al., 2014). Given the lack of terrestrial studies to date, it is necessary to use our
349 knowledge of microplastics in the freshwater environment, notably sediments, to infer the presence
350 and behaviour of microplastics in soils and to inform future sampling efforts.

351 A study carried out on lake beaches by Imhof et al (2013) measured microplastics found in
352 sediments of two beaches on the north and south shores of Lake Garda (Italy). Particle numbers
353 between these sites were significantly different, with these differences attributed to the prevailing
354 southerly wind direction transporting plastics either directly or by surface water movement to the
355 opposite shore (Imhof et al., 2013). The number of local sources, together with factors including water
356 surface area, depth, wind, currents and density of particles are all factors determining transport and
357 fate of particles within these aquatic systems and can lead to large variation, even within a relatively
358 small area (Castañeda et al., 2014; Eriksen et al., 2014; Eriksen et al., 2013; Fischer et al., 2016; Free
359 et al., 2014). Another significant factor influencing particle presence and abundance is urbanisation of
360 the area surrounding and influencing the waterbody. Eriksen et al. (2013) conducted a study in the
361 Great Lakes (USA) and found that downstream of highly populated Detroit and Cleveland metropolitan

areas, particle concentrations ranged from 280,947-466,305 particles km^{-2}. In Lake Huron, where the shorelines are less influenced by the presence of major urban centres, particle concentrations estimated from sampling were generally orders of magnitude lower, ranging from 456-6541 particles km^{-2}, with one trawl finding no particles (Eriksen et al., 2013). A similar study of the remote lake Hovsgol (Mongolia) also found microplastics present in all samples at concentrations comparable to those found in the Great Lakes (Table 1). Although the area surrounding Lake Hovsgol has a low population density, poor local waste management and inputs of wastewater are blamed for the presence of microplastic particles in the lake (Free et al., 2014). Additionally, the smaller volume of Lake Hovsgol, compared to the Great Lakes of the USA, may be an important reason for microplastic concentrations being comparable between these two studies.

Urbanisation has also been observed to be a significant factor influencing presence of microplastics in riverine environments, with plastics being introduced from a variety of sources including effluent, road runoff, littering and atmospheric deposition (discussed further in Section 3.1). Mani et al. (2015) and Yonkos et al. (2014) are among those who have found microplastics in higher abundances at sites in close proximity to urban areas than at more remote sites. However, although particle numbers are regularly found to be high near urban areas, this is not the only factor influencing presence of microplastic particles. For example, Horton et al. (2016), in addition to finding high numbers of particles downstream of urban discharge points, also found particles in rural areas where few human-associated inputs would be expected.

Given the growing need to make comparative assessments in order to identify regional, national and global trends in microplastic distribution, it would be desirable to be able to collate the available data to conduct meta-analyses. However, a major challenge to this is that no standard protocol for collecting particles from environmental samples exists, with different authors using different approaches. While many studies use broadly similar techniques to extract microplastics from environmental samples, including size fractionation, digestion of organic matter and density

separation, the specific parameters of methods differ between studies regarding volume of sample studied, upper and lower particle size limits, density separation media and particle identification criteria (Besley et al., 2016; Hidalgo-Ruz et al., 2012). Given that many methods currently rely on visual identification, there are also many opportunities for the introduction of sampling error, bias or omission of particles of certain size or density, leading many results to be qualitative rather than quantitative (Ivleva et al., 2016). Although many studies have established 'standard methods' for particle extraction in an effort to introduce consistency across studies, these methods are in fact quite disparate. Moreover, studies are still identifying new and reportedly more effective criteria. Thus no standardised methods have yet been agreed (Hidalgo-Ruz et al., 2012; Syberg et al., 2015). An additional issue is the use of non-standard units of measurement for reporting microplastic concentrations. In order to compare studies where units are not consistent, units must be transformed to units per volume, either as particles per litre of sampled water or as particles per kilogram of sediment (see Table 1). It is therefore of utmost importance that authors detail results in all units, or provide sufficient detail on the sampling methodology to do so (Phuong et al., 2016; Van Cauwenberghe et al., 2015). These differences between studies highlight the need for continued efforts to standardise methods for microplastic extraction and quantification, as has been recognised in environmental nanomaterial research (Delay et al., 2010).

Table 1. Summary of selected freshwater microplastic environmental sampling studies, covering a range of freshwater environments (water, plus benthic and shore sediments of lakes and rivers). Selected studies were those which quantified specifically microplastics and provided sufficient methodological detail to allow for conversion of units, to standardise by volume or mass for comparability.. Converted units for water and sediment were calculated by multiplying area sampled by sampling depth to estimate total volume, then converting this volume into litres or kg (dry weight). For sediment this calculation is based on typical dry sediment bulk density of 1.3 g cm^{-3} (Sekellick et al., 2013) Conversion was not required where the study already reports results as particles L^{-1} or kg^{-1}. For details of additional freshwater studies, refer to (Dris et al., 2015b).

Water body type	Sample type	Sample location and description	Study findings (reported units)	Study findings (converted units)	Study
Lake	Water	Great Lakes (USA) 16 cm sampling depth	Average particle concentration 43,000 km^{-2}	Average 0.00027 particles L^{-1}	Eriksen et al. (2013)
Lake	Water	Lake Hovsgol (Mongolia), sampling depth 16 cm	Average particle concentration 20,264 km^{-2}	Average 0.00012 particles L^{-1}	Free et al. (2014)
Lake	Benthic sediment	Lake Ontario (Canada) sampling depth 8 cm	26 particles in 42.2 g (station 403) 9 particles in 103.2 g (station 208)	616.1 particles kg^{-1} (station 403) 87 particles kg^{-1} (station 208)	Corcoran et al. (2015)
Lake	Shore sediment	Lake Garda (Italy), sampling depth 5 cm	Average particle abundance 1108 and 108 m^{-2} (north and south shores respectively)	Average 17 particles kg^{-1} (north) 1.7 particles kg^{-1} (south)	Imhof et al. (2013)
Lake	Shore sediment	Lake Garda (Italy), sampling depth 5 cm	Average particle abundance 75 m^{-2}	Average 1.2 particles kg^{-1}	Imhof et al. (2016)
Lake	Shore sediment	Various lakes (Switzerland), sampling depth 5 cm	Average particle abundance 1300 m^{-2}	Average 20 particles kg^{-1}	Faure et al. (2015)
Lake	Water and shore sediment	Lake Chiusi and Lake Bolsena (Italy)	Average particle abundance 234 kg^{-1}, 3.02 m^{-3} surface water (Chiusi) Average particle abundance 112 kg^{-1}, 2.51 m^{-3} surface water (Bolsena)	Average 0.03 particles L^{-1} surface water (Chiusi) Average 0.025 particles L^{-1} surface water (Bolsena)	Fischer et al. (2016)
Lake	Water and benthic sediment	Taihu Lake (China)	Particle abundance range: 3.4 – 25.8 L^{-1} surface water 11 – 234.6 kg^{-1} benthic sediment	-	Su et al. (2016)
Lake	Benthic and shore sediments	Lake Ontario (Canada)	Average particle abundance 980 kg^{-1} lake benthic 140 kg^{-1} lake beach	-	Ballent et al. (2016)
River	Water	Great Lakes tributaries (USA)	Particle abundance range: 0.05 – 32 m^{-3}	0.00005 – 0.032 particles L^{-1}	Baldwin et al. (2016)
River	Water	River Seine, urban area (Paris, France)	Average particle abundance 30 m^{-3} (plankton trawl) Average particle abundance 0.35 m^{-3} (manta trawl)	Average 0.03 particles L^{-1} Average 0.00035 particles L^{-1}	Dris et al. (2015a)
River	Water	Various rivers (Switzerland)	Average particle abundance 7 m^{-3}	Average particles 0.007 L^{-1}	Faure et al. (2015)
River	Water	River Danube (Austria)	Average particle abundance 316.8 m^{-3}	Average 0.32 particles L^{-1}	Lechner et al. (2014)
River	Water	River Rhine (various) sampling depth 18 cm	Average particle abundance 892,777 km^{-2}	Average particles 0.005 L^{-1}	Mani et al. (2015)

River	Water	Nine different rivers, Chicago area (USA)	Average particle abundance 2.4 m^{-3}, upstream sewage treatment works (STW) Average particle abundance 5.7 m^{-3}, downstream STW	Average particles 0.002 L^{-1} Average particles 0.006 L^{-1}	McCormick et al. (2014)
River	Water	Rivers: Papatsco Corsica Rhode Magothy Sampling depth 15 cm	Average particle abundance 155,374 km^{-2} 40,852 km^{-2} 67,469 km^{-2} 112,590 km^{-2}	Average particles 0.001 L^{-1} 0.00027 L^{-1} 0.00045 L^{-1} 0.00075 L^{-1}	Yonkos et al. (2014)
River	Shore sediment	Rivers Rhine and Main (Germany)	Particle abundance range: 228 - 3763 kg^{-1}	-	Klein et al. (2015)
River	Benthic sediment	Lake Ontario tributaries (Canada)	Average particle abundance 610 kg^{-1}	-	Ballent et al. (2016)
River	Benthic sediment	St Lawrence river sediments, sampling depth 10-15 cm (Canada).	Average particle abundance 13,759 m^{-2}	Average approx. 70.6-105.8 particles kg^{-1} (depending on depth sampled)	Castañeda et al. (2014)
River	Benthic sediment	River Thames Basin (UK), sampling depth approx. 10cm	Average particle abundance range: 185 kg^{-1} to 660 kg^{-1} depending on site.	-	Horton et al. (2016)
River	Benthic sediment	Beijiang River (China)	Particle abundance range: 178 - 554 particles kg^{-1}	-	Wang et al. (2016)

The numbers of particles reported in marine and freshwater surface waters are extremely variable. Concentrations of microplastics in marine surface waters have been reported from 0.0005 particles L^{-1} (Carson et al., 2013) (calculated as per Table 1) to 16 particles L^{-1} (Song et al., 2014) with a range of intermediate concentrations reported (Lusher et al., 2014; Zhao et al., 2014). Studies of freshwater surface samples generally show concentrations comparable to the lower end of the reported marine surface concentrations such as those seen by Carson et al. (2013) (see Table 1). Dris et al. (2015a) highlight the consequence of using different mesh sizes when determining the number of particles observed. When sampling with a plankton net (80 µm mesh), up to 100-fold more particles can be collected compared to use of a manta net (330 µm mesh). This effect of mesh size is an important consideration when comparing surface water studies, as differences in sampling method and equipment may lead to inconsistencies that prohibit the comparability of datasets (Cole et al., 2011). However, despite this variation, it remains possible that freshwater concentrations comparable to the higher marine concentrations will be found, likely within urban areas.

Studies in river sediments consistently report abundances of microplastics in the tens to hundreds of particles kg^{-1} (Table 1), values that are broadly comparable to those reported in marine sediment studies. For example, Dekiff et al. (2014) and Nor and Obbard (2014) reported marine microplastic concentrations in the range from individual particles to tens of particles per kilogram of dry sediment, consistent with a study of the sediments of the St Lawrence River (Castañeda et al., 2014). Hundreds of particles per kilogram of dry sediment were reported by Horton et al. (2016) in UK river sediments, values also reflected by Laglbauer et al. (2014) in coastal sediments in Slovenia. At the highest concentrations, thousands of particles kg^{-1} of dry sediment have been reported in river sediments in Germany (Klein et al., 2015), comparable to the 2000-8000 particles kg^{-1} reported by Mathalon and Hill (2014) in coastal sediments in Canada.

Efforts in colloid science and nanotoxicology have shown the value of working towards standard methods for key measurements of colloid and nanomaterial characteristics, such as size, stability and surface properties (Hassellov et al., 2008; Montes-Burgos et al., 2009). Similar efforts seem warranted in the microplastic community with respect to environmental sampling and qualification. Currently in the field of microplastics research, there are two widely accepted methods of polymer identification – Fourier transform infra-red (FTIR) spectroscopy and Raman spectroscopy, although both have drawbacks. Alternative identification methods such as differential scanning calorimetery (DSC) and thermo-gravimetric analysis (TGA) have been tested but not been widely applied (Dumichen et al., 2015). Of the sampling configurations available for FTIR, there are two that are most common: attenuated total reflectance (ATR) and or transmission (or absorbance). ATR is not effective for analysing very small particles due to the fact that the sample needs to be large enough to cover an 'ATR window' in order for a satisfactory spectrum to be obtained (typically > 1 mm). Additionally, while in transmission mode refractive or scattering artefacts can occur, most notably for particles with irregular surfaces (Harrison et al., 2012). Raman spectroscopy can be overridden by fluorescence from some polymer particles, while other interferences may occur if particles are dirty or contain larger amounts of filler, such as dyes or plasticisers (Löder and Gerdts 2015). These

limitations reduce the possibility of determining probable sources, fate and potential short and long-term environmental impacts of these microplastics as well as advising policy makers on how to regulate microplastic pollutants. It could be that in order to effectively identify environmental polymers, a combined and complementary approach is required, for example using both spectroscopy and thermal analysis (Gigault et al., 2016; Majewsky et al., 2016; Sgier et al., 2016). It will be important to use the experience of working with microplastics in aquatic environments, especially sediments, to inform methods for terrestrial studies.

3.3. Transport of microplastics within the environment

Estimating the quantity of plastic litter which is released to the environment is difficult due to a lack of data and international variations between plastic waste generation and disposal. These disparities arise as a result of international differences in societal attitudes, education and investment in waste management infrastructure. For example, in China in 2010, 76% of plastic waste (8.82 million metric tonnes) was considered to be mismanaged, compared with 2% (0.28 million metric tonnes) in the United States (Jambeck et al., 2015). Mismanaged waste accounts for plastic released to land by littering and wind-blown debris. The best available estimates for managed and mismanaged plastic waste worldwide are from Jambeck et al. (2015), who modelled how much plastic waste was emitted globally to the oceans from land-based sources during 2010. Our estimates presented in Table 2 focus on Europe and assume that the proportion of waste that is mismanaged in the European Union (EU) is equivalent to that of the United States (2%). This is a reasonable assumption based on similarities in national income and development of waste management infrastructure, evidenced by the application of EU wide policies governing waste management, such as the 1999 EC landfill directive (1999/31/EC) (European Council, 1999). Based on this assumption we estimate how much of this mismanaged waste, plus the additional source of

477 microplastics from sewage sludge application, is likely to remain on land annually within Europe
478 (Table 2).

479

480 **Table 2.** Waste management data and estimates of plastic waste released to terrestrial and freshwater
481 (continental) environments, based on figures for the European Union. Rows highlighted in grey are those directly
482 related to plastic within continental environments. ¤Values for specific waste management practises do not
483 account for mismanaged waste. *Managed and mismanaged waste figures are calculated based on the
484 proportion of waste categorised as managed or mismanaged in the United States: 2% (Jambeck et al., 2015).
485 ¥Values are calculated based on mismanaged waste to include plastics within sewage sludge, minus plastic that
486 is transported to the oceans. Some sources, such as atmospheric fallout have not been considered due to the
487 limited data available. [1]PlasticsEurope (2015) [2]Jambeck et al. (2015) [3]Nizzetto et al. (2016b)

Plastic handling/disposal	Plastic million metric tonnes/year
Plastic production (EU total, 2014)[1]	59
Plastic waste (EU total, 2014)[1]	25.8
Managed plastic waste (-2% mismanaged waste)*	25.28
Landfill (EU total)[1¤]	8
Recycling (EU total)[1¤]	7.6
Energy recovery (EU total)[1¤]	10.2
Mismanaged plastic waste (2% of plastic waste in the EU)*	0.52
Plastic in sewage sludge (EU total)[3]	0.063 - 0.43
Ocean input (EU total)[2]	0.04 - 0.11
Total mismanaged plastic waste remaining in continental environments (EU) ¥	0.47 - 0.91

488

489 Plastic materials used in consumer, domestic and agricultural products in Europe amounted
490 to 59 million metric tonnes in 2014 (PlasticsEurope, 2015). Mismanaged plastic waste within the EU is
491 calculated at 520,000 metric tonnes (plastic waste – managed waste).In addition to this, it is estimated
492 that between 63,000 and 430,000 metric tonnes of microplastics in sewage sludge are deposited on

493 land annually (Nizzetto et al., 2016b). As a result we calculate that in the EU between 473,000 and
494 910,000 metric tonnes of plastic waste is released and retained annually within continental
495 environments, between 4 and 23 times the amount estimated to be released to oceans (Table 2). With
496 the current lack of data on microplastics in soils, it is not possible to distinguish between particles that
497 are retained within terrestrial environments and those retained within freshwater systems. As plastic
498 production and thus environmental deposition increases, this will also result in greater accumulation,
499 and larger amounts being ultimately transferred to the marine environment. However, for a
500 considerable time into the future it remains likely that the amount of plastic deposited and retained
501 within continental environments will exceed that entering the oceans. It is important to note that the
502 study by Jambeck et al. (2015) considers all waste within the US to be well-managed, with the
503 exception of litter (2% of all waste). However, it is possible that some fraction of the waste that is
504 considered to be well-managed could enter the environment during waste processing (e.g. as wind-
505 blown debris or mechanical or human error). Therefore it remains plausible that the figures for
506 mismanaged waste may be higher than the stated value. When it is also considered that there may be
507 additional pathways of release that are poorly known, such as atmospheric deposition, then it may be
508 the case that the calculations presented here may be an underestimation of plastic releases.

509 Freshwater and soil systems are subject to both point and diffuse inputs of plastics and so
510 great research effort is warranted to understand transport, exposure and ecological effects of
511 microplastics in these systems. This knowledge will also inform our understanding of rivers and
512 freshwater bodies as transport pathways for plastics from land to oceans (Jambeck et al., 2015;
513 Lechner et al., 2014; Rillig, 2012). It has been estimated that between 70-80% of marine plastics are
514 transported to the sea through the conduits provided by rivers (Bowmer and Kershaw, 2010).
515 Recognising this need, freshwater environments have received more attention than terrestrial
516 environments thus far as they are seen as a direct link between land-based plastic waste and the open
517 oceans, as well as interest in the toxicological impact of microplastics on freshwater ecosystems (see

518 Table 1). Studies of microplastics in soil ecosystems are, however, notably lacking (Huerta Lwanga et
519 al., 2016; Zubris and Richards, 2005).

520 Figure 2 shows a conceptual diagram of the main flows of microplastics within and between
521 three environmental compartments: terrestrial, freshwater and marine. A key concept of the diagram
522 is partitioning of plastic particles between aquatic and terrestrial environments, highlighting that
523 plastic debris will not only be transported by rivers from land to sea, but that even once in the aquatic
524 environment, may also return to land during high tide or flooding events (Fig. 2). The extent of overall
525 deposition, retention and transport of microplastics will depend on many factors including human
526 behaviours, such as littering or recycling, particle characteristics such as density, shape and size,
527 weather, including wind, rainfall and flooding, and environmental topography and hydrology. This
528 variation can make predicting the spread of litter difficult (Zylstra, 2013). Transport of plastic particles
529 within river systems will be largely affected by the same factors affecting sediment transport, such as
530 hydrological characteristics and environmental conditions (Nizzetto et al., 2016a) . Conditions such as
531 low flows and change in river depth or velocity (for example, on a bend) may lead to deposition of
532 particulate matter, whereas high velocity flood conditions and erosion could lead to mobilisation of
533 previously sedimented particles, in addition to the introduction of particles via runoff (Milliman et al.,
534 1985; Naden et al., 2016; Walling, 2009). Surrounding land-use can also affect the dynamics of
535 sediment and particulate transport within a river due to erosion, use of soils, irrigation and runoff
536 (Chakrapani, 2005). Plastic residing in river systems may also be subject to in-situ degradation, either
537 by photodegradation or mechanical fragmentation (Williams and Simmons, 1999).

538 To date only scant attention has been paid to investigating sources, fate and transport of
539 microplastics in terrestrial environments. However it not unreasonable to suggest that microplastics
540 are widely present across land. Litter has been widely reported as a common observation, with many
541 studies commenting on land based (macro)plastic debris (Derraik, 2002; Hoellein et al., 2014; Jambeck
542 et al., 2015; Townsend and Barker, 2014; Williams and Simmons, 1999; Zylstra, 2013).

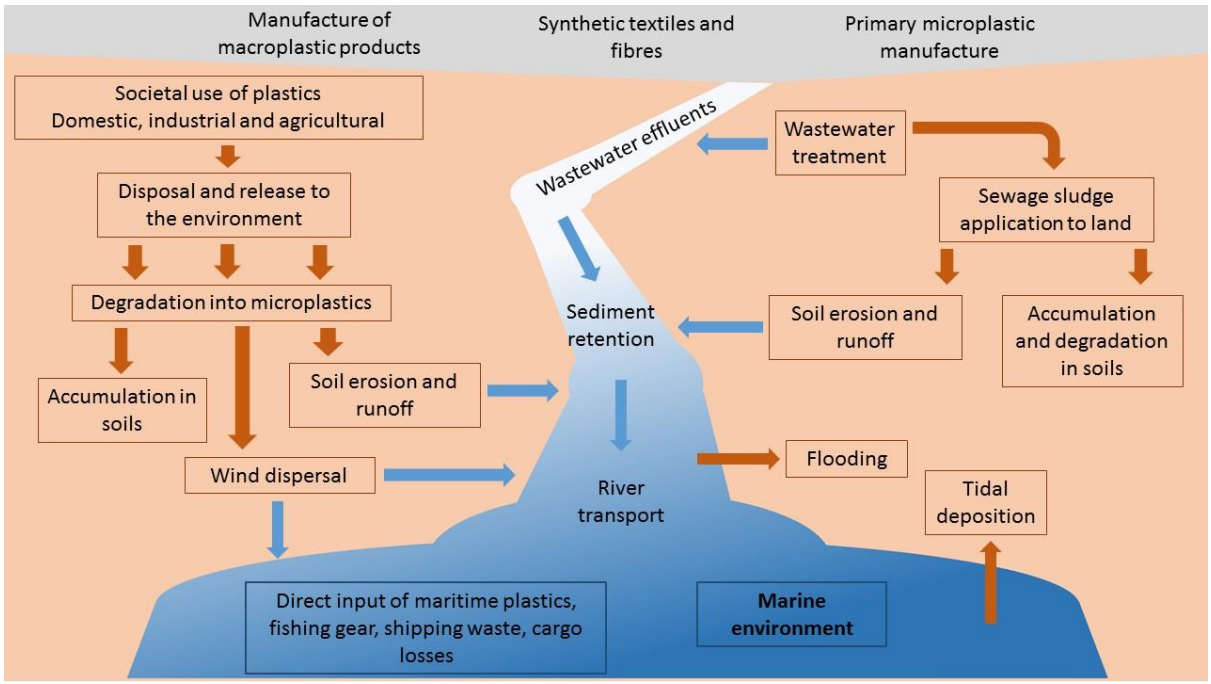

Figure 2. Conceptual diagram of microplastic sources and flows throughout and between anthropogenic, terrestrial, freshwater and marine environmental compartments.

4. Microplastics as an environmental hazard

4.1. Ecological impacts of microplastics

4.1.1. Organism interaction and ingestion of microplastics

Based on the evidence of widespread presence of plastics, it is highly likely that organisms in terrestrial and freshwater ecosystems will encounter microplastic particles. Depending on the particle size and the physiological and behavioural traits of the organism, there is an opportunity for the ingestion of these particles by invertebrates and vertebrates. Indeed such consumption has been widely observed in many marine species. Although plastic is largely excreted following ingestion, there is evidence to suggest that microplastics can be retained in the gut over timescales beyond those expected for other ingested matter (Browne et al., 2008). Further, there is evidence that particles may even cross the gut wall and be translocated to other body tissues, with unknown consequences

559 (Browne et al., 2008; Farrell and Nelson, 2013; von Moos et al., 2012). Given the similarity of some
560 phyla that are commonly found in freshwater and marine ecosystems (e.g. nematodes, annelids,
561 molluscs, arthropods) and indeed in soils, similar findings of ingestion in species in these ecosystems
562 are almost inevitable. Since many of these species, likely to take up microplastics, are important to
563 ecosystems (Lavelle, 1997; Sampedro et al., 2006) ecosystem processes such as decomposition and
564 nutrient cycling may be affected by microplastic exposure. Further, there is the potential for food web
565 effects either through effects on keystone species or possibly through the trophic transfer of
566 microplastics themselves.

567 Research to date, predominantly on marine species, has shown the ingestion of microplastic
568 particles in a wide range of species at many organisational levels and with different feeding strategies,
569 including detritivores, filter feeders and predators. In addition to accumulation of particles in
570 organisms at lower trophic levels (Browne et al., 2008), there is also evidence for the trophic transfer
571 of microplastic particles between marine species, especially bivalves and crustaceans (Farrell and
572 Nelson, 2013; Van Cauwenberghe and Janssen, 2014; Watts et al., 2014). This is also likely to occur in
573 terrestrial ecosystems in a similar manner to that of observed trophic transfer and accumulation of
574 gold nanoparticles between earthworms *Eisenia fetida* and bullfrogs *Rana catesbeina* (Unrine et al.,
575 2012). Gold nanoparticles are comparable to (nano)plastic particles in that are they are similarly
576 poorly soluble (Bouwmeester et al., 2015). There is also evidence that exposure to inert anthropogenic
577 particles can cause physical damage to body tissues (Lahive et al., 2014; Van Der Ploeg et al., 2013).

578 As far as we are aware, to date only three terrestrial species, the earthworms *Lumbricus*
579 *terrestris* (Huerta Lwanga et al., 2016) and *Eisenia andrei* (Rodriguez-Seijo et al., 2017) and the
580 nematode *Caenorhabditis elegans* (Kiyama et al., 2012), have been studied in the literature exposed
581 to microplastic particles under laboratory conditions and with ingestion being observed. Among
582 freshwater organisms, the filter feeder *Daphnia magna* has been observed to ingest microplastics
583 (Besseling et al., 2014; Casado et al., 2013; Rehse et al., 2016). Synthetic fibres have also been
584 observed in the digestive systems of freshwater fish collected from the wild, indicating consumption

585 either directly or in association with consumed prey items (Sanchez et al., 2014). Through such
586 consumption, mobile organisms such as fish, mammals and birds may also contribute to the dispersal
587 of microplastics over long distances following the ingestion and subsequent egestion of consumed
588 microplastics (Eerkes-Medrano et al., 2015). A major factor that is known to influence particle
589 ingestion by organisms is particle to mouth size ratio, with smaller particles having greater potential
590 to be ingested by a greater range of organisms. If ingested by lower tropic level organisms, this may
591 support further transfer and accumulation along food chains (Cole et al., 2013; Farrell and Nelson,
592 2013; Setälä et al., 2014).
593

594 **4.1.2. Observed toxicological effects of microplastics**

595 Ingestion of microplastic particles by marine invertebrates has been linked with a wide range
596 of sub-lethal effects including reduced reproduction, reduced growth of individuals and reduced
597 fitness. These are generally the result of the physical effects of ingested microplastics including
598 internal damage such as lacerations, inflammatory responses and plastic particles replacing digestible
599 food, causing individuals to reduce feeding hence resulting in lower energy intake, although effects
600 vary between species and plastic types (Moore, 2008; von Moos et al., 2012; Wright et al., 2013a;
601 Wright et al., 2013b). While there are fewer studies conducted to date with soil and freshwater
602 species, the studies that have been conducted generally confirm the potential for microplastics to
603 have detrimental effects on the physiology of species across many ecological niches.

604 In a recent soil study, Huerta Lwanga et al. (2016) observed mortality in *Lumbricus terrestris*
605 earthworms exposed to polyethylene particles; mortality was increased by 8% at a concentration of
606 450 g kg^{-1} polyethylene (in overlying leaf litter) and 25% mortality at 600 g kg^{-1}. Reduced growth and
607 negative effects on burrow construction were also observed. As the concentrations of plastic litter
608 micro-fragments found on soil surfaces are currently unknown, it is difficult to place the
609 concentrations that are used in this study within the range of possible microplastic concentrations

610 that may occurs in soils. The exposure concentrations would certainly seem high compared to
611 expected microplastic levels resulting from diffuse pollution. However, it remains possible that they
612 may be consistent with exposure around some point sources, especially following *in situ* degradation.
613 This finding that annelid worms can be affected by microplastics is consistent with a number of studies
614 conducted for marine species. For example, in a study of *Arenicola marina* exposed to uPVC
615 (unplasticised PVC) particles experienced weight loss and reduced lipid reserves were observed. A
616 uPVC treatment of 10 g kg^{-1} dry sediment reduced energy reserves by 30% while at a uPVC
617 concentration of 50 g kg^{-1} dry sediment, energy reserves were reduced by 50%. This effect overall
618 suggests that exposure to UPVC causes metabolic stress to marine benthic sediment worms (Wright
619 et al., 2013a). Due to the close relatedness of worm species in terms of morphology and how they
620 feed in sediment it is likely that similar effects would be observed in freshwater and terrestrial worm
621 species (Rillig, 2012). In the marine copepod, *Tigriopus japonicas*, Lee et al. (2013) found that although
622 acute exposure (96 hours) to three different particle sizes (0.05, 0.5 and 6 µm) of polystyrene
623 microbeads, had no impact on the survival rate of adults, in a two generation chronic exposure
624 experiment mortality was observed at concentrations above 12.5 µg ml^{-1}, with the second generation
625 observed to be much more sensitive than the first generation, especially when exposed to the nano-
626 scale particles (0.05 µm). Larger particles in contrast (6 µm) had no effect on survival even over two
627 generations, although fecundity was affected at concentrations above 25 µg ml^{-1}. Although the species
628 of copepod used in this study were marine, they are directly comparable to freshwater copepod
629 species and other planktonic filter feeding organisms like *Daphnia* sp. This implies that toxic effects of
630 microplastics may be size-dependent either as a result of particle ability to permeate body tissues or
631 to cause greater inflammatory response. Studies conducted with nanoplastics also highlight possible
632 size dependent influences on toxicity for both acute survival effects (Besseling et al., 2014; Nasser and
633 Lynch, 2016) and different reproductive effects observed in response to smaller particle fractions (Lee
634 et al., 2013).

635 It is also important to consider how alteration of particle characteristics over different
636 environmental timescales may affect toxicity. Exposure to artificially aged (nano)polystyrene has been
637 found to cause mortality, growth and reproduction effects to the standard test species *Daphnia*
638 *magna* over a 21 day period, whereas pristine nano-polystyrene particles caused no significant effects
639 on mortality. Mixtures of nano-polystyrene and fish kairomones (known to cause stress in *D. magna*)
640 produced an additive effect on body size and reproductive endpoints, indicating that exposure to
641 plastic particles can exacerbate existing environmental stress responses (Besseling et al., 2014). Many
642 studies investigating the toxicological impacts of microplastics have used virgin plastic particles.
643 However, if aged and contaminated, particles can have the potential for greater chemical transfer
644 than virgin particles (see section 4.2.2.). This use of pristine particles could thus lead to a potential
645 underestimation of the toxicological impacts of microplastic exposure under more realistic
646 environmental exposure scenarios. Recently the nanotoxicology research community have recognised
647 the need to conduct experiments with environmentally 'aged' nanomaterial forms (Christian et al.,
648 2008; Judy et al., 2015; Lahive et al., 2017). Common nanomaterial transformations, such as hetero-
649 and homo-aggregation, changes in surface charge and in particular the development of a surface
650 'corona' of associated macromolecules and chemicals may all occur for both nanoparticles and
651 microplastics (Syberg et al., 2015). Hence future studies with these 'aged' particle forms may be
652 needed to more accurately identify the possible effects of anthropogenic materials in real
653 environments (Schultz et al., 2015).

654 When considering microplastics and chemical co-transport, principles used in mixture
655 toxicology may be useful to assess these multifaceted stresses in the environment. Given that most
656 environmental microplastic studies quantify microplastics by number of particles rather than by
657 weight (as is more common for bioassays), and none to our knowledge have yet detected nanoplastics
658 in environmental samples, it is not yet possible to determine whether the concentrations used in these
659 studies are environmentally relevant. This is a similarly common criticism of microplastic studies in
660 that the concentrations of particles used are likely not environmentally realistic. Even though the

relationship between environmental concentrations and those used in toxicity bioassays is not fully established, it is likely that the concentrations used in laboratory tests are comparable to only the highest levels of environmental contamination. However, it is still valuable to understand the potential ecological implications of microplastic pollution at these high concentrations as a contribution to understanding of hazard and developing risk assessments. Further, given that environmental concentrations of microplastics are likely to increase with input and fragmentation of plastics already present in the environment, the future presence of higher concentrations can be expected (Phuong et al., 2016).

4.2. Microplastics as a chemical hazard

4.2.1. Leaching of plasticiser chemicals in freshwater and terrestrial environments

Plastic materials often contain a wide range of plasticiser chemicals to give them specific physical properties such as elasticity, rigidity, UV stability, flame retardants and colourings (Browne et al., 2013; Lithner et al., 2009; Moore, 2008; Teuten et al., 2009). Many of the chemicals associated with plastics have been identified as either toxic or endocrine disruptors including bisphenol-A, phthalates such as di-n-butyl phthalate and di-(2-ethylhexyl) phthalate, polybrominated diphenyl ethers (PBDEs) and metals used as colourings (Hua et al., 2005; Kim et al., 2006; Lithner et al., 2009; Oehlmann et al., 2009; Rochman et al., 2013b; Teuten et al., 2009). Additive chemicals like these are weakly bound, or not bound at all to the polymer molecule and as such these chemicals will leach out of the plastic over time. Such releases can be facilitated in environments where particle dispersal is limited and where plastics will experience UV degradation and high temperatures (Andrady, 2011). The locations where microplastics may accumulate in soil and surface waters are therefore likely to be subject to the possible release of these chemicals from plastics and their subsequent transfer to water, sediment and organisms. Lithner et al. (2009) showed that different plastic items can leach toxic chemicals into water that can cause varying effects on *Daphnia magna*. Different items made of

686 the same polymer may have varying toxicity effects following leaching, based on the type and amount
687 of plasticisers added during manufacture. This demonstrates that plastic materials can act as a source
688 of complex leachate mixtures to the environment.

689 As a major environmental sink for all types of plastic waste, landfill material and the leachates
690 arising from landfill sites are highly likely to contain high concentrations of plasticiser chemicals (do
691 Nascimento Filho et al., 2003; Slack et al., 2005; Yamamoto et al., 2001). Within a landfill site chemical
692 conditions change over time with regards to temperature fluctuation, oxygen presence, acid/alkaline
693 conditions and dissolved organic carbon all of which have the potential to change plasticiser leaching
694 (Teuten et al., 2009; Xu et al., 2011). Large scale chemical monitoring studies have identified the
695 presence of phthalate esters (plasticiser chemicals) in a wide range of agricultural and peri-urban soils
696 in various regions of China. Zeng et al. (2008) analysed soil samples from a range of field sites around
697 Guangzhou city, China. The study identified 16 phthalate compounds with concentrations for
698 individual phthalate found ranged from 0.195–33.5 mg kg^{-1} dry weight soil. The highest concentration
699 of phthalates were found in an agricultural soil, in close proximity to a water course into which
700 wastewater was discharged from nearby industrial activities including manufacture and disposal of
701 plastics and this was identified as the key source of phthalates in soil. Similarly Kong et al. (2012)
702 analysed soil samples from farmland finding concentrations of phthalates ranging from 0.05–10.4 mg
703 kg^{-1} dry weight. The highest concentrations were found in vegetable plots close to domestic rubbish
704 sites, from which phthalates could be expected to leach. High concentrations were found at sites close
705 to busy roads and at wasteland sites where plastic debris abundance was high. Further to these
706 studies, Wang et al. (2013) sampled soils used for vegetable production near Nanjing (east China).
707 Measured concentrations of phthalates ranged between 0.15–9.68 mg kg^{-1} dry weight; the highest
708 concentrations were found at sites where plastic mulches and polytunnels were in use. Proximity to
709 municipal solid waste sites and application of sewage sludge were also identified as major sources of
710 phthalates, indicating leaching of plasticiser chemicals from plastic particles deposited on land. Taken
711 together, the results suggest that plastic materials release chemicals to soil via a number of the

712 pathways and are a potential source of plasticisers to soils. This may have significant implications for
713 terrestrial locations where microplastic concentrations are high, although further studies are needed
714 to confirm this early evidence.

715

716 **4.2.2. Microplastic associations with organic pollutants**

717 Microplastics themselves are widely understood to bind to a range of different hydrophobic
718 organic chemicals (HOCs) within the environment, such as organochlorine pesticides, PAHs, PCBs,
719 PBDEs, dioxins and metals (Besseling et al., 2013; Mato et al., 2001; Rochman et al., 2013c). This may
720 be especially significant in continental freshwater and terrestrial environments, where concentrations
721 of these chemicals are expected to be higher than in marine systems, due to proximity to the use of
722 these chemicals (Dris et al., 2015b). HOCs are recognised as having high lipophilicity (i.e. high
723 octanol/water partition coefficient, Kow), determining whether a chemical will dissolve in water and
724 remain in solution). Chemicals with such a high K_{ow} will typically have a strong affinity for adsorption
725 to organic and particulate matter within water, soil and sediment. These same characteristics, in
726 addition to factors including hydrophobicity of polymer, large or abraded surface properties and
727 biofouling, mean that HOCs also have the potential for sorption to plastic materials (Karapanagioti and
728 Klontza, 2008; Teuten et al., 2007). Microplastics and representative chemicals from many POP classes
729 may become associated in waste streams (e.g. sewage effluent and sludge, landfill waste and leachate)
730 or in anthropogenically influenced environments. Hence, the interactions between microplastics and
731 organic pollutants are particularly pertinent in freshwaters inland, especially those in close proximity
732 to industrialised and populated areas with a high discharge of industrial and domestic wastewater,
733 where small dispersal areas can lead to high pollutant concentrations (Eerkes-Medrano et al., 2015;
734 Free et al., 2014). This will be especially relevant in agricultural areas where plastic products are used
735 in close proximity or in association with the application of hydrophobic chemicals such as some
736 pesticides.

737 Changes to environmental conditions will influence equilibrium dynamics between chemicals
738 and plastics, impacting on chemical accumulation and bioavailability (Bakir et al., 2016; Bakir et al.,
739 2014; Karapanagioti and Klontza, 2008; Koelmans et al., 2016). Additionally, particle size and texture
740 will affect the capacity of microplastics to either adsorb or leach contaminants and indeed plasticiser
741 additives. The greater surface area per unit of mass as particles decrease in size increases the potential
742 for surface chemical interactions and thus binding with hydrophobic chemicals. Physically weathered
743 particles are expected to have a larger surface area as a result of cracking and abrasion which increases
744 overall surface area (Ivar do Sul and Costa, 2014; Teuten et al., 2009). Such environmentally-induced
745 changes may be particularly relevant for terrestrial microplastics, which may be exposed to high levels
746 of UV radiation and wind. The ecological impacts of plastic-chemical associations are difficult to
747 predict due to the many interactions between polymers, plastic additives, adsorbent characteristics
748 and environmental conditions which will impact on bioavailability (Bakir et al., 2014; Koelmans et al.,
749 2016; Velzeboer et al., 2014).

750

751 **5. Future research recommendations**

752 As this review highlights, the largest gaps in current knowledge are in our understanding of
753 microplastic pollution in terrestrial ecosystems, especially environmental concentrations, sources and
754 ecological impacts. In freshwater systems, knowledge of concentrations of microplastics is rapidly
755 growing. However, in most instances this knowledge has yet to be related to ecological effects. Due
756 to the lack of quantitative data, it is difficult to assess quantitatively the exact nature of the
757 microplastic hazard in these systems and how the consequences of microplastic presence in these
758 ecosystems will manifest themselves. Indeed this is true of microplastics research as a whole, where
759 the long term implications of microplastics are still unclear compared to better-studied chemical
760 pollutants.

There is a large degree of uncertainty around the volume, composition and diversity of microplastic particles entering the environment. Information on the scale of production is available as is some data on plastic entry into major waste management systems, however current release rates from these streams either by deliberate or accidental release of refuse or wind action is not quantified. This route from accidental release and littering is, hence, one of the greatest uncertainties for emission predictions. This review highlights the complex challenge of understanding the dynamics and impacts of microplastics as an environmental pollutant, especially understanding microplastics in a freshwater and terrestrial context, but also demonstrates how information from marine studies can be used to infer or predict what may occur in these less studied systems. In a similar way, nanomaterial research can also provide insights into particulate behaviour and fate.

To progress the field of research, it is of utmost importance in the first place to define 'microplastics' clearly as an environmental contaminant, and thereafter to develop standardised methods for collecting, processing and analysing environmental samples. Such standardisation has the potential to reduce ambiguity and thus allow direct comparison between studies with a view to understanding sources and transport pathways. Spectroscopy methods have already been used to identify freshwater and terrestrial nanoparticles and the continued development of such methods, as well as alternatives such as differential scanning calorimetery (DSC) and thermo-gravimetric analysis (TGA), is important to provide additional information on the polymers present in terrestrial and freshwater ecosystems.

While an ideal scenario would be to reduce the amount of plastic entering the environment, the challenges of reduction from changes in manufacturer and consumer behaviour mean that releases can be expected to continue for some time. Given the volume of plastic currently present in the environment, and the likely increase of microplastics due to fragmentation, it therefore remains important to understand the potential effects of this ever-accumulating pollution (Nizzetto et al., 2016a; Phuong et al., 2016).

Based on the evidence presented in this review, it is clear that our understanding of microplastics in the environment is rapidly developing. However, there are still fundamental gaps in the knowledge and many questions still remain. In summary, the most important questions remaining are:

1) What is the current extent of microplastic pollution in terrestrial environments, and how does this compare to known contamination in aquatic environments? Which polymers are most abundant and does this vary across habitats and regions?
2) To what extent do environmental conditions and properties of different plastic materials affect microplastic behaviour and bioavailability under the conditions that are found in freshwater and terrestrial environments?
3) Are adverse effects primarily due to physical impacts of the particle itself, chemical toxicity or mixture effects, and does this vary between polymers and species? Are there parallels that can be drawn with what is known concerning mechanisms of action for some nanoparticles?
4) What are the likely ecological implications of plastics under realistic exposure conditions (i.e. microplastics of the type and concentrations likely to be encountered by organisms)?

6. Conclusions

The available literature reporting information on plastic use and release suggests that primary and certainly secondary microplastics are likely to be found ubiquitously across terrestrial and freshwater environmental compartments due to their proximity to most point and diffuse sources. Both primary and secondary microplastics entering the environment will persist and continue to fragment to smaller particles. These smaller fragments are likely to pose a greater risk to organism health due to their increased likelihood of uptake, increased surface area for interactions with chemicals and greater number of particles per unit of bulk mass (Jeong et al., 2016; Lee et al., 2013). The focus on nanoparticle hazards has recently generated a greater understanding of the behaviour

811 of particulate pollutants, as well as methods for their detection and hazard assessment. Clear parallels
812 exist from this work to future studies with nanoparticles, with collaboration between the disciplines
813 likely to improve understanding (Bouwmeester et al., 2015; Syberg et al., 2015). This takes the more
814 environmentally relevant approach that it is necessary to understand the fate, behaviour and impacts
815 of microplastics as an environmental pollutant and, therefore, their potential implications for keys
816 ecosystem components and processes.

817 As microplastics can act as both a direct (particulate) hazard and an indirect (chemical) hazard,
818 unravelling ecological effects may call for the application of approaches for mixture toxicity may be
819 beneficial for the analysis of combined plastic-chemical effects. Despite land being the least studied
820 environmental compartment, many of the ecological risks of microplastics identified in aquatic species
821 will also apply to terrestrial ecosystems due to the many ecological and taxonomic parallels that exist
822 between resident species. Studies on the dynamic interactions between plastic particles, plasticiser
823 additives and environmental contaminants is also a field that needs to be expanded to understand
824 how organic chemical partition coefficients to plastics are altered in the presence of sediment and
825 soil. Studies of chemical dynamics within the gut of organisms are also needed in order to better
826 understand the processes that govern bioaccumulation of plasticisers and co-transported chemicals.
827 Ultimately, studies are needed to link the finding in the field studies to laboratory results to better
828 understand both environmentally relevant scenarios of real-world risks posed by microplastics and
829 the underlying mechanisms.

830

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