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Microplastics in soils: an environmental geotechnics perspective

Brendan C. O'Kelly PhD, FTCD, CEnv, CEng, MICE
Associate Professor, Department of Civil, Structural and Environmental Engineering, Trinity College Dublin, Dublin, Ireland (Orcid:0000-0002-1343-4428) (corresponding author: bokelly@tcd.ie)

Abbas El-Zein PhD
Professor of Environmental Engineering, School of Civil Engineering, Faculty of Engineering, The University of Sydney, Sydney, Australia

Xiaoli Liu PhD
Professor, State Key Laboratory of Hydroscience and Engineering, School of Civil Engineering, Tsinghua University, Beijing, China (Orcid:0000-0002-6204-4485)

Anjan Patel PhD
Assistant Professor, Department of Civil Engineering, Visvesvaraya National Institute of Technology, Nagpur, India (Orcid:0000-0003-2645-613X)

Xunchang Fei PhD
Assistant Professor, School of Civil and Environmental Engineering, Nanyang Technological University, Singapore; Deputy Director, Residues and Resource Reclamation Centre, Nanyang Environment and Water Research Institute, Singapore

Susmita Sharma PhD
Assistant Professor, Department of Civil Engineering, National Institute of Technology Meghalaya, Shillong, India

Arif Mohammad BE
Research Scholar, Department of Civil Engineering, Indian Institute of Technology Bombay, Mumbai, India (Orcid:0000-0002-1815-5073)

Venkata Siva Naga Sai Goli MTech
Research Scholar, Department of Civil Engineering, Indian Institute of Technology Bombay, Mumbai, India (Orcid:0000-0003-1916-725X)

Jing Jing Wang PhD
Senior Photonics Scientist, Amber Research Centre and Centre for Research on Adaptive Nanostructures and Nanodevices, Trinity College Dublin, Dublin, Ireland

Dunzhu Li PhD
Research Scholar, Amber Research Centre and Centre for Research on Adaptive Nanostructures and Nanodevices, Trinity College Dublin, Dublin, Ireland

Yunhong Shi PhD
Research Scholar, Amber Research Centre and Centre for Research on Adaptive Nanostructures and Nanodevices, Trinity College Dublin, Dublin, Ireland

Liwen Xiao PhD
Assistant Professor, Department of Civil, Structural and Environmental Engineering, Trinity College Dublin, Dublin, Ireland (Orcid:0000-0003-3621-9568)

Ganaraj Kuntikana MTech
Research Scholar, Department of Civil Engineering, Indian Institute of Technology Bombay, Mumbai, India

Bettadapura Subramanyam Shashank PhD
Assistant Professor, Department of Civil Engineering, Birla Institute of Technology and Science (BITS) Pilani, Pilani, India

Theo S. Sarris MEng, MSc, PhD, CPEng
Science Leader, Health and Environment Division, Institute of Environmental Science and Research, Christchurch Science Centre, Christchurch, New Zealand (Orcid:0000-0002-1429-2185)

Bendadi Hanumantha Rao PhD
Assistant Professor, Geotechnical Engineering Division, School of Infrastructure, Indian Institute of Technology Bhubaneswar, Bhubaneswar, India

Abdel Mohsen O. Mohamed PhD
Professor of Geotechnical and Geo-environmental Engineering and General Managing Director, EX Scientific Consultants, Abu Dhabi, UAE; Senior Advisor, Uberbinder, Inc., Seattle, WA, USA (Orcid:0000-0002-0971-6940)

Evan K. Paleologos PhD
Professor of Civil Engineering, Department of Civil Engineering, Abu Dhabi University, Abu Dhabi, UAE (Orcid:0000-0002-3582-2288)

Mohaddeseh Mousavi Nezhad PhD
Associate Professor, School of Engineering, The University of Warwick, Coventry, UK

Devendra N. Singh PhD, FASCE, FNAE, FICE
D.L. Shah Chair Professor for Innovation, Department of Civil Engineering, Indian Institute of Technology Bombay, Mumbai, India (Orcid:0000-0003-3832-1507)

Microplastics (MPs) are emerging persistent contaminants in the terrestrial subsurface, and evidence has emerged for significant effects of MPs on the biological and ecosystem functions of soils. Main MP sources include land spreading of sewage sludge and biowaste composts, plastic mulching film used in horticultural fields, waste water irrigation and leachate from the landfills, among others. This updated state-of-the-art review paper describes recent experimental and numerical research and developments in understanding the accumulation and fate and effects of MPs in the soil environment (focusing on their storage, degradation, transportation, leaching to groundwater etc.), followed by mitigation and bioremediation measures, including MP-eating soil bacteria and fungi and the best management practices for reducing MP pollution of soil. Other areas covered are the combined effects of MPs and various other environmental contaminants (heavy metals, organic pollutants and antibiotics) in soil ecosystems and the standardisation of methods for detection, quantification and characterisation of MPs in soils, which is critical for MP research. The paper concludes by identifying knowledge gaps and presents recommendations on prioritised research needs.

Introduction

Pollution due to microplastics (MPs) is listed by the UN Environmental Programme (UNEP, 2014) as one of the top ten environmental issues due to their (*a*) presence in, and harmful impacts on, aquatic and terrestrial ecosystems (mainly soils) and (*b*) action as a transport vector for other potential pollutants, including human pathogens, organic contaminants and heavy metals (Qi *et al.*, 2020). Although no internationally agreed definition of the size below which a small piece of plastic should be called an MP presently exists (Hartmann *et al.*, 2019), MPs are generally defined as plastic debris with particle sizes between 0.1 and 5000 μm (Galgani *et al.*, 2013; Thompson *et al.*, 2004). Concern about MP pollution is due to their extremely small size, slow rate of degradation under ambient conditions and lack of efficient detection

techniques in environmental systems. Although numerous studies on the deleterious effect of MPs have been presented for the aquatic environment, much less is known on the fate, transport and harmful impacts of MPs in terrestrial ecosystems. Sources of MPs in terrestrial ecosystems are generally divided into primary and secondary. The primary sources of MPs are mainly due to the application of organic fertilisers, composts and sewage sludge (SS); use of plastic mulches in agriculture; flooding and irrigation with MP-contaminated waste water in agricultural practice; and atmospheric deposition, whereas secondary sources mainly include degradation of the macroplastics (MAPs) in the environment and leachate from landfills (Guo *et al*., 2020).

With the notable exception of karst aquifers (comprising terrain with distinctive hydrology and landforms that arise from a combination of high rock solubility and well-developed secondary porosity due to fracture), no significant amounts of MPs have yet been detected in groundwater. This is most likely due to filtration by the porous medium that separates it from surface sources of plastics (Bläsing and Amelung, 2018). Notable concentrations of MPs have been detected in karst aquifers because of the latter's macroporous structure, faster flow regimes and stronger connections to the ground surface (Panno *et al*., 2019). In any case, research on the prevalence of MPs in terrestrial ecosystems is in its infancy and the number of studies still limited, and it is therefore far too early to draw definitive conclusions regarding the risk of MPs to groundwater. On the other hand, three factors, not directly related to groundwater protection, currently drive interest in the transport and fate of MPs in soil. First, MPs entrained and weakly absorbed by soils can find their way into fluvial systems, oceans and the atmosphere (Campanale *et al*., 2020; Cook *et al*., 2020; Horton *et al*., 2017; Unice *et al*., 2019). For example, there is evidence that wind-driven soil erosion contributes to atmospheric transport of low-density MPs (Rezaei *et al*., 2019) and, in some cases, of almost total release into freshwater of MPs in SS applied to agricultural soils (Crossman *et al*., 2020). Second, soils can be a conduit for the entry of MPs into the food chain through fauna and flora capable of absorbing MPs (Huerta Lwanga *et al*., 2018; Li *et al*., 2020a). For instance, evidence of bioaccumulation was reported by Huerta Lwanga *et al*. (2017a), who assessed MP transfer from soil to earthworm and chicken in Mayan households in Mexico. They found that the number of MPs per gram increased from the soil (0.87 ± 1.9 particles/g) to earthworm casts (14.8 ± 28.8 particles/g), chicken faeces (129.8 ± 82.3 particles/g) and chicken gizzards (10.2 ± 13.8 particles/g) used for human consumption. Third, MPs can alter the behaviour of other contaminants, both organic (Hüffer *et al*., 2019; Zhang *et al*., 2020c) and inorganic (Hodson *et al*., 2017; Yu *et al*., 2020), potentially including priority pollutants such as plasticisers and flame retardants that enter in the manufacturing of plastics to enhance their engineering properties (van Praagh *et al*., 2018). Hence, MPs can become an important conduit for the migration of these contaminants in the subsurface.

Definition of MPs (sizes and compositions)

In this paper, plastic particles are classified into macro- (>25 mm), meso- (between 5 and 25 mm), micro- (between 100 nm and 5 mm) and nano-plastics (NPs) (<100 nm) (Alimi *et al*., 2018; Jahnke *et al*., 2017). Note that some authors refer to particles in the lower MP range (between 100 and 200 nm) as NPs (Hu *et al*., 2020; Keller *et al*., 2020). It is clear that this definition does not distinguish between different types (i.e. chemical composition) and origins (e.g. primary against fragmented) and physical properties (i.e. shape and texture) of plastic, all of which have a strong influence on their prevalence, transport and impacts on the environment. In terms of particle shape, MPs occur in different forms such as fibres, fragments, beads, pellets and film.

Six types of MP particles seem to account for most of MPs hitherto found in the environment – namely, polyethylene (PE), poly(vinyl chloride) (PVC), polyamide (PA), polypropylene (PP), poly(ethylene terephthalate) (PET) and polystyrene (PS) (Alimi *et al*., 2018; Imhof *et al*., 2013; Rochman *et al*., 2013). However, this is mostly based on studies in marine and freshwater environments, and it remains to be seen whether other plastic types are important in terrestrial ecosystems, including soils. Other plastic pollutants might be important in soils because soils are often the first recipient of MP contaminants and are closer to the source of emission than water bodies.

Degradation of plastics

Physical, chemical and biological degradation of plastics in terrestrial ecosystems can form a source of MPs by breaking down MAPs with particle sizes of >25 mm and meso-plastics (MEPs) with particle sizes between 5 and 25 mm. Furthermore, it can be stated that due to degradation of MPs, they are reduced to lower-sized plastics, such as NPs, or completely converted to carbon dioxide (CO_2) and water (H_2O) in soil ecosystems (Helmberger *et al*., 2020; Shah *et al*., 2008). Plastic degradation can be driven by photo-oxidative, thermal, ozone-induced, mechanochemical, catalytic and biological processes and is one of the major secondary sources of MPs in terrestrial ecosystems (Guo *et al*., 2020; Singh and Sharma, 2008; Wang *et al*., 2020; Zhu *et al*., 2019). These processes increase oxygen-containing functional groups, thereby producing changes in the surface properties and chemical structure of polymers (Zhu *et al*., 2019). However, among these degradation processes, photo-oxidative degradation, ozone-induced degradation and biodegradation are primarily responsible for the formation of MPs from MAPs and MEPs in soils, landfills and so on. The degradation of these plastics depends on (*a*) the type of polymers and their physico-chemical characteristics, such as molecular weight, size, chemical bonding, tacticity, mobility, crystallinity and hydrophobicity of the surface, and (*b*) environmental conditions, mainly temperature, pH, moisture content and availability of suitable microbes and enzymes (Shah *et al*., 2008; Zhu *et al*., 2019). Photodegradation that occurs under the action of ultraviolet (UV) radiation, within 290–400 nm wavelength, reduces the mechanical strength, extensibility and transparency of the polymer, depending on the type of polymer and its chemical bonds (Andrady, 2011). Although the process is slow, ozone present in the atmosphere can degrade the polymer under natural conditions (Singh and Sharma, 2008). Incidentally, the actions of abiotic agents (i.e. UV irradiation, thermal stress and ozone) break

down the polymer chain and make it more bioavailable, which subsequently enhances its biodegradation. For instance, in the review paper by Shah *et al.* (2008), it was reported that thermal actions on polymers mainly reduce their molecular weight and deteriorate their physical properties, such as ductility and embrittlement, leading to cracking. Also, Yoshida *et al.* (2016) reported that owing to the glass transition temperature of 75°C of PET, the polyester (PEST) chain present in it is in a glassy state at moderate temperature, conducive for the action of mesophilic enzymes. Heterotrophic microorganisms are involved in the degradation of polymers, which act as a source of organic carbon and energy for the microbes (Shah *et al.*, 2008). The biodegradation of polymer occurs due to the action of extracellular and intracellular enzymes, such as lipase, cutinases, proteinase, dehydrogenases, depolymerase and PETase, which are capable of biodegradation (Shah *et al.*, 2008; Yoshida *et al.*, 2016).

On the other hand, there are presently few studies on the degradation of MPs in soil, which is a very slow process (Cooper and Corcoran, 2010; Krueger *et al.*, 2015; Zhu *et al.*, 2019) and mainly occurs due to biodegradation, whereas the degradation effects of soil environment, mechanical crushing, high-temperature oxidation and UV radiation appear to be limited. In farmland soils, Briassoulis *et al.* (2015) found that PE MP fragments of plastic mulching film (PMF) are difficult to degrade, persisting for several years or even decades and finally forming small MP residues. It was found that the weight loss of MPs in soil was only 0.1–0.4% after 800 days for PE (Albertsson, 1980) and 0.4% after 1 year for PP (Arkatkar *et al.*, 2009). In contrast, PVC and PS did not exhibit any change 32 years after burial in soil (Zhu *et al.*, 2019). MPs may persist in soils for as long as 100 years due to low light and oxygen availability, which eventually suppress photo-oxidative degradation (Barnes *et al.*, 2009). Moreover, incomplete biodegradation of biodegradable polymers (oxo-biodegradable and compostable plastics used as carrier bags) (O'Brine and Thompson, 2010) is one of the significant sources of MPs in leachate generated by landfills and uncontrolled dumpsites (Chandana *et al.*, 2021; Qi *et al.*, 2020; Weinstein *et al.*, 2016). To the best of the authors' knowledge, no study in the literature has provided an in-depth account of the degradation products of MPs.

MPs in the soil environment

Sources of MPs in soils

An important source of MP pollution has been found to be waste water discharges and biosolids/SS, which are major by-products of waste-water-treatment plants (WWTPs) (Enfrin *et al.*, 2019; Mason *et al.*, 2016; Sun *et al.*, 2019). MPs have been detected in agricultural soils (Corradini *et al.*, 2019a; Dehghani *et al.*, 2017; Liu *et al.*, 2018; Ng *et al.*, 2018; Panno *et al.*, 2019; Piehl *et al.*, 2018; Rillig *et al.*, 2017a), particularly those to which sludge compost has been applied (Corradini *et al.*, 2019a; Li *et al.*, 2018a; van den Berg *et al.*, 2020); soils within suburban built environments (Liu *et al.*, 2018); coastal soils (Zhou *et al.*, 2018); forest soils adjacent to farmlands (Zhang and Liu, 2018); and landfill refuse and leachates (He *et al.*, 2019; Su *et al.*, 2019; van Praagh *et al.*, 2018). Apart from the aforementioned ones, MPs enter soil ecosystems through agricultural mulching layers and waste water irrigation (Conley *et al.*, 2019; Kleinteich *et al.*, 2018; Lares *et al.*, 2018; Talvitie *et al.*, 2017a). Other pathways include landfills, beach littering and run-off from farming and recreational, industrial and urban spaces (Hurley and Nizzetto, 2018). Several studies have shown that the migration of MPs through the food chain significantly changes the biological community and, with the decreasing particle size of MPs, leads to the accumulation and transmission of MPs in food chains, reaching humans (Farrell and Nelson, 2013; Kiyama *et al.*, 2012; von Moos *et al.*, 2012; Wright *et al.*, 2013a, 2013b).

Land application of organic fertilisers, composts, SS and waste water irrigation

Organic fertilisers and composts, which are derived from different wastes such as unsegregated municipal solid waste (MSW), SS and animal manure, are a potential source of MPs in agricultural lands due to the nature of initial feedstocks for composting/digestion (Ng *et al.*, 2018; Qi *et al.*, 2018). It appears that the majority of reported studies in this field investigated SS application to agricultural soils. It has been estimated that annually about 2800–19 000, 63 000–430 000 and 44 000–300 000 t of MPs are being released into Australian, European and North American agricultural fields, respectively, through SS application (Ng *et al.*, 2018; Nizzetto *et al.*, 2016). The presence of MPs has also been reported in agricultural soils that have not been amended with composts/organic fertilisers, which may be attributed to the disintegration of PMFs.

WWTPs, although not specifically designed to treat MPs, can remove up to 99% of MPs from waste water (Carr *et al.*, 2016). Owing to the low biodegradability of plastics, MPs are not eliminated during the waste-water-treatment processes but are transferred from the waste water to the sludge residues. Subsequent SS-treatment processes, such as thermal stabilisation and anaerobic digestion, cannot efficiently remove these MPs. Since land-spread municipal SS could be important MP sources for soil, a total of 18 studies have been recently performed to investigate the MPs in SSs from 65 WWTPs in 13 countries (refer to Table 1). These studies have found that the MP abundance in SSs ranges between 1×10^3 and 3.14×10^5 MP particles/kg dry soil, with these limiting values deduced from reported data for the UK (Murphy *et al.*, 2016) and Spain (Edo *et al.*, 2020), respectively. The variation may be due to differences in MP testing methods/protocols employed and (or) the WWTP parameters, including source waste water.

Studies showed that land application of SS could increase MP abundance in soil (Corradini *et al.*, 2019a; Li *et al.*, 2018a; Talvitie *et al.*, 2017b; van den Berg *et al.*, 2020; Zubris and Richards, 2005). van den Berg *et al.* (2020) compared MP concentrations in 16 agricultural fields, before and after SS application, and found that MP abundance in the top 30 cm soil layer increased by about 7.1×10^2 particles/kg soil when

Table 1. Estimated total MP transfer from municipal SS to agricultural soil in various countries (continued on next page)

Country	MP abundance: particles/kg dry sludge	Number of WWTPs	MP particle size: μm	MP particle shape	Average MP abundance: particles/kg dry sludge	Total sludge production rate: t/annum (dry weight)	Agricultural application: %	Total sludge to agricultural land: t/annum	Estimated total MPs in sludge: MP particles/annum[d]	Estimated MPs from sludge to agricultural soil: MP particles/annum[e]
Argentina	2.74×10^5: mixed sludge from primary and secondary tanks (Reis, 2019)	1	>0.5	38.14–57.9% fragments, 25.2–48.86% foams, 7.0–30.5% fibres	2.74×10^5	—	—	—	—	—
Canada	1.49×10^4: sludge from primary tanks 4.4×10^3: secondary tanks (Gies *et al.*, 2018)	1	>1	65% fibres, 34% fragments 82% fibres, 21% fragments	9.65×10^{3a}	660 000 (CCME, 2012)	40 (Ryerson University, 2015)	264 000[c]	6.37×10^{12}	2.55×10^{12}
Chile	3.4×10^4 ($1.8–4.1 \times 10^4$): dewatered sludge (Corradini *et al.*, 2019a)	—	>8	90% fibres	3.4×10^4	250 000 (Ujang and Henze, 2006)	9 (Cartes *et al.*, 2018)	22 500[c]	8.5×10^{12}	7.65×10^{11}
China	2.27×10^4 ($1.6–56.4 \times 10^3$): dewatered sludge (Li *et al.*, 2018a)	28	>37	62.5% fibres, 15% shafts, 14% film, 7.3% flakes, 1.3% spheres	2.93×10^{4b}	6 250 000 (Yang *et al.*, 2015)	2.4 (Yang *et al.*, 2015)	150 000[c]	1.83×10^{14}	4.4×10^{12}
China	2.4×10^5: mixed sludge from primary and secondary tanks (Liu *et al.*, 2019a)	1	>0.8	33.5–56.7% fibres, 30.4–45.6% fragments						
China	4.01×10^3: anaerobic digested sludge (Xu *et al.*, 2020a)	1	>5	18.68% fibres, 34.07% pellets/microbeads						
Finland	1.867×10^4: anaerobic digested sludge (Talvitie *et al.*, 2017b)	1	>20	—	9.48×10^{4b}	146 000 (Eurostat, 2020)	41 (Kangas, 2017)	59 860[c]	1.38×10^{13}	5.67×10^{12}
Finland	1.709×10^5: anaerobic digested sludge (Lares *et al.*, 2018)	1	>0.8	82.8% fibres						
Germany	1.0×10^4 ($0.1–2.4 \times 10^4$): sludge from primary tanks (Mintenig *et al.*, 2017)	6	>0.2	—	1.0×10^4	1 794 443 (Eurostat, 2020)	—	423 497 (Eurostat, 2020)	1.79×10^{13}	4.23×10^{12}
Ireland	3.95×10^3: anaerobic digested sludge (Mahon *et al.*, 2017)	7	1.2–4000	76.8% fibres, 18.6% fragments	3.95×10^3	58 773 (Eurostat, 2020)	—	46 485 (Eurostat, 2020)	2.32×10^{11}	1.84×10^{11}
Italy	1.13×10^5: sludge from secondary tanks (Magni *et al.*, 2019)	1	>8	51% film, 34% fragments, 15% fibres	1.13×10^5	1 102 700 (Eurostat, 2020)	—	315 600 (Eurostat, 2020)	1.25×10^{14}	3.57×10^{13}

Table 1. Continued

Country	MP abundance: particles/kg dry sludge	Number of WWTPs	MP particle size: μm	MP particle shape	Average MP abundance: particles/kg dry sludge	Total sludge production rate: t/annum (dry weight)	Agricultural application: %	Total sludge to agricultural land: t/annum	Estimated total MPs in sludge: MP particles/annum[d]	Estimated MPs from sludge to agricultural soil: MP particles/annum[e]
Norway	6.08×10^3 ($1.701–19.837 \times 10^3$): stabilised and dewatered sludge (Lusher *et al.*, 2017)	8	>2.7	37.6% beads, 31.8% fragments, 28.9% fibres	6.08×10^3	121 300 (Eurostat, 2020)	—	66 000 (Eurostat, 2020)	7.37×10^{11}	4.01×10^{11}
Spain	5.007×10^4: SS (van den Berg *et al.*, 2020)	4	>11	82% fragments, 15% fibres, 2% film	1.03×10^{5b}	1 082 690 (Eurostat, 2020)	—	754 740 (Eurostat, 2020)	1.11×10^{14}	7.76×10^{13}
Spain	3.14×10^5: heated sludge (Edo *et al.*, 2020)	1	>25	62% fibres, 38% fragments						
Sweden	1.67×10^4: sludge from secondary settling tanks (Magnusson and Norén, 2014)	1	>300	70% fibres, 30% fragments and flakes	1.67×10^4	204 300 (Eurostat, 2020)	—	69 500 (Eurostat, 2020)	3.41×10^{12}	1.16×10^{12}
UK	1×10^3: mixed sludge from primary and secondary tanks (Murphy *et al.*, 2016)	1	>11	—	1×10^3	1 136 700 (Eurostat, 2020)	—	844 400 (Eurostat, 2020)	1.14×10^{12}	8.44×10^{11}
USA	4×10^3: dewatered sludge (Zubris and Richards, 2005)	1	>0.45	Fibres	2.5×10^{3b}	6 500 000 (Zubris and Richards, 2005)	—	4 000 000 (Zubris and Richards, 2005)	1.63×10^{13}	1.0×10^{13}
USA	1×10^3: biosolids (Carr *et al.*, 2016)	1	>10	—						

[a] Average MP abundance = average of MP abundance in primary and secondary sludge
[b] Average MP abundance = Σ(MP abundance × number of WWTPs)/Σ(number of WWTPs)
[c] Total sludge to agricultural land = total sludge production rate × % agricultural application
[d] Estimated total MPs in sludge = average MP abundance × total sludge production rate
Estimated MPs from sludge to agricultural soil = average MP abundance × total sludge to agricultural land

22–25 t/ha of SS with an average MP abundance of about 5.0×10^4 particles/kg sludge had been applied. Similarly, Corradini *et al*. (2019a) studied MPs in the topsoil (0–25 cm) of 31 agricultural fields and found that MPs accumulated in soils with successive sludge application. By considering MP abundances in SS and the amount of sludge applied to agricultural land, the amount of MPs transferred from SS sources to agricultural soil for the 13 countries considered could be estimated (refer to Table 1), ranging between 1.84×10^{11} and 7.76×10^{13} MP particles/annum, with the limits of this range corresponding to Ireland and Spain, respectively. Aside from agricultural land spreading, SSs are mainly disposed of by compost/soil amendment, landfilling and incineration, although the proportion treated by the last approach in Europe is low (Eurostat, 2020). Hence, it is reasonable to assume that most MP particles accumulated in SSs are eventually transferred to soils.

The use of treated waste water from WWTPs for irrigation of arable land has also increased over the past few decades (Lee *et al*., 2013; Sun *et al*., 2019). However, variations in treatment methodologies adopted in the WWTPs have also resulted in changes in the concentration of MP particles in irrigable waste water. Researchers have reported that up to 40% of MPs persist in the effluent discharge of WWTPs (Conley *et al*., 2019; Lares *et al*., 2018; Li *et al*., 2018a; Talvitie *et al*., 2017a) – some of which enter agricultural soils through irrigation.

Mulching activities

Extensive use of plastic or mulch film, as covering material, in agriculture consumes 2% of global production, amounting to approximately 6.96 Mt of plastic annually (Espi *et al*., 2006). PMFs are a water-efficient agricultural method that enhances crop growth, reduces weeds and pests, conserves moisture and soil nutrients and leads to higher crop yields (Bläsing and Amelung, 2018; Cook *et al*., 2006; Egley, 1983; Halley *et al*., 2001). It has been reported that more than 128 652 km^2 of agricultural land around the world is covered with PMFs (Bläsing and Amelung, 2018; Briassoulis and Giannoulis, 2018), which has produced large amounts of PMF residue, owing to its low recovery from agricultural fields. In the mechanised cultivation approach, thin sheets of PMF are spread over the farm field, with PMF thicknesses of <0.008 mm used in China, 0.020 mm in the USA and EU and 0.015 mm in Japan (Cook *et al*., 2006; Kyrikou and Briassoulis, 2007). The PMF material is subjected to seasonal weathering, which causes embrittlement and disintegration of the sheet (Kasirajan and Ngouajio, 2012; Kyrikou and Briassoulis, 2007), with macroscopic plastic debris eventually fragmenting into MPs. Since PMF is mainly composed of PE, the residual PMF does not readily degrade in soil (Halley *et al*., 2001). Even small degradation of PE-based PMF residue (or MPs) would result in leaching from the mulch film of the plasticising agents, such as phthalate, which is carcinogenic and a widely studied soil contaminant (Hüffer and Hofmann, 2016; Kijchavengkul and Auras, 2008; Liu *et al*., 2018; Wang *et al*., 2018). Thus, the existence of MPs with associated harmful counterparts may pose a significant risk to the sustenance of essential ecosystem services in agricultural landscapes. Such contaminants with MPs may disseminate in the environment by plant uptake and run-off into the surface and groundwater sources. For instance, MPs smaller than 0.5 µm accumulate in biota, which do not 'ingest' their food, such as filamentous bacteria and yeast; this may indicate potential MP magnification in soil and the food chain (Hodson *et al*., 2017; Huerta Lwanga *et al*., 2016; Schmid and Stoeger, 2016). Studies undertaken by Albertsson (1980) indicated that biodegradation of low-density PE (LDPE) buried in soil might take more than 10 years. Otake *et al*. (1995) conducted a study on 60 µm thin LDPE film buried in the soil for 32 years, only to observe partial degradation of the film. Under such conditions, MP concentrations as high as 78.0 and 62.5 particles/kg in shallow and deep agricultural soils, respectively, observed in Shanghai, China (Liu *et al*., 2018), or as low as 0.34 ± 0.36 particles/kg of dry soil in agricultural farmland in Germany (Piehl *et al*., 2018) may result in soil degradation and pose a potential threat to both aquatic and terrestrial life in the long run (Fuller and Gautam, 2016).

Landfills and landfill leachate

Three recent studies reported the detection of MPs in landfilled solid waste (Su *et al*., 2019), landfill-mined-soil-like-fractions (LFMSFs) obtained from landfill mining (Zhang and Liu, 2018) and landfill leachate (He *et al*., 2019), but without tracing their origins. The fate of MAPs (Levis *et al*., 2017) and MPs (Hale *et al*., 2020) in solid waste in landfills, and their transport beyond the waste, is even less understood than MP and NP transport in soil. Yadav *et al*. (2020) present a framework for quantifying environmental losses of plastics from landfills, although the transport of MPs is not considered explicitly. Key environmental transport pathways for landfilled plastic waste are flooding, manual scavenging (rag-picking), surface run-off, wind-driven transport and animal scavenging. Waldschläger *et al*. (2020) include landfills as reservoirs of secondary MPs in their source–pathway–receptor model, despite the lack of actual measurements of MP transport from solid waste.

Su *et al*. (2019) reported that oxidative degradation of plastics present in landfill refuse could be one of the pathways for the presence of MPs in landfill leachate. To substantiate it, Su *et al*. (2019) determined the change in carbonyl index (CI), which is defined as the ratio of the absorption peak area of carbonyl moieties to the reference peak of methylene moieties at 1780–1600 and 1490–1420 cm^{-1}, respectively, measured through Fourier transform infrared (FTIR) spectroscopy. For instance, it was reported that CI increased from 0.18 (for virgin PE) to 1.3 for MPs over time in landfills (Su *et al*., 2019). Based on this finding, Su *et al*. (2019) considered that one of the main indicators of PE biodegradation in landfill is the presence of oxidised carbonyl group at wave number 1780–1600 cm^{-1} of FTIR spectra. Analysing leachates from six MSW landfills, He *et al*. (2019) identified 17 types of MPs in the leachates, including PE, PP, PVC, PS, PET, acrylonitrile butadiene styrene and polyurethane

(PUR), with the MP concentration ranging between 0.42 and 24.58 particles/l of leachate. The majority (~77%) of the MP particles were in the 0.1–1.0 mm size range and were extracted and identified by slightly modifying the National Oceanic and Atmospheric Administration laboratory methods (Masura *et al*., 2015). The smallest detectable size in their study was 100 µm. He *et al*. (2019) reported that PE and PP MPs were found in the majority of leachate. A study of MPs in landfills in Finland, Iceland and Norway detected concentrations of up to about 5 MP particles/l of leachate and found evidence that treatment of leachate is effective in reducing MP concentrations by more than one order of magnitude (van Praagh *et al*., 2018). Further, the type of MPs in refuse and leachate depends on the state of degradation, which is generally correlated to the age of the landfill, with Su *et al*. (2019) reporting that the average abundances of MPs in refuse were over 80 MP particles/g (maximum 102 particles/g) detected in new refuse, reducing to an average of 35 MP particles/g in old refuse. Remarkably, the quoted values of MPs in refuse are comparable with, or higher than, MP concentrations in SS reported by Li *et al*. (2018a) (ranging from 1.6 to 56 particles/g of dry sludge) and around one order of magnitude higher than concentrations found by van den Berg *et al*. (2020) in agricultural soils to which MP-rich SS has been applied (5.19 particles per gram of soil). No study appears to have assessed the extent of penetration of MPs into base or cover liners in landfills or the soils beneath landfill sites. Hence, from an environmental geotechnics perspective, there is now a need to quantify plastic sources in landfills, processes of plastic fragmentation in the waste and the extent of migration of MPs into the wider environment around landfills.

Coastal beaches and deep-sea sediments

Coastal shorelines are highly dynamic environments, and MP deposition depends on various factors, which include (Gesamp, 2019) the proximity and type of human activity; the geology, geomorphology and vegetation of the coast; rainfall and prevailing wind speed and direction; and tides, ocean currents and sea temperature, among others. Depending on these factors, plastics may be transported towards the sea or the land. Plastic fragmentation results from exposure to UV radiation, followed by abrasion and/or biodegradation taking place on the shore or the sea (Andrady, 2011). Studies have reported that MP accumulation has occurred in deep-sea sediments (Kane *et al*., 2020; Pohl *et al*., 2020). Van Cauwenberghe *et al*. (2013) revealed the presence of MP particles in the top centimetre of sediment samples collected from the Porcupine Abyssal Plain (North Atlantic Ocean), distal lobe of Congo Canyon (Gulf of Guinea, South Atlantic Ocean) and the Nile deep-sea fan (Eastern Mediterranean Sea), with depths ranging from 1100 to 5000 m. Woodall *et al*. (2014) collected sediment samples from the Atlantic Ocean, Mediterranean Sea and Indian Ocean, at water depths from 300 to 2200 m, and found that MP fibres ranged between 28 and 800 pieces/l. MP abundance in deep-sea sediments (per unit volume) is hence four orders of magnitude greater than that found in contaminated sea-surface waters, indicating that the deep sea is a major sink for MP debris (Woodall *et al*., 2014). Bergmann *et al*. (2017) found MP abundance of 44–3464 pieces/l in Arctic deep-sea sediment samples from depths of 2340–5570 m. Peng *et al*. (2018) reported that MPs were found at depths of up to 11 000 m, at the deepest point on Earth, at the southern Mariana Trench, with MP abundance ranging from 200 to 2200 pieces/l in the sediment samples. Eleven different polymers were identified in this study, and the abundance of each polymer (pieces/l) at various depths in the sediments is shown in Figure 1. These authors hypothesised that the MPs found in Mariana bottom water and sediments may be derived from industrialised regions in the Northwest Pacific and the North Pacific Subtropical Gyre, where the Pacific surface circulation may lead to long-distance transport of MPs to the Mariana Trench. Except for PP and PE, all the polymer types recorded in this study are negatively buoyant and would eventually sink. Colonisation by organisms, adherence to phytoplankton and aggregation with organic debris and small organic particles will eventually enhance settling. Deposition of MPs in the Mariana Trench may have occurred because of fast vertical transport of surface-derived material or may be due to erratic downslope sediment transport triggered by occasional earthquakes and/or repeated resuspension and deposition of material. The narrow V-shaped topography of the trench may also enhance the downslope flux of MPs into the hadal zone, and bottom currents together with propagating internal tides, may further enhance the downwelling of particles and foster the accumulation of MPs in the Mariana Trench (Peng *et al*., 2018).

Groundwater systems

Despite the increasing number of studies investigating the presence of MPs in surface waters, only a few address their presence in groundwater (e.g. Bouwman *et al*., 2018; Mintenig *et al*., 2019; Panno *et al*., 2019; Połeć *et al*., 2018). Not surprisingly, MPs in groundwater are found at lower counts and particle sizes than those of surface waters, because of the longer timescales of the groundwater cycle and the potential filtration through the finer sediments. As Re (2019) points out, however, the presence of non-naturally occurring fibres such as asbestos in groundwater suggests that other similarly sized emerging contaminants, such as MP fragments and fibres, should also be expected to be transported to, and through, aquifers.

Referring to Figure 2, the MPs can be transported or introduced into the soil geoenvironment in numerous ways, such as (*a*) transportation through the vadose zone during recharge events (Bläsing and Amelung, 2018), (*b*) migration through the activity of earthworms (Huerta Lwanga *et al*., 2016; Rillig *et al*., 2017b), (*c*) transportation of the MPs in the soil matrix caused by tilling operations and crop rotation, (*d*) domestic septic tank discharges in rural areas (Panno *et al*., 2019) and/or (*e*) discharges of leachate through leaking landfill liners. MPs can also be introduced directly into an aquifer from losing streams, where fast-flowing stream water (often contaminated with MP-rich industrial discharges) recharges the aquifer (Re, 2019). Finally, MPs can be introduced much deeper in an aquifer, by managed aquifer recharge or aquifer storage and recovery systems (Re, 2019). These systems use surface run-off, stream water or treated WWTP effluent to recharge the aquifer, often at great depths in fully confined conditions, by means of injection.

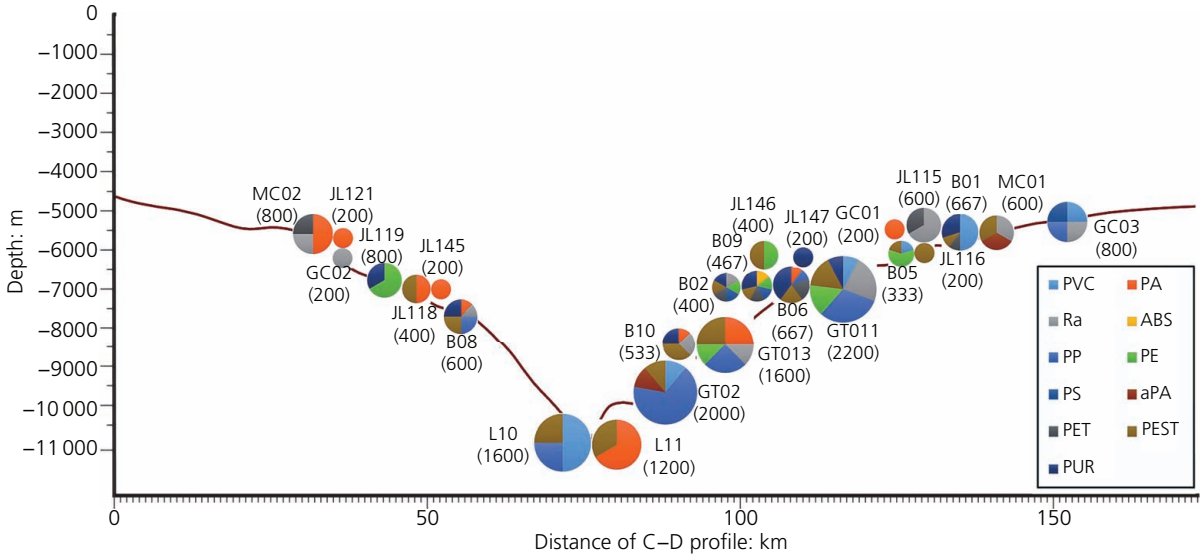

Figure 1. Profile of MP abundances and compositions in sediment samples from the Mariana Trench. Pie charts represent the MP compositions, and numbers in the brackets are the MP abundances (pieces/l). ABS, acrylonitrile butadiene styrene; aPA, aromatic polyamide; PA, polyamide; PE, polyethylene; PEST, polyester; PET, poly(ethylene terephthalate); PP, polypropylene; PS, polystyrene; PUR, polyurethane; PVC, poly(vinyl chloride); Ra, rayon. The *x*-axis corresponds to the cross-line from point C (12° north, 141.9° east) to point D (10.5° north, 141.3° east) (adapted from Peng *et al*. (2018))

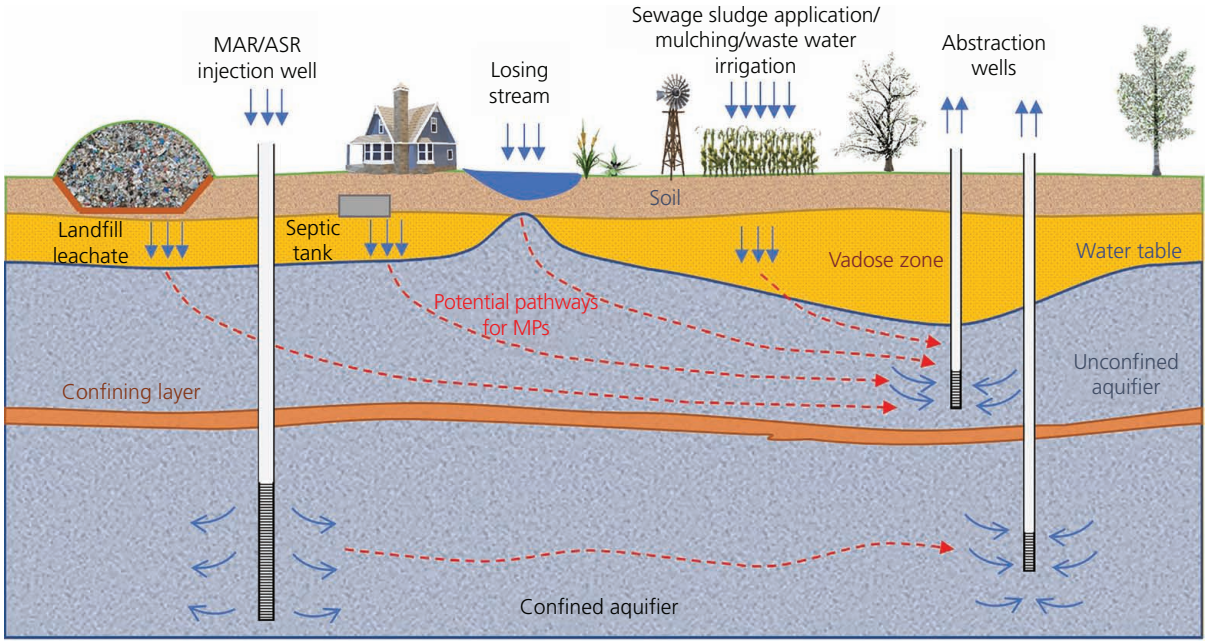

Figure 2. Schematic representation of possible pathways for MPs to enter and be transported through groundwater. ASR, aquifer storage and recovery; MAR, managed aquifer recharge

Fate and effects of MPs on the soil environment

As elaborated in the following sections, the published literature on the fate of MPs in soils and their effects on soil function is building up but remains limited, and this review is based on existing research.

Processes affecting the fate of MPs in soil

The migration, partitioning and degradation of MPs in soil environments are affected by the characteristics of the MP particles (size, shape, density and polymer type), climate (wind, rainfall and temperature), soil physics and biochemistry (e.g. soil

biota) and other environmental factors (e.g. mechanical disturbance) (Bläsing and Amelung, 2018). Moreover, MP surfaces could be positively or negatively charged due to the weathering of friction with soil particles and oxidation of sunlight to produce an electric charge, which will affect the properties and migration processes of the MPs (Mammo *et al*., 2020).

The vertical and horizontal distribution of MPs in the soil can be influenced by soil physical properties, soil biota and agronomic practices (Rillig, 2012). The migration of MPs in the horizontal direction is assisted by wind erosion and/or surface run-off (Nizzetto *et al*., 2016). It is found that the migration depth of MPs increases significantly with an increase in number of wetting–drying (w-d) cycles. O'Connor *et al*. (2019) studied MP migration in sand column experiments and reported the mobility of five different MPs, including PE and PP, of various particle sizes and densities. They reported that the smallest-sized PE MPs had the greatest movement potential, and when these MPs were subjected to greater numbers of w-d cycles, the migration depth significantly increased. Increasing the volume of infiltration liquid or the surface MP concentration had only weak effects on migration depths. Further, based on the observed w-d cycle trend, they forecasted 100-year migration depths using weather data for 347 cities across China and suggested that currently accumulated MPs in sand soils could potentially penetrate into the subsurface to depths that, in the long term, could expose subterranean fauna or aquifer system receptors.

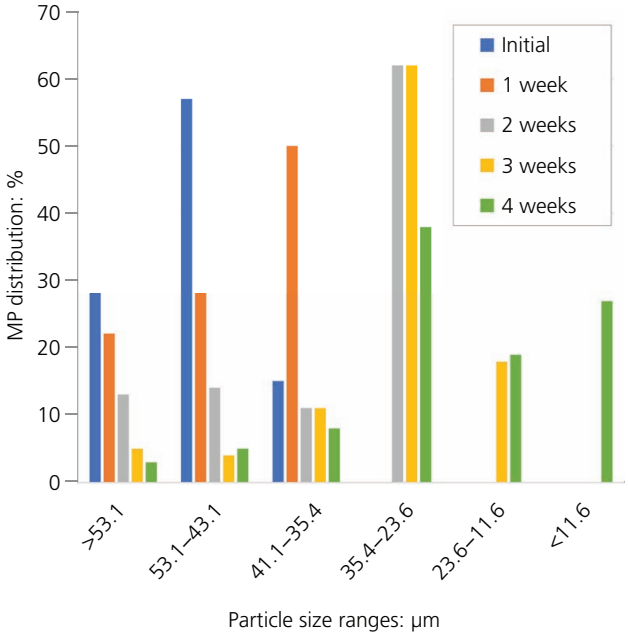

Figure 3. Particle-size distribution of LDPE MPs due to biodegradation in the earthworm gut Gram-positive bacteria, belonging to phyla Actinobacteria and Firmicutes, during a treatment period of 4 weeks (adopted from Huerta Lwanga *et al.* (2018))

Soil microorganisms can greatly affect the migration and transformation process of MPs. These are transported downwards from the ground surface by soil biota, such as earthworms and indigenous fungal mycelia, affecting the various soil horizons (Huerta Lwanga *et al*., 2016; Rillig *et al*., 2017b; Zhu *et al*., 2018). Earthworms can move MPs from the surface of the soil into its pores, horizontally and vertically; the smaller the MP particle size, the easier the migration (Huerta Lwanga *et al*., 2016; Rillig *et al*., 2017b). Zhu *et al*. (2018) investigated the effects of three soil arthropods on the migration and transformation of soil MPs. This migration increases the risk of other microorganisms contacting MPs, affects the retention time of MPs in soil, and increases the risk of MPs entering groundwater (Huerta Lwanga *et al*., 2016). Several studies (Huerta Lwanga *et al*., 2016, 2018) have shown that earthworm gut can decompose MPs in soil, but the specific mechanism needs to be verified further by experiments. Huerta Lwanga *et al*. (2016) found that earthworms selectively absorbed LDPE MPs (<150 μm sized), which accumulated in their body and were then transferred to other soil organisms. Figure 3 shows LDPE MP particle-size distribution due to Gram-positive bacterial biodegradation in the earthworm gut for a period of 4 weeks (Huerta Lwanga *et al*., 2018). From this figure, it can be seen that there are significant reductions in biodegradation time occurring for the larger-sized particles and an increase in the smaller particle ranges measuring an average of 53.1–41.3 to 35.4–23.6 μm. The authors have reported that detection of NPs and some long-chain alkanes compounds, such as octadecane, eicosane, docosane and tricosane, after 4 weeks indicates that the long-chain alkanes are products of the breakdown of the long-carbon chains of the LDPE MPs, since they were not detected in sterile soil with LDPE MPs or in soils without bacteria. Hence, it is possible that soil biota not only acts as a vector of transport of MPs in soil but can also enhance their further mobility by reducing their size.

Within the soil matrix, MPs may also bind to microorganisms and other pathogens in the soil ecosystem. Since the microbial population is smaller in deeper parts of the soil profile, decomposition/degradation of MPs by native microorganisms will be very slow, thus leading to higher retention of MPs in deeper soil layers (Kleinteich *et al*., 2018; Nizzetto *et al*., 2016). Native fungal mycelia may also aid in the downward migration of MPs through air-filled pores (Mammo *et al*., 2020).

It is clear from the above that the focus in the literature so far has been on agricultural soils, with little to no research on the fate of MPs in non-agricultural soils, such as landfill systems and their surroundings, which appear to be a major sink of MPs.

Effects of MPs on soil

MPs have a profound impact on the material cycle and energy flow of terrestrial ecosystems. Due to their adsorption characteristics, not only do entering MPs absorb organic pollutants (Beckingham and Ghosh, 2017), but they also act as carriers of heavy metals, increasing the bioavailability of the latter (Hodson *et al*., 2017) and their accumulation in the food chain through the ingestion of animals (Huerta Lwanga *et al*., 2016, 2017b). High MP levels may change soil chemistry by interfering with the degradation of soil organic

matter (SOM) (Liu *et al*., 2017; Rochman *et al*., 2014). Besides, the presence of MPs can alter the soil physical structure (i.e. bulk density and water-stable aggregate), thereby affecting water dynamics in the pores (de Souza Machado *et al*., 2018a; Piehl *et al*., 2018) and impacting on soil function and biodiversity (Rillig, 2012). For instance, PMF residue can decrease soil porosity and air circulation, change microbial communities and potentially lower farmland fertility by increasing the salt content in the topsoil (Huerta Lwanga *et al*., 2017a). MPs have been found to affect soil aggregation and, hence, water retention and hydraulic conductivity (Zhang *et al*., 2020c) and desiccation potential (Wan *et al*., 2019). de Souza Machado *et al*. (2018b) reported laboratory experiments in which PEST MPs (sized between 1.54 and 6.3 mm, with an average of 5 mm) increased the water-holding capacity of a loamy sand soil, while PEST, PE and polyacrylic MPs (average sizes between 0.643 and 3.76 mm) reduced its bulk density and microbial activity. However, adding MPs to the soil did not cause significant changes in hydraulic conductivity. Liu *et al*. (2017) found that the addition of MPs to loess soil led to a decrease in the rate of decomposition of dissolved organic matter and was hence considered beneficial for soil nutrient content. Zhang and Zhang (2020) reported that adding PEST MP fibres (mean size 2–3 mm) to a reconstituted and aggregated clayey silt led to an increase in soil organic carbon in the small soil macroaggregates (0.25–2 mm) but not the total soil samples. This increase cannot be accounted for by the distribution of the fibres themselves among the aggregates.

Effects of MPs on organisms, plants and the biogeochemical cycle

The presence of MPs in the soil alters the biotic component of the terrestrial ecosystem. The interaction of soil microorganisms with MPs leads to alteration of their metabolic pathways through changes in their enzymatic activity (Huang *et al*., 2019) and the biogeochemical cycle. The surface charges on MPs give rise to electrostatic interaction between them and microorganisms – that is, a positively charged surface of an MP could adsorb onto the surface of the microbes, or vice versa, and, in turn, be ingested when these microorganisms are fed upon by their predators (Wright *et al*., 2013b). Further, size-selective ingestion of MPs by soil-burrowing organisms, such as earthworms, contributes to the creation of MPs with reduced sizes (Rillig, 2012), which accumulate in the body of these organisms (Gaylor *et al*., 2013; Huerta Lwanga *et al*., 2016), and/or egestion of MPs (<50 μm) in the casts. Leaching of xenobiotic compounds, such as phthalates and bisphenol, from MPs and the bioaccumulation of toxic chemicals within the earthworm body significantly disrupt the endocrine system of these organisms (de Souza Machado *et al*., 2018a; Hodson *et al*., 2017; Huerta Lwanga *et al*., 2016). For instance, investigations by Gaylor *et al*. (2013) and Hodson *et al*. (2017) on the toxicological impact of MP ingestion on *Eisenia fetida* and *Lumbricus terrestris* earthworm species found that at lower MP concentrations (≤0.5% (w/w)), exposure of *E. fetida* to PS MPs (58 μm) in soils showed negligible effects, whereas higher concentrations (i.e. 1 and 2%) led to stunted growth and higher mortality rates, presumably due to the damage incurred on the self-defence mechanism (Cao *et al*., 2017). Similar observations were made on *L. terrestris* exposed to PE MPs (<150 μm). Sublethal harm to soil organisms (earthworms, springtails and nematodes) includes reproductive dysfunction (Lei *et al*., 2018), stunted growth (Galloway *et al*., 2017), weakened adaptability, internal damage (e.g. laceration), influence of substitute food on nutrient intake (Wright *et al*., 2013a, 2013b), inflammatory reactions, liver pressure and oxidative stress (Alomar *et al*., 2017). An adverse effect on the digestive system functioning of the terrestrial isopod *Porcellio scaber* exposed to PE MPs was reported by Kokalj *et al*. (2018). Further, the hydrophobic surfaces of MPs act as binding sites for various organic matter, such as lipids, proteins and nucleic acid (Shashank *et al*., 2020). The higher surface-area-to-volume ratio of MPs provides an arena for adsorption of organic matter and hence biological activity, and colonisation of microorganisms on MPs leads to the formation of MP biofilms – that is, the incorporation of MPs into the microbial cells and the extracellular polymeric substance matrix. These biofilms alter the metabolic activities of the microbial communities, leading to the development of pathogenicity, antibiotic resistance and so on among these communities, aside from the development of metabolic pathways aiding in the degradation of the MPs.

To date, the influence of MPs on plants has seldom been studied in the literature (Ebere *et al*., 2019; Li *et al*., 2020b). In the recent past, a few studies have demonstrated the influence of MPs on different plant species, such as *Lepidium sativum*, *Triticum aestivum* and *Vicia faba* (Bosker *et al*., 2019; Jiang *et al*., 2019; Qi *et al*., 2018). Bosker *et al*. (2019) demonstrated the reduction in germination rates of *L. sativum* seeds exposed to different sizes of MPs, while Qi *et al*. (2018) established the adverse effects of LDPE and biodegradable PMF residues on the root and shoot parts of the wheat plant *T. aestivum* during vegetative and reproductive growth. Significant changes were observed in plant biomass, elemental tissue composition, root traits, leaf traits and soil microbial activities for *Allium fistulosum* grown in the presence of various MPs – namely, PEST fibres, PA beads, PE, PP, PS and PEST terephthalate (de Souza Machado *et al*., 2019). Another study performed by seeding and planting *Lolium perenne* (perennial ryegrass) in soils containing biodegradable poly(lactic acid) (PLA), high-density PE and MP-clothing fibres observed a significant influence on shoot lengths, dry root biomass, dry root/shoot ratio and chlorophyll *a*/*b* ratio. Comparatively less seed germination success was achieved in the presence of MP-clothing fibres or PLA, as compared with the control soil (Boots *et al*., 2019). The study conducted by Taylor *et al*. (2020) observed no uptake of MPs in the internal root structure of *Arabidopsis* and wheat plant species, rather the accumulation of PS beads at the root surface for both species. Conversely, the study conducted by Li *et al*. (2019a), using fluorescent markers of PS, established the uptake, distribution, transportation and accumulation of 0.2 μm size PS microbeads in an edible plant species (*Lactuca sativa*). Further, microscopic examination of the shoot system also revealed the movement of PS microbeads through intercellular spaces of the vascular system, driven along the transpiration stream. Similarly, the roots of *V. faba* exposed to fluorescent MPs of various sizes have shown that the accumulation of these polymers in their

roots resulted in reduced growth (Jiang *et al*., 2019). The latter studies have contradicted the conventional understanding – that polymer molecules, being larger in size compared with plant cells, should not accumulate in plants (Li *et al*., 2020b) – and opened a new horizon in the interaction of MPs with plants from the perspective of their fate and transmission in the food chain. While the accumulation of MPs in different regions of a plant species poses serious risks and challenges from the standpoint of transmission through the food chain, it is encouraging from the perspective of a remediation strategy, in that the plants could potentially be used as means of harvesting the MPs from a contaminated soil, although the plants themselves would still pose a risk. These issues necessitate further research to understand the mechanisms of interactions and the level of susceptibility of different plant species to MP contamination.

Tools and techniques for quantification and characterisation of MPs in soils and associated challenges

Precise quantification of MPs presence in soils is critical not only for mapping their distribution and understanding their impacts but also for developing suitable remediation strategies. However, no widely accepted testing protocol exists for identifying the level of MP contamination in soils. Focusing on the studies of MPs in natural soil, manufactured polluted soil, landfill refuse, landfill leachate and compost/fertiliser, Table 2 summarises detection details in terms of currently available identification equipment, spectral range, filter material/size, quality control test and MP type/concentration in the top 21 studies published since 2016.

Based on this literature review and the authors' own experiences, the following steps, or different combinations thereof, are usually employed:

(*a*) determination of target MPs
(*b*) sample preparation
(*c*) visual inspection
(*d*) destructive and non-destructive testing
(*e*) fast detection methods
(*f*) reporting of results.

These steps are discussed in turn next.

Determination of target MPs

Identifying test targets, including plastic type, shape and size, is an important first step because, collectively, they will guide the choice of test methods as described below. In typical soil samples, PE and PP are the most commonly found MPs, while others (e.g. PVC and PET) are also widely reported.

Sample preparation

After representative soil samples were collected, they were dried, disaggregated, sieved, floated, filtered, density-separated and/or pretreated depending on their SOM content (He *et al*., 2018a). Although sieving and density separation appear to be effective in isolating MPs in soil, concern may arise due to the presence of

- SOM that may potentially embed MPs and hence interfere with extraction efficiency (Zhang *et al*., 2018) and chemical identification accuracy (Bläsing and Amelung, 2018)
- higher-density MPs (e.g. PVC, PET and nylon) that settle out, even in density separation columns filled with saturated salt solutions, such as sodium chloride (NaCl), calcium chloride ($CaCl_2$) or zinc chloride ($ZnCl_2$).

To address the first issue, an intermediate stage of oxidising the SOM can be performed, either by addition of oxidising chemicals (e.g. acids, alkalis or hydrogen peroxide) or through enzymatic digestions. Among these pretreatment methods, treatment with hydrogen peroxide has been the most preferred because of its minimal influence on MP surfaces compared with other methods. For separating higher-density MPs from sediments, another methodology has been proposed that consists of a number of extraction cycles using saturated sodium chloride solution, followed by ultrasonic treatment prolonging the time of floatation (Liu *et al*., 2018).

Pretreated samples are usually filtered through a filter membrane before visual inspection and chemical identification. The filters used for MPs identification in previous soil studies vary and include filters based on nylon, glass fibre, cellulose nitrate and aluminium oxide (see Table 2). To detect MPs accurately, it is vital to ensure that the filter has a smooth surface, good contrast and low spectroscopic background. According to the authors' own experiences, a filter with a very rough surface, such as glass fibre filter, is not suitable for MP detection. This is particularly true for the detection of small, thin and nearly transparent MPs (<100 μm). For the test of MPs larger than 20 μm, cellulose nitrate and aluminium oxide filters are suggested due to their relatively smooth surface. For MPs between 1 and 20 μm, metal-coated polycarbonate (PC) filters (e.g. gold (Au)-coated PC or aluminium (Al)-coated PC) are suggested because of their very smooth surface and low background Raman signal (Oßmann *et al*., 2017; Schymanski *et al*., 2018). Table 2 shows that a wide range of filter pore sizes have been reported in the literature (from 0.2 to 20 μm), which makes comparability a challenging task.

Besides filter requirements, it is also necessary to ensure that samples are well mixed to avoid potential floating or sedimentation of MPs. When high quantities of MPs are present in the sample, care must be taken to avoid aggregation or overlapping particles in the filtering process.

Visual inspection

Visual inspection of MPs by particle shape, size, texture and colour using a microscope is suitable for the detection of large MPs (ranging 0.5–5 mm) (Doyle *et al*., 2011), due to the significant differences of plastics from other organic/inorganic soil particles – that is, apart from aluminium silicates, which have identical visual appearance (i.e. illuminating surface) as that of the MPs and, if present in the soil, would lead to misidentification and inaccurate assessment. Combined with a high-resolution camera, it is possible to detect smaller MPs and determine their size. This method is non-

Table 2. Summary of MPs detection in soil media, landfill refuse and leachate (continued on next page)

Sample type	MP type	MP size and shape	Identification method	Filter (material, pore size)	Blank sample	Recovery efficiency test	Determined spectral range	MP concentration[a]	Reference
Industrial site soil	PVC	NA	GC-MS, FTIR	Glass fibre filter	Y	Glass bead, recovery efficiency: 101–111%	4000–600 cm^{-1}	0.3–67 mg/kg soil	Fuller and Gautam (2016)
Home garden soil	NA	10–50 μm	Microscopy	NA	N	N	No need	870 ± 1900 particles/kg soil	Huerta Lwanga *et al.* (2017a)
Costco beach soil	PE, PP, PS, PEUR, PE + PP blend	<1 mm	ATR-FTIR and SEM analysis	NA	Y	97%	4000–650 cm^{-1}, at a spatial resolution of 4 cm^{-1}	1.3–14 712.5 particles/kg dry soil	Zhou *et al.* (2018)
Floodplain soil	PE	<500 μm, particles	Raman, FTIR	Aluminium oxide filter, 0.2 μm	Y	N	Transmission mode, 4000–1250 cm^{-1}	Up to 55.5 mg/kg and up to 593 particles/kg soil	Scheurer and Bigalke (2018)
Agricultural soils: fruit field, greenhouse field	PP, LDPE	>100 μm	Microscopy	Glass slide	Y	Irregularly shaped particles PP and LDPE, 90%	No need	<0.54 mg/kg soil	Zhang *et al.* (2018)
Vegetable field soil	PP, PE	NA	μ-FTIR	Nylon net filter, 20 μm	Y	PP, PE, PA, PC, ABS, PMMA and PS particles, up to 90%	4000–675 cm^{-1}	62.5–78.0 particles/kg soil	Liu *et al.* (2018)
Greenhouse field/buffer zone soil	NA	50 μm–1 mm, fibres	Microscopy	NA	Y	N	No need	7100–42 960 particles/kg soil	Zhang and Liu (2018)
Agricultural soil	PE	>2 mm, fragments and films	ATR-FTIR	NA	Y	N	4000–400 cm^{-1}	0.34 ± 0.36 g/kg dry soil	Piehl *et al.* (2018)
Rice–fish culture field soil	PP, PE	<1 mm, fibres	μ-FTIR	Net filter, 20 μm	Y	N	4000–675 cm^{-1}	10.3 ± 2.2 particles/kg soil	Lv *et al.* (2019)
Agricultural soil without/with sludge application	NA	Fibres	Microscopy	Ashless paper, 2.5 μm	Y	Y	No need	200–600 particles/kg without sludge application 1100–3500 particles/kg with sludge application, dry soil	Corradini *et al.* (2019a)
Agricultural soil	PEST	NA	μ-FTIR	Glass fibre filter	Y	PE, PVC, tyre particles, PET, 80–97.3%	4000–650 cm^{-1}, at a spatial resolution of 4 cm^{-1}	4–541 particles/kg soil	Crossman *et al.* (2020)
Agricultural soil without/with sludge application	PP, PVC	150–250 μm, fragments	μ-FTIR	Cellulose, 11 μm	N	N	3500–1000 cm^{-1}, at a spatial resolution of 4 cm^{-1}	930–1100 particles/kg without sludge application 2130–3060 particles/kg with sludge application	van den Berg *et al.* (2020)
Vegetated wetland soil	PS, PE, synthetic rubber	NA	FTIR	NA	Y	N	4000–525 cm^{-1}	1270 ± 150 particles/kg soil	Helcoski *et al.* (2020)

Table 2. Continued

Sample type	MP type	MP size and shape	Identification method	Filter (material, pore size)	Blank sample	Recovery efficiency test	Determined spectral range	MP concentration[a]	Reference
Agricultural soil without/with sludge application	PP	>0.2 mm	Microscope and μ-FTIR	0.45 μm glass fibre filter	Y	N	NA	5.0 ± 0.4 particles/kg without sludge application 87.6 ± 9.6 to 545.9 ± 45.7 particles/kg soil with sludge application	Zhang *et al.* (2020a)
Lab-made polluted soil	LDPE, PET, PVC	0.5–1 mm	Vis-NIR	No need	N	Y	Near-infrared range, 350–2500 nm	Detection limit 15 g/kg soil	Corradini *et al.* (2019b)
Lab-made polluted soil	PE, PP, PET, PS	<125 μm	High-throughput NIR	No need	N	Y	Near-infrared range, 667–2500 nm	Detection range 5–50 g/kg soil	Paul *et al.* (2019)
Lab-made polluted soil	PET, LDPE	<5 mm	Vis-NIR	No need	N	Y	Near-infrared range, 350–2500 nm	Detection range 5–50 g/kg soil	Ng *et al.* (2020)
Landfill refuse	PE	0.23–4.97 mm, fragments, MPs dominated (accounting for 59.82%)	μ-FTIR and stereomicroscopy	Nylon filters (pore size of 20 μm)	Y	88.3 ± 4.2%	Wave number range of 4000–600 cm^{-1}, resolution of 4 cm^{-1}	20–91 particles/g	Su *et al.* (2019)
Landfill leachate	PE and PP	77.48% MPs were between 100 and 1000 μm, fragments	FTIR	0.45 μm filter membrane	Y	N	NA	0.42–24.58 particles/l	He *et al.* (2019)
Landfill leachate	Cellophane	0.07–3.67 mm, fibres	μ-FTIR and stereomicroscopy	Nylon filters (pore size of 20 μm)	Y	91.7 ± 5.9%	Wave number range of 4000–600 cm^{-1}, resolution of 4 cm^{-1}	4–13 particles/l	Su *et al.* (2019)
MSW compost and amended soil	NA	NA	TEM and GC-MS	0.45 μm	N	N	Scan range *m/z* 40–700	NA	Watteau *et al.* (2018)
Organic fertilisers from biowaste fermentation	Styrene-based polymer	2–5 mm	SEM and ATR-FTIR	NA	N	N	3940–800 cm^{-1}	20–146 particles/kg dry organic fertiliser	Weithmann *et al.* (2018)

[a] Only four of the listed studies clearly stated the soil sample moisture condition (dry) at result report; the remaining cases did not specify the soil samples as natural or dry state

ABS, acrylonitrile butadiene styrene; ATR, attenuated total reflection; FTIR, Fourier transform infrared spectroscopy; GC-MS, gas chromatography–mass spectrometry; LDPE, low-density polyethylene; NA, not available; PA, polyamide; PC, polycarbonate; PE, polyethylene; PEST, polyester; PET, poly(ethylene terephthalate); PEUR, polyether urethane; PMMA, poly(methyl methacrylate); PP, polypropylene; PS, polystyrene; PVC, poly(vinyl chloride); SEM, scanning electron microscopy; TEM, transmission electron microscopy; vis-NIR, visible–near-infrared spectroscopy

destructive, particle number countable and easy to perform by an operator. Hence, it is a relatively fast method to obtain preliminary results. Image technology, such as scanning electron microscopy (SEM), can also be used to obtain detailed surface morphology. The SEM technique has been employed to obtain magnified and clearer micrographs of MP particles, although the time required in sample preparation and the need for coating the samples with conductive paints make the identification of MPs based on surface texture and colour a difficult task (Shim *et al*., 2017). Furthermore, the size, shape and surface area of the determined MPs can be analysed by using image-processing software, such as ImageJ.

The main disadvantage of visual inspection is its potentially low accuracy because the results are influenced by operators' experience and quality of eyesight, sample type, location of the observation point and microscope quality (e.g. resolution and contrast). Furthermore, visual inspection does not allow the operator to distinguish different types of plastics.

Destructive and non-destructive testing

The limitations of visual inspection can be overcome by combining it with non-destructive (FTIR or Raman spectroscopy) and/or destructive (gas chromatography–mass spectrometry (GC-MS)) chemical identification analyses. FTIR-based methods can screen out MPs larger than 20 μm, while the Raman method can detect MPs larger than 1 μm (Araujo *et al*., 2018; Käppler *et al*., 2016; Li *et al*., 2020c; Schwaferts *et al*., 2019). With a high-numerical-aperture objective and a short excitation wavelength, the spatial resolution of Raman spectroscopy can be extended to around 0.5 μm. SEM, mentioned earlier, can provide clear images up to nanometre resolution. Overall, as the size of MPs decreases, the test time and the cost of characterisation increase. Both FTIR and Raman spectroscopy are accurate and reliable in determining the chemical identity and lateral size of MPs. However, both methods are time consuming, although FTIR is relatively more efficient than Raman for MPs larger than 20 μm.

There are usually three different spectral ranges used to distinguish MPs in Raman spectroscopy: 759–709 cm^{-1} (symmetric stretching vibration of CF_2), 1640–1580 cm^{-1} (aromatic bending vibration) and 2780–2980 cm^{-1} (stretching vibrations of $CH/CH_2/CH_3$ groups). For FTIR spectroscopy, there are five different spectral ranges: 1174–1087 cm^{-1} (CF_2 stretching vibration), 1480–1400 cm^{-1} (CH_2 bending vibration), 1760–1670 cm^{-1} (C=O stretching vibration), 1800–1740 cm^{-1} (C=O stretching vibration) and 2980–2780 cm^{-1} (stretching vibrations of $CH/CH_2/CH_3$ groups) (Käppler *et al*., 2016). It is also necessary to consider the potential filter background interference when choosing the detection spectral range.

Moreover, during testing of MPs using Raman spectroscopy, a low excitation laser power should be applied on the particles to avoid damaging or decomposing small-sized MPs (Schymanski *et al*., 2018). To obtain the clear spectra of small MPs, increasing accumulation and exposure times is suggested rather than directly increasing laser intensity.

The GC-MS method can rapidly and precisely quantify PE, PP, PS and PET contents (Dümichen *et al*., 2017) However, melting polymeric particles would lead to loss of information about particle-size distribution, which is critical information for pollution assessment. Hence, if data on the size and number of MPs are required, GC-MS is not recommended. Other recent advancements for detecting MPs in soil include the thermal extraction desorption and hyperspectral imaging technology with the potential to assess directly the presence of MPs in the soil surface (Shan *et al*., 2018). Finally, energy-dispersive X-ray analysis gives the atomic composition of the specimen.

It is noteworthy that the analysis time of soil samples varies dramatically depending on sample complexity, MP content and size, test area of the filter, chemical determination method and personal experience. Given a typical soil sample and the current technology, the digestion and separation time is usually around 1 to 10 h (Liu *et al*., 2018), although it could be weeks for organic-rich samples (Scheurer and Bigalke, 2018). After proper filtration, the visual inspection and chemical detection for a whole filter (e.g. 47 mm in diameter) ranges from approximately 9 h to a few days using FTIR (Chen *et al*., 2020; Renner *et al*., 2020; Tagg *et al*., 2015). Hence, the total analysis time, including proper sample preparation and chemical identification, of a single soil sample could be at least approximately 10 to 100 h.

Fast detection methods

In addition to the methods discussed earlier, extraction-free methods for rapid MP detection in soil have also been reported. The near-infrared (NIR)-based method has been attracting increasing attention in the past 2 years (Corradini *et al*., 2019b; Ng *et al*., 2020; Paul *et al*., 2019). Using a portable spectroradiometer over the range 350–2500 nm (vis-NIR), the concentration of spiked MPs (PET and LDPE) can be successfully determined (Ng *et al*., 2020). Although it is still in the early stage of laboratory testing, the NIR-based method is a noteworthy technology for cost-effective detection of MPs.

Reporting of results

Due to the differences in measurement approaches and units reporting results, it is difficult to compare between soil studies. For example, in terms of units, 'mass of MPs per mass of soil', 'number of MP particles per mass of soil' and both of these have been adopted in 2, 11 and 1 studies, respectively, out of the top 14 studies shown in Table 2. By combining size distribution and number of MPs, it is possible to assess roughly the mass of MPs in soil. However, it is impossible to do the reverse. The water content of soil is another important information, but only three of the 14 studies referred to specify whether the test soil is dry. Hence, standardisation of the reporting format is critical to increase the comparability of studies. For instance, the number of particles/kg wet soil for critical size ranges marked by agreed classification points (e.g. 100, 20 and 1 μm) should be an agreed part of result reporting, along with measured soil water content and calculated MP particles/kg dry soil values. Furthermore, it is important to supply the details of a blank sample and recovery efficiency test, which can be used for study comparison

and for uncertainty assessments. Figure 4 shows the suggested analytical protocol for the determination of MPs in soil media.

Table 3 summarises the advantages/disadvantages of the main inspection and detection methods discussed earlier.

It can be seen from Tables 2 and 3 that the most commonly used test procedures are collection and pretreatment (e.g. density separation and digestion) for MP extraction, followed by MP characterisation and identification. In terms of MP detection, out of the 21 cases summarised in Table 2, four studies have used visual inspection alone (e.g. microscopy) to determine large-sized MPs. In comparison, 14 studies have used FTIR spectroscopy (usually equipped with a microscope) to determine the chemical identity of MPs. Among these, four studies combined FTIR with GC-MS, SEM or Raman spectroscopy in order to obtain more data on the MPs (e.g. morphology and chlorine concentration). There is a clear tendency of moving away from visual inspection of large-sized MPs towards accurate chemical identification of small-sized MPs. For NPs and MPs smaller than a 500 nm size, however, there is no robust method to determine simultaneously the size and chemical properties due to the limits of current detection methodologies. Some studies

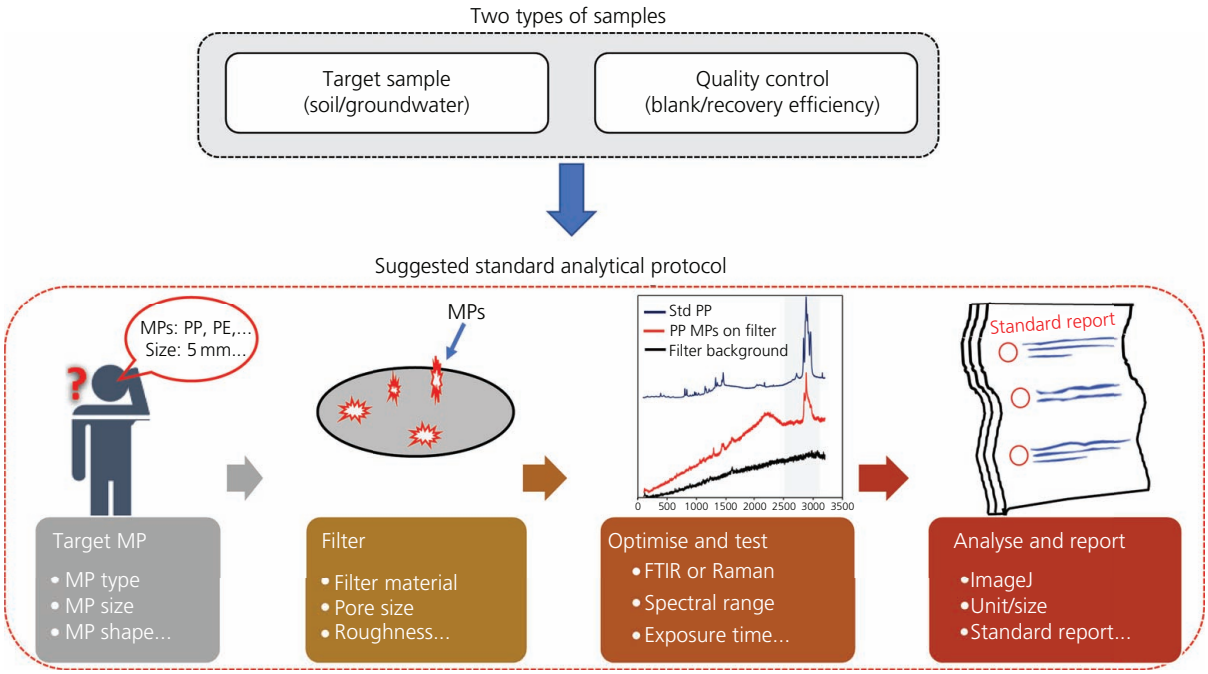

Figure 4. Suggested standard analytical protocol for determination of MPs in soil/groundwater

Table 3. Comparison of MPs detection methods

Advantage	Typical methods			Extraction-free methods		
	Visual/microscopy	FTIR	Raman	GC-MS	SEM/SEM-EDX	Vis-NIR/HT-NIR
Low cost	☺	😐	😐	😐	😐	😐
Low professional training requirement	☺	☹	☹	☹	☹	😐
Fast test	😐	😐	😐	☹	☹	😐
Particle number countable	☺	☺	☺	☹	☺	☹
Non-destructive	☺	😐	😐	☹	😐	😐
Suitable for large MPs (0.5–5 mm)	☺	☺	☺	😐	☺	☹
Suitable for median MPs (20–500 μm)	☹	☺	☺	😐	☺	☹
Suitable for small MPs (1–20 μm)	☹	☹	☺	😐	☺	☹
Suitable for nano-sized MPs (<1 μm)	☹	☹	☹	😐	☺	☹
Suitable for chemical identification	☹	☺	☺	☺	😐	☺
Wide range of particle sizes	☺	😐	😐	☹	☺	☹
High accuracy	☹	😐	☺	☺	☺	☹

☺, has this advantage; 😐, potentially has this advantage; ☹, does not have this advantage
EDX, energy-dispersive X-ray analysis; FTIR, Fourier transform infrared spectroscopy; GC-MS, gas chromatography–mass spectrometry; HT-NIR, high-throughput near-infrared spectroscopy; Raman, Raman spectroscopy; SEM, scanning electron microscopy; Vis-NIR, visible–near-infrared spectroscopy

(Hernandez *et al*., 2019; Schwaferts *et al*., 2019) have been successful in detecting these particles in pure and simple samples using light scattering, microscopy imaging, spectroscopy or pyrolysis GC-MS methods. For instance, the sizes of NPs and small MPs released from a teabag in hot water (Hernandez *et al*., 2019) were determined by SEM, while the chemical properties were determined by X-ray photoelectron spectroscopy, separately. Since the majority of these particles in the samples are NPs, the detected size can be assumed to be highly representative of the actual NP particle-size distribution. However, this is not the case for NPs in soil samples mixed with many other types of interference particles. The nature of NP aggregation is also a challenge to analyses. Hence, robust test methods for NPs and small-sized MPs in complex media, such as soil, are required. Further, no discussions have been found in the literature on how to avoid physical breakdown of degraded MP particles into smaller-sized MPs during sample preparation or how to adapt extraction procedures specifically to coarse- and fine-grained soil samples.

In summary, there is no single perfect method for MP detection in soil media. The combined application of different methods (visual inspection and chemical detection) seems a better choice for MP research, although the balance of cost, accuracy and time must be taken into consideration.

MPs as carriers for other contaminants

Role of MPs as carriers into and within aquifers

Although MPs are considered chemically inert, they can absorb persistent organic pollutants (POPs) and metals and thereby become carriers of harmful substances in the subsurface (Połeć *et al*., 2018). As they are transported through varying terrestrial subsurface chemistry conditions (Sarris *et al*., 2019; Wilson *et al*., 2020), points or locally distributed sources of MPs can form at locations further along the flow path. In the marine environment, MPs have been shown to function as vectors for the extended survival and dispersal of pathogens (Kirstein *et al*., 2016). Similar effects should be expected in the terrestrial subsurface environment, potentially resulting in significantly reduced pathogen die-off rates and increased travel distances, with consequent significant implications for the safety of groundwater drinking supplies and the characterisation of wells as safe sources of drinking water.

Notably, in manufacturing of plastics, toxic additives are included to enhance their final functional properties, such as plasticisers, flame retardants, antioxidants, acid scavengers, light and heat stabilisers, lubricants, pigments, anti-static agents, slip compounds and thermal stabilisers (Hansen *et al*., 2013). Only the reactive organic additives – for example, some flame retardants – are polymerised with the plastic molecules and become part of the polymer chain, while most additives are not chemically bound to the plastic polymer (Hansen *et al*., 2013). Release of toxic additives from various plastic products has been reported in earlier studies – for instance, phthalates (Rijk and Ehlert, 2001; Tønning *et al*., 2010); brominated flame retardants (Kim *et al*., 2006); bisphenol-A dimethacrylate (Olea *et al*., 1996); lead (Pb), tin and cadmium (Al-Malack, 2001); formaldehyde and acetaldehyde (Mutsuga *et al*., 2006); 4-nonylphenol (Fernandes *et al*., 2008; Loyo-Rosales *et al*., 2004); methyl *tert*-butyl ether; benzene (Skjevrak *et al*., 2003); and many other volatile organic compounds (Hennesuse-Boxus and Pacary, 2003).

Combined effect of MPs with other pollutants in soil ecosystem

Due to their small particle size, large specific surface area (SSA) and strong hydrophobicity, MPs can increase the mobility of other pollutants in the terrestrial subsurface by

- sorbing and carrying them
- changing soil properties that lead to more release and transport.

Heavy metals, POPs and antibiotics have been detected on the surfaces of MPs (Hartmann *et al*., 2017; Rochman *et al*., 2013; Teuten *et al*., 2009). As a suitable carrier of these substances, MPs have a complex effect on the soil environment, which, depending on the setting, may contain a large number of clay minerals, metal oxides and hydroxides, humus and microorganisms along with other natural substances that can be combined with environmental pollutants. MPs can also trigger changes in the soil properties that affect the uptake of other soil pollutants present in the geoenvironment, altering the chemical forms and bioavailability of these soil contaminants (see the section headed 'Effects of MPs on soil'). However, far more research is still required to generate solid evidence on these interactions and their effects on the environment.

Affinity of heavy metals towards MPs in soil

The intrusion of MPs affects the bioavailability of heavy metals in the soil environment and their mobility into deep soil layers (or groundwater) by way of cracks in the soil and/or its porous matrix. There is no doubt that MPs can adsorb heavy metals from soil matrices and act as a vector for their transfer to soil biota and/or groundwater. Hence, it is important to understand the mechanisms driving the affinity of heavy metals towards MPs in soils and soil–groundwater. The authors' review of the literature has revealed that, while several studies have been conducted on the interaction between MPs and heavy metals in various environments, knowledge pertaining specifically to such interactions in the terrestrial subsurface remains sparse, sporadic and not always consistent (Boots *et al*., 2019; Brennecke *et al*., 2016; de Souza Machado *et al*., 2018a; Ebere *et al*., 2019; Hodson *et al*., 2017; Liu *et al*., 2018; Rillig *et al*., 2019; Rochman *et al*., 2014; Tang *et al*., 2018; Wang *et al*., 2017, 2018, 2020; Wirnkor *et al*., 2019; Xu *et al*., 2020b; Zhou *et al*., 2019; Zon *et al*., 2018). This may be due to the large number of influencing factors, with each reported experimental study investigating only a limited few.

Accumulation of metals may proceed through interactions between divalent cations and oxyanions with charged or polar sites of the plastic surface and through non-specific interactions between neutral metal–organic complexes and the hydrophobic

MP surfaces (Holmes *et al*., 2012; Wang *et al*., 2017). For example, Zou *et al*. (2020) found that lead exhibited significantly stronger sorption than did copper (II) (Cu^{2+}) and cadmium (Cd^{2+}) because of strong electrostatic interactions. Furthermore, a clear trend of higher concentration of MP-sorbed heavy metals (aluminium and lead) with a higher temperature has been established based on experiments performed on PET, PA and ethylene vinyl acetate granules for temperatures ranging from 25 to 55°C (Oz *et al*., 2019). However, the effect of pH on interactions between MPs and heavy metals is not yet clear.

Possible attributes affecting the tendency of MPs to attract heavy metals can be grouped into two categories:

- those inherent to the MP particles (e.g. size, surface properties, porosity, morphology, material type, manufacturing process, pigments/masterbatch used for colour, natural ageing of the MP material)
- environmental conditions (e.g. dissolved organic matter, ionic concentration and salinity of the soil pore water, properties of the soil solids, contact time, pH value, temperature, outdoor weathering effects, formation of biofilm on MP surfaces).

In view of the different physical and chemical properties of MPs, the adsorption rate of heavy metals by MPs will vary greatly. Further, the surface structure of MPs changes due to oxidation and weathering by sunlight, and surfaces easily obtain charge, such that they readily adsorb metal ions to achieve charge balance (Hodson *et al*., 2017; Massos and Turner, 2017). The residence time is another important factor. For example, after a long time of UV irradiation of PVC MP fragments, the amount of copper (II) and zinc (Zn^{2+}) ions adsorbed on MPs has been found to increase (Bandow *et al*., 2017). Besides, metal cations are adsorbed by combining with polar regions or oxygen anions on the surface of plastics and forming complexes with organic compounds. For convenience, a list of several of the aforementioned factors investigated with respect to metal and plastic types is presented in Table 4. A note of caution here is that given the affinity of heavy metals towards MPs is dependent on many factors, it may be misleading to estimate accumulation based on one-to-one correlations.

The affinity of heavy metals towards MPs in the terrestrial subsurface can be significantly altered through the bioaccumulation process (Dobaradaran *et al*., 2018; Rochman *et al*., 2014; Wang *et al*., 2017). The reactivity of the MP surfaces is further enhanced by a change in their inherent properties due to ageing/weathering or, as shown in Figure 5, by the formation of a biofilm and chemical precipitates (Holmes *et al*., 2012). In fact, biofilm becomes the strongest predictor variable of metal accumulation, dominating over the substrate material and locations from where the MPs were collected. Accumulation of biofilms on MP surfaces alters their physical properties (Rummel *et al*., 2017), such that they attract higher concentrations of heavy metals. Similarly, aged MPs have a greater affinity towards heavy metals, as shown in Figure 5(b), on account of their higher SSA and roughness caused by the degradation of plastics mainly due to long-term photo-oxidation, corrosion, friction or other processes (Brennecke *et al*., 2016; Lang *et al*., 2020; Wang *et al*., 2020).

Furthermore, pigments used in the manufacturing of plastic contain certain components, such as titanium and iron oxide, which favour the formation of negatively charged surface complexes that subsequently have a greater affinity for metal cations. The adsorption of MPs is greatly enhanced with an increase in SSA and polarity. For example, accumulation of copper (II) ions is significantly greater in PVC than in PS, probably due to higher SSA and higher polarity (Brennecke *et al*., 2016). Finally, although the sorption coefficients of heavy metals on MPs are rather low compared with those of hydrophobic organic compounds (HOCs) on MPs, considering a similar initial concentration range, aged MPs of smaller sizes have a greater affinity towards heavy metals (Xu *et al*., 2020b).

Affinity of organic pollutants towards MPs in soil

Organic pollutants, such as polycyclic aromatic hydrocarbons (PAHs), hexachlorinated hexanes, polybrominated diphenyl ethers and polychlorinated biphenyls (PCBs), in the geoenvironment may sorb to MPs depending on the type of polymer (Kleinteich *et al*., 2018; Rochman *et al*., 2013; Yu *et al*., 2019). As such, MPs in the soil are ideal carriers of PAHs, PCBs, pesticides, herbicides and other hydrophobic organic substances, which can directly affect the distribution of POPs in the soil environment and directly threaten the health of the soil ecosystem. Several studies (Horton *et al*., 2017; Hüffer and Hofmann, 2016; Seidensticker *et al*., 2018) have found that the concentration of organic pollutants on MPs in the soil is much higher than that in the surrounding soil environment, indicating the synergistic threat of MPs and POPs on the soil regional system. Any synergetic or antagonistic effects between POPs and MPs depend on the molecular polarity of persistent organic compounds. HOCs usually lead to more serious synergetic pollution. The main factors affecting physical adsorption are the SSA and van der Waals forces, while the affinity of organic pollutants to the hydrophobic surface of MPs is the key factor affecting chemical adsorption. The hydrophobicity, crystallinity, functional groups and electrostatic attraction between MPs and organic compounds belong to their own physico-chemical properties (Mato *et al*., 2001), which are internal factors affecting the adsorption of MPs. Most compounds in the environment have antagonistic or synergistic effects. When adsorbed by MPs, there will be different adsorption capacities and competitive adsorption may occur. Hüffer and Hofmann (2016) studied the adsorption behaviours of four kinds of MPs – namely, PS, PVC, PA and medium-density PE – with seven kinds of aliphatic substances – namely, *n*-hexane, cyclohexane, benzene, toluene, chlorobenzene, ethyl benzoate and naphthalene – and found that the adsorption coefficient of MPs was closely related to their hydrophobicity. Similarly, studies undertaken by Seidensticker *et al*. (2018) reported that the adsorption of hydrophobic compounds was stronger than that of neutral substances.

Table 4. Factors possibly affecting the affinity of heavy metals towards MPs in soils and soil–groundwater

Attribute	Metal	MPs	MP particle size	Reference
MP SSA and polarity, surface properties and ageing	Copper (Cu), zinc (Zn)	PS beads, PVC	PS beads (0.7–0.9 mm dia.); PVC (1.6 × 0.8 mm)	Brennecke *et al*. (2016)
MP particle size and colour	Aluminium, cadmium (Cd), chromium (Cr), copper, iron (Fe), manganese (Mn), nickel (Ni), lead	Randomly collected MPs along Persian Gulf	2–5 and <0.25 mm	Dobaradaran *et al*. (2018)
MP SSA, porosity and morphology, dissolved organic matter, pH	Cadmium, cobalt (Co), chromium, copper, nickel, lead, zinc	PE, PP, PS, PVC	<5 mm (shredded into fragments, with irregular shapes and thicknesses)	Godoy *et al*. (2019)
MP SSA, weathering, formation of organic complexes on surfaces	Cadmium, cobalt, chromium, copper, nickel, lead	PE pellets	<1 mm (sieved through 1 mm nylon mesh)	Holmes *et al*. (2014)
MP types, pH, duration of contact, initial concentration of heavy metals, temperature	Aluminium, lead	EVA granules, PA, PET	0.5–1 mm	Oz *et al*. (2019)
Formation of biofilm on MPs	Aluminium, barium (Ba), cobalt, caesium (Cs), copper, iron, gallium (Ga), potassium (K), magnesium (Mg), manganese, nickel, lead, rubidium (Rb), uranium (U)	Plastic pellets (PLA, LDPE)	Spherical PLA pellets (4 mm dia.) and cylindrical LDPE (3 mm long, 5 mm dia.)	Richard *et al*. (2019)
MP SSA, weathering and formation of biofilm	Silver (Ag), aluminium, calcium (Ca), cadmium, cobalt, chromium, copper, iron, mercury (Hg), manganese, nickel, lead, zinc	LDPE, HDPE, PET, PP, PVC	Cylindrical PET pellets (3 mm long, 2 mm dia.) and spherical pellets of LDPE, HDPE, PP and PVC (3 mm dia.)	Rochman *et al*. (2014)
MP ageing, temperature, pH	Copper, zinc	PET debris	<5 mm (secondary MPs produced by decomposition of larger plastics due to external forces)	Wang *et al*. (2019)
MP particle size, pH	Cadmium	HDPE	48–58 μm, 100–154 μm, 0.6–1 mm, 1–2 mm	Zhang *et al*. (2020b)
MP weathered surface, formation of biofilm	Toxic metals	PE, PEUR, PP, PS, polymer blend of both PE and PP	Majority of MPs (about 60%) composed of particles <1 mm, with 100–250 μm size fractions accounting for about 50% of those particles	Zhou *et al*. (2018)
MP SSA and hydrophobicity	Silver, cadmium, chromium, copper, iron, mercury, manganese, lead, antimony (Sb)	PA, PE, PP, PS, PVC	81.7% of MPs measured in the range of 10–100 μm	Zhou *et al*. (2019)
Chemical structure and electro-negativity of the sorbents, crystallinity of MPs, pH, electrostatic interaction	Cadmium, copper, lead	CPE, PVC, LDPE and HDPE	<5 mm (defined as MPs)	Zou *et al*. (2020)

CPE, chlorinated polyethylene; EVA, ethylene vinyl acetate; HDPE, high-density polyethylene; LDPE, low-density polyethylene; PA, polyamide; PE, polyethylene; PET, poly(ethylene terephthalate); PEUR, polyether urethane; PLA, poly(lactic acid); PP, polypropylene; PS, polystyrene; PVC, poly(vinyl chloride); SSA, specific surface area

The presence of MPs in soils may make organic pollutants less available to soil biota and restrict their uptake in plants (Kleinteich *et al*., 2018), although long-term exposure of these MPs in the natural environment may lead to the release or desorption of their sorbates.

Affinity of antibiotics towards MPs in soil

Li *et al*. (2018b) investigated the adsorption of five kinds of antibiotics (sulfadiazine, amoxicillin, tetracycline, ciprofloxacin and trimethoprim) to five kinds of MPs (PE, PS, PP, PA and PVC). They found that PA had the strongest adsorption capacity for antibiotics, concluding that pore structure development and hydrogen bond formation were two main mechanisms. Studies undertaken by Zhang *et al*. (2017) on the adsorption mechanism of antibiotics onto MP surfaces demonstrated that pH value, ionic strength, temperature and other ageing factors have little effect on the adsorption capacity of tetracycline. The adsorption of antibiotics by MPs may lead to a compound effect – that is, they may interact with each other, causing greater harm to the soil ecosystem. Antibiotics in soil can also change the degradation process of MPs because antibiotics can reduce soil microbial community diversity (Kong *et al*., 2006).

MP transport in soil

Experimental investigations of MP transport in soil

In the past few years, there has been a surge in the studies of MP and NP transport in porous media, mainly in the field of environmental science, with a systematic summary and comparison of 20 available experimental studies on this topic presented in

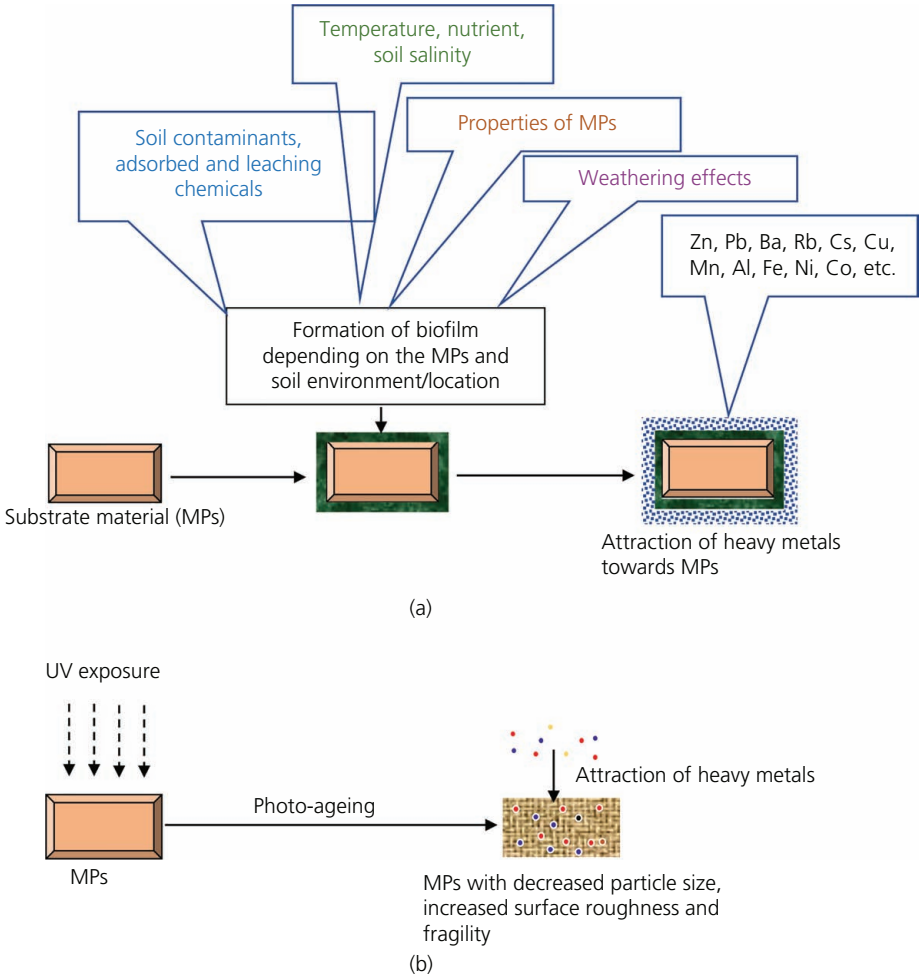

Figure 5. Accumulation of heavy metals on MP surfaces: (a) biofilm formation; (b) example of UV ageing effects

Table 5. The focuses of these studies can be categorised under the following general objectives: (*a*) transport behaviour of MPs in clean sand and gravel; (*b*) transport behaviour of NPs in clean sand and natural soil; (*c*) cotransport of MPs and NPs with other microparticles in clean sand; and (*d*) interaction between MPs, NPs and other constituents in the soil matrix. A few studies cover more than one of these objectives.

As evident from Table 5, existing studies have adopted a combination of experimental conditions selected from narrow ranges of factors and parameters. The transporting particles in these studies are typically pristine MPs and/or NPs with a few uniform sizes and arbitrary concentrations. Sometimes cotransport particles were added at concentrations that are uncommon in the natural environment. Johnson *et al*. (2020) reported the first set of field experimental results by releasing MPs to the clean sand and gravel sections of a constructed channel. Their measurements and data analysis techniques are similar to laboratory studies. Out of the 19 laboratory studies, only two simulated unsaturated rainwater infiltration (O'Connor *et al*., 2019; Waldschläger and Schüttrumpf, 2020), while the others used saturated flow. Moreover, only one laboratory study used three types of natural soils (desert soil, black soil and red soil) containing both sand and clay (Wu *et al*., 2020), while the others either purchased standard clean sand or sampled local sand for washing and sieving. All the test soils were packed loosely in medium-sized columns. Among the 17 laboratory studies conducted under saturated conditions, only two studies investigated multiple flow rates (Hou *et al*., 2020; Zhao *et al*., 2020), while the other 15 studies each picked one arbitrary flow rate. The concentrations of the migrating fluid varied from freshwater to seawater, although mostly the latter. The general conclusions from the available studies are, therefore, pertinent to the impacts of the aforementioned factors and conditions on MP and NP transport in soil, while the mechanisms are mostly explained by the interaction energy using the Derjaguin–Landau–Verwey–Overbeek (DLVO) theory.

It is worth highlighting that some aspects and factors that are conventional and intuitive to geotechnical and geoenvironmental researchers have not been considered at all. Specifically, a few

Table 5. Summary of experimental studies on the transport of MPs in soil media (continued on next page)

Reference	General objectives	Transporting MP particles	Soil matrix and test specimen dimensions	Fluid condition
Johnson *et al*. (2020)	MP transport field test	PS, D = 0.2 μm, $C = 1.3 \times 10^9$ particles/ml D = 1 μm, $C = 7.4 \times 10^7$ particles/ml PMMA, D = 5.5 μm, $C = 1.1 \times 10^8$ particles/ml	Unsaturated constructed wetland channel, 12.7 cm depth, 12.7 cm basal width, 40.6 cm surface width Clean gravel, L = 19.5 m, D_{50} = 4.2 mm, n = 0.35 Clean sand, L = 4.9 m, D_{50} = 0.42 mm, n = 0.25	Natural water, 1 conc., 1 pulse input
Cai *et al*. (2019)	MP transport cotransport	PS, D = 0.2, 1 and 2 μm, C = 4 mg/l Nano-titanium dioxide (TiO_2), C = 50 mg/l	Saturated quartz sand, D = 0.4–0.6 mm, n = 0.42 ϕ = 2 cm, L = 10 cm	Salt solution, 3 conc., 2 pH, 1 flow rate
Chu *et al*. (2019)	MP transport	PS, D = 1.0 μm, C = 20 mg/l	Saturated glass bead, D = 0.25–0.3 mm, n = 0.36 ϕ = 3 cm, L = 10 cm	Salt solution, 4 conc., 1 flow rate
Dong *et al*. (2018)	MP transport	PS, D = 0.1, 0.4, 0.6, 0.8, 1.5 and 2.0 μm, C = 30 mg/l	Saturated sea sand, D_{50} = 0.45 mm, n = 0.45 ϕ = 1 cm, L = 10 cm	Artificial seawater, 4 conc., 1 flow rate
Hou *et al*. (2020)	MP transport interaction	PE, D = 40–48 μm, C = 300, 400 and 500 mg/l	Saturated quartz sand, D = 1–2 and 2–4 mm, n = 0.42 ϕ = 3 cm, L = 20 cm	Salt solution with fulvic acid, 5 conc., 4 flow rates
Zhao *et al*. (2020)	MP transport	PS, D = 4.5 μm, $C = 4.6 \times 10^8$ particles/ml	Saturated glass bead, D = 0.5–0.6 mm, n = 0.38 ϕ = 1.6 cm, L = 15 cm Plate chamber 5 cm × 1 cm	Salt solution, 4 conc., 3 flow rates
O'Connor *et al*. (2019)	MP transport	PE, D = 21, 181, 349 and 535 μm, C = 3.2% of sand by weight PP, D = 29 μm, C = 3.2% of sand by weight	Unsaturated clean sand, D_{50} = 0.38 mm, n = NA ϕ = 4 cm, L = 25 cm	Artificial rainwater, 1 conc., 6 rainfall rates
Waldschläger and Schüttrumpf (2020)	MP transport	MP mixture consists of ten pieces of the following: PET, D = 1.1, 1.4, 1.5, 2.3 and 2.5 mm PVC, D = 1.5 and 3.2 mm SBR, D = 0.6 mm PA, D = 0.8, 1.0, 1.1, 1.4 and 2.2 mm PS, D = 4.8 mm PE, D = 1.0 and 2.2 mm PP, D = 1.1, 1.4, 3.0, 3.9 and 5.0 mm	Unsaturated glass bead, D = 1.5, 2, 3, 4, 6, 8 and 11 mm, n = 0.32–0.37 ϕ = 19 cm, L = 30 cm	Pure water, 1 flow rate
Dong *et al*. (2019a)	NP transport cotransport	Nano-PS, D = 200 nm, C = 1.5, 5 and 15 mg/l Nano-C_{60}, C = 15 mg/l	Saturated sea sand, D_{50} = 0.45 mm, n = 0.45 ϕ = 1 cm, L = 10 cm	Artificial seawater, 3 conc., 1 flow rate
Dong *et al*. (2019b)	NP transport	Nano-PS, D = 200 nm, C = 30 mg/l, 4 surface functionalities	Saturated sea sand, D_{50} = 0.45 mm, n = 0.45 ϕ = 1 cm, L = 10 cm	Artificial seawater, 3 conc., 1 flow rate
Dong *et al*. (2020)	NP transport interaction	Nano-PS, D = 200, 500 and 1000 nm, C = 30 mg/l	Saturated sea sand, D_{50} = 0.45 mm, n = 0.45 ϕ = 1 cm, L = 10 cm	Artificial seawater with 2 proteins, 9 conc., 1 flow rate
Hu *et al*. (2020)	NP transport interaction	Nano-PS, D = 122 nm, C = 10, 50 and 100 mg/l	Saturated quartz sand, D_{50} = 0.6 mm, n = NA ϕ = 2.5 cm, L = 10 cm	Salt solution with naphthalene, 4 conc., 1 flow rate
Liu *et al*. (2019b)	NP transport interaction	Nano-PS, D = 487 nm, C = 15 mg/l, 3 ages	Saturated loamy sand, D_{50} = 0.3 mm, n = 0.45 ϕ = 1 cm, L = 10 cm	Salt solution with 2 non-polar organic contaminants, 8 conc., 1 flow rate
Pradel *et al*. (2020)	NP transport	Nano-PS, D = 200, 350, 430 and 460 nm, C = 5 mg/l	Saturated Fontainebleau sand, D_{50} = 0.21 mm, n = 0.4 ϕ = 2.6 cm, L = 12 cm	Salt solution, 1 conc., 1 flow rate
Wu *et al*. (2020)	NP transport	Nano-PS, D = 100 nm, C = 5 mg/l	Saturated natural soil, 3 types, n = 0.3, 0.42 and 0.45 ϕ = 2.5 cm, L = 10 cm	Salt solution, 7 conc., 1 flow rate

Table 5. Continued

Reference	General objectives	Transporting MP particles	Soil matrix and test specimen dimensions	Fluid condition
He *et al.* (2018b)	MP and NP transport interaction	PS, D = 0.2 and 2 μm, C = 4 mg/l Nano-PS, D = 20 nm, C = 4 mg/l Subsequent *Escherichia coli*, $C = 1.3 \times 10^7$ particles/ml	Saturated quartz sand, D = 0.3–0.43 mm, n = 0.42 ϕ = 2 cm, L = 10 cm	Salt solution, 4 conc., 1 flow rate
He *et al.* (2020)	MP and NP transport interaction	PS, D = 0.2 and 2 μm, C = 4 mg/l Nano-PS, D = 20 nm, C = 4 mg/l	Saturated quartz sand, D = 0.3–0.43 mm, n = 0.42, coated with biofilm ϕ = 2 cm, L = 10 cm	Salt solution, 2 conc., 1 flow rate
Li *et al.* (2019b)	MP and NP transport cotransport	PS, D = 0.2 and 2 μm, C = 4 mg/l Nano-S, D = 20 nm, C = 4 mg/l Goethite, C = 3.5 mg/l Haematite, C = 3.5 mg/l	Saturated quartz sand, D = 0.3–0.43 mm, n = 0.42 ϕ = 2 cm, L = 10 cm	Salt solution, 2 conc., 1 flow rate
Tong *et al.* (2020a)	MP and NP transport interaction	PS, D = 0.2 and 2 μm, C = 4 mg/l Nano-PS, D = 20 nm, C = 4 mg/l	Saturated quartz sand, D = 0.3–0.43 mm, n = 0.42, mixed with two types of biochar ϕ = 2 cm, L = 10 cm	Salt solution, 2 conc., 1 flow rate
Tong *et al.* (2020b)	MP and NP transport cotransport	PS, D = 0.2 and 2 μm, C = 4 mg/l Nano-PS, D = 20 nm, C = 4 mg/l Biochar, C = 100 mg/l	Saturated quartz sand, D = 0.3–0.43 mm, n = 0.42 ϕ = 2 cm, L = 10 cm	Salt solution, 2 conc., 1 flow rate

conc., concentration; D, particle size; D_{50}, mean particle size; L, soil column length; n, porosity; NA, not available; PA, polyamide; PE, polyethylene; PET, poly (ethylene terephthalate); PMMA, poly(methyl methacrylate); PP, polypropylene; PS, polystyrene; PVC, poly(vinyl chloride); SBR, styrene-butadiene rubber; ϕ, soil column diameter

major missing points are (*a*) testing of natural soils with different mineral compositions and soil classifications; (*b*) studies with varied and realistic flow regimes, both saturated and unsaturated; (*c*) soil status and properties, including confining stress, temperature gradient, multiphase flows, porosity, permeability and fabric; and (*d*) relevant mechanistic and numerical models. Clearly, experimental complexity makes it difficult to assess the prevalence and validate models. The available studies ignored the aforementioned conditions and factors either because these are irrelevant for environmental concerns or simply for convenience. Consequently, the available results are applicable to limited situations – for example, shallow soil with a high groundwater table, coastal soil and riverine and marine sediments. Overall, there is a need for more diverse and interdisciplinary studies incorporating knowledge and objectives from geotechnics, geochemistry and hydrogeology to be conducted on this topic.

Computational modelling of MP transport in soil (soil–groundwater)

About 50% of land-based plastic wastes reach oceans through rivers alone (Atwood *et al.*, 2019). Most of the research on MP transport is focused on the movement of MP particles in water. Less investigated are the transportation of MPs in sediments, coastal aquifers and the terrestrial subsurface more generally. Engdahl (2018) presented a numerical model for describing the transport of idealised MPs (the particle is defined solely by its size and shape) and other micro-sized fibre-like objects in saturated soil. The migrating particles are treated as discrete bead–rod chains, and the transport process is simulated as a random walk of particles in the porous media. Johnson (2020) used mechanistic pore-scale simulations and continuum-scale simulations to explain MP transport in gravel. To the authors' knowledge, these are the only available modelling works targeting specifically the transport of MPs in porous media. There are also a number of reviews that include sections on MP transport in soil, although their major focuses are diverse and environmentally oriented (de Souza Machado *et al.*, 2018a; Guo *et al.*, 2020; Hale *et al.*, 2020; Li *et al.*, 2020d; Qi *et al.*, 2020; Wang *et al.*, 2020; Xu *et al.*, 2020b).

Modelling considerations and challenges

Modelling contaminant migration in soil requires accurate quantification of sources and sinks of MPs in soil as well as processes governing their fate, as described in the section headed 'MPs in the soil environment'. Once in soils, MP fate is driven by water flow, mechanical disturbance (ploughing, harvesting, mammalian digging), ingestion and excretion by epigean fauna, elongation of plant roots, competitive sorption and differentiation by soil aggregation, soil cracking and interaction with organic and inorganic contaminants and nutrients (Chae and An, 2018; Guo *et al.*, 2020; Qi *et al.*, 2020). The transport of MPs into and through subsurface environments (soil, groundwater and surface

water) strongly depends on the MP properties (Farré *et al*., 2010; Geissen *et al*., 2010) and the environment. The challenge, overall, is to be able to characterise the relative importance of different processes mentioned earlier, under pertinent physico-chemical conditions, while taking into account the effects of plastic size, polymer type and soil structure and chemistry. Soil structure, in particular, appears to play a critical role in the distribution of MPs in soil (Zhang and Liu, 2018).

MPs are colloidal particles usually present in water as suspensions – hence, models of transport must take into account colloidal interactions at micro- and nanoscales, including patterns of competitive sorption that are often difficult to capture. The DLVO theory is the most widely used framework for quantifying double-layer electrostatic forces and van der Waals interactions between particles (Ohshima, 2012). Chu *et al*. (2019) used the DLVO theory and the dispersion–advection equation to simulate the transport and retention of the MPs in a saturated glass-bead column. The approach was successful in explaining the observed behaviour at small pore volumes but failed to capture non-monotonic breakthrough curves at higher pore volumes, likely due to detachment and reattachment of colloids. Extending the type of studies conducted by Chu *et al*. (2019) to actual soils and enriching transport models with other processes mentioned earlier is clearly needed. Furthermore, given that most MPs are found in topsoil, models of MP fate in it ought to consider partial saturation and capillarity.

Another challenge is to develop an understanding of the influence of hydrodynamic processes on the fate of MPs. River sediments and coastal aquifers are complex heterogeneous porous systems in which non-linear flows take place and fluctuations of the flow field typically define the smallest length scales governing the mixing. At pore scales, it is expected that molecular diffusion becomes the dominant mechanism controlling the mixing rates from pore to field scales (Meyer, 2014). Reviewing recent investigations on the effects of flow rates on the mixing, it is observed that the chemical residence time is controlled by hydrodynamics, which in turn controls the fate of MPs, not just by the advective transport but also with the non-Fickian (anomalous) mixing process (Baioni *et al*., 2020). Therefore, reliable modelling of the soil–MP interactions needs incorporating the effects of such complex non-Fickian mixing on the MP pollution in the coastal zone, as investigations show that an estimated value of 250 Mt plastic components reached marine environments in 2015 (Wright and Kelly, 2017).

Another key research interest – alongside the fate of MPs – is the effect of the presence of NPs and MPs on the transport of other contaminants. Hu *et al*. (2020) found that naphthalene is more mobile in the presence of very small PS MPs (average size of 120 nm) but that the relationship depended on the ionic strength of the solution. They also reported that, conversely, these particles have lower mobility in the presence of naphthalene. Another study, also using a glass-bead analogue, found evidence of co-mobility between the mobile organic fraction and very small MP particles (Keller *et al*., 2020). Hüffer *et al*. (2019) showed that PE MPs reduce the capacity of soil to sorb hydrophobic pesticides. Hence, it is clear that, while the mobility of MPs in soils remains an open question, their effects on the mobility of other contaminants of high relevance to environmental geotechnics are equally, if not more, important.

Mitigation–remediation measures

Strategies for reduction and removal of MPs in the geoenvironment

Separation of MPs from the soil has proven to be a major challenge in efforts to remediate soils contaminated with MPs. Unlike other contaminants such as heavy metals and organic pollutants, there are no 'established' techniques for remediation of MPs (Bhattacharya and Khare, 2020). However, various strategies for prevention, reduction and/or removal of MPs from the environment are being researched. These can be categorised as pre-emptive strategies, control or management strategies and remediation strategies.

Pre-emptive and control/management strategies

Pre-emptive strategies include social awareness measures, transition towards use of biodegradable plastics and regulatory measures to limit the use of plastics (McDevitt *et al*., 2017; Silva *et al*., 2018; Steensgaard *et al*., 2017; Wu *et al*., 2017). Regulatory measures, such as limiting the use of MPs in certain products; disincentivising the use of plastic items, packaging and carrier bags; and restricting the landfilling of plastics, would aid in reducing some of the sources of MPs. Reducing the amount of MPs introduced into agricultural soils through the application of composts and organic fertilisers requires a special effort. This includes avoidance of the overuse of compost and fertilisers, including sludge, and waste water irrigation, as well as better characterisation of their MP concentrations. Guidelines (standards) for the application of PMF in farm-specific conditions and practices need to be evaluated further to help farmers adopt sustainable practices. The separation of MPs at the source (e.g. plastic waste from unsegregated MSW feedstocks) is another possible measure. Such separated municipal plastic waste can be utilised as an energy source in waste-to-energy generation (Sai *et al*., 2018) and as construction materials for manufacturing of civil engineering composites (Goli *et al*., 2020). Moreover, LFMSF (also known as soil-like materials), generated during landfill mining activities, could be a potential source of MPs in agricultural soils, when they are utilised as fertilisers (Chandana *et al*., 2021; He *et al*., 2019). Hence, proper care should be taken in (*a*) quantification of MPs and the associated risk and (*b*) monitoring of the application of LFMSF in agricultural fields.

Use of biodegradable plastics has also been considered as a pre-emptive, more sustainable alternative (Lambert and Wagner, 2017; Rujnić-Sokele and Pilipović, 2017). However, questions have been raised about the actual degradability of 'biodegradable' plastic in real environments, as opposed to laboratory environments (Harding *et al*., 2017), particularly with antioxidant additives used to increase the

design life and/or antimicrobial resistance of plastics (Harding *et al.*, 2017; Lambert and Wagner, 2017).

Control or management strategies refer to the use of engineering or technological interventions to remove MPs from sources. These include measures implemented at WWTPs (Wu *et al.*, 2017) and interventions to remove and reuse MPs from sediments and freshwater bodies (Eriksen *et al.*, 2018). Carr *et al.* (2016) suggest that the majority of MPs are removed from the effluents of WWTPs during primary and secondary treatment stages. More effective MP removal processes generate higher MP concentration in the sludge. However, without special treatment for MP removal from WWTP effluent and relying only on standard processes of treatment, a large proportion of the MPs remain in the effluent. Various processes such as biologically active filtration (Talvitie *et al.*, 2017b), membrane bioreactors (MBRs) (Lares *et al.*, 2018; Talvitie *et al.*, 2017a), dissolved air floatation, rapid sand filtration and disc filters (Talvitie *et al.*, 2017a) have been evaluated for their efficiencies in removing MPs from the effluents of these treatment plants. While each of these techniques has been successful in reducing MPs from effluents, their level of efficiency has been shown to be highly variable, with MBRs being the most efficient and disc filters being the least (Lares *et al.*, 2018; Talvitie *et al.*, 2017a).

Remediation strategies

Several remediation strategies based on physical degradation, chemical degradation, catalytic degradation and biodegradation have been developed and are being adopted. Among these, bioremediation based on the microbial degradation of soil contaminated with MPs has become popular because the soil is a habitat for a wide range of microorganisms, which, as elaborated below, are sometimes quite efficient in the degradation of plastics (Pathak and Navneet, 2017). The majority of plastics have low biodegradability (Wei and Zimmermann, 2017), although a few such as PLA, polyhydroxyalkanoate and polyhydroxybutyrate are highly biodegradable. Several biodegradation mechanisms are at play (see Figure 6), but two, in particular, have been prominently investigated for degradation of complex polymers in soils: a direct action, in which the degraded plastic fragments provide a nutritional source for microbial growth, and an indirect action, in which microbial metabolic products such as enzymes degrade the plastic structure, both operating under aerobic and anaerobic conditions (Ahmed, 2018; Ghosh *et al.*, 2013; Shah *et al.*, 2008). Although several studies have been conducted on the microbial degradation of MPs (Guo *et al.*, 2020; Sarker *et al.*, 2020; Yuan *et al.*, 2020), most are laboratory investigations. Hence, in situ investigations are needed that would better reflect the diverse and complex characteristics of different types of soils.

When microbes are introduced to MPs, they first adhere to exposed polymer surfaces, colonising them to produce enzymes (Lam *et al.*, 2008), which prompt hydrolysis of polymers. As such, this hydrolytic division splits polymer chains by ester bond formation, which in turn causes degradation of polymers into oligomers, dimers and monomers. The degraded products are taken up by microorganisms through their semipermeable membranes and are eventually converted to carbon dioxide and water by mineralisation (Tokiwa *et al.*, 2009).

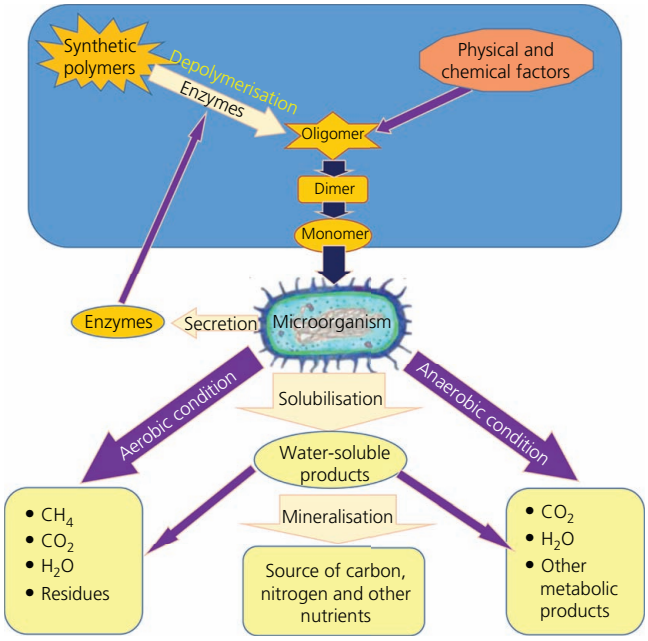

Figure 6. Mechanisms of biodegradation of plastics by microorganisms under aerobic and anaerobic conditions

Under aerobic conditions, microbes use oxygen as an electron acceptor and produce methane (CH_4), carbon dioxide, water and other residues as end products (Priyanka and Archana, 2012). Under anaerobic conditions, polymers are degraded in the absence of oxygen by microorganisms through the catalytic activity of enzymes. Sulfate, nitrate, iron and manganese are used as electron acceptors, and carbon dioxide, water and other metabolic products are produced (Priyanka and Archana, 2012).

In the pure-culture method (ex situ approach), specific bacteria/fungi that are isolated and cultured using different nutrient media are added to the MP-contaminated soil, which is then incubated in a reactor under optimal conditions for a specific period (Hadad *et al.*, 2005). Samples are later retrieved and assessed for degradability of plastics. In the compost method, a known weight of dry plastic is admixed with a definite amount of mature compost and the mixture is incubated at a particular temperature and moisture. Evidence in the literature indicates that, among the different soil MP-bioremediation techniques, the pure-culture method appears to be the most effective (Shahnawaz *et al.*, 2019).

Concluding remarks and way forward

MPs have emerged as one of the top environmental issues due to their prevalence in, and impacts on, aquatic and terrestrial ecosystems – the vast scale of which is only just becoming apparent. Impacts of MPs in soils include their entry into the food

chain through fauna and flora capable of absorbing MPs, which is exacerbated by their slow rate of degradation under ambient in situ conditions. Lastly, MPs can influence soil bio-physico-chemical properties and the mobility of other contaminants in soil, with potentially significant implications for groundwater quality.

In this paper, the authors have reviewed available evidence for, and knowledge of, MP contamination of the terrestrial subsurface and its effects on key soil ecosystem functions. Through the review, the authors have identified five key research gaps in the literature (presented below in no particular order of importance), with particular emphasis on aspects of soil and groundwater behaviour of interest to environmental geotechnics:

(*a*) better analytical characterisation of MPs and NPs in the soil
(*b*) mapping of MP prevalence in soils and groundwater and its effects on soil physical, chemical and biological conditions and properties
(*c*) better understanding and quantification of the fate of MPs in soils, including their effects on the mobility of other soil contaminants
(*d*) better understanding and quantification of MP fate in landfills and their environments
(*e*) better remediation strategies for MP pollution in soils.

The above research goals require concerted, multidisciplinary effort. Nevertheless, with the exception of goal (*a*), they are ones in which geotechnical and geoenvironmental concepts, approaches and protocols – theoretical, experimental and computational – are paramount.

Analytical characterisation of MPs and NPs

The lack of established analytical protocols for measuring concentrations of NPs and smaller-sized MPs is a serious obstacle. Analytical methods used in most studies found in the literature are capable of detecting only those particle sizes greater than typically 1 μm, such that the extent of the prevalence of NPs and smaller-sized MPs in the environment is not known. The ability to detect NPs and smaller MPs is, therefore, clearly an important methodological gap and a research priority. As shown in this paper, the lack of protocol for result reporting is an obstacle to the comparability of studies, which hampers scientific progress on MPs. The precise detection and quantification of the types and extents of MPs present in soils using standardised tools/techniques is required not only to understand the level of contamination but also to inform the adoption of suitable remediation strategies. The combined application of visual inspection and chemical detection methods seems a better choice for MP research, although the balance of cost, accuracy and time must be taken into consideration. The proposed standard analytical protocol of MP detection in soil media, elaborated earlier in the paper, provides focused direction for the research community in this regard.

MP prevalence and effects in soils

Many studies mention groundwater as a potential receptor of MP fibres and fragments, but only a very limited number address their presence in aquifers. The development of accurate and groundwater-specific MP detection and quantification standards will boost research efforts, minimise biases and increase understanding of MP existence in aquifers. To that end, including MPs in existing emerging contaminant-monitoring programmes would be vital for the development of this new research area. Further, it is recommended that local, regional and global pollution maps be generated highlighting land use–land change patterns and showing the extent of MP contamination. This is essential as part of an environmental risk assessment framework required to manage this new anthropogenic pollutant.

To the best of the authors' knowledge, no attempt has been made at studying the effect of MPs on soil mechanical properties and slope stability, and given the widespread prevalence of MPs, there is a need, from an environmental geotechnics perspective, to characterise better the effects of MPs on the mechanical and hydraulic properties of soils. From a biological point of view, the role of MPs in transforming soil-microbial ecology and, hence, its implications for geoenvironmental physico-chemical processes have not been investigated either. Such changes can affect soil–plant interactions and soil biodiversity, which have far-reaching consequences on all living species. Of particular interest is the exposure of soil biota to different levels of MP contamination in soil, considering the coexistence of other contaminants and, given the persistence of MPs, transgenerational effects.

Fate of MPs in soils

Understanding and quantifying transport mechanisms, degradation and transformation processes governing MPs in soil and groundwater environments are urgently required. Equally important is an understanding of the role of MPs as contaminant carriers and their long-term cumulative and synergistic pollution effects on soil ecosystem functions, including biodiversity at different scales and subsurface water quality. This includes efforts to understand the direct and indirect influence of the MP shape, size, composition and concentration on the key processes of soil aggregation and soil physico-chemical and hydromechanical properties.

There is ample evidence of sorption of heavy metals onto MPs, which can then act as pollutant carriers. However, far more work is needed in this area, as well as a better understanding of the interaction of MPs with organic contaminants and the effect of MPs on the mobility of organic and inorganic soil contaminants through ways other than direct carrying. For example, it is still not clear whether the characteristics of MPs or environmental conditions play a more important role in heavy metal sorption on MPs. Based on such knowledge, it may be possible to modify sources of MPs during the manufacturing process so as to reduce future heavy metal mobility in soil.

Interdisciplinary studies incorporating knowledge and objectives from geotechnics, geochemistry and hydrogeology are required in this regard. Computational modelling frameworks of MP transport in soil need to be developed, extended and calibrated to consider

complex interactions between MPs and other chemicals under complex environmental conditions pertinent to specific field scenarios.

MPs in landfill environments

The number of studies of MPs in municipal waste landfills is still limited but already strongly indicates that they constitute major sinks for MPs and potentially major sources of MP pollution in soils. This is unsurprising given the amount of plastic of different sizes and of SS received by landfills. To the best of the authors' knowledge, not a single study can be found in the literature investigating the extent to which MP's pollution has affected environments adjacent to municipal waste landfills (base liners, soil buffers, aquifers and neighbouring sites). Quantification of the fluxes of MPs in landfill waste (input, discharge by leachate treatment, escape into the wider environment) is urgently required, as well as an understanding of the capacity of conventional base liners to prevent MP contamination of the subsurface.

New treatments

To address the MP terrestrial pollution problem, concentrated efforts are required in developing comprehensive pre-emptive strategies (e.g. transiting towards the use of biodegradable plastics), control or management strategies (including use of engineering or technological interventions to remove MPs from sources that contribute these pollutants into the geoenvironment) and green and sustainable remediation strategies. New treatment and/or methods for removing or reducing MP contamination at the source or within the soil are therefore needed. For example, given the poor efficacy and slow degradation rate of many plastics, there is a great need for the evolution of new microbes, enzymes and degradation pathways that can convert recalcitrant plastics into harmless monomers with a focus on beneficial end products. Use of 'active microbial consortia', 'Blue Technology' and 'plastisphere' seem to have greater potential for natural bioremediation processes. Another example is research and development of biodegradable PMF and multifunctional mulch recovery machinery that can help promote effective management and control of residual mulch pollution.

This is, however, a rapidly changing field, and the paper is intended for providing guidance to researchers and policymakers with interest in this field. It is a contribution to a mounting worldwide effort at tackling a widespread environmental problem that threatens lives and livelihoods.

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