CRITICAL REVIEW

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A review of microplastic fibres: generation, transport, and vectors for metal(loid)s in terrestrial environments†

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The laundering of synthetic fabrics has been identified as an important and diffuse source of microplastic (<5 mm) fibre contamination to wastewater systems. Home laundering can release up to 13 million fibres per kg of fabric, which end up in wastewater treatment plants. During treatment, 72–99% of microplastics are retained in the residual sewage sludge, which can contain upwards of 56 000 microplastics per kg. Sewage sludge is commonly disposed of by application to agricultural land as a soil amendment. In some European countries, application rates are up to 91%, representing an important pathway for microplastics to enter the terrestrial environment, which urgently requires quantification. Sewage sludge also often contains elevated concentrations of metals and metalloids, and some studies have quantified metal(loid) sorption onto various microplastics. The sorption of metals and metalloids is strongly influenced by the chemical properties of the sorbate, the solution chemistry, and the physicochemical properties of the microplastics themselves. Plastic–water partition coefficients for the sorption of cadmium, mercury and lead onto microplastics are up to 8, 32, and 217 mL g^{-1} respectively. Sorptive capacities of microplastics may increase over time, due to environmental degradation processes increasing the specific surface area and surface density of oxygen-containing functional groups. A range of metal(loid)s, including cadmium, chromium, and zinc, have been shown to readily desorb from microplastics under acidic conditions. Sorbed metal(loid)s may therefore become more bioavailable to soil organisms when the microplastics are ingested, due to the acidic gut conditions facilitating desorption. Polyester (polyethylene terephthalate) should be of particular focus for future research, as few quantitative sorption studies currently exist, it is potentially overlooked from density separation studies due to its high density, and it is by far the most widely used fibre in apparel textiles production.

Environmental significance

This paper critically reviews the evidence for microplastic fibres released during home laundering becoming a vector for metal(loid)s from wastewater to terrestrial environments. Quantitative data on fibre shedding from textiles were collated and critically evaluated. A conceptual framework is presented which discusses the transport of microplastic fibres from laundry water, through wastewater treatment plants, to agricultural soils through the application of sewage sludge. Distribution coefficients that quantify the sorption of aqueous metal(loid)s to microplastics were compared and critically analysed. The scarcity of both soil concentration and metal(loid) sorption data for common textiles polymers, such as polyester and nylon, is highlighted as an important knowledge gap. The bioavailability of microplastic-sorbed metal(loid)s to organisms living in agricultural soils is critically discussed.

1. Introduction

The annual global total production of plastics exceeded 400 million metric tonnes (MMT) per year in 2015.[1] In 2015, the

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seven plastics with the highest demand in the EU (excluding fibres) were polypropylene (PP) > low-density polyethylene (LDPE) > high-density polyethylene (HDPE) > polyvinyl chloride (PVC) > polyurethane (PUR) > polyethylene terephthalate (PET) > polystyrene (PS).[2] Plastics offer many advantageous properties such as corrosion resistance, low cost of raw materials and production, and general durability, meaning they have become increasingly favourable over traditional materials (metal, paper, wood) since the 1950s.[1] Plastics are now routinely manufactured for a wide variety of end uses, including food packaging, synthetic textile fibres, building insulation, and protective coatings. The most commonly manufactured plastics are not

readily biodegradable, and thus accumulate in the environment. The rate of plastic waste recycling to the original product is below 10%.[3] These factors, together with the continuously increasing global plastic demand at 8.7% annual growth rate,[1] have resulted in a global plastic pollution issue.

Microplastics are typically defined as measuring 5 mm or less across their longest dimension, and have become a global pollutant of concern due to their persistence and ubiquity in terrestrial and marine environments.[4–6] Microplastics have been detected worldwide in oceans,[4,7] lakes and rivers,[8,9] Arctic sea ice,[10] marine and freshwater sediments,[11–13] sewage sludge,[14–18] and agricultural soils.[19–21] Published literature on the environmental impact microplastics has increased exponentially since 2010. However, the majority of studies have focused on the impacts to marine environments.[22] From January 2004 to June 2018, only 4% of published literature on microplastics focused on terrestrial sinks such as soil and sludge.[22] Consequently, current knowledge on the scale, environmental fate, and ecological impacts of microplastic pollution in terrestrial environments is limited.

This knowledge gap is concerning as it has been estimated that, in the EU, terrestrial environments could receive 4–23 times more microplastic pollution than oceans.[23] It has also been estimated that up to 48% of this microplastic pollution is due to the direct application of contaminated sludge to agricultural soils.[24] The shedding of textiles during laundering is thought to be a considerable source of microplastic pollution.[25] Synthetic fabrics, such as polyethylene terephthalate (PET), and nylon (polyamide-6,6) shed recalcitrant, non-biodegradable fibres during laundering; the majority of which are 0.20 to 2.75 mm in length.[25,26] Between 72% and 99.9% of microplastics (by number) are removed during the sewage treatment process,[27] and approximately 78% are retained in the semi-solid sludge fraction.[28] This sludge is collected and disposed of in a number of ways, including application to agricultural land, incineration or landfill. Recycling to agricultural land is often a preferred option across much of Europe, USA, and China

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because treated sludge poses little risk to human and animal health, and contains essential plant nutrients and organic matter which improve soil fertility and physicochemical properties.[29] Sludge use in agriculture is heavily regulated. Before application to agricultural land, sludge must be treated to reduce pathogen content, odour, and attraction of potential disease vectors such as rats. This treatment may be biological (*e.g.* aerobic or anaerobic digestion), chemical (*e.g.* lime stabilisation), or physical (*e.g.* thermal drying), or any combination of the three. Sludge that has been treated and stabilised for land application purposes is referred to as biosolids.[29,30]

In the UK, approximately 79% of municipal sewage sludge is recycled to agricultural soils as biosolids.[31] In other European countries, application rates vary from 0–91%.[24] Therefore, the application of biosolids may constitute a very significant route for the entry of microplastic fibres derived from laundered fabrics, into agricultural soils. This conceptual pathway is illustrated in Fig. 1 (Section 2). Moreover, during sewage treatment, synthetic fibres are exposed to elevated levels of metals and metalloids,[32] and organic contaminants, including antibiotics, endocrine-disruptors and polycyclic aromatic hydrocarbons,[33,34] which may sorb to the surfaces of the fibres. The study of the sorption of metals and metalloids to microplastics is still in its infancy. Nevertheless, metals including cadmium (Cd), copper (Cu), nickel (Ni), and lead (Pb) have been shown to sorb to both virgin and beached microplastic pellets.[35,36] The bioavailability and environmental fate of microplastic-bound metals and metalloids is poorly understood. However, microplastics may act as vectors for metals and metalloids that would otherwise have been discharged in the effluent. Therefore, metal and metalloid sorption to microplastics may ultimately increase their exposure to important soil organisms such as earthworms.[37]

Throughout this review, microplastic particles (≤5 mm in length) with fibrous morphologies are referred to as 'microplastic fibres', and the uniform adoption of this term is suggested for future research. In textiles science, the term microfibre has been clearly defined as a single thread with a denier of 0.3–1.[38–40] A denier is a unit of linear density equal to the mass in grams of 9000 m of fibre.[38] In environmental science however, the term 'microfibre' has not been appropriately defined and is sometimes used to refer to only synthetic fibres, or to synthetic and natural fibres.[7,25,41,42] We suggest that 'microplastic' and 'microfibre' are not used interchangeably, and that the distinction between natural and synthetic fibres should be retained, given that fibres are likely to have different sources, environmental partitioning behaviours, and environmental impacts than natural fibres or microplastics with other morphologies.

This review aims to (i) synthesise and critically evaluate recent qualitative research on fabric shedding as a source of microplastic fibre pollution; (ii) provide a conceptual framework for, and comment on the environmental significance of, the transfer of microplastic fibres from laundry wastewater to agricultural soils through the application of sewage sludge as a soil amendment, and finally (iii) critically evaluate existing data concerning the sorption of metals and metalloids to microplastics.

2. Microplastic fibres from synthetic textiles

The laundering of synthetic textiles was first evidenced as a diffuse source of microplastic fibre pollution by Habib *et al.* (1998),[14] who used polarised light microscopy to qualitatively identify synthetic fibres in dewatered sewage sludge, biosolid pellets, and wastewater effluent from a wastewater treatment plant (WWTP) in Long Island, New York. These fibres were hypothesised to come from the shedding of apparel textiles
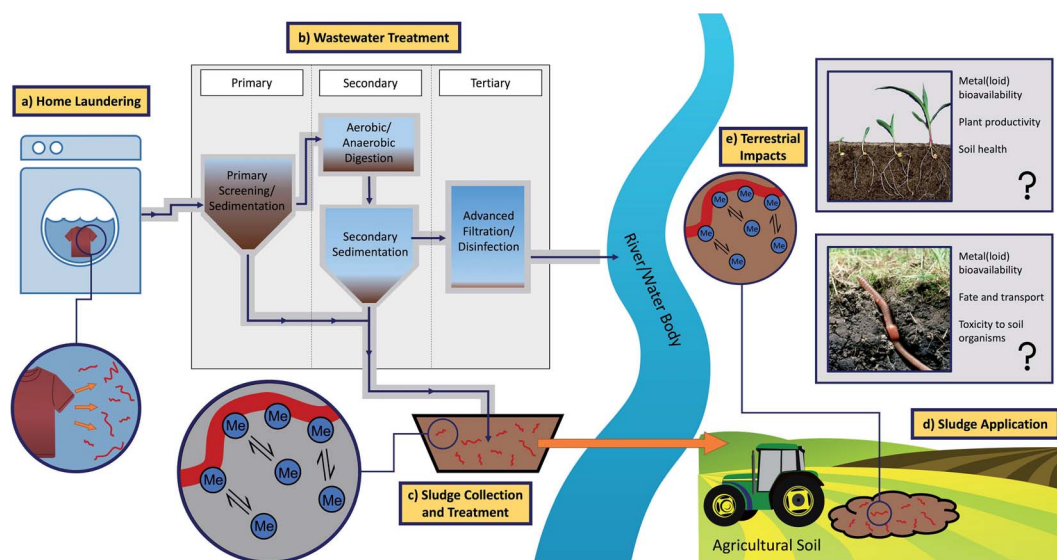

Fig. 1 Conceptual pathway visualising the transport of microplastic fibres from laundry effluent to agricultural soils through the production and application of sewage sludge (Me = metal(loid) ion).

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during laundering (Fig. 1a).[14] Shedding refers to the detachment and release of fibres from the surface of the fabric and primarily occurs during laundering, where the rotational force and mechanical action of the washing machine drum cause fibres to break and enter the water.[42] Zubris and Richards (2005)[19] detected synthetic fibres in soil samples up to 15 years after the application of biosolids, implicating sludge disposal to soil as an important pathway for the terrestrial transport of fibres. Browne *et al.* (2011)[4] found that sediments from sewage sludge disposal sites and wastewater treatment plant effluent contained proportions of synthetic fibres which resembled those used in apparel textiles (78% polyester, 9% polyamide, 7% polypropylene, and 5% acrylic),[43] suggesting that the fibres were largely derived from the shedding of clothing during laundering.

Several factors may influence the propensity of fabrics to shed during laundering, including the physicochemical properties of the fibres,[25] yarns,[44,45] and fabric.[41,42,46–48] Fabric shedding is also influenced by laundering conditions, such as washing machine type (front-loading or top-loading), water temperature, and the presence of surfactants and fabric softeners.[49,50]

We highlight 25 research papers, published between 2011 and 2021, that report quantitative data on fabric shedding and summarise key data in Table 1.[4,25,41,42,46–66] Quantifying the number of fibres shed from a fabric during laundering is practically difficult due to the small size and vast numbers of fibres generated, and the lack of standardised methodologies. Moreover, the fabric types and construction, reported units, laundering apparatus, laundering conditions, and fibre characterisation techniques all differ between studies, making comparison of the results difficult. Hernandez *et al.* (2017),[50] Almroth *et al.* (2018),[42] De Falco *et al.* (2018)[56,57] Jönsson *et al.* (2018),[59] Zambrano *et al.* (2019),[25] Haap *et al.* (2019),[58] Kelly *et al.* (2019),[61] Frost *et al.* (2020),[48] Raja Balasaraswathi and Rathinamoorthy (2021),[65] Cai *et al.* (2020),[52] and Özkan and Gündoğdu (2021),[63] all used standard laundry testing apparatus, while the remaining studies used commercially available washing machines to generate fibres. Fabric shedding varied from 900–110 000 fibres per garment,[4,42] although the sizes and masses of the garments were unspecified. On a number per mass basis, fibre shedding ranged from 8809–72 000 000 per kg of fabric,[52,66] and on a mass per mass basis, 7–1507 mg fibres per kg of fabric.[54,65]

The extreme range in literature values highlights the need for standardisation in the quantification of fabric shedding during laundering to make meaningful comparisons between fabrics. Pirc *et al.* (2016)[47] reported among the lowest number of fibres per mass of fabric, but also used the largest filter size (200 μm). Zambrano *et al.* (2019)[25] reported that shed fibres 25–200 μm in length were more numerous than fibres 200 μm–2.75 mm, shed from PET, cotton, and PET-cotton blend fabrics, suggesting that the shedding data from Pirc *et al.* (2016)[47] may have underestimated total fibre release, due to smaller fibres passing through the filter. Raja Balasaraswathi and Rathinamoorthy (2021)[65] and Vassilenko *et al.* (2021)[66] found shedding propensity significantly increases with fabric thickness. This is

thought to be due to an increased density of fibre ends per unit of surface area.[65] A higher stitch density (number of stitches per unit area) results in less fibre release, as friction between constituent fibres is increased.[65] Dalla Fontana *et al.* (2021)[54] compared the shedding of two 100% polyester fabrics with differing constructions, and observed significantly different fibre release during conventional laundering experiments. Differences were attributed to the differing linear densities of the constituent fibres, which influences tensile properties, and the stitching used to finish the fabric edges.[54] Fabrics composed of natural fibres, such as cotton, generally have a higher shedding propensity then fabrics constructed with synthetic fibres, such as polyester.[25] This may be due to the lower tensile strength of natural fibres, or the shorter fibre length of cotton, resulting in more fibre breakages overall.[25,66] It is important to note that chemical identification of the shed fabrics with spectroscopic techniques (IR/Raman), was only performed in 5 of the 25 studies (Table 1). In the remaining studies, fibres were counted and/or weighed without confirming their composition. 10 studies investigated the shedding of fabrics with mixed fibre compositions, and of these, only 3 confirmed the chemical composition of the shed fibres with FTIR. Chemical confirmation of shed fibres is of particular importance in studies using mixed composition fabrics, so that the relative proportions of shed fibres can be assigned to each fibre composition. Haap *et al.* (2019)[58] investigated the shedding of fibres from a 50% polyester, 50% cotton woven fabric. After quantification, chemical separation was performed by using sulphuric acid to digest the cotton fibres. 86% by mass, of fibres released from the fabric were cotton and 14% were polyester, which can be attributed to the higher tensile strength and abrasion resistance of polyester, and the shorter constituent fibre lengths of cotton.[25,58,66] Kelly *et al.* (2019)[61] found that increasing the water volume in accelerated laundering experiments, from 300 mL to 600 mL, resulted in an increase in fibre shedding, from 54 mg kg^{-1} to 120 mg kg^{-1}. Several studies in Table 1 do not report the total volume of water used, and the fabric weight, density and surface area. It is recommended in future that these parameters are quantified and reported, as they also influence shedding propensity.

Microscopy and manual or computational counting from micrographs was by far the most common method for fibre characterisation, used in 18 of the 25 studies included in Table 1. However low spatial resolution of optical microscopes and image analysis techniques mean that underestimations are likely. McIlwraith *et al.* (2019)[62] used ImageJ (image processing software) to quantify fibres from a series of micrographs. However, the limit of detection was 100 μm in length, resulting in the exclusion of fibres <100 μm. Hernandez *et al.* (2017)[50] also analysed micrographs with ImageJ; ascertaining a 40 μm limit of detection from the minimum visible number of covered pixels in each micrograph (between 2 and 5 pixels). In this study, it was found that shed fibres from a PET single-knit jersey and interlock fabrics were typically 100–800 μm in length. However, size distributions revealed a general increase in fibre frequency as fibre length decreased. Moreover, fibres above 1 mm in length represented only 2–5% of the total shed fibre

Table 1 Review table of fabric shedding data from laboratory studies. Fabric type, fibre shedding, and general experimental parameters are shown. NS = not specified; LOD = limit of detection

Reference	Fibres: Fabric type	Fabric mass (g)	Fibres shed	Units	Laundering: Type/apparatus	Experimental/washing conditions	Analysis: Filter pore-size (μm)	Fibre characterisation	LOD (μm)	Chemical confirmation method
Browne et al. (2011)[4]	100% polyester blankets, fleeces and shirts	NS	>1900	Fibres per garment	Conventional: Bosch WAE24468GB, John Lewis JLWM1203 and Siemens Extra Lasse XL 1000 washing machines	Speed 600 rpm; time unspec.; 40 °C	NS	Number: counted manually	NS	None
Bruce et al. (2016)[49]	100% polyester; 100% nylon; 97% polyester; 3% Spandex fleece and non-fleece jackets	NS	100.5–1081.9	mg per garment	Conventional: Whirlpool WET3300XQ1 washing machine	Speed unspec.; 30 min; temp. Unspecified; 43 L capacity, front-load and top-load machines; artificially aged and new garments; 'regular warm wash cycle'	333 and 20	Mass: gravimetric analysis	NS	None
Napper and Thompson (2016)[46]	100% polyester jumper; 100% acrylic jumper; 65% polyester; 35% cotton jumper	NS	82 671; 121 465; 22 992	Fibres per kg fabric; Fibres per kg fabric; Fibres per kg fabric	Conventional: Whirlpool WWDC6400 washing machine	1400 rpm; 75 min; 30 and 40 °C; 20 × 20 cm swatches; no detergent; with bio/non-bio detergent; with/without conditioner	25	Mass: gravimetric analysis; number: estimated; length: light microscope and image analysis; width: SEM and image analysis	NS	None
Pirc et al. (2016)[47]	100% polyester fleece blanket	NS	22 600; 12	Fibres per kg fabric; mg per kg fabric	Conventional: Bosch model Maxx7 VarioPerfect	600 rpm; 15 min; 30 °C; 120 × 70 cm blankets; without additives; with detergent; with detergent and fabric softener	200	Mass: gravimetric analysis; relative release: estimated	NS	None
Sillanpää and Sainio (2017)[41]	100% polyester blankets and T-shirts; 100% cotton jeans and shirts	NS	211 000–13 100 000	Fibres per fabric	Conventional: Bosch WAE28477SN washing machine	1200 rpm; 75 min; 40 °C; 50 mL liquid detergent; 27–39 L of washing water	0.7	Number: counted manually, light microscopy; mass: gravimetric analysis	NS	None
Belzagui et al. (2019)[51]	Commercial garments: 100% polyester; 80% polyester; 20% elastane; 70% acrylic, 30% nylon	74–728	166 667–1 083 333	Fibres per kg of fabric	Conventional: FAGOR innovation F-2810, Spain	1000 rpm; 15 min; ambient temp.; 22 L water, commercially available detergent used as specified; water hardness of 349 mg L⁻¹ as CaCO₃	20	Number: counted manually, light microscopy; mass: estimated; morphology: SEM	NS	None
De Falco et al. (2019)[55]	100% polyester T-shirts and blouses	2000–2500	48.6–307.6; 641 000–1 500 000	mg per kg fabric	Conventional: Bosch WLG24225i	1200 rpm; 107 min; 40 °C; liquid detergent	400; 60; 20 and 5	Mass: gravimetric analysis; number: estimated; length and width: light microscopy, SEM and image analysis	NS	FTIR

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Table 1 (Contd.)

Reference	Fibres				Laundering		Analysis			
	Fabric type	Fabric mass (g)	Fibres shed	Units	Type/apparatus	Experimental/washing conditions	Filter pore-size (μm)	Fibre characterisation	LOD (μm)	Chemical confirmation method
McIlwraith et al. (2019)[62]	100% polyester fleece blanket	545	233 395	Fibres per kg fabric	Conventional: SDL Atlas M6 Vortex washing machine	660 rpm; 30 min; 16 °C; 26.5 L capacity	10	Mass: gravimetric analysis; number: light microscopy and image analysis; length: light microscopy and image analysis	100	None
Dalla Fontana et al. (2020)[53]	100% polyester knit	2000	32.5–38.6 (sum of 5 consecutive washes)	mg per kg fabric	Conventional: Bosch WAW286H8	600, 1400 rpm; 43, 90 min; 30 °C, 40 °C; with commercial detergent (45 mL)	40	Mass: gravimetric	NS	FTIR
Dalla Fontana et al. (2021)[54]	100% polyester	NS	7–19.5	mg per kg fabric	Conventional: Bosch WAW286H8	1400 rpm; 90 min; 40 °C; with commercial detergent (45 mL)	40	Mass: gravimetric	NS	None
Kärkkäinen and Sillanpää (2021)[60]	Various polyester, nylon, and acrylic commercially available garments	184–1052	100 000–6 300 000	Fibres per kg fabric	Conventional: Bosch WAE28477SN	1200 rpm; 75 min; 40 °C; with commercial detergent (50 mL)	0.7	Number: counted manually, light microscopy; mass: gravimetric; length: light microscopy and image analysis	NS	None
Periyasamy (2021)[64]	Jeans, 97% polyester, 3% lycra; 70% polyester, 27% cotton, 3% lycra; 50% polyester, 50% cotton	453–462	2 300 000–4 900 000	Fibres per kg fabric	Conventional: Whirlpool FRESH CARE	1200, 1400 rpm; 60, 75, 90 min; 30 °C, 45 °C, 60 °C; with and without 40 mL of fabric conditioner	200; 5	Number: counted manually, light microscopy; length and width: SEM and image analysis	NS	FTIR
Vassilenko et al. (2021)[66]	Fabrics of various compositions, constructions, treatments and finishes	500	8809–6 877 000; 9.6–1240	Fibres per kg fabric; mg per kg fabric	Conventional: SDL Atlas Vortex M6	645 rpm; 18 min; 41 °C; 34.1 L capacity	20	Mass: gravimetric; number: estimated, light microscopy	NS	FTIR
Hernandez et al. (2017)[50]	100% polyester knitted interlock, 98% polyester and 2% Spandex single knit Jersey fabrics	7	25–100	mg per kg fabric	Accelerated: Washtex-P Roaches laboratory washing machine	40 rpm; 45 min; 40 °C; 7 g of fabric, with/without liquid or powder detergent (4 g L^{-1}); 200 mL liquid; 10 × 6 mm diameter metal beads	0.45	Number: counted manually, light microscopy; mass: estimated; length normalised distribution: light micros and image analysis	40	None
Almroth et al. (2018)[42]		NS	900		Accelerated: Gyrowash one bath 815/8	Speed unspec.; 30 min; 60 °C; 10 × 10 cm fabric swatches, 125 mL water;	1.2	Number: counted manually, light	NS	None

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Table 1 (*Contd.*)

Reference	Fibres				Laundering		Analysis			
	Fabric type	Fabric mass (g)	Fibres shed	Units	Type/apparatus	Experimental/washing conditions	Filter pore-size (µm)	Fibre characterisation	LOD (µm)	Chemical confirmation method
	100% polyester, acrylic and nylon knits; 100% polyester fleece		110 000	Fibres per garment; Fibres per garment	standardised laboratory washer	0.375 mL liquid detergent; 25 × 6 mm diameter metal beads; with/without detergent		microscopy; length: light microscopy, image analysis		Accelerated: Linitest apparatus
De Falco et al. (2018)[57]	polyester weave and double-knit Jersey, and 100%; 5	100%	1 000 000	Fibres per garment; NS	polypropylene weave; None	1.05–1.67; Speed unspec.; 45 min; 40 °C; 9.3 × 9 cm swatches; with/without household and industrial detergent; 10 steel beads		Number: count. Manual SEM; length: SEM and image analysis; diameter: SEM and image analysis; mass: estimated	Fibres per kg fabric	None
Jönsson et al. (2018)[59]	Recycled 100% polyester tricot; Virgin 100% polyester tricot; Ultrasonic cut recycled 100% polyester tricot; Scissor cut recycled 100% polyester tricot; Recycled 100% polyester fleece; Virgin 100% polyester fleece	NS	9318; 10 886; 12 618; 27 370; 7075; 10 489	Fibres per 17 × 9 cm fabric swatch	Accelerated: Gyrowash standardised laboratory washer	Speed unspec.; 60 min; 40 °C; 17 × 9 cm fabric swatches; edge welded to form a bag with 25 steel beads inside; 75 mL water	0.65	Number, length and width counted manually, light microscopy and image analysis	Number, length and width: 100 length, 5 number	None
Haap et al. (2019)[58]	50% polyester, 50% cotton woven fabric	6	368 000	Fibres per kg fabric	Accelerated: Labomat lab-scale washing machine	40 rpm; 30 min; 40 °C; 8.5 × 16.5 edge-sealed fabric pillows; 50 steel beads; 160 mL liquid containing 2 mL liquid detergent	8	Number, length and width: NS SEM, dynamic image analysis; morphology: SEM	NS	Cotton removal by sulphuric

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Table 1 (Contd.)

Reference	Fibres: Fabric type	Fabric mass (g)	Fibres shed	Units	Laundering: Type/apparatus	Experimental/washing conditions	Analysis: Filter pore-size (μm)	Fibre characterisation	LOD (μm)	Chemical confirmation method
Zambrano et al. (2019)[25]	100% cotton, 100% rayon, 100% polyester and 50%/50% polyester–cotton weft knitted interlock fabrics	2–2.5	5 000 000–15 000 000	Fibres per kg fabric	Accelerated: SDL Atlas LaunderOmeter	Speed unspec;; 16 min; 25 and 44 °C; 10 × 10 cm fabric swatches; with/without AATCC standard reference liquid detergent (1.47 g L⁻¹); 150 mL liquid; 25 × 6 mm diameter metal beads	1.2	Number: FQA; mass: gravimetric analysis; length and width normalised distributions (FQA)	25	acid digestion None
Frost et al. (2020)[48]	100% cotton knit (virgin, 20% recycled and 40% recycled) 100% polyester knit (virgin, 40% recycled and 70% recycled) 30% polyester 70% cotton twill weave (virgin, 7% recycled cotton, 21% recycled cotton)	2.1–3.6	8 500 000–11 500 000 17 000 000–32 000 000 3 000 000–4 500 000	Fibres per kg fabric	Accelerated: SDL Atlas LaunderOmeter	Speed unspec;; 16 min; 20 °C; 10 × 10 cm fabric swatches; with AATCC standard reference liquid detergent (1.47 g L⁻¹); 150 mL liquid; 25 × 6 mm diameter metal beads	1.2	Number: FQA; mass: gravimetric analysis; length and width normalised distributions (FQA)	25	None
Cai et al. (2020)[52]	100% polyester, various knitted and woven fabrics	1.5–2.3	210 000–72 000 000	Fibres per kg fabric	Accelerated: Gyrowash standardised laboratory washer	Speed unspec;; 45 min; 40 °C; with surfactant (linear alkylbenzene sulfonic acid; 0.75 g L⁻¹; 0, 10 and 20 metal beads	0.45	Number: counted manually, light microscopy; mass: estimated; length and width: light microscope and image analysis	90	None
Özkan and Gündoğdu (2021)[63]	100% virgin polyester 100% recycled polyester	NS	167 437–368 094	Fibres per kg fabric	Accelerated: Gyrowash standardised laboratory washer	Speed unspec;; 45 min; 40 °C; 4 × 10 cm swatches; 150 mL water; 10 metal beads	0.45	Number: counted manually, light microscopy; length and width: unspec	0.45	None
Raja Balasaraswathi and Rathinamoorthy (2021)[65]	100% polyester weft-knitted fabrics	3.15–6.3	905 000–5 675 000 131.9–1507.2	Fibres per kg fabric mg per kg fabric	Accelerated: SDL Atlas LaunderOmeter	Speed unspec;; 45 min; 30 °C; 15 × 15 cm fabric swatches; with detergent (4 g L⁻¹); 150 mL liquid; 10 metal beads	11	Number: estimated; mass: estimated, light microscopy	NS	None
De Falco et al. (2018)[56]	100% polyester knit T-shirt	2500	549 913	Fibres per kg fabric	Conventional: Bosch WLG24225	1200 rpm; time unspecified; 40 °C; liquid detergent	400; 60; 20	Mass: gravimetric analysis; number: estimated; length and width: light microscopy, SEM and image analysis	NS	FTIR
	NS		1 733 000	Fibres per kg fabric	Accelerated: Gyrowash standardised laboratory washer	Speed unspec;; 45 min; 40 °C; 9 × 9 cm fabric swatches, edges thermally sealed; 10 steel beads; 150 mL liquid	5			

Table 1 (Contd.)

	Fibres				Laundering		Analysis			
Reference	Fabric type	Fabric mass (g)	Fibres shed	Units	Type/apparatus	Experimental/washing conditions	Filter pore-size (μm)	Fibre characterisation	LOD (μm)	Chemical confirmation method
Kelly et al. (2019)[61]	100% polyester knit T-shirts	NS	86.63–206.49	mg per kg fabric	Conventional: Miele front-loading washing machine W3622	600 or 1600 rpm; 30, 59 or 85 min; 13–15 or 30 °C; various wash cycles including cotton short cycle, cold express cycle and delicate cycle; 30–69 L; with/without 35 mL of commercial liquid detergent; 1.5 kg of T-shirts	22	Mass: gravimetric analysis; number: estimated; length: light microscopy and image analysis; width: SEM and image analysis	NS	None
			618 724–1 474 793	Fibres per kg fabric						
			53.84–119.83	mg per kg fabric	Accelerated: Tergotometer benchtop washing simulation device	100 or 200 rpm; 15 or 60 min; 15 or 30 °C; 5 × 5 cm laser-cut fabric swatches; no steel beads				
			384 545–855 884	Fibres per kg fabric						

mass. Size distributions constructed by Zambrano et al. (2019),[25] for fibres shed from PET, cotton, and PET-cotton blend fabrics also revealed this trend – fibre frequency increased as fibre length decreased, until the lower limit of detection for fibre length (200 μm) was reached.[25] Since the widths of synthetic fibres are usually very uniform and have a typical mean diameter of 11–16 μm,[25,46] it could be reasonably assumed that the majority of shed fibres would be captured by a filter with a pore-size of 10 μm or below. In Table 1, 9 of the 25 studies used filters with a pore-size above 10 μm, meaning fibres may have been lost even before analysis. Furthermore, it is important to highlight that only six studies provided a lower limit of detection for the measurement of fibre length. This is an essential parameter to evaluate the suitability of the methodology, and to contextualise results in ecotoxicology, since fibre size influences environmental fate and transport,[67] ingestion rates by organisms,[68] and specific surface area.[69]

The laundering method and apparatus used also likely contributed to the variation in shedding rates reported in the literature. Laundering speed varied from 40–1600 rpm,[50,61] and was unspecified in 10 of the 25 studies listed in Table 1. Laundering speed is thought to greatly influence fibre shedding because it determines the mechanical action exerted on the fabric.[42,49] Accelerated laundering methods were used in 9 studies, whereas commercial, or laboratory-scale washing machines were used in the other 8 studies. Accelerated laundering refers to the laboratory-scale simulation of home laundering, by placing the fabric and water/detergent solution under continuous agitation, often with the addition of metal beads to increase mechanical abrasion. This allows multiple experimental treatments and replicates to be performed simultaneously and reduces the total volume of water to be filtered and analysed. Cai et al. (2020)[52] reported that up to 72 000 000 fibres per kg of fabric could be released, but accelerated laundering was used in their study. Moreover, Zambrano et al. (2019)[25] found accelerated laundering generated approximately 40 times more fibres per unit mass of fabric, compared to conventional washing machines. This likely accounts for some variation in shedding rates between studies adopting conventional and accelerated laundering, and highlights that results from accelerated laundering studies should not be used to estimate fibre emissions during home laundering.

Control strategies at various levels of intervention have been proposed to reduce microplastic fibre emissions (Ramasamy and Subramanian, 2021).[70] At the individual level, commercially available capture devices may be used during laundering to reduce microplastic fibre emissions at the source. McIlwraith et al. (2019)[62] compared the microplastic fibre reduction efficiencies of two such devices; the Cora ball (a plastic ball with hooked arms) and the Lint LUV-R (retrofitted filter), when laundering a 100% polyester fleece. Where no device was used, 4800 fibres per litre were released. Fibre release decreased to 3580 fibres per litre with the use of the Cora ball, and only 648 fibres per litre with the Lint LUV-R filter,[62] representing fibre capture rates of 25.4% and 86.5% respectively. Napper et al. (2020)[71] tested the efficiency of several capture devices, finding the XFiltra retrofitted filter to be the most effective, reducing

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fibre emissions by 78% compared to a control where no devices were used. This was attributed to the fine pore-size of the filter (60 μm). However, two mesh bags, the Guppyfriend and the Fourth Element washing bag, resulted in reductions of 54% and 21% respectively, despite both having a mesh pore-size of 50 μm, so other design variables appear to influence the efficiency of these devices.[71] Using lower water volume laundering cycles has also been shown to significantly reduce the mass of shed fibres.[61] Fabric production may also be altered to reduce fibre shedding propensity. Generally, fibres with higher tensile strength and tenacity result in yarns with a lower hairiness (number of protruding loops and ends), meaning the final fabric has a higher abrasion resistance and therefore a lower shedding propensity.[25,44] Increasing the number of yarn twists per unit length, and the stitch density of fabrics can also decrease their shedding propensity.[42,65] Fleece fabrics, which are mechanically cut after construction, have an increased shedding propensity compared to similar, non-fleece knitted fabrics.[42] Current legislation aimed at reducing microplastic fibre emissions is non-existent in most countries.[70,72] Laws implemented in New York, and California, state that clothing containing more than 50% synthetic fibres must be labelled as a contributor to microplastic fibre pollution, aim to impart consumer knowledge to facilitate gradual consumer behavioural changes.[70]

3.   Microplastics in sewage sludge

Shed fibres are typically carried through municipal drainage systems to a wastewater treatment plant (WWTP) (Fig. 1b). Processes of wastewater treatment vary between facilities, however they typically begin with primary screening and sedimentation to remove coarse grit and suspended solids. This stage is followed by secondary treatment, which involves aerobic or anaerobic microbial incorporation to remove suspended or dissolved organic matter, often aided by the addition of flocculants in a secondary sedimentation tank. Secondary treatment usually also involves a disinfection stage to remove pathogens.[17] Tertiary treatment involves additional specialised mechanisms to improve effluent quality before discharge into the environment, such as additional filtration or the removal of nitrates and phosphates.[73] The solid residue, or sewage sludge, is collected and typically dewatered to reduce its volume, before being chemically, aerobically or anaerobically stabilised.[74,75] In the UK, the majority of sewage sludge is then utilised as a fertiliser (biosolids) in agriculture (79%), incinerated for energy recovery (18.4%), or disposed of in landfill (0.6%).[32] Throughout Europe, biosolids application rates to land vary greatly between countries, from over 90% in Ireland and Lithuania, to less than 5% in the Netherlands, Slovenia and Malta.[76] In the USA and China, 49–60% and 14–45% of sludge produced respectively, is applied to agricultural land as biosolids.[77–80]

Several studies have quantified microplastic contamination through the different stages of the wastewater treatment process at specific WWTPs. Comparison of microplastic concentrations in the influent to the effluent has revealed that 72–99.9% of microplastics (by number) are removed during the wastewater treatment process.[81,82] For example, Murphy et al. (2016)[16] sampled concentrations of microplastics at four progressive stages of wastewater treatment, at a WWTP in Scotland, finding that microplastic concentrations decreased from 15.7 microplastics L^{-1} in the influent, to 0.25 microplastics L^{-1} in the final effluent; a reduction of 98%. Microplastics were identified visually using a dissection microscope and characterised using FT-IR (Fourier Transform Infrared) spectroscopy, although specific details of spatial resolution and limits of detection for size were not reported. It has been estimated that approximately 78% of microplastics (by number) entering a WWTP are removed during wastewater treatment and are present in the sewage sludge.[29] Primary wastewater treatment has been shown to remove 92–93% of textiles fibres (both natural and synthetic fibres), with secondary treatment resulting in only a further 0.2% reduction.[83] In the same study, microplastics removal after primary treatment was 32%, with 76% of remaining microplastics being removed after secondary treatment.

While there is some published data on the fate of microplastics in WWTPs, there is a lack of information concerning the fate of microplastic fibres specifically. The differing behaviours of textiles fibres and microplastics may be explained by differences in density, which affects the settling velocity of the particles.[84] Polyethylene terephthalate (polyester) is the most widely used synthetic polymer in the manufacturing of apparel textiles,[44] and has a density of 1.32–1.41 g mL^{-1},[85] which is higher than that many other polymer types commonly identified in sewage, including polyethylene ($\rho = 0.89$–0.97 g mL^{-1}) and polypropylene ($\rho = 0.85$–0.92 g mL^{-1}).[86] It is yet unclear how microplastic morphology influences wastewater separation efficiency, although for spherical microplastics, a larger diameter will increase settling velocity.[84] Nonetheless, methods of sampling, microplastic extraction from sludge, and characterisation are still currently in development and often differ widely between studies.

Chemical oxidation is often employed to digest organic matter, allowing an easier separation of microplastics from the solid sewage sludge fraction.[27] It also aids the removal of any organic substances impregnating/coating the surfaces of the microplastics, which would hinder the spectral characterisations and chemical classification with techniques such as FT-IR or Raman spectroscopy. Mason et al. (2016)[17] used 30% hydrogen peroxide (H_2O_2) to digest sludge samples from 17 WWTPs, before analysing microplastics visually under a dissection microscope. The dominant shape fraction (fibres, fragments, films, pellets, foams) in this study was found to be fibres; accounting for 46% and 80% of total microplastics in the 125–355 μm and >355 μm size fractions respectively.[17] However, sampling bias and misidentification of microplastics can occur where only visual identification is employed.[27] Talvitie et al. (2017)[28] found that only 34% of fibres separated from sludge were composed of synthetic polymers (PET – 33%; polyacrylic – 1%) when analysed with FT-IR, with the remainder being natural fibres such as cotton, or regenerated fibres such as rayon. Eriksen et al. (2013)[87] and Lenz et al. (2015)[88] found 20% and 32% of particles were visually identified erroneously as

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microplastics, after verification with scanning electron microscopy (SEM) and Raman microscopy, respectively. Mason *et al.* (2016)[17] may therefore have overestimated the presence of microplastic fibres in sewage sludge samples, as only visual identification based on morphology was employed. The use of H_2O_2 may also result in the oxidation and subsequent destruction of polymers such as nylon in the digested sample.[89] Nylon is a common synthetic fibre used in apparel textiles,[44] and the shedding propensity of nylon garments during laundering is comparable to that of polyester (Table 1).[42,49,51] It is possible, therefore, that the relative proportion of nylon fibres is underestimated when H_2O_2 is used as a chemical oxidant in sludge processing. Other methods used for processing of microplastics from environmental or biological samples include ultrasonic extraction and the use of alkaline (*e.g.* KOH or NaOH) dissolution and/or acid (*e.g.* HNO_3 or HCl) digestion of samples.[90–94] However, these methods have not been tested for the analysis microplastic fibres in sewage sludge.

Microplastic extraction from sludge in more recent studies typically involves a density separation step to remove inorganic debris such as sand, grit and aluminosilicate minerals.[27] This separation involves the agitation and prolonged settling of the sludge matrix in a high density, saturated salt solution, such as sodium chloride (NaCl) ($\rho = 1.2$ g mL^{-1}) or zinc chloride (ZnCl) ($\rho = 1.6$ g mL^{-1}),[85,94] or mixtures of water, sucrose, and ethanol.[95] Plastics typically have a density of 0.89–1.2 g mL^{-1}. However, PET and PVC can have densities up to 1.41 and 1.70 g mL^{-1}, respectively.[86] Li *et al.* (2018)[96] extracted microplastics by density separation from sewage sludge from 28 WWTPs in China using saturated NaCl solution, followed by H_2O_2 oxidation to digest remaining organic matter. Microplastics were visually sorted by morphology and subsamples were qualitatively analysed by FT-IR and SEM microscopy. Li *et al.* (2018)[96] identified between 1565 and 56 386 microplastics per kg of sludge (37 μm–5 mm in size), 62.5% of which were fibres. The total number, and proportion of fibrous microplastics were potentially underestimated in this study due to the higher density of PET ($\rho = 1.32$–1.41 g mL^{-1}) than the NaCl solution ($\rho = 1.2$ g mL^{-1}).[85] This underestimation is a significant limitation of the methodology because PET is by far the most widely used synthetic fibre in apparel textiles production.[43] During quality control experiments by Liu *et al.* (2018),[20] soils were spiked with microplastics including polypropylene (PP), polyethylene (PE), nylon (polyamide), PET and PVC, before density separation with NaCl. Recovery of both PET and PVC ($\rho = 1.32$–1.7 g mL^{-1})[86] from the soil samples was 0%, highlighting the importance of using higher density salts such as ZnCl ($\rho = 1.6$ g mL^{-1}).[94]

There is general agreement between studies investigating the mass balance of microplastics entering WWTPs that microplastics are removed from wastewater very effectively during treatment. Carr *et al.* (2016),[15] Magnusson and Norén (2014),[82] Gies *et al.* (2018)[97] and Leslie *et al.* (2017)[81] reported removal rates of 99.9%, 99.9%, 97–99%, and 72%, respectively, by quantifying the number of microplastics in the influent, effluent and sludge. The majority of microplastics are retained in the collected sewage sludge (Fig. 1c) where they are exposed to elevated concentrations of metals and metalloids (Table S1 in

ESI materials†).[98–106] Despite practical limitations of microplastic extraction from solid media such as sludge and soils, reported concentrations in sewage sludge samples have ranged from 1–56 386 microplastics per kg of sludge.[17,19,82,96,107] Their small size and high specific surface area, coupled with the consistently elevated concentrations of metals and metalloids in sludge, means that sorption of metals and metalloids on microplastic surfaces may occur during the wastewater or sludge treatment processes.

4. Metal and metalloid sorption onto microplastics

The presence of metals and metalloids such as arsenic (As), copper (Cu), chromium (Cr), mercury (Hg), nickel (Ni), lead (Pb), and zinc (Zn) in sewage sludge has been well studied, and regulations for sludge use in agriculture are enforced worldwide, including in the UK, EU and USA (Table S2 in ESI material†), to reduce their potential harm to organisms.[29,108–111] Many studies have quantified the sorption of a variety or organic pollutants such as polycyclic aromatic hydrocarbons (PAHs),[112] antibiotics,[113] and phthalate esters[114] onto microplastic surfaces, and this topic has been extensively reviewed.[115,116] However, the sorption of metals and metalloids to microplastic surfaces is far less studied. Furthermore, the sorption of metals and metalloids to microplastic fibres has rarely been studied, and data are extremely scarce. As a result, inferences on the sorption of metals and metalloids to microplastic fibres need to be made, based on observations made on the sorption of metals and metalloids to microplastic pellets (Table 2). There are several limitations to this inference that should be acknowledged. The surface area to mass ratio of microplastic fibres is likely to be greater than pellets. Therefore, a fairer comparison of the sorption of metals and metalloids to microplastics should be based on mass of adsorbate to surface area of adsorbent. However, because the surface area of microplastics is rarely reported in sorption experiments, this is currently not possible with the data available in Table 2.

Reported plastic–water partition coefficients (K_d values), representing the distribution of the respective metal or metalloid between the plastic-bound phase and the aqueous phase, range widely between metal or metalloid type, plastic type, plastic aging, and aqueous matrix type (Table 2). For example, the partitioning of aqueous Cu to virgin plastics decreases in the order PVC > PS > PE,[36,118] and is higher in seawater than in freshwater.[35,36] Cr is sorbed more strongly to aged, rather than virgin, PE microplastics by an order of magnitude.[35] Cu sorption was not significantly affected by plastic age in seawater whereas, in freshwater, sorption was greater on the aged, rather than virgin, PE microplastics.[35] Tang *et al.* (2021)[119] investigated the sorption of Cu, Ni and Zn onto nylon microplastics collected from the environment. Langmuir modelling of isotherm data revealed maximum sorption capacities for Cu, Ni and Zn were 16.7 μg g^{-1}, 10.6 μg g^{-1} and 12.7 μg g^{-1} respectively, however, the data are not reported for the virgin nylon microplastics. Chen *et al.* (2021)[120] compared the sorption of Cu onto virgin

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Table 2 Adsorption of metals to microplastic pellets under various conditions (K_d = plastic–water partition coefficient; PS = polystyrene, PVC = polyvinyl chloride, HDPE = high-density polyethylene, PE = polyethylene, PP = polypropylene)

Metal	Polymer	K_d (mL g^{-1})	Matrix	Type of microplastic	Experimental conditions (solid to liquid ratio, $S:L$; agitation; contact time)	Ref.
Ag	PE	1.18	Freshwater (river Plym, UK)	Virgin	1 : 96 ($S:L$); 150 rpm; 48 h	Turner and Holmes (2015)[36]
	PE (>90%) and PP	96.3		Aged (beached)	1 : 86 ($S:L$); 150 rpm; 48 h	
Cd	PE	1.24	Seawater	Virgin	1 : 75 ($S:L$); 150 rpm; 48 h	Holmes et al. (2012)[35]
		7.94		Aged (beached)		
	PE	0.347	Freshwater (river Plym, UK)	Virgin	1 : 96 ($S:L$); 150 rpm; 48 h	Turner and Holmes (2015)[36]
	HDPE	242	Distilled water	Virgin	1 : 500–1 : 4000 ($S:L$); 20 rpm; <24 h	Zou et al. (2020)[117]
	LDPE	345				
	PVC	1748				
	CPE	7485				
Co	PE	0.834	Seawater	Virgin	1 : 75 ($S:L$); 150 rpm; 48 h	Holmes et al. (2012)[35]
		4.03		Aged (beached)		
Cu	PE	0.304	Freshwater (river Plym, UK)	Virgin	1 : 96 ($S:L$); 150 rpm; 48 h	Turner and Holmes (2015)[36]
	PE	45.0	Seawater	Virgin	1 : 75 ($S:L$); 150 rpm; 48 h	Holmes et al. (2012)[35]
		45.2		Aged (beached)		
	PE	3.62	Freshwater (river Plym, UK)	Virgin pellets	1 : 96 ($S:L$); 150 rpm; 48 h	Turner and Holmes (2015)[36]
	PE (>90%) and PP	61.0		Aged (beached)	1 : 86 ($S:L$); 150 rpm; 48 h	
	PS	659	Seawater (Madeira, Portugal)	Virgin, with anti-fouling paint	1 : 2500 ($S:L$); no agitation; 336 h	Brennecke et al. (2016)[118]
	PVC	849			1 : 1000 ($S:L$); no agitation; 336 h	
	HDPE	385	Distilled water	Virgin	1 : 500–1 : 4000 ($S:L$); 20 rpm; <24 h	Zou et al. (2020)[117]
	LDPE	56				
	PVC	431				
	CPE	3868				
Cr	Nylon	172–432	Unspecified	Aged (aquaculture)	1 : 333 ($S:L$); 185 rpm; 72 h	Tang et al. (2021)[119]
	PS	394.8	Seawater (Shenzhen, China)	Virgin	1 : 2000 ($S:L$); 140 rpm; 24 h	Chen et al. (2021)[120]
		761.3		Aged (1 month in seawater)		
		591.3		Aged (3 months in seawater)		
	PE	331.2		Virgin		
		755		Aged (1 month in seawater)		
		448.4		Aged (3 months in seawater)		
	PE	11.6	Seawater	Virgin	1 : 75 ($S:L$); 150 rpm; 48 h	Holmes et al. (2012)[35]
		221		Aged (beached)		
	PE	1.01	Freshwater (river Plym, UK)	Virgin	1 : 96 ($S:L$); 150 rpm; 48 h	Turner and Holmes (2015)[36]
	PE (>90%) and PP	4.60		Aged (beached)	1 : 86 ($S:L$); 150 rpm; 48 h	
Cs	HDPE	80.3	Freshwater (NSW, Australia)	Gamma-irradiated, aged	Unspecified ($S:L$); 50 rpm; 48 h	Johansen et al. (2018)[121]
	PE	9.30	Estuarine water (NSW, Australia)			
Hg	PE	5.84	Freshwater (river Plym, UK)	Virgin	1 : 96 ($S:L$); 150 rpm; 48 h	Turner and Holmes (2015)[36]
	PE (>90%) and PP	31.8		Aged (beached)	1 : 86 ($S:L$); 150 rpm; 48 h	
Ni	PE	1.60	Seawater	Virgin	1 : 75 ($S:L$); 150 rpm; 48 h	Holmes et al. (2012)[35]
		8.87		Aged (beached)		
Pb	PE	0.491	Freshwater (river Plym, UK)	Virgin	1 : 96 ($S:L$); 150 rpm; 48 h	Turner and Holmes (2015)[36]
	Nylon	48–183	Unspecified	Aged (aquaculture)	1 : 333 ($S:L$); 185 rpm; 72 h	Tang et al. (2021)[119]
	PE	0.245	Seawater	Virgin	1 : 75 ($S:L$); 150 rpm; 48 h	Holmes et al. (2012)[35]
		149		Aged (beached)		

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Table 2 (Contd.)

Metal	Polymer	K_d (mL g^{-1})	Matrix	Type of microplastic	Experimental conditions (solid to liquid ratio, $S:L$; agitation; contact time)	Ref.
PE	PE	5.80	Freshwater (river Plym, UK)	Virgin	1:96 (S:L); 150 rpm; 48 h	Turner and Holmes (2015)[36]
	PE (>90%) and PP	217		Aged (beached)	1:86 (S:L); 150 rpm; 48 h	
	HDPE	2316	Distilled water	Virgin	1:500-1:4000 (S:L); 20 rpm; <24 h	Zou et al. (2020)[117]
	LDPE	590				
	PVC	2518				
CPE		45 306				
Sr	HDPE	6.90	Freshwater (NSW, Australia)	Gamma-irradiated, aged	Unspecified (S:L); 50 rpm; 48 h	Johansen et al. (2018)[121]
		5.00	Estuarine water (NSW, Australia)			
Zn	PS	304	Seawater (Madeira Island, Portugal)	Virgin, with anti-fouling paint	1:2500 (S:L); no agitation; 336 h	Brennecke et al. (2016)[118]
	PVC	195			1:1000 (S:L); no agitation; 336 h	
	Nylon	87-191	Unspecified	Aged (aquaculture)	1:333 (S:L); 185 rpm; 72 h	Tang et al. (2021)[119]

and aged PS and PE microplastics, finding aged microplastics to have a higher sorption capacity (Table 2). This was partly attributed to the increased specific surface area, and more negative zeta potentials of the aged microplastics.[120]

The sorption of metals and metalloids onto microplastic surfaces is strongly influenced by the chemical properties of the metal or metalloid and the physicochemical properties of the microplastic surfaces.[35,36,118–120] Previous studies investigating metal and metalloid sorption onto microplastics have mainly focussed on microplastic types commonly found in marine environments, such as PE, PS and PVC. Data for metal and metalloid sorption onto PET microplastic surfaces are scarce in the literature in comparison with other polymer types. Cojocariu et al. (2017)[122] quantified the sorption of Pb and Cu to recycled PET fibres, reporting Pb and Cu sorption to be 48.9 mg g^{-1} and 30.7 mg g^{-1} respectively. These sorption values are several orders of magnitude higher than others reported thus far in the literature,[35,36,118–120] although it should be noted that a physicochemical characterisation of the PET fibres was not provided in this study, so fibre size and specific surface area are not known. Recently, Han et al. (2021)[123] investigated the sorption of Pb, Cu and Cr onto PET microplastics of varying size fractions (<0.9 mm; 0.9–2 mm; 2–5 mm). Sorption increased in the order Cr < Cu < Pb, and decreased with increasing microplastics size. Kinetics experiments revealed that at equilibrium the sorbed concentrations of Pb, Cu and Cr on the PET fibres, calculated by fitting the pseudo-second order model, were 1.04, 0.488 and 0.385 μg g^{-1} respectively. These values are similar to those previously reported for virgin and beached PE pellets.[35,36] PET is often underrepresented in density separations due to its high density,[85] yet is the most commonly used material in the manufacturing of apparel textiles,[43] which have been shown to shed large quantities of fibres into the wastewater system (Table 1). The laundering of synthetic textiles therefore represents a considerable diffuse source of PET microplastics into sewage sludge where they are exposed to elevated concentrations of metals and metalloids (Table S1†). Unlike the previously investigated plastic types (PE, PVC and PS), PET contains hydroxyl (–OH) groups at the ends of the polymer chains, which are approximately 100 monomer units in length.[124] These hydroxyl groups may increase the sorption capacity of PET for metals and metalloids, as they become deprotonated at circumneutral pH, which is often characteristic of sewage sludges.[125] Deprotonated hydroxyl groups (–OH–) are negatively charged and therefore may facilitate the sorption of cationic metals, such as Cu^{2+}, Pb^{2+}, and Cd^{2+}.[126,127] Zou et al. (2020)[117] found that the sorption capacity of chlorinated-PE for Cd, Cu and Zn, was at least one order of magnitude higher than HDPE and LDPE microplastics, which was partly attributed to the high electronegativity of chlorine. The high electronegativity the oxygen atoms in PET may therefore increase its sorption capacity, particularly for cationic metals. Further study of PET is required to understand the role of its unique functional groups in the sorption of metals and metalloids.

Environmental water samples often contain humic substances; high molecular weight organic macromolecules, with heterogeneous branching structures and oxygen-

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containing functional groups,[36,126] which may also influence the sorption of metals and metalloids onto microplastics. Li *et al.* (2019)[128] reported decreased sorption of Cd to PP and PE microplastics with increasing concentrations of humic substances. This observation was attributed to the complexation of Cd by the acidic functional groups of the humic substances, and the subsequent decrease in free Cd^{2+} ions in solution.[128] Turner and Holmes (2015),[36] however, hypothesised that the observed increase in the sorption capacity of beached PE pellets over virgin PE pellets could be in part due to the gradual accumulation of organic matter on the surfaces of the beached pellets. It was suggested that this organic matter provided an increased number of charged functional groups, and therefore increased the sorption capacity of the plastic surface.[36] These conflicting results are difficult to resolve, because the concentrations of humic substances in the water samples used by Turner and Holmes (2015)[36] were not quantified. Biofilm formation may also alter the sorption capacity of microplastics over time. Biofilms are formed by the extracellular polymeric substances, such as polysaccharides, of colonising bacteria on surfaces. Biofilms may change the surface properties, and therefore sorption capacity of microplastics by introducing new functional groups, such as amines, hydroxyls, and carboxylic acids.[129] Biofilms may also enrich the microplastic surfaces with particular elements, for example strontium and sulphur, present in the radiolarian protozoa *Acantharea*.[130,131]

The accumulation of metals and metalloids to microplastics represents an important knowledge gap, and more work is necessary in order to provide a more systematic quantification of the retention of metals and metalloids onto microplastic surfaces in general, and microplastic fibres in particular. Nonetheless, the current data show that microplastics are capable of sorbing considerable amounts of metals and metalloids which may in turn influence the biogeochemical cycling of these metals and metalloids in agricultural soils when the microplastics are applied in biosolids.

5. Sewage sludge application and impacts to terrestrial environments

Sewage sludge, the solid residual product of wastewater treatment, contains elevated concentrations of microplastics, and is widely applied to agricultural soils as biosolids. Using density separation, followed by filtration and visual identification with a stereo microscope, Corradini *et al.* (2019)[21] quantified microplastic concentrations in agricultural soils with differing biosolids application rates. The median estimated mass of microplastics in the soil increased with each successive biosolids application, from 1.37 to 4.38 mg kg^{-1} in soils that had received 1 and 5 biosolids applications, respectively, and 97% of these microplastics were categorised as fibres. Fibres were also found to be the dominant morphology of microplastics in agricultural soils in north-western and south-western China, accounting for 49% and 92% of all microplastics respectively.[132,133] As highlighted by Corradini *et al.* (2019)[21] microplastic morphologies were categorised at the operator's

discretion, further necessitating a need for a strict and appropriate definition of microplastic fibres. Nevertheless, Corradini *et al.* (2019)[21] provide strong *in situ* evidence that the application of sewage sludge derived biosolids provides an important pathway for the transfer of microplastics, particularly microplastic fibres, to agricultural soils (Fig. 1d).

Like most other widely manufactured polymers, PET is highly resistant to degradation under typical environmental conditions, however, is susceptible to hydrolytic degradation due to its ester bonds, particularly at extreme pH values.[86,134] Over time, water reacts with the ester bonds of the PET backbone, forming two shorter polymer chains ending in alcohol and carboxylic acid groups. UV radiation from sunlight exposure will also initiate photooxidation of PET, where the ester bonds are cleaved, leaving carboxylic acid groups on the polymer surface. Degradation products include CO, CO_2, terephthalic acid and other carboxylic acids.[134] Environmental exposure of PET can significantly decrease tensile strength after approximately one year, depending on UV intensity, temperature, and precipitation.[135] Shape also influences degradation rate, as highlighted by Chamas *et al.* (2020),[134] who estimated that HDPE films, fibres and spheres weighing 2.75 g each, will take approximately 1.8, 465 and 2000 years respectively to completely degrade. Microplastic fibres are likely to degrade in the environment more rapidly than films, but let rapidly than spheres, as degradation is controlled largely by the size of the surface area exposed.[134] Biological degradation of PET fibres is typically slow. Zambrano *et al.* (2019)[25] assessed the biodegradation of various yarns, by measuring the total oxygen demand in a closed respirometer over 243 days, after inoculation with aerobic microorganisms from activated sludge. Biodegradation of the polyester yarn was only 4.1% compared with 75.9% for the cotton yarn, revealing that PET is a relatively unavailable carbon source for microorganisms.[25] Yoshida *et al.* (2016)[136] isolated a bacterium, *Ideonella sakaiensis* 201-F6, that was capable of biodegrading PET by adhering to its surface and secreting two hydrolytic enzymes. The bacterium was capable of almost completely degrading a 60 mg PET film after 60 days, and catabolised 75% of the total carbon after 15 days.[136] The main biodegradation product was mono(2-hydroxyethyl) terephthalic acid, which was rapidly metabolised further into the two monomers of PET, terephthalic acid and ethylene glycol.[136,137] The co-polymer poly(ethylene terephthalate-*co*-lactate) can be effectively degraded under laboratory composting conditions,[138] although further research is needed before bioremediation strategies are implemented for PET fibres in sludge and soils.

The impacts of microplastics on agroecosystems are still relatively unknown. However, emerging research is indicating that microplastic contamination may influence the physical, chemical, and biological properties of soils. Machado *et al.* (2018)[139] reported a reduction in bulk density, and an increase in water holding capacity for soils spiked with polyester fibres, that was not observed in soils spiked with polyethylene fragments and polyamide beads. This observation is thought to be due to the more efficient entanglement of soil aggregates by the fibres, creating more air-filled pore spaces in the soil. Kim and

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An (2019)[140] observed marked behavioural changes and a significant reduction in mobility of springtails in soils due to the presence of PS microplastics, even at relatively low concentrations (8 mg kg^{-1}). PS nanoplastics at concentrations of 10 μg L^{-1} were shown to induce toxicity in nematodes (*Caenorhabditis elegans*).[141] Earthworms (*Lumbricus terrestris*) were observed ingesting PE microplastics (150–2800 μm in diameter) in mesocosm experiments,[67,142,143] however the ecotoxicological implications of this were not studied.

Sewage sludge contains elevated concentrations of metals and metalloids (Table S1†), which have been shown to sorb onto microplastic surfaces (Table 2). In the UK and EU, legislation stipulates the maximum concentrations of metals and metalloids such as Cd, As, Hg, Pb and Zn in agricultural soils and maximum permissible loadings in sludge applications (Table S2†). However, there are currently no regulatory limits on microplastic additions to agricultural soils. As a result, the occurrence and accumulation of microplastics in agricultural soils has gone largely unmonitored in recent years. In the EU, it is estimated that up to 4.7 times more microplastics per year are released into agricultural soils than to surface waters,[24] despite terrestrial microplastics research continuing to lag behind marine and freshwater microplastics research.[22]

Metals and metalloids sorbed onto the surfaces of ingested microplastics may have an altered bioavailability and bioaccessibility to soil organisms, compared to dissolved ions. Pollutants sorbed onto the surfaces of ingested microplastics may readily desorb in acidic gut environments.[37,144,145] Microplastics may therefore act as a vector, facilitating an increased exposure of the sorbed pollutant to the organism (Fig. 1e).[144] For example, desorption of persistent organic pollutants from PE and PVC microplastics was shown to be up to 30 times greater under simulated gut conditions, than in seawater alone.[145] Synthetic earthworm gut extraction tests of HDPE microplastics and soil aggregates by Hodson *et al.* (2017),[37] revealed that Zn desorption was 4–30 times greater from the microplastics than from the soils. Despite this, in the same study, Zn-loaded microplastics induced no statistically significant effects in Zn bioaccumulation, weight or mortality on earthworms. Exposure studies have shown that ingestion can facilitate the fragmentation of microplastics, creating smaller particles. In a study by Kwak and An (2021),[146] earthworms (*Eisenia andrei*) were exposed to polyethylene microplastics (180–300 μm diameter) for 21 days, before microplastics were extracted from earthworm intestines and casts. Using SEM, the researchers identified smaller microplastics on the surfaces of the ingested and excreted microplastics, measuring as small as 182 nm in diameter.[146] This is thought to be primarily due to microbial degradation by bacteria in the gut microbiome of earthworms.[147] Similar results were reported by Dawson *et al.* (2018),[148] who observed that microplastics ingested by Antarctic krill (*Euphausia superba*) were on average 78% smaller in diameter (7.1 μm) than the original microplastics (31.5 μm). Ingestion of microplastics by soil organisms may therefore result in an increase in the specific surface area of the microplastics as they are fragmented. Experiments by Khan *et al.* (2017),[144] using polyethylene microplastics loaded with silver

(Ag), revealed that desorption of Ag was significantly higher under lower pH conditions, reaching 98% after 3 hours at a pH of 4.1. However, such experiments have not been performed with polymers commonly used in textiles, such as PET, nylon, and polyurethanes. These polymers are likely to contribute significantly to the microplastics entering agricultural soils that receive municipal sewage sludge.

6. Conclusions

Microplastics are ubiquitous, highly recalcitrant emerging contaminants of concern. Current data suggest microplastics, particularly fibres, are pervasive in sewage sludge and agricultural soils. Further work is required to develop more robust, efficient, and consistent analytical procedures to quantify their abundance. The laundering of synthetic fibres provides an important diffuse source of microplastic fibres, which are transported into wastewater systems. Microplastic fibres from laundering have a typical diameter of approximately 20 μm. An inverse relationship between frequency and length is often observed. Values reported in the literature for the number of fibres shed during conventional laundering experiments are 1900–110 000 fibres per garment, 22 600–13 100 000 fibres per kg of fabric, and 7–1240 mg of fibres per kg of fabric. During wastewater treatment, 78% of microplastics are retained in the sewage sludge. Municipal biosolids can reportedly contain up to 56 000 microplastics per kg, although values reported in the literature vary greatly. This sludge is commonly applied to agricultural land as a soil amendment, creating a pathway for microplastic fibres generated during laundering to contaminate agricultural soils. Therefore, microplastic fibres may cause deleterious effects to the soil biophysiochemical environment which require further investigation.

The surfaces of microplastics may also be loaded with elevated concentrations of metals and metalloids due to sorption during the wastewater treatment process. Research on the sorptive properties of microplastics is still in its infancy. However, available data reveals that sorption of metals and metalloids is largely dependent on solution chemistry (pH, ionic strength, presence of humic substances), and the physicochemical properties of the microplastics (functional groups, degree of surface oxidation, specific surface area, point of zero charge). Therefore, the application of sewage sludge to agricultural land may represent an important vector for metals and metalloids, sorbed on the surfaces of microplastics, to soil organisms.

Microplastic fibres are an important subcategory of microplastics, due to their prevalence in sludge and agricultural soils, and high specific surface area. However, the sample processing method used to isolate microplastics from soils and sludge should be carefully considered, particularly during density separation to avoid excluding high-density microplastics, such as PET, from being quantified. Research on the sorptive properties of synthetic polymers commonly used in textiles, such as PET and poly(amide), is extremely scarce, representing an important knowledge gap. This gap requires immediate attention because textiles-derived microplastic fibres are likely to

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represent a significant proportion of microplastics applied to agricultural soils, in biosolids produced from municipal sewage sludge.

Author contributions

H. Frost: conceptualisation, investigation, writing – original draft, writing – review & editing, visualisation; T. Bond: conceptualisation, writing – review & editing, supervision, funding acquisition; T. Sizmur: conceptualisation, writing – review & editing, supervision, funding acquisition; M. Felipe-Sotelo: conceptualisation, writing – review & editing, supervision, funding acquisition.

Conflicts of interest

There are no conflicts to declare.

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