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Review

Variation in microplastic concentration, characteristics and distribution in sewage sludge & biosolids around the world

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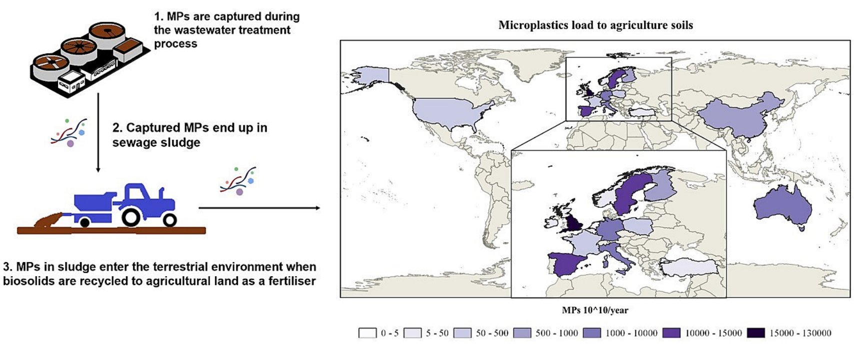
HIGHLIGHTS

- Systematic analysis of 65 studies reporting on microplastics in sludge and biosolids
- Microplastic in sludge, biosolids and soil quantified across 25 countries
- Microplastic concentrations varied widely and spanned 6 orders of magnitude.
- 6.44×10^8 to 1.67×10^{12} microplastics were present per 1000 t sludge.
- Up to 6430 t of plastic are released to land globally through biosolid recycling per year.

GRAPHICAL ABSTRACT



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ABSTRACT

Microplastics have been reported in wastewater treatment works across the world. The majority of microplastics are removed during the wastewater treatment process, with removal efficiencies between 57 % to 99 %. What happens to the microplastics removed from the wastewater, and how they accumulate in sewage sludge and biosolids (by-products of the wastewater treatment process), remains a topic of high interest. Here we systematically reviewed the current state of knowledge on the presence, concentration, and characteristics of microplastics in sewage sludge and biosolids globally to understand how biosolids may act as a pathway for microplastic pollution to soils. A systematic search was performed on the Web of Science and Science Direct databases. Sixty-five studies reporting on microplastic pollution in sewage sludge and biosolid products were identified, spanning twenty-five countries. Reported microplastic concentrations varied considerably from 0.193 microplastics/g to 1.69×10^5 microplastics/g with a median microplastic concentration of 22.41 microplastics/g, illustrating how many microplastics are captured during the wastewater treatment process, and retained in the sewage sludge. The extent to which biosolid recycling pollutes the terrestrial environment was compared between countries. High numbers of microplastics were estimated to reach fields via biosolid application with a wide variation of 8.2×10^{10} to 1.29×10^{15} microplastics/year between sixteen countries, although there was no significant difference in microplastic concentration between fields with a history of biosolid applications and control fields. The comparative risk this delivery of approx. 0.4 to 6430 tonnes of microplastics poses compared to the environmental benefits of nutrient and carbon recycling associated with biosolids reuse, or compared to other sources of microplastic pollution remains a global research imperative. The next step in scientific research needs to focus on solutions to the biosolid and circular economy conundrum – biosolids are a valuable source of nutrients but contain high concentrations of microplastics, which are ultimately entering the terrestrial environment.

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Contents

1. Introduction

Microplastics are a contaminant of emerging concern (Wagner and Lambert, 2018), and are found in various environmental settings at increasing concentrations (Catarino et al., 2021). Despite most research on microplastics being concentrated in the marine environment, 80 % of marine plastic debris is transported to the oceans from land-based sources such as rivers (Li et al., 2016; Horton et al., 2017). To tackle microplastic contamination in freshwater and marine environments we need to understand their sources, pathways, and fate in wastewater systems as well as spatial distributions and concentrations (Woodward et al., 2021), this is crucial to determining environmental exposure and associated risks, as well as enabling efficient intervention measures.

Microplastics can act as a vector for chemical and biological contaminants, creating a potential exposure route for the transfer of contaminants from the environment to organisms (Xie et al., 2021; McCormick et al., 2014). Microplastic components such as monomers and additives (plasticizers, flame retardants, antioxidants), which can be hazardous to human and environmental health, may be released when microplastics enter the environment (Yu et al., 2020). Currently, there is limited information regarding the leaching of chemicals from microplastics into the environment and uptake into the food chain. The risks posed to human health by microplastics, and their chemical additives, are largely unknown (Catarino et al., 2021; Galloway, 2015), however, for a thorough understanding of the risks posed by microplastics, their distribution patterns and pathways to the environment need further study (Roscher et al., 2022).

Microplastics, plastic particles or fibres <5000 μm, is a general term used to describe a large and complex suite of contaminants, originating from many different product types and sources and which consist of varying shapes, sizes, molecular structures, and characteristics (Rochman et al., 2019). Microplastics can be intentionally manufactured to <5000 μm (primary microplastics) or formed from the fragmentation, of larger plastic or plastic-containing items (Harley-Nyang et al., 2022) through physical, chemical, and biological processes during exposure to the environment (Liu et al., 2022).

1.1. Microplastics in wastewater treatment works

Wastewater treatment works (WwTWs) are an important pathway for the release of microplastics to the terrestrial environment via final effluent release, combined sewer overflows, or through the recycling of sewage sludge (biosolids) to agricultural land (Harley-Nyang et al., 2022).

Microplastics have been reported in wastewater influent, with concentrations ranging considerably between 0.631 particles/l (Magnusson et al., 2016) to 1.3×10^5 microplastics/Litre (Hansen and Vollertsen, 2017). These microplastics can originate from different sources such as domestic, industrial, or surface water runoff when sewerage systems are combined.

Despite the high concentration identified in the influent, WwTWs have been found to be (unintentionally) effective at removing microplastics from the wastewater stream with removal efficiencies of 57 %–99 % reported following secondary treatment processes (Carr et al., 2016; Gies et al., 2018; Liu et al., 2019). The high removal efficiency of microplastics from wastewater indicates that most are captured during wastewater treatment and retained in the sludge (Sun et al., 2019). Sewage sludge is the solid by-product generated from the treatment of wastewater and consists of primary sludge (faecal matter) and activated secondary sludge (biomass) (UK Water, 2010).

1.2. Management and use of biosolids

In the European Union (EU), sludge produced from the wastewater treatment process can enter the terrestrial environment through the three main disposal routes; incineration, landfill, and recycling (Aubain et al., 2002). Globally, sludge can be treated before disposal either partially (dewatering), fully (stabilisation) or in some cases it is not treated at all. Within Europe, when sewage sludge is used in agriculture, it is treated prior to being applied to land (unless it is injected or worked into soils under certain conditions) to minimise the risk to human health (Council Directive 86/278/EEC, 1986; Department for Environment, Food and Rural Affairs, 2018).

Once sewage sludge has undergone a treatment process (to reduce the water and pathogen content), a material known as a biosolid is produced. This can be used as a replacement for manufactured fertilisers and recycled onto agricultural land. Biosolids contain organic matter essential for healthy, productive soils and their application to land can improve soil quality, structure, drainage, and available water capacity. Biosolids are also a valuable source of nutrients, such as nitrogen and phosphorous, for soils, especially those depleted of nutrients or subject to erosion (Assured Biosolids Ltd, 2019). The recycling of treated sludge, or biosolids, to agricultural land is promoted by many countries. The European Union (EU) and United Kingdom (UK) Government considered it the best environmental option in most circumstances (Assured Biosolids Ltd., 2019). In addition, the recycling of biosolids to land is an integrated part of the circular economy concept (Collivignarelli et al., 2019). Globally only a small proportion of sludge is regulated for reuse as a biosolid, and this can be found mainly in developed countries (Drechsel et al., 2015). Around 77 % of sewage sludge is reused in the EU (either through recycling to land or energy generation) with 50 % of sewage sludge recycled to agricultural soils (European Commission, 2020a, 2020b), however, how much each country recycles varies from 0 %–100 % (European Commission, 2020a, 2020b). Due to concerns regarding the presence of contaminants in biosolids, the majority of all sewage sludge produced is incinerated in the Netherlands (Leslie et al., 2017; Rolsky et al., 2020; Eurostat, 2022). In the UK 96 % of sewage sludge is treated and recycled to land (GOV.UK, 2020), while in China <3 % is recycled and >80 % is mismanaged or improperly dumped (Drechsel et al., 2015; Yang et al., 2015).

1.3. Microplastics in sewage sludge and biosolids

In Europe, the main sources of nitrogen input to agricultural soils are manufactured inorganic mineral fertilisers (40 %), followed by livestock manure (17 %) while biosolids represent <2 % of nitrogen inputs to soils (Misselbrook et al., 2019). As well as containing beneficial nutrients, sludge and biosolids can contain pollutants including heavy metals, pathogens, and emerging contaminants such as microplastics (Aubain et al., 2002). It is suggested the recycling of biosolids to land can create a pathway for microplastic contamination of agricultural soils, with soils acting as a likely reservoir of microplastic pollution (Hurley et al., 2018a, 2018b).

Despite these concerns, there is a need for robust and comparable research on the concentration, composition, and diversity of microplastics entering the environment, while research on the fate and behaviour of microplastics in biosolids and subsequent concentrations in soils are lacking (Horton et al., 2017). There is limited knowledge of the pollution pathways of microplastics in biosolids, levels of exposure and risks to terrestrial habitats and human health (Mahon et al., 2017b). Consequently, there is a lack of sufficient evidence to assess the comparative risk of recycling biosolids to land compared to the benefits of resource recycling from a regulatory perspective (Assured Biosolids Limited, 2018; Nicholson et al., 2018).

Further research is required to determine the characteristics, concentration, and fate of microplastics in biosolids (Assured Biosolids Limited, 2018), as a source of microplastic pollution to agricultural soils. Further understanding of the fate and behaviour of microplastics in agricultural soils is required, and their subsequent mobilisation and transfer to nearby waterbodies (Horton et al., 2017; House of Commons Environmental Audit Committee, 2016).

1.4. Aim of review

The aim of the study was to conduct a systematic review of the literature and provide a summary of the current state of knowledge on the presence, concentration, characteristics, and distribution of microplastics in sewage sludge globally, and identify evidence gaps and areas requiring future research. An overview of the current extent of microplastic pollution in sewage sludge and biosolids, provided by data gathered from an extensive collation of a wide range of research published globally, will present details on biosolids as a potential environmental exposure pathway for terrestrial microplastic pollution and associated risks. To the best of our knowledge, an extensive systematic search such as this has not previously been performed.

A systematic literature review presents a meticulous summary of all primary research in response to a question and assists in establishing the state of existing knowledge of a topic. It involves a detailed and transparent plan and search strategy, with the aim to reduce bias by identifying, selecting, and analysing all relevant studies on a particular topic. Selected studies are synthesised, and data or findings are objectively presented. A systematic review is based on a peer-review protocol so that it can be replicated (Bettany-Saltikov, 2012; Clarke, 2011; Uman, 2011).

1.5. Research questions

The main research question the review aims to address is:

- What is the current state of knowledge regarding the presence, concentrations, characteristics, and distribution of microplastics in sewage sludge and biosolids globally, and can a robust data set be produced from data obtained from research studies, to provide evidence on the concentration of microplastics in sludge and biosolids?

In addition to the main research question, we aim to establish further detailed knowledge by addressing the following questions:

- What role do biosolids play in acting as a pathway for microplastics to enter the terrestrial environment during biosolids recycling?
- Is the presence of microplastics in sewage sludge and biosolids a significant source of microplastics to the terrestrial environment?

2. Method

The literature search process was based on methods by Koelmans et al. (2019) and Foley et al. (2018). Two different databases were searched: Science Direct and Web of Science. A set of criteria were developed to assist the search for all relevant articles and research. Full criteria statements are presented in Supplementary Information (S2).

2.1. Search process

To ensure no relevant studies were missed, a two-step search process was performed.

1. The first step involved searching the databases through the 'Advanced Search' option and using specific keywords. The title and abstract were reviewed for each result and the article was accepted if it met the established criteria. If it was unclear whether the article met the criteria from the title and abstract search alone, a more thorough search of the article's contents was performed.
2. The second step was performed on the reference list of all the accepted articles, if the title looked appropriate, the article was searched and the abstract and content reviewed if required. The article was accepted if it met the criteria. This step was repeated on all reference lists of new articles accepted from previous reference searches until no new articles were discovered. This repeated reference search is known as 'backward snowballing' (Wohlin, 2014).

The search was performed using a set of strings in each data base:

1. *Science Direct*: In 'Advanced Search' the following search was performed for all years up until present in the 'Title, abstract or author-specific keywords' search; "('microplastics' OR "PLASTICS") AND ('SLUDGE' OR 'sewage' OR 'BIOSOLIDS')".
2. *Web of Science*: In 'Advanced Search' the following search was performed for all years up until present; Title = ("microplastics" OR "plastics") AND Title = ("sludge" or "sewage" or "wastewater" or "biosolids").

2.2. Abstract and reference screening results

A search was carried out in August 2020. Following the keyword search, Science Direct and Web of Science delivered 272 and 111 hits, respectively. Twenty-nine articles were accepted based on the articles meeting the criteria set out in the supplementary information. Following the reference search, another seven articles were accepted generating a total of thirty-six articles accepted following both searches. In March 2022, the search was updated, and an additional twenty-nine new papers were accepted following a new database and reference list search.

2.3. Data extraction

All accepted articles were read thoroughly, and relevant information and data were extracted for further analysis to address the questions and aims. If results from a study were reported as a bar graph (rather than exact results), data mining software was used to extract the data from the graph. In this case, Automeris Web Plot Digitizer (Version 4.3) was used to extract underlying numerical data from graphs (Rohatgi, 2020).

2.4. Data synthesis

All data on microplastic concentration in different sludge samples were collated together in Table 1, a more detailed table including additional relevant information such as microplastic characteristics, sample location, sludge treatment before collection and wastewater microplastic concentrations (when reported) is included in the supplementary information, Table S1. The microplastic concentrations in sludge were mostly either reported in number/kg (microplastics/kg) or number/g (microplastics/g), however, a small number of studies reported microplastic number/L (microplastics/L). For comparison purposes during data analysis, the microplastic number/kg was converted to microplastic number/g, while studies that reported values of microplastic number/L were included in the results Table 1 but excluded from the data analysis due to difficulties in comparing the results. Each study was allocated a number for future reference and comparison between the graphs and Table 1.

All graphs, statistical analysis and descriptive statistics were carried out using GraphPad Prism version 9.2.0 (332) for Windows, GraphPad Software, San Diego, California USA, www.graphpad.com. Graphs presenting results with large variations were plotted on a $\log_{10}$ scale, this is for easier visual interpretation. Due to the large range in reported values between studies, which span multiple orders of magnitudes, a logarithmic graph better presents data on one graph and improves transparency (GraphPad Software, LLC, 2021). Scatter plot column graphs were used to present the data for microplastic concentration. For each of these graphs, individual values were plotted to improve transparency and allow for individual interpretation (The Company of Biologists Ltd., 2021), the columns represent the median values, and error bars (when present) present the 95 % confidence levels. The median (geometric mean) was chosen to be represented over the mean because it is a more robust measurement of the central value, being less sensitive to outliers and more suitable for asymmetric distributions (The Company of Biologists Ltd., 2021). The error bars present on the graphs depict the 95 % confidence level, this is recommended (over the standard error mean) to show a more realistic summary of variation (The Company of Biologists Ltd., 2021). The 95 % confidence levels also allow visible interpretation of statistical difference, if the error bars do not overlap, this indicates statistical difference (The Company of Biologists Ltd., 2021).

3. Results and discussion

The data on the microplastic concentration and characteristics were gathered from sixty-five studies, spanning twenty-five different countries. The methods adopted for sample collection, processing, and the reporting of data, varied considerably between studies. Microplastics are complex materials originating from many different products and composed of a diverse range of polymers (of different densities) and additives. In addition, the size of microplastics can range from 1 μm to 5000 μm and come in a variety of shapes (Koelmans et al., 2020). Because of this the definition of microplastic differs between studies and is usually subject to methodological limitations. The analytical techniques, and hence size range, adopted by studies varied widely creating discrepancies between data and increasing the complexities in interpreting the data. This creates challenges in assessing the risk posed by microplastics to the environment (Koelmans et al., 2020).

As shown in Table 1, the result reported by Vollertsen and Hansen (2017) (a microplastic concentration of 1.69×10^5 MPs/g dry weight (dw) of sludge), is elevated compared to all other results, being 22× higher than the next highest individual concentration of 7.65×10^3 MPs/g (dw) reported by Horton et al. (2021). The elevated concentration may be down to the (semi-automated) methodology adopted by these studies, such as the use of Fourier Transform–Infrared (FT-IR) spectroscopy combined with a microscope equipped with a Focal Plane Array (which is able to scan a slide and produce a spectral map), or the addition of a cellulose digesting enzyme prior to the digestion process to improve the clarity of samples during analysis. However, the higher microplastic concentrations reported may be influenced by other, unknown, variables. In the study by Vollertsen and Hansen (2017), a relatively low number of microplastic particles (twenty-nine) were identified across five samples from the study, and due to the limited sample size, observed differences could be down to random variability (Vollertsen and Hansen, 2017).

The high microplastic concentration reported by Vollertsen and Hansen (2017) significantly affected the data analysis in this review, by elevating the mean values considerably, because of this, results including and excluding the microplastic concentration identified by Vollertsen and Hansen (2017) were reported.

3.1. Study location

Sixty-five studies spanning twenty-five countries across the world investigated microplastics in sewage sludge, either as the focus of the research or as an additional investigation when researching microplastics in wastewater or environmental samples. Of these, 29 % were carried out in China, 8 % in Spain and 6 % in Australia.

The number of studies per country is visually represented in the map in Fig. 1. Research on microplastics in sewage sludge is more common in China and Europe with a lack of studies in Central and South America, Asia, the Middle East, and Africa. Of all the countries where microplastic research has taken place, 19 % have conducted only one study (this includes many European countries). The lack of studies carried out in some countries provides a limited indication as to whether sludge and biosolids are a significant source of microplastic in the terrestrial environment.

Most studies (nineteen) were carried out in China, where sludge production per capita is low and sludge management is poor, with >80 % of sludge disposed of improperly and only 2.4 % recycled to land (Yang et al., 2015). Formal recycling of sludge as a biosolid for agriculture is generally only documented in developed countries, where data for research purposes is more widely available (including data on sludge production and end usage/disposal). In the drive for sustainable sludge treatment and management options, the recycling of sewage sludge to land as a biosolid is often encouraged by governments and unions (European Commission, 2020a, 2020b). Where sludge reuse is encouraged, there is a rise in concern regarding the presence of emerging contaminants and persistent and toxic chemicals in biosolids. This may lead to implications on the quality of the biosolid regarding the potential environmental and human health risks. The implications and the risk to the bioresource market may be behind the drive in microplastic research in biosolids in these countries (Nicholson et al., 2018; Assured Biosolids Ltd., 2018).

Table 1
All studies identified in the literature search, and microplastic concentration reported. Additional information on country, microplastic size and sludge type is included.

Study number	Study	Country	MP reporting size (μm)	Sludge type	MP concentration in sludge
1	Hongprasith et al. (2020)	Thailand	300–4750	Secondary sludge	103.4 pieces/L
2	Murphy et al. (2016)	Scotland	<5000	Sludge cake	0.8 MPs/g
				Sludge	1.6 MPs/g
3	(Mahon et al., 2017a)	Ireland (REP)	250–4000	Thermally dried sludge	10.012 MPs/g (dw)
					6.504 MPs/g (dw)
					15.396 MPs/g (dw)
					4.197 MPs/g (dw)
				Anaerobically digested sludge	2.743 MPs/g (dw)
					5.156 MPs/g (dw)
				Lime stabilised sludge	14.064 MPs/g (dw)
					10.012 MPs/g (dw)
4	Zhang et al. (2020)	China	200–5000	Dewatered sludge	Avg. 2.53 items/g
				Raw sludge compost	0.353 items/g
				Semi-finished sludge compost	0.708 items/g
				Finished sludge compost	0.246 items/g
5	Sujathan et al. (2017)	Germany	20–500	Return activated sludge	495 particles/g (dw).
6	Wiśniowska et al. (2018)	Poland	109–<5000	Digested sludge	62.6 particles/g (dw)
				Stabilised sludge	15.8 particles/g (dw)
				Stabilised sludge	28.3 particles/g (dw)
				Stabilised sludge	6.7 particles/g (dw)
				Stabilised sludge	27.7 particles/g (dw)
				Stabilised sludge	52.6 particles/g (dw)
7	Brandsma et al. (2013)	Netherlands	1–5000	–	0.76 particles/g (ww)
					0.51 particles/g (ww)
					0.66 particles/g (ww)
8	Lusher et al. (2017)	Norway	>50 μm	Stabilised and dewatered sludge	19.898 particles/g (dw)
				Stabilised and dewatered sludge	8.237 particles/g (dw)
				Stabilised and dewatered sludge	2.475 particles/g (dw)
				Stabilised and dewatered sludge	2.78 particles/g (dw)
				Dewatered raw sludge	2.949 particles/g (dw)
				Stabilised and dewatered sludge	7.966 particles/g (dw)
				Dewatered sludge	1.695 particles/g (dw)
				Dewatered raw sludge	2.915 particles/g (dw)
9	Magnusson and Norén (2014)	Sweden	>300	Dewatered sludge	16.7 MPs/g (dw)
10	Van Echelpoel et al. (2014)	Belgium	>15	–	289 plastics/g (dw)
11	Vollertsen and Hansen (2017)	Denmark	20–500	Anaerobically digested sludge	169,000 particles/g (dw)
12	Chen et al. (2020)	China	<300–5000	Dewatered raw sludge	74 MPs/g (dw)
				Hyperthermophilic composted sludge	41.565 MPs/g (dw)
				Conventional thermophilic composted sludge	70.783 MPs/g (dw)
13	Li et al. (2019a)	China	–	Dewatered sludge	13.787 particles/g (dw)
				Dewatered sludge	15.08 particles/g (dw)
				Dewatered sludge	37.463 particles/g (dw)
14	Corradini et al. (2019)	Chile	>8	Dewatered, dried sludge (end product)	34 particles/g (dw)
15	Edo et al. (2020)	Spain	25–5000	Anaerobically digested (wet) sludge	314 MPs/g (dw)
				Heat dried anaerobically digested sludge	302 MPs/g (dw)
16	Talvitie et al. (2017)	Finland	100–>3000	Anaerobically digested (wet) sludge	76.3 ML particles/g
				Dried/dewatered sludge	186.7 ML/particles/g
17	Mintenig et al. (2017)	Germany	<500–>500	Drained sludge	1 MP/g (dw)
					8.22 MP/g (dw)
					3.162 MPs/g (dw)
					5.1 MPs/g (dw)
					24 MPs/g (dw)
					13.75 MPs/g (dw)
18	Raju et al. (2020)	Australia	>1.5–>1000	Waste activated sludge (primary & secondary sludge)	7.91 MPs/L
19	Jiang et al. (2020)	China	20–5000	Rag sludge (does not enter WwTWs)	24.2 particles/g
				Raw sludge	5.8 particles/g
				Return activated sludge	36.3 particles/g
				Sludge cake	46.3 particles/g
20	Xu et al. (2020)	China	>5	Anaerobically digested dehydrated sludge	Avg. 4.044 particles/g (dw)
21	Leslie et al. (2017)	Netherlands	–	–	0.51 particles/g (ww)
					0.76 particles/g (ww)
					0.66 particles/g (ww)
22	Hayany et al. (2020)	Morocco	<500–>2000	Fresh sludge	40.5 particles/g (dw)
				Dewatered sludge	36 particles/g (dw)
23	Lv et al. (2019)	China	>25–>500	Excess raw sludge	0.72 MP/L
				Excess raw sludge	4 MP/L
24	Li et al. (2018)	China	>37	Dewatered sludge	Avg. 22.7 particles/g (dw). Range 1.6–56.4 particles/g
25	Naji et al. (2021)	Iran	3–5000	–	6.57 MPs/g (dw)
					5.57 MPs/g (dw)
26	Lares et al. (2018)	Finland	<250–>5000	Activated sludge	23.0 MPs/g (dw)
				MBR sludge	27.3 MPs/g (dw)
				Digested sludge	170.9 MPs/g (dw)

(continued on next page)

Table 1 (*continued*)

Study number	Study	Country	MP reporting size (μm)	Sludge type	MP concentration in sludge
27	Gies et al. (2018)	Canada	>1	Primary sludge	14.9 MPs/g
				Secondary sludge	4.4 MPs/g
28	Bretas Alvim et al. (2020)	Spain	150–5000	Activated sludge	112 MPs/g (dw)
29	Q. Li et al. (2019)	China	2–5000	–	Avg. 8.7 items/g
30	Van den Berg et al. (2020)	Spain	50–1000	–	73 MPs/g
					4.83 MPs/g
					21.84 MPs/g
					22.65 MPs/g
31	Kazour et al. (2019)	France	<20–>500	–	16.13 suspected MPs/g (dw)
32	Magni et al. (2019)	Italy	10–5000	Recycled activated sludge	113 MPs/g (dw)
33	Liu et al. (2019)	China	20–5000	Raw activated sludge (primary + secondary)	240.3 MPs/g (dw)
34	Crossman et al. (2020)	Canada	>1.6	Biosolid	Avg. 14.1 MPs/g (dw)
					Avg. 8.68 MPs/g (dw)
35	Carr et al. (2016)	USA	–	WRP 1: Return Activated sludge	0.05 MPPs/L
				WRP 1: Primary tank skimmings	5 MPP/g
				WWTP: Primary and secondary tank skimmings	4 MPPs/g
				WWTP: Biosolid	1 MPP/g
36	Lee and Kim (2018)	Korea	106 > 300	Dehydrated sludge cake	14.895 MPs/g
				Dehydrated sludge cake	9.475 MPs/g
				Dehydrated sludge cake	13.2 MPs/g
37	Rasmussen et al. (2021)	Sweden	>10	Digested sludge	Avg. 1413 MPs/g (dw)
38	X. Ren et al. (2020)	China	8–1000	Dewatered sewage sludge	220 particles/g (dw)
39	P.J. Ren et al. (2020)	China	80–5000	Dewatered digested sludge	2.92 MP/g
40	Tagg et al. (2022)	Germany	>100	Sewage sludge	97.66 MPs/g (dw)
41	Ziajahromi et al. (2021)	Australia	>25	Primary sludge	Avg. 31.1 particles/g (dw)
				Activated sludge	Avg. 41.3 particles/g (dw)
				Digested sludge	Avg. 52.1 particles/g (dw)
42	Yang et al. (2021a)	China	1–5000	Primary sludge	10.12 No./g (dw)
				Final dewatered sludge	1.02 No./g (dw)
43	Wei et al. (2022)	China	<5000	Preliminary sludge	Avg. 0.4915 n/g
				Primary sludge	Avg. 0.0595 n/g
				Secondary (cyclic activated sludge) sludge	Avg. 0.0275 n/g
44	Okoffo et al. (2020)	Australia	<5000	Biosolid	75.3 mg/g (dw)
45	Li et al. (2022)	China	–	Raw sludge	7.6 n/g (dw)
				Anaerobically digested sludge	7.5 n/g (dw)
				Raw sludge	11.1 n/g (dw)
				Thermally dried sludge	10.6 n/g (dw)
				Raw sludge	4.2 n/g (dw)
				Hydrolysed sludge	5.4 n/g (dw)
				Composting sludge	7.2 n/g (dw)
					1.7 n/g (dw)
					1.9 n/g (dw)
					1.9 n/g (dw)
				Fully composted sludge	1.5 n/g (dw)
46	Vardar et al. (2021)	Turkey	<2000	Biosolids	32 MPs/g
47	Schell et al. (2022)	Spain	>50	Final biosolid	7 MPs/g (dw)
48	Harley-Nyang et al. (2022)	England	50–5000	Raw sludge	107.5 MPs/g (dw)
				Thickened	50.2 MPs/g (dw)
				Anaerobically digested sludge	180.7 MPs/g (dw)
				Secondary digested sludge	286.5 MPs/g (dw)
				Anaerobically digested biosolid	97.2 MPs/g (dw)
				Dewatered raw sludge	74.7 MPs/g (dw)
				Lime stabilised biosolid	37.7 MPs/g (dw)
49	Yuan et al. (2022)	China	–	S1; Preliminary sludge, S2; primary sludge, S3; secondary sludge, S4; Excess sludge, S5; dehydrated sludge cake	S1; 6.74 MP/g (dw), S2; 11.04 MP/g (dw), S3; 6.78 MP/g (dw), S4; 6.32 MP/g (dw), S5; 13.06 MP/g (dw). Avg. 7.4 MPs/g (dw)
				S1; preliminary sludge, S2; primary/excess sludge after cyclic activated sludge technology tank, S3; secondary/excess sludge, S4; tertiary sludge, S5; final dehydrated sludge	S1; 6.31 MP/g (dw), S2; 19.47 MP/g (dw), S3; 10.34 MP/g (dw), S4; 7.69 MP/g (dw), S5; 14 MP/g (dw). Avg. 11.38 MPs/g (dw)
				S1; preliminary sludge, S2; primary sludge, S3; secondary/excess sludge, S4; secondary sludge, S5; final dehydrated sludge	S1; 12.52 MP/g (dw), S2; 15.63 MP/g (dw), S3; 13.79 MP/g (dw), S4; 9.84 MP/g (dw), S5; 22.36 MP/g (dw). Avg. 12.902 MPs/g (dw)
				S1; preliminary sludge, S2; secondary sludge, S3; tertiary sludge, S4; thickened sludge, S5; dehydrated sludge	S1; 9.21 MP/g (dw), S2; 13.33 MP/g (dw), S3; 5.88 MP/g (dw), S4; 12.04 MP/g (dw), S5; 21.25 MP/g (dw). Avg. 8.84 MPs/g (dw)
				S1; preliminary sludge, S2; secondary sludge, S3; secondary sludge, S4; raw sludge S5; dehydrated sludge	S1; 16.62 MP/g (dw), S2; 17.91 MP/g (dw), S3; 14.53 MPs/g (dw), S4; 13.04 MPs/G (dw), S5; 29.66 MPs/g (dw). Avg. 16.21 MPs/g (dw)
50	Tadsuwan and Babel (2022a)	Thailand	50–5000	Raw return activated sludge	26.3 particles/g (dw)
51	Tadsuwan and Babel (2022b)	Thailand	50–5000	Raw secondary sludge	8.12 particles/g (dw)

Table 1 (*continued*)

Study number	Study	Country	MP reporting size (μm)	Sludge type	MP concentration in sludge
52	Chand et al. (2022)	Denmark	>10	Dewatered sludge	810 N/g (dw)
53	Hayany et al. (2020)	Morocco	–	Fresh sludge	40.5 MPs/g
				Dewatered sludge	36 MPs/g
54	Ragoobur et al. (2021)	Mauritius	>250	Raw secondary sludge	10.9 particles/g (ww)
				Raw primary sludge	2.6 particles/g (ww)
				Raw secondary sludge	8.49 particles/g (ww)
55	Yang et al. (2021b)	China	21–4996	Fresh sludge	441 MPs/g (ww)
				Fresh sludge	291 MPs/g (ww)
				Mixed Sludge	239 MPs/g (ww)
				Mixed sludge	111 MPs/g (ww)
				Heat dried biosolids	224 MPs/g (ww)
56	Pittura et al. (2021)	Italy	30–5000	Excess fresh sludge	1.67 MPs.gTS^{-1}
				Activated fresh sludge	5.3 MPs.gTS^{-1}
				Final anaerobically digested sludge	4.7 MPs.gTS^{-1}
57	Tang et al. (2020)	China	20–5000	–	13.4 number/L
					63.4 number/L
58	Zhang et al. (2021a)	China	68–>900	Dewatered sludge	12.73 particles/g (dw)
59	Zhang et al. (2021b)	China	–	Dehydrated dewatered sludge	6.91 items/g (dw)
				Dehydrated dewatered sludge	
				Dehydrated dewatered sludge	2.19 items/g (dw)
				Dehydrated dewatered sludge	0.23 items/g (dw)
60	Salmi et al. (2021)	Finland	20–5000	Raw primary sludge	1560 MPs/L
				Return activated sludge	142 MPs/L
				Anaerobically digested sludge	102 MPs/L
				Dewatered anaerobically digested sludge	9379 MPs/g (dw)
61	Okoffo et al. (2020)	Australia	–	–	Range 0.1–9.6 mg/g (dw). Median 0.7 mg/g (dw).
62	Horton et al. (2021)	England	25–178	Advanced anaerobically digested sludge	7652 MPS/g (dw)
				Advanced anaerobically digested sludge	500 MPs/g (dw)
				Advanced anaerobically digested sludge	2600 MPs/g (dw)
				Conventionally digested sludge	2062.8 MPs/g (dw)
				Lime stabilised sludge	2541 MPs/g (dw)
63	Hernández-Arenas et al. (2021)	Spain	>300	–	17.8 particles/g (dw)
					27.8 particles/g (dw)
					47.13 particles/g (dw)
64	Chand et al. (2021)	Sweden	10–5000	Primary sludge/grease	311 number/g (dw)
				Thickened primary + secondary sludge	4080 number/g (dw)
				Digested sludge	6360 number/g (dw)
65	Petroody et al. (2021)	Iran	>37	Primary sludge	214 microplastics/g (dw)
				Secondary/activated sludge	206 microplastics/g (dw)
				Thickened sludge	200 microplastics/g (dw)
				Aerobically digested sludge	238 microplastics/g (dw)
				Dewatered aerobically digested sludge	129 microplastics/g (dw)

dw = dry weight, ww = wet weight MBR = membrane bioreactor ML = micro-litter, n/g = number/g.

3.2. Microplastic concentration in sewage sludge

As evident from Table 1, the microplastic concentrations varied widely between studies from a study mean of 0.193 MPs/g (Zhang et al., 2020) to 1.69 × 10^5 MPs/g (Vollertsen and Hansen, 2017). A comparison of the values for microplastic concentration in sludge for each study is presented as a graph in Fig. 2 (only studies reporting microplastic number/g or microplastic number/kg are included in this graph, with number/kg converted to number/g for comparison purposes). The majority of microplastic concentrations are reported as values per dry weight (dw) of sludge, while one study reported values per wet weight (ww) (Yang et al., 2021b). Some studies did not indicate whether microplastic concentration was reported as being per dry weight or wet weight (Zhang et al., 2020; Murphy et al., 2016; Talvitie et al., 2017; Jiang et al., 2020; Gies et al., 2018; Q. Li et al., 2019; Van den Berg et al., 2020; Carr et al., 2016; Lee and Kim, 2018; P.J. Ren et al., 2020; Wei et al., 2022; Vardar et al., 2021).

The number of individual measurements (or data points) varied between studies (depending on the number of samples collected and whether this was reported as individual values or a mean value) from one value (an average of twenty individual sludge samples) (Vollertsen and Hansen, 2017) to eleven different values reported in one study (Li et al., 2022). Yuan et al. (2022) collected samples from five different locations, from five different WwTWs, contributing a total of twenty-five values. For ease of data handling, the mean for each treatment works was calculated from this study and used in the data analysis. Due to the varying ways in which each study was conducted, data analysis was performed on the mean values for each study when required.

The values for each study are indicated in Fig. 2 (individual points), along with the median value (bar). When including the study by Vollertsen and Hansen (2017), the maximum value was 1.69 × 10^5 MPs/g, the median was 22.41 MPs/g and the mean value of the sixty-five study means was 3.12 × 10^3 MPs/g. When excluding the study by Vollertsen and Hansen (2017), the max study value was 3.58 × 10^3 MPs/g, the median was 22.1 MPs/g and the overall mean was 208.3 MPs/g. The high concentrations reported by Vollertsen and Hansen (2017) produced an elevated mean value but did not influence the median to such an extent.

Many variables could influence microplastic concentrations including; the characteristics of the wastewater (and microplastic concentration in influent), serving population size and characteristics (demographic and economic status), catchment and local environment characteristics, sewage collection system type, weather conditions, wastewater treatment processes, sludge treatment process, sample location, sample collection procedure, analytical procedures, quality of data reporting, quality control, temporal/seasonal changes etc. This makes identifying a single, influential variable affecting microplastic concentration in sludge more complex. Variations in reported microplastic concentrations may be because of the varying temporal and spatial distribution of microplastics in sludge which can

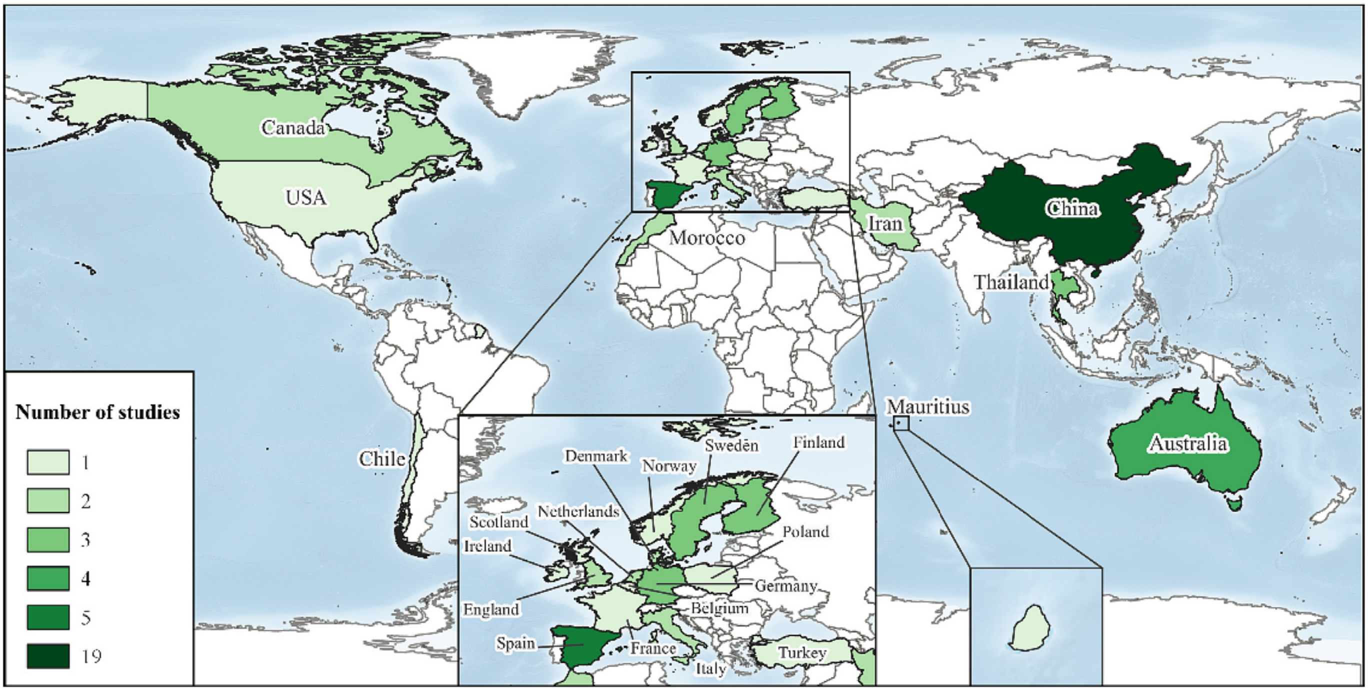

Fig. 1. Number of studies per country.

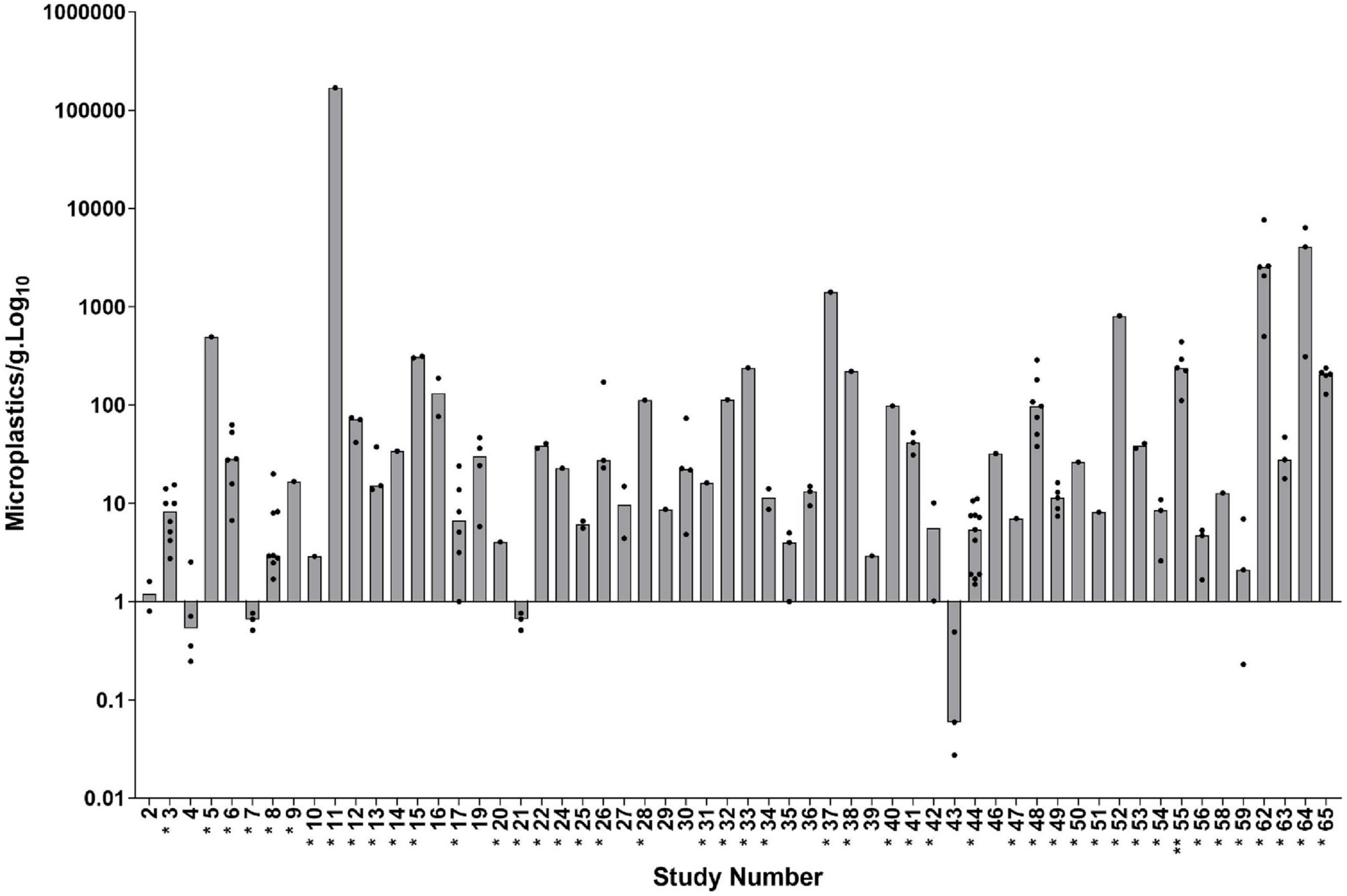

Fig. 2. Microplastic concentration in sludge and biosolid samples.
* Values reported as dry weight of sludge
**Values reported as wet weight of sludge
If not indicated with a ‘*’ or ‘**’, the study did not state if microplastics were reported per wet weight or dry weight of sludge.

occur within treatment works, between samples collected locally, and between locations. Grab sampling is the most widely adopted sampling technique but represents a relatively small quantity when compared to the volume of sludge produced (Salmi et al., 2021). There is a high spatial-temporal variation in microplastic concentrations in wastewater and hence sludge (Blair et al., 2019) increasing the complexity of obtaining comparable datasets between studies and across locations. Currently, no data on spatial-temporal variations of microplastics in sludge exist.

The reported treatment of sludge before sample collection and the type of sludge sample collected also varies widely between studies; from untreated raw primary/secondary/activated sludge (Gies et al., 2018; Liu et al., 2019; Ragoobur et al., 2021; Wei et al., 2022; Tadsuwan and Babel, 2022a; Tadsuwan and Babel, 2022b), to partially treated or dewatered sludge (Li et al., 2018; Magnusson and Norén, 2014; Zhang et al., 2021a; Chand et al., 2022; P.J. Ren et al., 2020; X. Ren et al., 2020). This may influence the microplastic concentration and characteristics recorded (Mahon et al., 2017a), as well as the extraction method employed, and the efficiency of extraction. The sludge type is indicated in Table 1, while the location and type of treatment experienced prior to sample collections are indicated in Table S1.

The presence of microplastics in sewage sludge arises from the capture and retention of microplastics during the wastewater treatment process, with microplastics being detected in orders of magnitude higher in sludge samples than numbers found in wastewater (Gatidou et al., 2019). Despite the large range in microplastic concentrations reported in sewage sludge, it has been suggested that only a small fraction of microplastics removed from the wastewater stream, are actually accounted for in the sewage sludge, with up to 96 % of microplastics entering WwTWs remaining undetected in sludge samples with the use of current methodologies (Koutnik et al., 2021).

3.3. Sludge treatment

There have been suggestions that sludge treatment (biological, chemical and heat treatment required to treat sludge to reduce risk to human health) may influence the microplastic concentration and characteristics. In some instances, microplastic characteristics were found to differ depending on the sludge treatment applied. Microplastics in alkaline-stabilised sludge are shorter, more brittle and exhibit erosion (Zubris and Richards, 2005) and possess a more shredded, flaked appearance, compared to microplastics in sludge subjected to alternative treatments (thermal drying and anearobic digestion) (Mahon et al., 2017a). These characteristics were thought to arise from elevated pH, temperature, and mechanical mixing within the process (Zubris and Richards, 2005). In addition, differences in microplastic characteristics have been observed between those in wastewater and those captured in sludge (Vollertsen and Hansen, 2017) with a significantly lower number of nylon fibres present in anaerobically digested sludge samples compared to wastewater (from the same treatment plant). Anaerobic digestion could promote the break-up of the microplastics into smaller particles which are not easily detected through the methodologies employed, whilst biological degradation of the Nylon particles could be decreasing the concentration of the microplastics in the sludge compared to the wastewater (Vollertsen and Hansen, 2017). Uncertainties arise as to whether this difference is down to random variability or due to the small sample size of the study.

A difference between the microplastic concentrations in sludge having undergone different treatments (anaerobic digestion, thermal drying, and lime stabilisation) has been observed. A significantly lower number of microplastics have been found in sludge having undergone anaerobic digestion compared to thermal drying at the same site (Mahon et al., 2017a). The same study found the average number of microplastics at the anaerobically digested sites was lower than that of thermal drying and lime stabilisation sites (Mahon et al., 2017a). The lime stabilisation processes (high pH, temperature, and mechanical mixing) could result in an elevated number of smaller particles identified in the matrix (lime stabilised samples contained a significantly higher number of smaller particle sizes compared to anaerobic digestion and thermal drying). Pre-treatment samples were not taken during this study against which to compare, and further investigation into the biological breakdown of microplastic within anaerobic digestion is needed.

Harley-Nyang et al. (2022) collected samples across the whole sludge treatment stream from one wastewater treatment works and from two different biosolid products. They identified a difference in microplastic concentration across the treatment stream from 286.5 MPs/g following secondary digestion in open lagoons, to 37.7 MPs/g in limed stabilised sludge. The lime stabilised sludge, one of the biosolids produced at the works, had a lower microplastic concentration compared to the digested sludge cake, 97.2 MPs/g. This is in contradiction to results published by Mahon et al. (2017a), who found a lower microplastic concentration in anaerobically digested sludge (average 3.9 MPs/g) compared to lime stabilised sludge (12 MPs/g). Comparing the two studies proves difficult with different limits of detection adopted, and extraction methods employed. Harley-Nyang et al. (2022) did advise caution when interpreting their results, despite samples collected along the whole sludge treatment stream, they were collected in one day. This did not consider the retention time of the sludge treatment process which totals 30 days (sludge at the end of the anaerobically digested treatment stream is 30 days older than sludge at the start).

Discrepancies across the literature occur with contradictory findings suggesting there is no significant difference in microplastic abundance in samples from sludge treated by anaerobic digestion and lime stabilisation (Hurley et al., 2018a, 2018b). Elevated numbers of microplastics were not observed in anaerobically digested sludge compared to alkaline stabilised or composted sludge (Zubris and Richards, 2005) or activated sludge (Lares et al., 2018). Hyperthermophilic composting was found to reduce microplastic concentration by 43.7 % compared to the conventional thermophilic composting treatment (4.5 % reduction) (Chen et al., 2020) with the reduction of microplastics following hyperthermophilic composting processes deemed 'significant' by the authors, suggesting an excellent removal of microplastics (Chen et al., 2020). This is not a widely used method for the treatment of sludge, with only three studies identified in the review reporting on composting as a sludge treatment process, and only one reporting specifically on hyperthermophilic composting.

The artificial environment of wastewater treatment works can increase the degradation and fragmentation of microplastics, changing the physiochemical properties by increasing the surface area and enhancing sorption characteristics (Teuten et al., 2007). Changes in microplastic characteristics during wastewater treatment and sludge treatment processes can cause variations in the adsorption potential of microplastics for metal pollutants and possibly enhance their adsorption capacity (Q. Li et al., 2019). The rougher surfaces (resulting from erosion and oxidative degradation) of sludge-based microplastics can increase the surface area and increase potential adsorption sites. Fragmentation of particles increases the surface area to volume ratio, thus increasing adsorption rates. Uneven and rough surfaces, enhanced by sludge treatment processes such as lime stabilisation, allow microorganisms, chemical and heavy-metal contaminants (all potentially present in wastewater and sludge) to concentrate on the surface thus potentially increasing toxicity. Smaller-sized microplastics may increase bioavailability via ingestion (Li et al., 2018; Mahon et al., 2017b), therefore, in ecological terms, particle number and size are more important compared to mass, whereas mass can be used to determine treatment efficiencies in wastewater treatment plants (Vollertsen and Hansen, 2017).

Data collected from the systematic review was analysed to identify if sludge treatment influenced the microplastic concentration (data from studies reporting dry weight only (excluding Vollertsen and Hansen (2017)) were included). A comparison was made between the treatment of sludge and the reported microplastic concentration (in microplastics/g dry weight of sludge) (Fig. 3). The sludge treatment was classified as, '*raw sludge*' (untreated sludge), '*dewatered/thickened raw sludge*' (sludge that has undergone a thickening/dewatering process only) and '*stabilised/further treatment*' (sludge that has undergone a form of stabilisation or additional treatment to dewatering/thickening). The median values for the 'raw

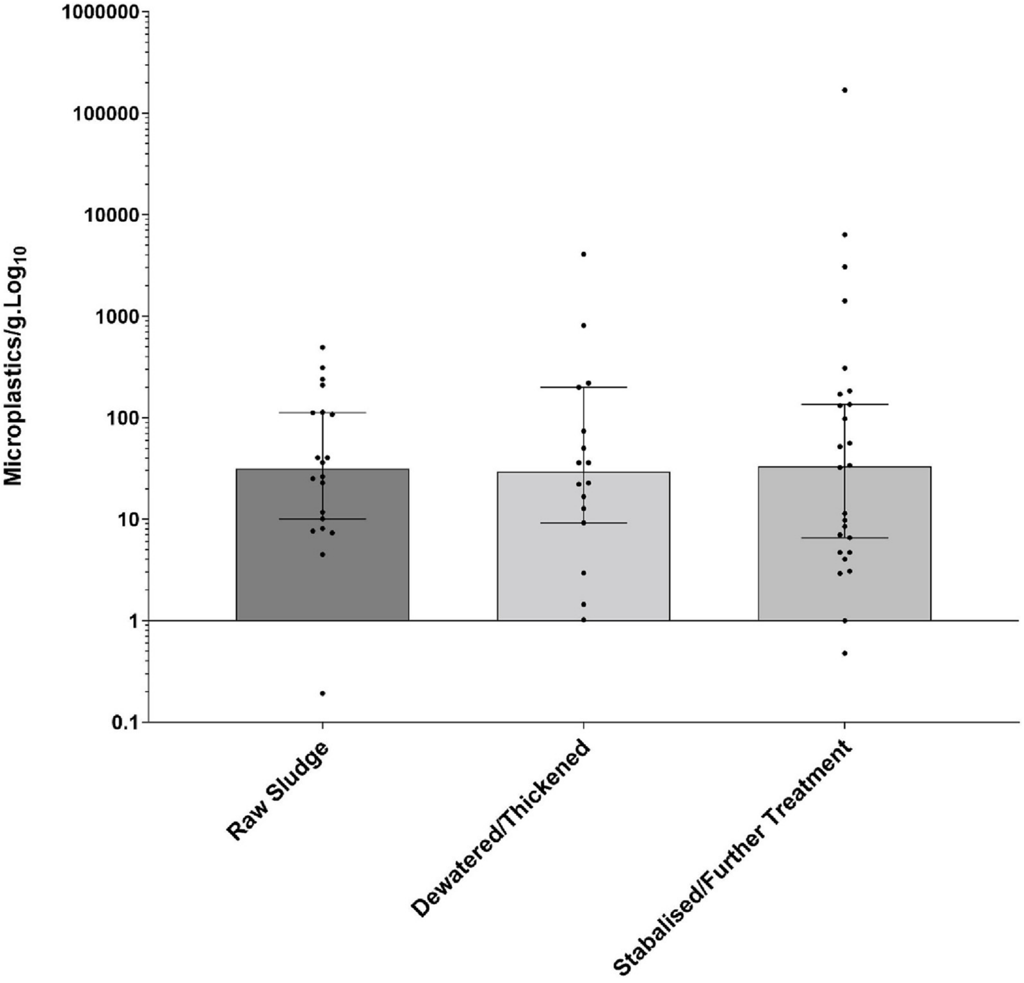

Fig. 3. Microplastic concentration and sludge treatment.

sludge', 'dewatered/thickened sludge' and 'stabilised/further treatment sludge' was 31.24 MPs/g, 29.35 MPs/g and 33.17 MPs/g respectively.

The difference in median values between the three groups is small while the difference between values within each group is large, with a range of 4.95×10^2 MPs/g, 4.08×10^3 MPs/g and 1.69×10^5 MPs/for 'raw sludge', 'dewatered/thickened sludge' and 'stabilised sludge' respectively. A Kruskal Wallis test indicates no significant difference between the means with a *p*-value of 0.9727, suggesting current data synthesised from the studies identified in the search demonstrates no significant difference in microplastic concentrations in sludge and biosolid samples collected after varying levels of treatment (Fig. 3). It is difficult to compare the different groups due to the variations in how studies were conducted and reported (reporting value, methodology etc.). The extent to which sludge treatment influences microplastic concentration in samples may not be apparent because of additional variables that can influence microplastic concentration (as previously discussed), and the difference in analytical procedures adopted by each study.

3.4. Microplastic concentration in biosolids and soils

By considering the quantity of sludge and biosolids produced, and the number of microplastics reported in sludge and biosolids across literature, the potential microplastic load per year released through the application of biosolids to agricultural can be estimated for each country. Based on the extrapolation of data from the literature search, the number of microplastics potentially entering the soil from the application of biosolids to land is presented for sixteen different countries. The weight of sludge disposed (1000 tonnes (t) per year), and the percentage recycled to land is presented in Fig. 4 along with the mean concentrations of microplastics in sludge (MPs/g) (the mean value for each country was calculated from the studies associated with that country and which reported concentrations per dry weight of sludge). The number of microplastics per 1000 tonnes of sludge and potential load to agricultural soils (MPs/year) is presented in Table 2 (as well as the number of studies the mean microplastic number per gram was calculated form for each particular country).

The United States of America (USA) recycled around 2.09×10^6 tonnes of sludge to land in 2018 (data provided in tons and converted to metric tonnes for comparison with other countries), this is 36 % of the 5.28×10^6 tonnes of sludge (dw) produced annually (Ned et al., 2022). Based on the study carried out in the USA which found 1 microplastic particle/g biosolid (Carr et al., 2016) (it was not stated if this was per dry or wet weight but microplastic concentration was reported for a biosolid), there are potentially 1.00×10^9 microplastics per 1000 tonnes sludge (sludge produced and sludge recycled is generally reported internationally as 'number per 1000 tonnes of sludge'). This equated to a load of 2.09×10^{12} microplastics potentially introduced to agricultural land in 2018 in the USA. The mean microplastic concentration for Spain (based on five studies that reported data as dry weight of sludge) was 97.7 MPs/g (dw) (Bretas Alvim et al., 2020; Edo et al., 2020; Hernández-Arenas et al., 2021; Schell et al., 2022; Van den Berg et al., 2020). Spain annually recycles 1.05×10^6 tonnes of sludge (dw) for agricultural use (data from 2018, (Eurostat, 2022)), potentially releasing 1.03×10^{14} microplastics to land annually. England recycles 8.06×10^5 tonnes of sludge (dw) to land, while Scotland recycles 6.9×10^4 tonnes (dw) for agricultural use. When considering the application of sludge (or biosolids) for England and Scotland, and assuming the studies are representative across the countries,

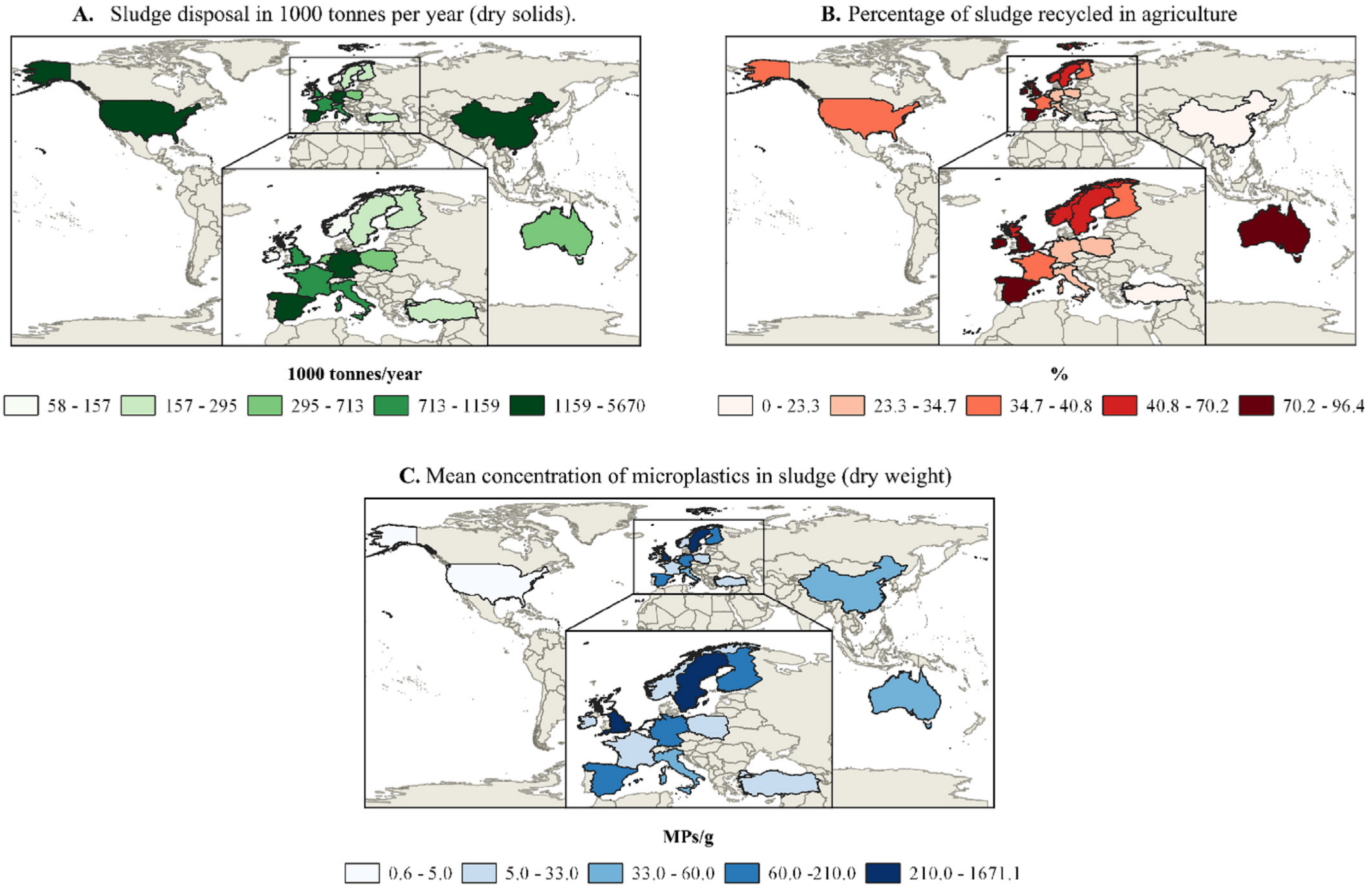

Fig. 4. Distribution of sludge disposal, reuse and microplastic concentrations.

Table 2

Estimated number of microplastics entering the soils of various countries based on data gathered from studies identified in the literature search, and data on sludge production and use.

Country	Sludge recycled to land (ds) (1000 t) b	Date of data	Sludge data reference	Average MPs/t sludge (dw) c	Average MP/1000 t sludge (dw)	MP load to soils * (b*1000) *(c). (per year)	Number of studies
Ireland	51.79	2020	(Eurostat, 2022)	8.51×10^6	8.51×10^9	4.41×10^{11}	1
[a]China	136.08	2013	(Yang et al., 2015)	5.36×10^7	5.36×10^{10}	7.30×10^{12}	19
Germany	423.497	2019	(Eurostat, 2022)	2.01×10^8	2.01×10^{11}	8.50×10^{13}	3
Poland	137.77	2020	(Eurostat, 2022)	3.23×10^7	3.23×10^{10}	4.45×10^{12}	1
Netherlands	0	2020	(Eurostat, 2022)	6.44×10^5	6.44×10^8	0.00	2
Norway	68.74	2020	(Eurostat, 2022)	6.11×10^6	6.11×10^9	4.20×10^{11}	1
Sweden	82.3	2018	(Eurostat, 2022)	1.67×10^9	1.67×10^{12}	1.38×10^{14}	3
Spain	1052.7	2018	(Eurostat, 2022)	9.77×10^7	9.77×10^{10}	1.03×10^{14}	5
France	299	2017	(Eurostat, 2022)	1.61×10^7	1.61×10^{10}	4.82×10^{12}	1
[c]England	806.2896	2022	[a]Sources for England	1.60×10^9	1.60×10^{12}	1.29×10^{15}	2
Scotland	69.03	2021	Scottish Water, 2021	1.20×10^6	1.20×10^9	8.28×10^{10}	1
[a]Australia	254.77	2021	(Australian and New Zealand Biosolids Partnership, 2020)	4.15×10^7	4.15×10^{10}	1.06×10^{13}	4
Finland	64.07	2019	(European Commission, 2020a, 2020b)	1.03×10^8	1.03×10^{11}	6.57×10^{12}	3
Italy	315.6	2010	(WISE Freshwater, 2022)	5.84×10^7	5.84×10^{10}	1.84×10^{13}	2
Belgium	35.95	2020	(Eurostat, 2022)	2.89×10^6	2.89×10^9	1.04×10^{11}	1
Turkey	3.51	2020	(Eurostat, 2022)	3.20×10^7	3.20×10^{10}	1.12×10^{11}	1
[b]USA	2088	2018	(Ned et al., 2022)	[b]1.00×10^6	1.00×10^9	2.09×10^{12}	1

Microplastic concentrations per gram were obtained from data reported in studies identified in the literature search. Of the seventeen countries shown, an average of all individual microplastic values (values given as per dry weight sludge only) was calculated. All values in microplastics/g were converted to microplastics/t (value/g $*$ 10^6). The number of studies for each country is displayed in the last column. The types of sludge samples varied, and values are not always representative of biosolids only.

[a] Data only available on sludge produced rather than sludge disposal.

[b] It was not clear if the value was reported as dry weight or wet weight of sludge, however, data from the study was reported as 'biosolids', so is included in the analysis (assuming dw). Values are recorded as microplastics/g dry weight of sludge unless stated.

[c] Data on sludge production and end-use for England was calculated from data provided by each individual water company and combined to give total values (South West Water Limited, 2022; Anglian Water Services Limited, 2022; Northumbrian Water Group Limited, 2022; Severn Trent Water Limited, 2021; Southern Water Services Limited, 2022; Thames Water Utilities Limited, 2022; United Utilities Group PLC, 2022; Wessex Water Services Limited, 2022; Yorkshire Water Services Limited, 2022). Data from English water companies was provided as 'sludge produced' and 'sludge disposed' (in thousands of tonnes of dry solids), Figures for 'sludge disposed' was used in calculations of microplastic loads to land, this is generally less than 'sludge produced'. This distinction was not always available for other countries and therefore 'sludge produced' was used in calculating microplastic load to land.

it is estimated 1.29×10^{15} and 8.28×10^{10} microplastics could be released to the environment in England (Horton et al., 2021; Harley-Nyang et al., 2022) and Scotland (Murphy et al., 2016) respectively with 1.6×10^{12} microplastics/1000 tonnes of sludge (dw) in England and 1.2×10^{9} MPs/1000 tonnes of sludge (dw) in Scotland. One study from the literature search found 495 particles/g dry weight of (return activated) sludge (Sujathan et al., 2017), and estimated 80×10^{4} microplastic particles per m^2 are entering soils in Lower Saxony (Germany) based on an application rate of 1.6 tons/ha. While in Norway, the average microplastic concentration in sludge samples (from ten different sludge samples) was 6.1 microplastics/g sludge (dw) (Lusher et al., 2017). It was estimated in the study, that over 4.46×10^{11} microplastics were released into the environment through the recycling of biosolids to agricultural land every year (Lusher et al., 2017).

According to the microplastic concentrations obtained from the literature search, and scaling data up to fit country-wide sludge application rates (Table 2) the potential total number of microplastics entering soils from biosolid application could be in the billions or trillions per year.

Based on the extrapolations, England, Sweden, Spain, and Germany are associated with the highest number of microplastics released to terrestrial soils with 1.29×10^{15}, 1.38×10^{14}, 1.03×10^{14} and 8.5×10^{13} respectively potentially entering agricultural soils across each country annually. With respect to the number of microplastics per 1000 tonnes sludge, the top countries are Sweden, England, Germany, Finland, and Spain with 1.67×10^{12}, 1.6×10^{12}, 2.10×10^{11}, $1.03 \times 10^{11}$and 9.77×10^{10} microplastics per 1000 tonnes of sludge, respectively.

According to this analysis, the recycling of biosolids to land can be considered a pathway for the release of a high number of microplastics into terrestrial environments, even when considering the lower values reported for microplastic concentrations. A study conducted in Ireland reported 8.5 microplastics/g sludge (dw), assuming this value represents the microplastic concentration in sludge across Ireland (Mahon et al., 2017a), 8.51×10^{9} microplastics can be found in 1000 tonnes of sludge (dw). When considering the biosolid application rates in Ireland, 3.96×10^{11} microplastics could be released to agricultural soils every year. Apart from the Netherlands (where biosolids are not recycled to agricultural land), Scotland and Belgium are associated with the lowest number of microplastics released, with potentially 8.28×10^{10} and 1.04×10^{11} released respectively to soils each year. The USA is ranked second (after the Netherlands) for average microplastics per 1000 tonnes of sludge, at 1.00×10^{9} MPs/1000 tonnes, followed by Scotland, with 1.20×10^{9} MPs/1000 tonnes, and then Belgium, with 2.89×10^{9} MPs/1000 tonnes. In terms of sludge recycled to land, Belgium ranks third lowest (after Netherlands and Turkey), while Scotland ranked seventh lowest. The values are given as a total number of microplastics released to soils and the unit area of application is not specified, therefore a lack of unites (e.g., microplastic number or mass per area of land) results in data not being easily interpretable. Besseling et al. (2019) produced a mass per particle factor of 5 μg/particle based on the weight of an average microplastic particle on shores. When converting the number of microplastics to mass, based on this conversion factor, we can produce more interpretable data; potentially 6430 tonnes, 687.7 tonnes, 514 tonnes and 424 tonnes of microplastics are spread onto

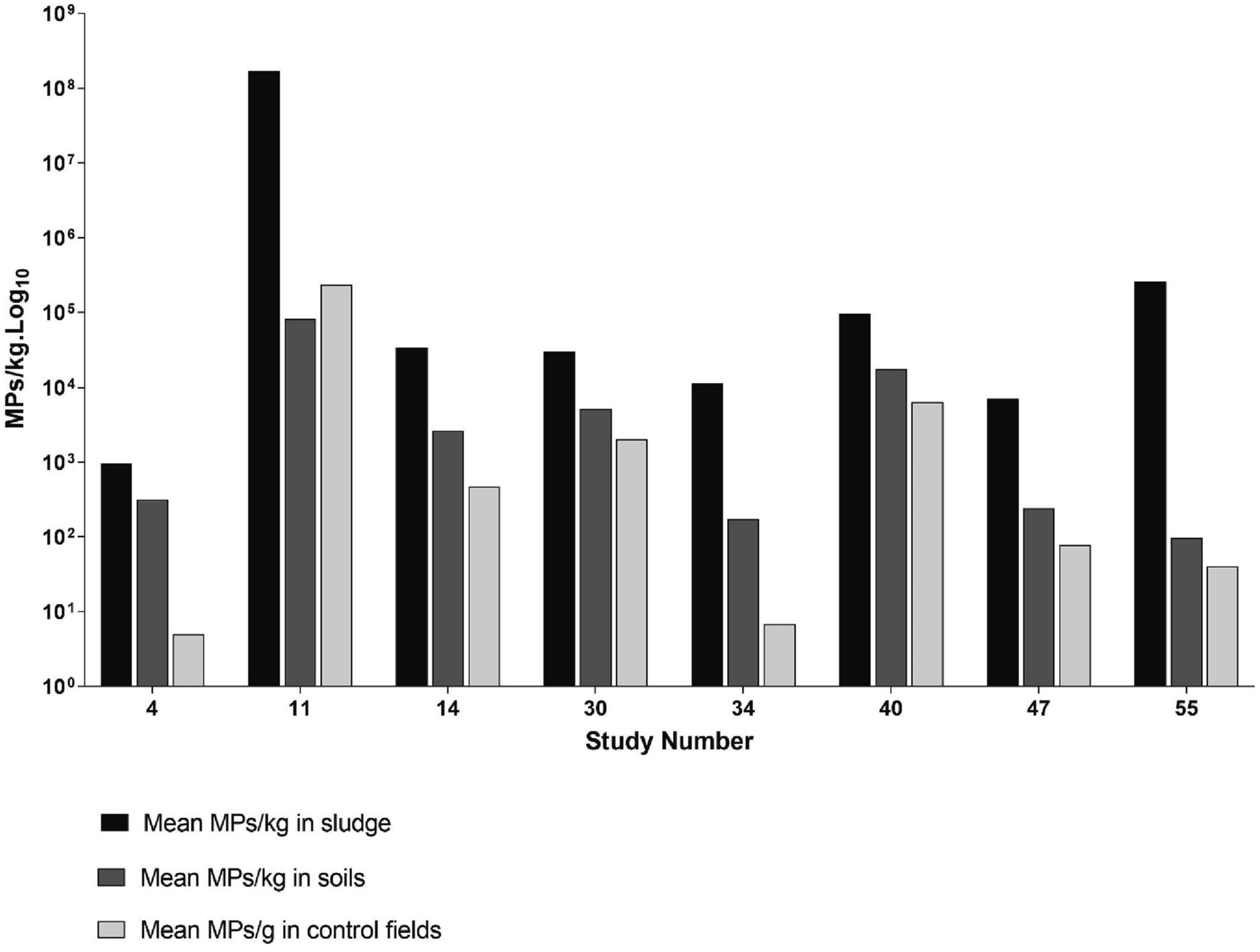

Fig. 5. Microplastic concentration in sludge and soils.

agricultural lands in England, Sweden, Spain and Germany respectively. The total land area in the UK used for agriculture (utilised agriculture land) is 9.05 million hectares (Defra, 2019), about 37 % of the total UK land mass of 24.2 million hectares (Ministry of Defence, 2020). Biosolids are recycled to 1.3 % of the UK's agricultural land (Assured Biosolids Limited, 2021), which equates to 1.18 × 10^5 hectares. Assuming the mean microplastic load to soils for Scotland and England (6.43 × 10^2 trillion) is representative of the load to soils for the whole UK, it is estimated 5.5 billion microplastics could be released per hectare of land.

A Spearman's rank correlation was performed based on sludge recycled to land, and average microplastic concentration per tonne, to identify if there is a relationship between each variable and microplastic numbers in soils. A strong positive relationship was identified between both average microplastic concentration per tonne of sludge ($r_s = 0.94$) and of sludge recycled to land ($r_s = 0.82$), with little difference between the degree of strength of the relationships. However, microplastic concentration per tonne may vary greatly on methodologies and microplastic detection size limits adopted within each study. The highest microplastic concentration per tonne is associated with Sweden and England (average 2.44 × 10^3MPs/g (dw) and 1.35 × 10^3 MPs/g (dw)). These higher values are associated with studies adopting semi-automated FT-IR imaging methods, Rasmussen et al. (2021) and Chand et al. (2021) conducted studies in Sweden, and both employed a very similar method for the identification and confirmation of microplastics. Both adopted the use of Agilent Cary 620 FT-IR microscope equipped with a 128 × 128-pixel Focal Plane Array, permitting an average microplastic concentration of 1.4 × 10^3 MPs/g (dw) and 3.58 × 10^3 MPs/g (dw) respectively. While Horton et al. (2021) adopted a semi-automated method where-by all microplastics present on a filter paper were identified, quantified and polymers analysed simultaneously using a μFTIR spectrometer, thereby identifying an average of 3.07 × 10^3 MPs/g (dw).

Results from the country-wide extrapolations demonstrate that high numbers of microplastics are entering the terrestrial environment through the recycling of biosolids and given the concerns regarding the risks posed by the presence of microplastics in sludge and soils, a more comprehensive data set is required to accurately evaluate the human and environmental risk.

3.4.1. Soil microplastic concentration attributed to biosolid recycling

Eight studies reported microplastic concentration in agricultural soils as well as sludge and biosolids (Corradini et al., 2019, 2020; Schell et al., 2022; Tagg et al., 2022; Van den Berg et al., 2020; Vollertsen and Hansen, 2017; Yang et al., 2021b; Zhang et al., 2020), the average microplastic concentration in soil samples, sludge/biosolid samples and control field samples (per kg) for each study are displayed in Fig. 5. Vollertsen and Hansen (2017) reported a higher concentration of microplastics in control fields compared to fields that had a history of biosolid application. Their results indicate an increase of 35 % in control field soil microplastic concentration (Vollertsen and Hansen, 2017). This higher value for control fields may be due to the low number of microplastic particles detected in soil samples (a total of 13 and 24 microplastics detected in soil samples with and without sludge). Apart

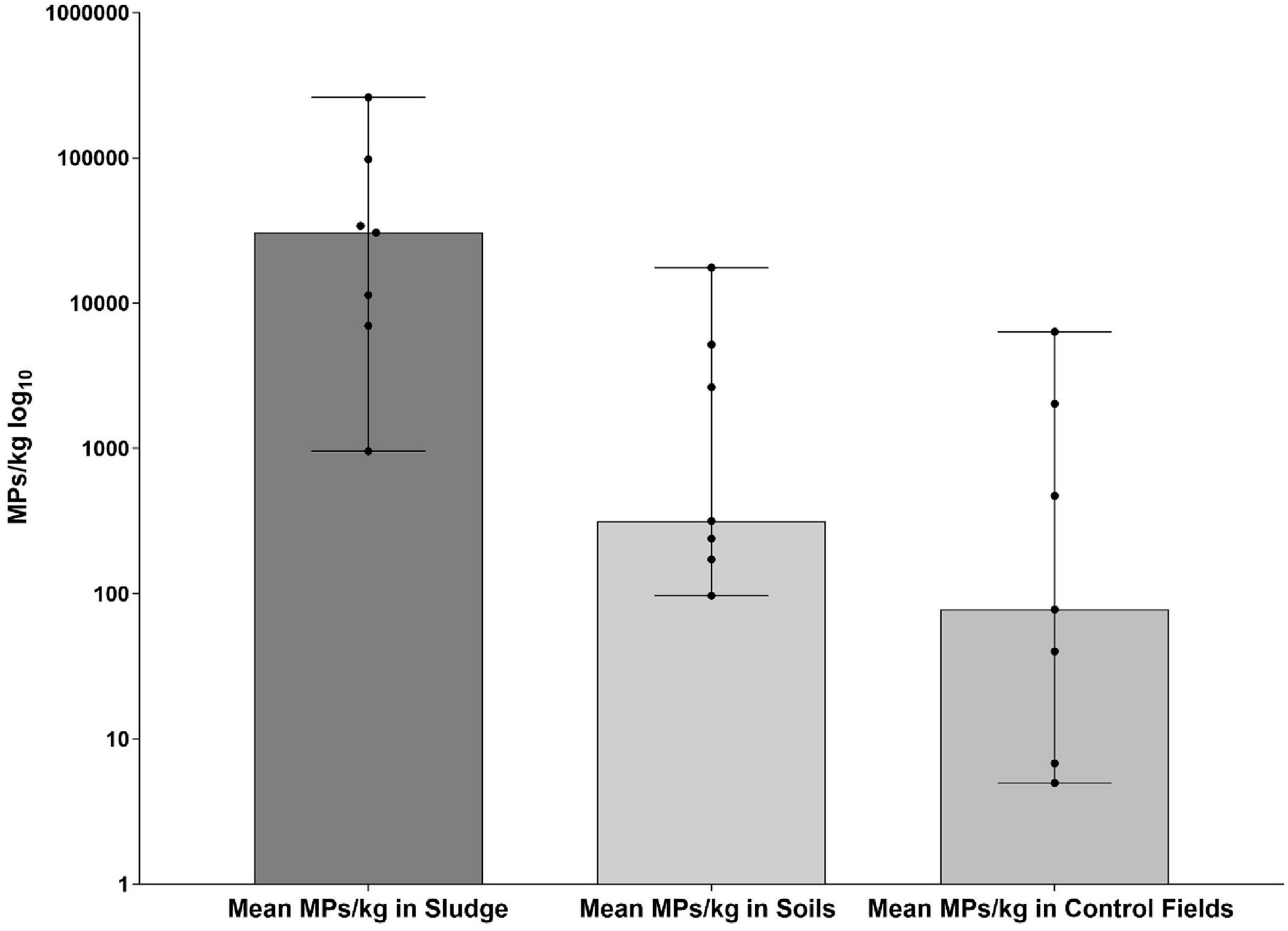

Fig. 6. Comparison of microplastic concentration in sludge and soils. A comparison of the microplastic concentrations in sludge samples, soil samples taken from fields with a history of biosolid application, and control fields (excluding outlying data from Vollertsen and Hansen (2017)). Individual vales, median and 95 % confidence intervals are displayed in the Figure. The overlap between the 95 % confidence intervals demonstrates there is no significant difference between the different categories.

from the study by Vollertsen and Hansen (2017), all other studies reporting on microplastics in soils as well as the sludge, reported a higher mean microplastic concentration in soil samples collected from fields with a history of biosolid application compared to control fields. With values ranging from 97 MPs/kg (Yang et al., 2021b) to 1.76 × 10^4 MPs/kg (Tagg et al., 2022) (or 8.2 × 10^4 MPs/kg including Vollertsen and Hansen (2017)) with a mean of 3.74 × 10^3 MPs/kg (or 1.35 × 10^4 MPs/kg including data from Vollertsen and Hansen (2017)) reported in fields with a history of sludge application. The microplastic concentration in control fields (fields with no history of biosolid application) was generally lower, with values ranging from 6.8 MPs/kg (Crossman et al., 2020) to 6.36 × 10^3 MPs/kg (Tagg et al., 2022) (or 2.36 × 10^5 MPs/kg including Vollertsen and Hansen (2017)) with a mean value of 1.29 × 10^3 MPs/kg (or 3.06 × 10^4 MPs/kg including Vollertsen and Hansen (2017)). The Mann-Whitney test comparing microplastic concentrations in fields with and without a history of sludge application found differences were not significant (with *P*-values of 0.208 or 0.328 including results from Vollertsen and Hansen, 2017). The data on microplastic concentration in sludge, soils with a history of sludge application, and control fields are presented in Fig. 6. It must be noted the data comparing microplastic concentration in sludge and soils was extracted from studies adopting widely different methodological approaches and from different locations.

The mean microplastic concentration for sludge samples were higher than microplastic concentrations reported in soils for the same study, with a mean of 6.33 × 10^4 MPs/kg (or 2.12 × 10^7 MPs/kg including Vollertsen and Hansen (2017)).

3.4.2. Microplastic fate and behaviour in soils

There is evidence that microplastics accumulate in soils over time and with successive sludge applications (Corradini et al., 2019; Van den Berg et al., 2020; Zhang et al., 2020). Questions regarding the mobility of microplastic fibres vs particles are also raised as the ratio of fibres to particles was higher in soil samples compared to sludge samples (Corradini et al., 2019; Crossman et al., 2020); indicating microplastic particles could have higher mobility in soils compared to fibres, or fibres from other sources (atmospheric) are favourably retained in soil. An eventual decline of microplastic fragments in soils following biosolid application suggests fragments are more readily transported from the soil matrix while fibres are more readily retained (Crossman et al., 2020). The mobilisation (and subsequent fate and behaviour) of microplastics in soils, following biosolid application, is influenced by several different variables and complex relationships including rainfall, time of year of biosolid application, soil characteristics and vegetation cover (Crossman et al., 2020). Saturated soils, those with a higher wet density, were less able to retain microplastics, and a loss of microplastics from soils was observed where run off events (high rainfall events) occurred (Crossman et al., 2020). This suggests biosolids, and hence microplastics, were removed during early runoff events (such as high rainfall events following biosolid application) and lateral movement of microplastics may dominate over vertical movement. Soils exposed to less rainfall, and lower soil saturation, experienced an increase in microplastic concentration in soils following biosolid application, and infiltration of microplastics into deeper soil layers. Microplastics demonstrated vertical movement when biosolids were applied to fields with established crops, suggesting root growth can enhance the vertical movement of microplastics. An increase in microplastic concentration directly following biosolid applications has been observed, with this increase being retained over successive months but moving to deeper soil layers, indicating vertical movement of microplastics over time. This vertical movement dominated in fields where crops were pre-established, which is suggested to provide preferential vertical pathways through root growth and transport of

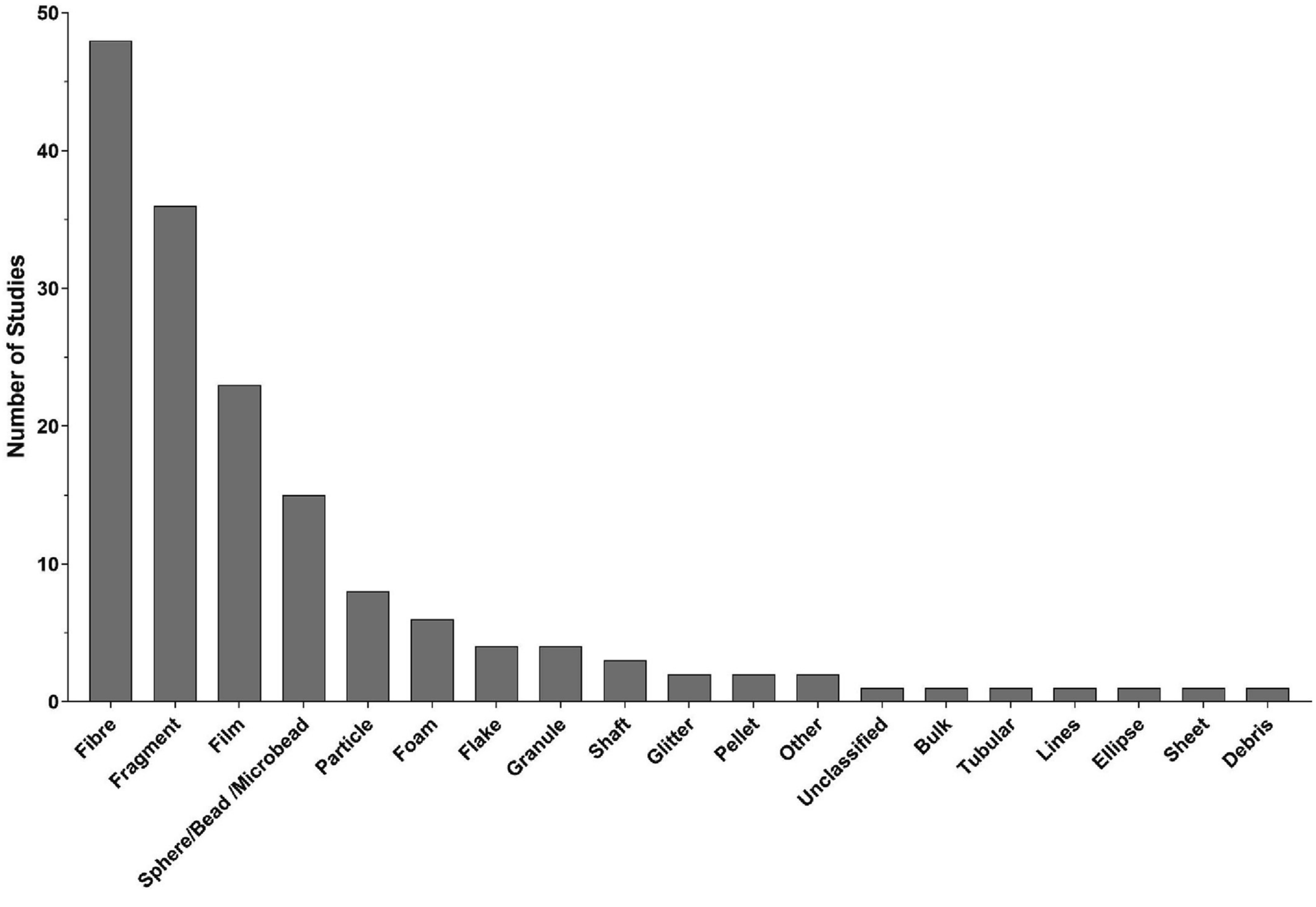

Fig. 7. Frequency of microplastic shape.

microplastics via bioturbation (Crossman et al., 2020). In another case, the initial increase in microplastic concentration in soils directly following biosolid application was observed which was not retained in successive months with a loss of microplastics from the topsoil layer (and no increase in the bottom layer) indicating limited vertical movement of microplastics and removal during runoff events (Crossman et al., 2020).

3.4.3. Summary of microplastics in soils

Understanding microplastic concentration, distribution and fate in soils following activities such as biosolids application assists in the understanding of environmental and ecological risks posed by microplastics. Research carried out on the mobilisation of microplastics in soils following sludge application demonstrates soil can act as a store for microplastics, especially when infiltration occurs. Soils can exhibit a limited capacity to retain microplastics (following biosolid application), especially during times of high surface flushing events. This demonstrates soils do not always act as a sink to microplastics, but rather soils are part of the pathway for microplastic contamination of surface water bodies. Understanding the concentration and mobilisation of microplastics in soils will allow for a greater understanding of the transfer of microplastics in the terrestrial environment and what role biosolid application plays in microplastic pollution of the terrestrial environment.

3.5. Microplastic shape categorisations

Microplastic shape can be used to determine microplastic source and origin (Sun et al., 2019). The presence of fibres may originate from textiles, clothing and carpets and typically represent domestic sources (Li et al., 2018; Lares et al., 2018). One item of clothing can release ~1.9×10^3 fibres/wash (Browne et al., 2011) while the number of fibres potentially lost during a 6 kg load could be as high as 1.38×10^5 for polyester-cotton blend, 4.96×10^5 for polyester and 7.29×10^5 for acrylic (Napper and Thompson, 2016). The presence of microbeads can indicate domestic origins such as personal care and cosmetic products, or industrial origins, where microbeads are used in air blasting (Hurley et al., 2018a, 2018b). The microplastic shape can also influence its removal efficiency from wastewater (Sun et al., 2019) with fibres being more readily retained in sewage sludge with higher proportions of fibres to particles reported (Magnusson and Norén, 2014; Magnusson et al., 2016; Lares et al., 2018). This is not always observed in the data with several studies having identified microplastic particles are the dominant microplastics type in sewage sludge (Liu et al., 2019; Hurley et al., 2018a, 2018b; Magni et al., 2019; Crossman et al., 2020). Microplastics in sludge can accumulate higher concentrations of metals compared to surrounding sewage sludge (X. Li et al., 2019), and microplastic characteristics such as shape can influence this.

Forty-nine studies provided enough data on microplastic type or shape for comparison between studies. Nineteen different descriptions were used to characterise microplastic type and/or shape, Fig. 7. ‘Fibre’ was the most frequently reported classification, with forty-eight studies reporting ‘fibre’ as a microplastic shape (in one study, ‘line’ was reported instead of ‘fibre’ (Magni et al., 2019)), this was followed by ‘fragment’ (thirty-six studies) and ‘film’ (twenty-three studies). Fig. 8 presents the variation in the particle-to-fibre ratio for a number of countries.

3.6. Microplastic polymer

Microplastic confirmation and polymer identification are achieved via chemical analysis, typically through the use of spectroscopic techniques

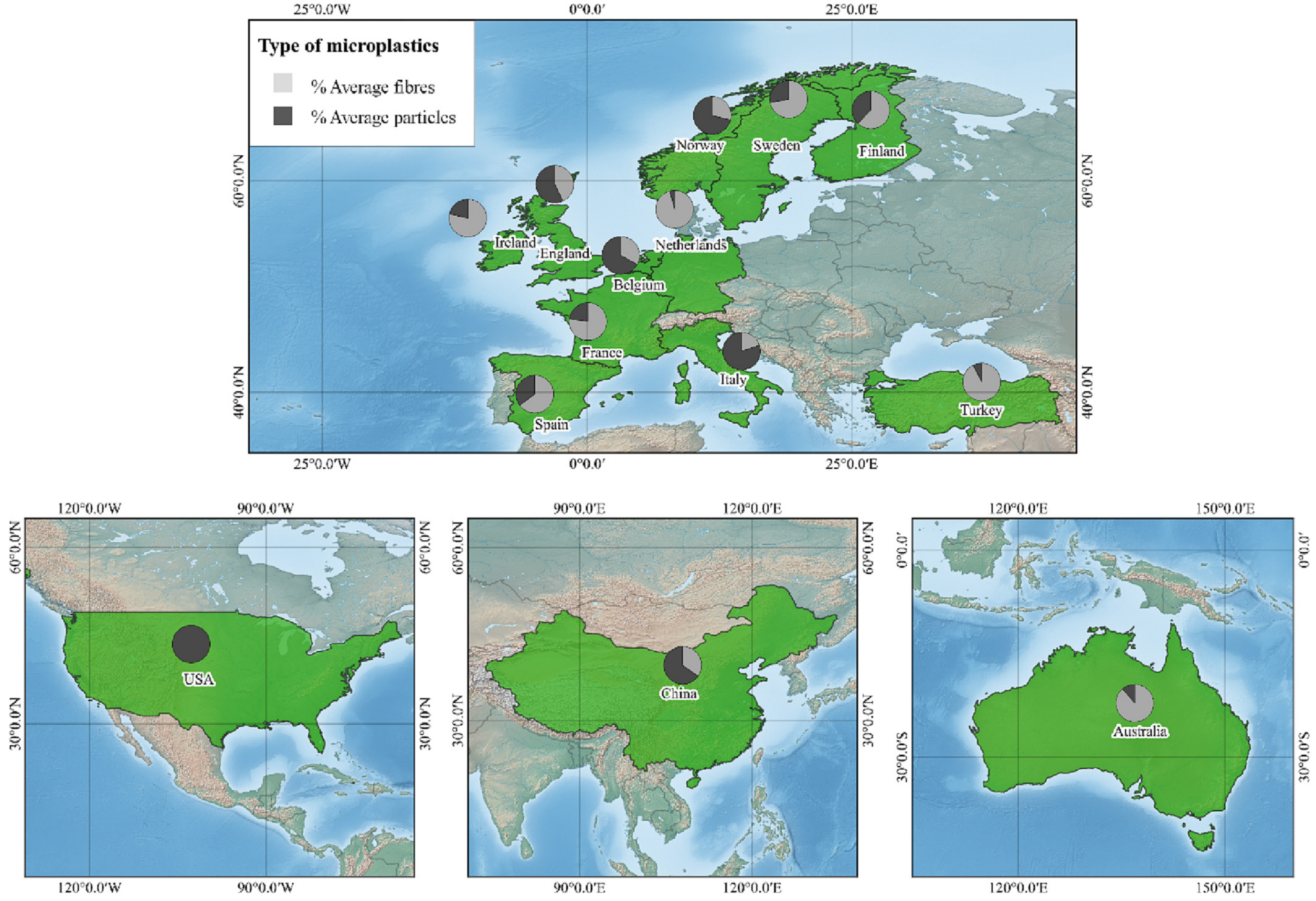

Fig. 8. Ratio of fibre to particle for several countries.

such as μFT-IR or Raman spectroscopy, and less typically through the use of gas chromatography coupled to mass spectrometry (GC–MS), including pyrolysis-GC–MS and thermal extraction desorption-GC–MS (Sun et al., 2019). There are inconsistencies in the way in which polymers were reported between studies; with either the percentage of each polymer identified reported (Hongprasith et al., 2020; Sujathan et al., 2017; Chen et al., 2020; Bretas Alvim et al., 2020), or just the polymer type reported (rather than the extent to which that polymer was identified) (Mahon et al., 2017a; Van Echelpoel et al., 2014; Li et al., 2018; Naji et al., 2021; Vardar et al., 2021; Okoffo et al., 2020; Ziajahromi et al., 2021; Tagg et al., 2022; Rasmussen et al., 2021). In some cases, chemical analysis was not carried out on microplastics found in sludge samples (Wiśniowska et al., 2018; Brandsma et al., 2013; Magnusson and Norén, 2014; Van Echelpoel et al., 2014; Corradini et al., 2019; Talvitie et al., 2017; Leslie et al., 2017). The quality of the chemical analysis, polymer identification and reporting varied widely. For example, in some cases, a small number of microplastics were subsampled for chemical analysis, with subsamples as low as 5.9 % and even 0.6 % of suspected microplastics subjected to chemical analysis (Jiang et al., 2020; Naji et al., 2021). This is lower than the suggested minimum proportion of 10 % of suspected microplastics subjected to chemical analysis (Lusher et al., 2020). A statistical approach conducted to present a theoretical evaluation of the accuracy of the subsample as a function of the size of the population sampled, found that in samples with potentially thousands of particles, accuracy did not increase with increasing sample size(Kedzierski et al., 2019) and 3 % of the subsample size would provide sufficient accuracy (Kedzierski et al., 2019).

Fifty-six studies reported chemical analysis (with varying quality and consistency of reporting). At least forty-three different descriptions/names of polymers were reported. The most frequently reported polymers across the literature are presented in Fig. 9- which portrays the number of studies reporting the different polymers. Polyethylene (PE) was reported in over 90 % of studies followed by polypropylene, reported in 80 % of studies, and polyamide/Nylon, reported in 59 % of studies. These percentages reflect the number of times a polymer was reported, not the extent to which the polymer was reported (this was harder to compare between studies due to the inconsistent reporting of the chemical analysis), so even though PE is the most widely reported polymer, the extent to which it was observed will vary (for example, PE accounted for 47 % of polymers in sludge reported by Mintenig et al. (2017) while it only represented 3.2 % of polymers identified by Kazour et al. (2019)). Fig. 9 presents the top twenty-eight reported polymers only, the category ‘Other’ represents at least another twenty-six polymer names/categories.

The different sources of wastewater (runoff, domestic and industry) can influence the polymer characteristics present in wastewater while identifying polymer type can assist in identifying sources of plastic pollution. Polyethylene can represent a number of possible sources, such as fragments from cleansing scrubs or personal care products (Lares et al., 2018; Sun et al., 2019), packaging (plastic bags, films, bottles etc.) (Vollertsen and Hansen, 2017) and agricultural greenhouses/film (Bayo et al., 2020). Polyester or Nylon (generally found in the form of fibres) can originate from synthetic clothing, textiles, and carpets (Mahon et al., 2017a; Vollertsen and Hansen, 2017). Polypropylene can be found in fibrous material and textiles (Liu et al., 2019), or in food packaging, car parts, electrical goods, films and automobile parts (PlasticsEurope, 2018). Acrylonitrile butadiene styrene (ABS) (Novodur) is a polymer that can originate from the automobile industry and is used in the manufacture of automotive parts including hub caps, tyre manufacture, electro-plates, decorative parts, dashboard components, instrument panels, radiator and bumper grill, mirrors, exterior trim, rear lighting, interior applications (INEOS Styrolution Group, 2016). The presence of these polymers could be entering combined sewerage system from urban water runoff. Similar polymers have been recorded in influent samples, including copolymers acrylonitrile butadiene and ethylene

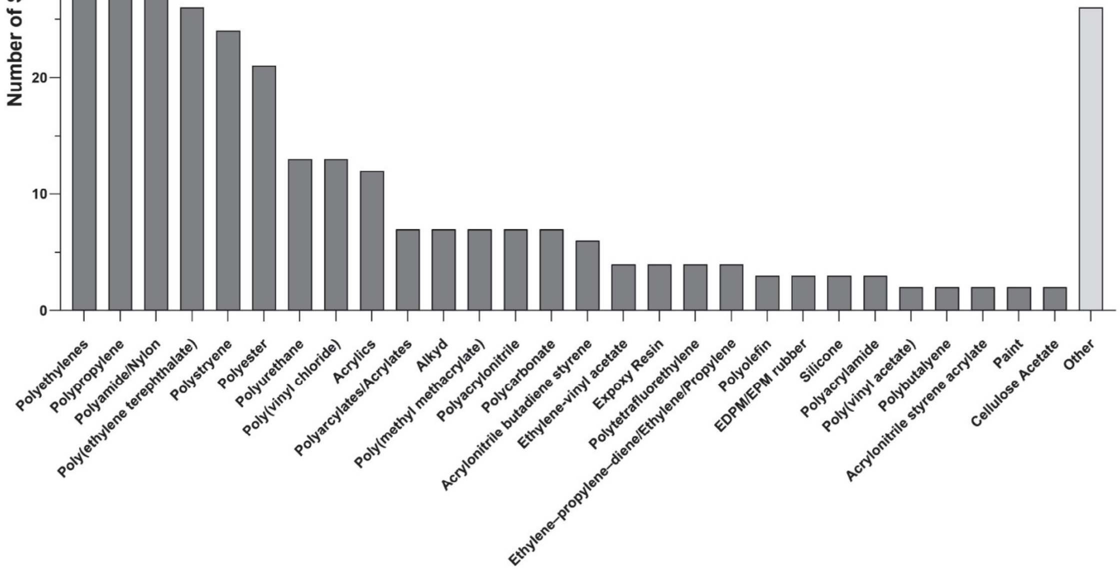

Fig. 9. Most frequently reported polymer.

propylene, as well as polyester and polyethylene being the most abundant polymer identified (Magni et al., 2019).

The data follows the distribution of global polymer demand; polypropylene was the resin in most demand (data from 2016) representing 26 % of the share of demand, while high- and low-density PE together also represented 26 % share of resin demand (Garside, 2020). These two polymers are the top-reported polymers across the studies. Polyethylene terephthalate, poly(vinyl chloride), polystyrene and ABS are also listed as the major polymers in demand worldwide (from 2016 data), all these polymers are widely reported as being present in sludge samples.

4. Conclusion

The review provides a summary of the characteristics of microplastics in sludge and biosolids from a range of different studies. The review presented a summary of the characteristics of microplastics in sludge and biosolids reported across several studies. Domestic sources of microplastics in sludge and biosolids are evident by the dominance of fibres as the most frequently reported microplastic shape reported across several studies. While polymer characteristics suggest a range of sources influencing microplastics in sludge and biosolids, with polymers associated with the automobile industry indicating influences from surface water runoff. The polymers of highest global demand, polyethylene, and polypropylene, were the most frequently reported polymers.

The review established the current state of knowledge of the presence, concentrations, and distribution of microplastics in sludge and biosolids. The mean data on microplastic concentration in sludge and biosolids varied considerably across the sixty-five studies included in the literature search and review: from 0.193 microplastics/g to 1.69×10^5 MPs/g with a median microplastic concentration of 22.4 MPs/g. This variation could be attributed to many different variables influencing microplastic concentration in sludge and subsequent samples, but due to the difference in methodologies and reporting of data between studies, attributing which variables contribute to the difference in microplastic concentration was not possible. Inconsistencies in methodologies and reporting of data across literature have also resulted in a lack of a robust and comparable evidence-based global dataset.

Our data reveals a disproportional spread of research, globally. There is a considerable lack of research in low-income countries which may reflect the lack of formal regulations and reuse of sewage sludge. Nineteen studies on microplastic in sludge were carried out in China, the greatest number of studies attributed to any one country, despite the low percentage of sludge recycled to land in China, while the UK recycles 87 % of sewage sludge and only three studies have been carried out in the UK.

The initial calculations give an idea of the number of microplastics potentially entering agricultural soils each year for different countries. From the data, the recycling of sewage sludge and biosolids to land is a pathway for microplastics to enter the terrestrial environment, especially where large quantities of sewage sludge are recycled to land. In England and Sweden, results indicate 1290 trillion and 138 trillion microplastics respectively could enter agricultural soils annually. Assuming an average weight of 5 μ per particle, this equates to potentially 6430 tonnes and 687.7 tonnes of microplastics released, respectively, to the terrestrial environment per year in these countries. In Spain and Germany, 103 trillion and 85 trillion microplastics could be released every year through biosolids recycling. Further research is needed to identify the total area of agricultural land receiving biosolids in different countries globally, and the subsequent microplastic concentration potentially entering these soils.

The significance of biosolids as a pathway for microplastic contamination of the terrestrial environment compared to other sources (aerial deposition or other agricultural applications such as compost application or anaerobic digestate) is unknown. A higher mean microplastic concentration for fields which had a history of biosolid application was found compared to control fields, however, this difference was not significant.

A complex picture regarding the fate of microplastics in agricultural soils is likely, with mobilisation dependent on soil characteristics, precipitation, crop cover, bioturbation processes etc. indicating that soils are not just a sink for microplastics, but rather microplastics can become mobilised and transferred from the point of biosolid application.

Laws governing the management of sewage sludge can be complicated and often out-of-date when it comes to addressing microplastic pollution. The data presented here add to the evidence base on the contribution of biosolids to overall microplastic pollution. Further research is required on the fate and behaviour of microplastics in soils and the risk to the environment and human health to create a reliable evidence base to support changes in policy.

The recycling of waste as a biosolid is an important element of the circular economy concept, providing a renewable source of nutrients to agricultural fields. However, the increased confirmation and certainty of high numbers of microplastics in biosolids presents a conundrum for the different stakeholders involved in the supply chain (from the water industry/companies to farmers, to consumers). Comparing the risks posed by the presence of microplastics in biosolids, to the benefit of reusing what otherwise would be a waste product, needs to be established and questions need to be asked – despite what we know, does the risk of introducing microplastics pollutants to the terrestrial and freshwater environments outweigh the benefits of recycling biosolids to land? Despite the importance of the recycling of biosolids in terms of closing the loop, the process is ultimately not fully closed, with huge numbers of microplastics pollutants leaking into the environment.

Further research is required on the effects of sludge treatment on microplastic concentration, characteristics, and fate. In addition, comparing microplastic concentrations in biosolids having undergone different treatment processes may assist in assessing the risk the different biosolids pose to environmental health, and identify if biosolid products differ in terms of contribution to microplastic pollution to the terrestrial environment. Subsequently, biosolids products, having undergone different treatment methods, may be more desirable. Further research is required on the fate and behaviour of microplastics in soils and risk to the environment and human health to create a reliable evidence base to support changes in policy, as well as establish solutions to prevent and reduce microplastic pollution of the terrestrial and freshwater environments.

CRediT authorship contribution statement

Daisy Harley-Nyang: Conceptualization, Formal analysis, Investigation, Writing – original draft, Project administration. **Fayyaz Ali Memon:** Writing – review & editing. **Andrea Osorio Baquero:** Formal analysis. **Tamara Galloway:** Writing – review & editing, Supervision.

Data availability

The research data supporting this publication is available from the University of Exeter's institutional repository at doi:10.24378/exe.XXX.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2023.164068.

References

Anglian Water Services Limited, 2022. Annual Performance Report 2022. [Online]Anglian Water Services Limited, Huntingdon Available at: https://www.anglianwater.co.uk/about-us/our-reports/reports-search/ [Accessed 14 December 2022].

Assured Biosolids Limited, 2018. Assured Biosolids Limited - Microplastics Position Statement. [Online]Assured Biosolids Limited, Huntingdon Available at: https://assuredbiosolids.co.uk/wp-content/uploads/2018/07/Microplastics-Position-Statement-27th-July-2018.pdf [Accessed 05 July 2021].

Assured Biosolids Limited, 2019. Biosolids Agricultural Good Practices Guidance Leaflet. [Online]Assured Biosolids Limited, Huntingdon Available at: https://assuredbiosolids.co.uk/wp-content/uploads/2019/01/Biosolids-Agric-Good-Practice-Guidance-January-2019.pdf [Accessed 28 January 2019].

Assured Biosolids Limited, 2021. About Biosolids [Online]. Assured Biosolids Limited, Huntingdon Available at: https://assuredbiosolids.co.uk/about-biosolids/ [Accessed 06 September 2021].

Assured Biosolids Ltd, 2019. Biosolids Agricultural Good Practices Guidance Leaflet. [Online] Assured Biosolids Limited, Huntingdon Available at: https://assuredbiosolids.co.uk/wp-content/uploads/2019/01/Biosolids-Agric-Good-Practice-Guidance-January-2019.pdf [Accessed 28 January 2019].

Aubain, P., Gazzo, A., Le Moux, J., Mugnier, M., Brunet, H., Landrea, B., 2002. Disposal and Recycling Routes for Sewage Sludge; Synthesis Report. [Online]European Commission, Toruń Available at: https://ec.europa.eu/environment/archives/waste/sludge/pdf/synthesisreport020222.pdf [Accessed 14 December 2022].

Australian and New Zealand Biosolids Partnership, 2020. Australian Biosolids Statistics. [Online]Australian Water Association Available at: https://www.biosolids.com.au/guidelines/australian-biosolids-statistics/ [Accessed 11 September 2022].

Bayo, J., Olmos, S., López-Castellanos, J., 2020. Microplastics in an urban wastewater treatment plant: the influence of physicochemical parameters and environmental factors. Chemosphere 238, 124593. https://doi.org/10.1016/j.chemosphere.2019.124593.

Besseling, E., Redondo-Hasselerharm, P., Foekema, E., Koelmans, A., 2019. Quantifying ecological risks of aquatic micro- and nanoplastic. Crit. Rev. Environ. Sci. Technol. 49, 32–80.. https://www.tandfonline.com/doi/full/10.1080/10643389.2018.1531688.

Bettany-Saltikov, J., 2012. How to Do a Systematic Literature Review in Nursing; a Step-by-steo Guide. Open University Press, New York.

Blair, R., Waldron, S., Gauchotte-Lindsay, C., 2019. Average daily flow of microplastics through a tertiary wastewater treatment plant over a ten-month period. Water Res. 163, 114909. https://doi.org/10.1016/j.watres.2019.114909.

Brandsma, S., Nijssen, P., Velzen, M., Leslie, H., 2013. Microplastics in River Suspended Particulate Matter and Sewage Treatment Plants [Online]. IVM Institute for Environmental Studies, Amsterdam Available at: https://www.noordzeeloket.nl/publish/pages/122253/microplastic_river_suspended_particulate_and_sewage_treatment_plants_ivm-vu_2013_5087.pdf [Accessed 14 December 2022].

Bretas Alvim, C., Bes-Piá, M., Mendoza-Roca, J., 2020. Separation and identification of microplastics from primary and secondary effluents and activated sludge from wastewater treatment plants. Chem. Eng. J. 402, 126293. https://doi.org/10.1016/j.cej.2020.126293.

Browne, M., Crump, P., Niven, S., Teuten, E., Tonkin, A., Galloway, T., Thompson, R., 2011. Accumulation of microplastic on shorelines worldwide: sources and sinks. Environ. Sci. Technol. 45 (21), 9175–9179. https://doi.org/10.1021/es201811s.

Carr, S., Liu, J., Tesoro, A., 2016. Transport and fate of microplastic particles in wastewater treatment plants. Water Res. 91, 174–182. https://doi.org/10.1016/j.watres.2016.01.002.

Catarino, A., Kramm, J., Völker, C., Henry, T., Everaert, G., 2021. Risk posed by microplastics: scientific evidence and public perception. Curr. Opin. Green Sustain. Chem. 29, 100467. https://doi.org/10.1016/j.cogsc.2021.100467.

Chand, R., Rasmussen, L., Tumlin, S., Vollertsen, J., 2021. The occurrence and fate of microplastics in a mesophilic anaerobic digester receiving sewage sludge, grease, and fatty slurries. Sci. Total Environ. 798, 149287. https://doi.org/10.1016/j.scitotenv.2021.149287.

Chand, R., Kohansal, K., Toor, S., Pedersen, T., Vollertsen, J., 2022. Microplastics degradation through hydrothermal liquefaction of wastewater treatment sludge. J. Clean. Prod. 335, 130383. https://doi.org/10.1016/j.jclepro.2022.130383.

Chen, Z., Zhao, W., Xing, R., Xie, S., Yang, X., Cui, P., Liao, H., Yu, Z., Wang, S., Zhou, S., 2020. Enhanced in situ biodegradation of microplastics in sewage sludge using hyperthermophilic composting technology. J. Hazard. Mater. 384, 121271. https://doi.org/10.1016/j.jhazmat.2019.121271.

Clarke, J., 2011. What is a systematic review? [online]. Evidence-based Nursing. BMJ Journals, Aukland https://doi.org/10.1136/ebn.2011.0049 Available at:. [Accessed 21 April 2023].

Collivignarelli, M., Abbà, A., Frattarola, A., Miino, M., Padovani, S., Katsoyiannis, I., Torretta, V., 2019. Legislation for the reuse of biosolids on agricultural. Sustainability 11 (6015), 1–22. https://doi.org/10.3390/su11216015.

Corradini, F., et al., 2019. Evidence of microplastic accumulation in agricultural soils from sewage sludge disposal. Sci. Total Environ. 671, 411–420. https://doi.org/10.1016/j.scitotenv.2019.03.368.

Council Directive 86/278/EEC of 12 June 1986 on the protection of the environment, and in particular of the soil, when sewage sludge is used in agriculture. Official Journal of the European Communities. (86/278/EEC) 12 June. Available at: https://eur-lex.europa.eu/eli/dir/1986/278/oj [Accessed 15 December 2022].

Crossman, J., Hurley, R., Futter, M., Nizzetto, L., 2020. Transfer and transport of microplastics from biosolids to agricultural soils and the wider environment. Sci. Total Environ. 724, 138334. https://doi.org/10.1016/j.scitotenv.2020.138334.

Department for Environment, Food and Rural Affairs, 2018. Sewage Sludge in Agriculture: Code of Practice for England, Wales and Northern Ireland. [Online]Department for Environment, Food and Rural Affairs, London Available at: https://www.gov.uk/government/publications/sewage-sludge-in-agriculture-code-of-practice/sewage-sludge-in-agriculture-code-of-practice-for-england-wales-and-northern-ireland [Accessed 01 May 2019] Drechsel, P., Qadir, M. & Wichelns, D., (2015) Wastewater; Economic Asset in an Urbanizing World. New York: Springer.

Department for Environment, Food and Rural Affairs, 2019. Farming Statistics Land Use, Livestock Populations and Agricultural Workforce at 1 June 2019 – England. [Online]Department for Environment, Food and Rural Affairs, York Available at: https://assets.publishing.service.gov.uk/government/uploads/system/uploads/attachment_data/file/868945/structure-jun19-eng-28feb20.pdf [Accessed 14 December 2022].

Drechsel, P., Qadir, M., Wichelns, D., 2015. Wastewater; Economic Asset in an Urbanizing World. Springer, New York.

Edo, C., González-Pleiter, M., Leganés, F., Fernández-Piñas, F., Rosal, R., 2020. Fate of microplastics in wastewater treatment plants and their environmental dispersion with effluent and sludge. Environ. Pollut. 259, 113837. https://doi.org/10.1016/j.envpol.2019.113837.

European Commission, 2020a. Sewage Sludge. [Online]European Commission Available at: https://ec.europa.eu/environment/waste/sludge/ [Accessed 21 January 2021].

European Commission, 2020b. Feedback from: Finnish Water Utilities Association. [Online] European Commission, Helsinki Available at: https://ec.europa.eu/info/law/better-regulation/have-your-say/initiatives/12328-Sewage-sludge-use-in-farming-evaluation/F544631_en [Accessed 11 08 2022].

Eurostat, 2022. Sewage sludge production and disposal from urban wastewater (in dry substance (d.s)).[Online]. Available at:Eurostat https://ec.europa.eu/eurostat/web/environment/water [Accessed 20 January 2021].

Foley, C., Feiner, Z., Malinich, T., Höök, T., 2018. A meta-analysis of the effects of exposure to microplastics on fish and aquatic invertebrates. Sci. Total Environ. 631-632, 550–559.. https://www.sciencedirect.com/science/article/pii/S0048969718307630.

Galloway, T., 2015. Micro- and nano-plastics and human health. In: Bergmann, M., Gutow, L., Klages, M. (Eds.), Marine Anthropogenic Litter. Springer International Publishing AG, Switzerland, pp. 343–366.

Garside, 2020. Distribution of Polymer Demand Worldwide in 2016 by Major Polymer. [Online]Statista, New York Available at: https://www.statista.com/statistics/756728/worldwide-distribution-of-polymer-demand-by-type/ [Accessed 28 January 2021].

Gatidou, G., Arvaniti, O., Stasinakis, A., 2019. Review on the occurrence and fate of microplastics in sewage treatment plants. J. Hazard. Mater. 367, 504–512. https://doi.org/10.1016/j.jhazmat.2018.12.081.

Gies, E., LeNoble, J., Noel, M., Etemadifar, A., Bishay, F., Hall, E., Ross, P., 2018. Retention of microplastics in a major secondary wastewater treatment plant in Vancouver, Canada. Mar. Pollut. Bull. 133, 553–561. https://doi.org/10.1016/j.marpolbul.2018.06.006.

Gov.UK, 2020. Policy Paper: Environment Agency Strategy for Safe and Sustainable Sludge Use. [Online]Environment Agency Available at https://www.gov.uk/government/publications/environment-agency-strategy-for-safe-and-sustainable-sludge-use/environment-agency-strategy-for-safe-and-sustainable-sludge-use [Accessed 20 December 2022].

GraphPad Software, LLC, 2021. Logarithmic Axes. [Online]Dotmatics, San Diego Available at: https://www.graphpad.com/guides/prism/latest/user-guide/logarithmic_axes.htm [Accessed 01 October 2021].

Hansen, A., Vollertsen, J., 2017. Microplastics in Danish Wastewater. Sources, Occurrence and Fate. [Online]Ministry of Environment and Food of Denmark, Environmental Protection Agency, Denmark Available at: http://mst.dk/service/publikationer/publikationsarkiv/2017/mar/mikroplast-i-renseanlaeg/ [Accessed 12 December 2022].

Harley-Nyang, D., Memon, F., Jones, N., Galloway, T., 2022. Investigation and analysis of microplastics in sewage sludge and biosolids: a case study from one wastewater treatment works in the UK. Sci. Total Environ. 823, 153735. https://doi.org/10.1016/j.scitotenv.2022.153735.

Hayany, B., El Fels, L., Quénéa, K., Dignac, M.-F., Rumpel, C., KumarGupta, V., Hafidi, M., 2020. Microplastics from lagooning sludge to composts as revealed by fluorescent staining-image analysis, Raman spectroscopy and pyrolysis-GC/MS. J. Environ. Manag. 275, 111249. https://doi.org/10.1016/j.jenvman.2020.111249.

Hernández-Arenas, R., Beltrán-Sanahuja, A., Navarro-Quirant, P., Sanz-Lazaro, C., 2021. The effect of sewage sludge containing microplastics on growth and fruit development of tomato plants. Environ. Pollut. 268, 115779. https://doi.org/10.1016/j.envpol.2020.115779.

Hongprasith, N., Kittimethawong, C., Lertluksanaporn, R., Eamchotchawalit, T., Kittipongvises, S., Lohwacharin, J., 2020. IR microspectroscopic identification of microplastics in municipal wastewater treatment plants. Environ. Sci. Pollut. Res. 27, 18557–18564.. https://link.springer.com/article/10.1007/s11356-020-08265-7.

Horton, A., Spurgeon, D., Lahive, E., Svendsen, C., 2017. Microplastics in freshwater and terrestrial environments; evaluating the current understanding to identify knowledge gaps and future research. Sci. Total Environ. 586, 127–141. https://doi.org/10.1016/j.scitotenv.2017.01.190.

Horton, A., Cross, R., Read, D., Jürgens, M., Ball, H., Svendsen, C., Vollertsen, J., Johnson, A., 2021. Semi-automated analysis of microplastics in complex wastewater samples. Environ. Pollut. 268, 115841. https://doi.org/10.1016/j.envpol.2020.115841.

House of Commons Environmental Audit Committee, 2016. Environmental Impacts of Microplastics; Fourth Report of Sessions 2016–2017. [Online]House of Commons, London Available at https://publications.parliament.uk/pa/cm201617/cmselect/cmenvaud/179/179.pdf [Accessed 21 January 2019].

Hurley, R., Lusher, A., Olsen, M., Nizzetto, L., 2018a. Validation of a method for extracting microplastics from complex, organic-rich, environmental matrices. Environ. Sci. Technol. 52 (13), 7409–7417. https://doi.org/10.1021/acs.est.8b01517.

Hurley, R., Nizzetto, L., Lusher, A., Vogelsang, C., Olsen, M., 2018b. Mapping Microplastics in Sludge. Norwegian Institute for Water Research, Oslo Available at: https://www.miljodirektoratet.no/publikasjoner/2018/april-2018/mapping-microplastics-in-sludge/ Available at:.

INEOS Styrolution Group, 2016. Driving Design Innovation. [Online]INEOS, Frankfurt Available at: https://www.ineos-styrolution.com/INTERSHOP/static/WFS/Styrolution-Portal-Site/-/Styrolution-Portal/en_US/industry/automotive/Styrolution_Automotive_2016.pdf [Accessed 07 May 2020].

Jiang, J., Wang, X., Ren, W., Cao, G., Xie, G., Xing, D., Liu, B., 2020. Investigation and fate of microplastics in wastewater and sludge filter cake from a wastewater treatment plant in China. Sci. Total Environ. 746, 141378. https://doi.org/10.1016/j.scitotenv.2020.141378.

Kazour, M., Terki, S., Rabhi, K., Jemaa, S., Khalaf, G., Amara, R., 2019. Sources of microplastics pollution in the marine environment: importance of wastewater treatment plant and coastal landfill. Mar. Pollut. Bull. 146, 608–618. https://doi.org/10.1016/j.marpolbul.2019.06.066.

Kedzierski, M., Villain, J., Falcou-Préfol, M., Kerros, M., Henry, M., Pedrotti, M., Bruzaud, S., 2019. Microplastics in Mediterranean Sea: a protocol to robustly assess contamination characteristics. PLoS One 2, 12. https://doi.org/10.1371/journal.pone.0212088.

Koelmans, A., Nor, N., Hermsen, E., Kooi, M., Mintenig, S., De France, J., 2019. Microplastics in freshwaters and drinking water: critical review and assessment of data quality. Water Res. 155, 410–422. https://doi.org/10.1016/j.watres.2019.02.054.

Koelmans, A., Redondo-Hasselerharm, P., Nor, N., Kooi, M., 2020. Solving the nonalignment of methods and approaches used in microplastic research to consistently characterize risk. Environ. Sci. Technol. 54 (19), 12307–12315. https://doi.org/10.1021/acs.est.0c02982.

Koutnik, V., Koutnik, S., Leonard, J., DePrima, F., Cao, S., Hoek, E., Mohanty, S., 2021. Unaccounted microplastics in wastewater sludge: where do they go? ACS EST Water 1, 1086–1097. https://doi.org/10.1021/acsestwater.0c00267.

Lares, M., Ncibi, M., Sillanpää, M., Sillanpää, M., 2018. Occurrence, identification and removal of microplastic particles and fibres in conventional activated sludge process and advanced MBR technology. Water Res. 133, 236–246. https://doi.org/10.1016/j.watres.2018.01.049.

Lee, H., Kim, Y., 2018. Treatment characteristics of microplastics at biological sewage treatment facilities in Korea. Mar. Pollut. Bull. 137, 1–8. https://doi.org/10.1016/j.marpolbul.2018.09.050.

Leslie, H., Brandsma, S., Velzen, M., Vethaak, A., 2017. Microplastics en route: field measurements in the Dutch river delta and Amsterdam canals, wastewater treatment plants, North Sea sediments and biota. Environ. Int. 101, 133–142. https://doi.org/10.1016/j.envint.2017.01.018.

Li, W., Tse, H., Fok, L., 2016. Plastic waste in the marine environment: a review of sources, occurrence and effects. Sci. Total Environ. 566-567 (1), 333–349. https://doi.org/10.1016/j.scitotenv.2016.05.084.

Li, X., Chen, L., Mei, Q., Dong, B., Dai, X., Ding, G., Zeng, E., 2018. Microplastics in sewage sludge from the wastewater treatment plants in China. Water Res. 142, 75–85. https://doi.org/10.1016/j.watres.2018.05.034.

Li, Q., Wu, J., Zhao, X., Gu, X., Ji, R., 2019a. Separation and identification of microplastics from soil and sewage sludge. Environ. Pollut. 254 (part B), 113076. https://doi.org/10.1016/j.envpol.2019.113076.

Li, X., Mei, Q., Chen, L., Zhang, H., Dong, B., Dai, X., He, C., Zhou, J., 2019b. Enhancement in adsorption potential of microplastics in sewage sludge for metal pollutants after the wastewater treatment process. Water Res. 157, 228–237. https://doi.org/10.1016/j.watres.2019.03.069.

Li, X.-Y., Liu, H.-T., Wang, L.-X., Guo, H.-N., Zhang, J., Gao, D., 2022. Effects of typical sludge treatment on microplastics in China—characteristics, abundance and micro-morphological evidence. Sci. Total Environ. 826, 154206. https://doi.org/10.1016/j.scitotenv.2022.154206.

Liu, W., Yuan, W., Di, M., Li, Z., Wang, J., 2019. Transfer and fate of microplastics during the conventional activated sludge process in one wastewater treatment plant of China. Chem. Eng. J. 362, 176–182. https://doi.org/10.1016/j.cej.2019.01.033.

Liu, Y., Wang, B., Pileggi, V., Chang, S., 2022. Methods to recover and characterize microplastics in wastewater treatment plants. Case Studies Chem. Environ. Eng. 5, 100183. https://doi.org/10.1016/j.cscee.2022.100183.

Lusher, A., et al., 2017. Mapping Microplastics in Sludge. Norwegian Institute for Water Research, Oslo.

Lusher, A., Brate, I., Munno, K., Hurley, R., Welden, N., 2020. Is it or isn't it: the importance of visual classification in microplastic characterization. Appl. Spectrosc. 74 (9), 1139–1153. https://doi.org/10.1177/0003702820930733.

Lv, X., Dong, W., Zuo, Z., Liu, Y., Huang, X., Wu, W.-M., 2019. Microplastics in a municipal wastewater treatment plant: fate, dynamic distribution, removal efficiencies, and control strategies. J. Clean. Prod. 225, 579–586. https://doi.org/10.1016/j.jclepro.2019.03.321.

Magni, S., Binelli, A., Pittura, L., Avio, C., Torre, C., Parenti, C., Gorbi, S., Regoli, F., 2019. The fate of microplastics in an Italian Wastewater Treatment Plant. Sci. Total Environ. 652, 602–610. https://doi.org/10.1016/j.scitotenv.2018.10.269.

Magnusson, K., Norén, F., 2014. Screening of Microplastic Particles in and Down-stream a Wastewater Treatment Plant. IVL Swedish Environmental Research, Stockholm. http://www.diva-portal.org/smash/get/diva2:773505/fulltext01.pdf.

Magnusson, K., Jorundsdottir, H., Noren, F., Lloyd, H., Talvitie, J., Setala, O., 2016. Microlitter in Sewage Treatment Systems; a Nordic Perspective on Waste Water Treatment Plants as Pathways for Microscopic Anthropogenic Particles to Marine Systems. Nordic Council of Ministers, Stranden. https://norden.diva-portal.org/smash/get/diva2:923936/FULLTEXT01.pdf.

Mahon, A., O'Connell, B., Healy, M., O'Conner, I., Officer, R., Nash, R., Morrison, L., 2017a. Microplastics in sewage sludge; effects of treatment. Environ. Sci. Technol. 51, 810–818. https://doi.org/10.1021/acs.est.6b04048.

Mahon, A., Offiver, R., Nash, R., O'Connor, I., 2017b. Scope, Fate, Risks & Impacts of Microplastic Pollution in Irish Freshwater Systems. Environment Protection Agency, Johnstown Castle. https://www.epa.ie/publications/research/water/research-210-scope-fate-risks-and-impacts-of-microplastic-pollution-in-irish-freshwater-systems.php.

McCormick, A., Hoellein, T., Mason, S., Schluep, J., Kelly, J., 2014. Microplastic is an abundant and distinct microbial habitat in an urban river. Environ. Sci. Technol. 48 (20), 11863–11871. https://doi.org/10.1021/es503610r.

Ministry of Defence, 2020. UK Land Holdings 2020. [Online]Ministry of Defence, Sutton Coldfield Available at: https://assets.publishing.service.gov.uk/government/uploads/system/uploads/attachment_data/file/886546/UK_Land_Holdings_Report_2020.pdf [Accessed 14 December 2022].

Mintenig, S., Int-Veen, I., Löder, M., Primpke, S., Gerdts, G., 2017. Identification of microplastic in effluents of waste water treatment plants using focal plane array-based micro-Fourier-transform infrared imaging. Water Res. 108, 365–372. https://doi.org/10.1016/j.watres.2016.11.015.

Misselbrook, T., Bittman, S., Cordovil, C., Rees, B., Sylvester-Bradley, R., Olesen, J., Vallejo, A., 2019. Field Application of Organic and Inorganic Fertilizers and Manure; Draft Section for a Guidance Document. [Online]European Commission., Brussels Available at: https://commission.europa.eu/system/files/2019-09/field_application_of_organic_and_inorganic_fertilizers_23sep19.pdf [Accessed 14 December 2022].

Murphy, F., Ewins, C., Carbonnier, F., Quinn, B., 2016. Wastewater treatment works (WwTW) as a source of microplastics in the aquatic environment. Environ. Sci. Technol. 50 (11), 5800–5808. https://doi.org/10.1021/acs.est.5b05416.

Naji, A., Azadkhah, S., Farahani, H., Uddin, S., Khan, F., 2021. Microplastics in wastewater outlets of Bandar Abbas city (Iran): a potential point source of microplastics into the Persian Gulf. Chemosphere 262, 128039. https://doi.org/10.1016/j.chemosphere.2020.128039.

Napper, I., Thompson, R., 2016. Release of synthetic microplastic fibres from domestic washing machines; effects of fabric type and washing conditions. Mar. Pollut. Bull. 112, 39–45. https://doi.org/10.1016/j.marpolbul.2016.09.025.

Ned, B., Beecher, J., Burke-Wells, J., Lono-Batura, M., Goldstein, N., Kester, G., Toffey, B., 2022. National Biosolids Data Project: Biosolids Management in the U.S. [Online]National Biosolids Data Project Available at: https://www.biosolidsdata.org [Accessed 14 December 2022].

Northumbrian Water Group Limited, 2022. Annual Performance Report. [Online]Northumbrian Water Group Limited, Durham Available at: https://www.nwg.co.uk/about-us/nwl/how-we-are-performing/annual-performance-report/ [Accessed 11 08 2022].

Okoffo, E., Tscharke, B., O'Brien, J., O'Brien, S., Riberio, F., Burrows, D., Choi, P., Wang, X., Mueller, J., Thomas, K., 2020. Release of plastics to Australian land from biosolids end-use. Environ. Sci. Technol. 54, 15132–15141. https://doi.org/10.1016/j.scitotenv.2021.152382.

Petroody, S., Hashemi, S., Gestel, C., 2021. Transport and accumulation of microplastics through wastewater treatment sludge processes. Chemosphere 278, 130471. https://doi.org/10.1016/j.chemosphere.2021.130471.

Pittura, L., Foglia, A., Akyol, C., Cipolletta, G., Benedetti, M., Regoli, F., Eusebi, A., Sabbatini, S., Tseng, L., Katsou, E., Gorbi, S., Fatone, F., 2021. Microplastics in real wastewater treatment schemes: comparative assessment and relevant inhibition effects on anaerobic processes. Chemosphere 262, 128415. https://doi.org/10.1016/j.chemosphere.2020.128415.

Ragoobur, D., Huerta-Lwanga, E., Somaroo, G., 2021. Microplastics in agricultural soils, wastewater effluents and sewage sludge in Mauritius. Sci. Total Environ. 798, 149326. https://doi.org/10.1016/j.scitotenv.2021.149326.

Raju, S., Carbery, M., Kuttykattil, A., Senthirajah, K., Lundmark, A., Rogers, Z., SCB, S., Evans, G., Palanisami, T., 2020. Improved methodology to determine the fate and transport of microplastics in a secondary wastewater treatment plant. Water Res. 173, 115549. https://doi.org/10.1016/j.watres.2020.115549.

Ren, X., Sun, Y., Wang, Z., Barceló, D., Wang, Q., Zhang, Z., Zhang, W., 2020a. Abundance and characteristics of microplastic in sewage sludge: a case study of Yangling, Shaanxi province, China. Case Studies Chem. Environ. Eng. 2, 100050. https://doi.org/10.1016/j.cscee.2020.100050.

Rasmussen, L., Iordachescu, L., Tumlin, S., Vollertsen, J., 2021. A complete mass balance for plastics in a wastewater treatment plant - macroplastics contributes more than microplastics. Water Res. 201, 117307. https://doi.org/10.1016/j.watres.2021.117307.

Ren, P.J., Dou, M., Wang, C., Li, G.Q., Jia, R., 2020b. Abundance and removal characteristics of microplastics at a wastewater treatment plant in Zhengzhou. Environ. Sci. Pollut. Res. 21, 36295–36305.. https://link.springer.com/article/10.1007/s11356-020-09611-5.

Rochman, C., Brookson, C., Bikker, J., Djuric, N., Earn, A., Bucci, K., Athey, S., Huntington, A., McIlwraith, H., Munno, K., De Frond, H., Kolomijeca, A., Erdle, L., Grbic, J., Bayoumi, M., Borrelle, S., Wu, T., Santoro, S., Werbowski, L., Zhu, X., Giles, R., Hamilton, B., Thaysen, C., Kaura, A., Klasios, N., Ead, L., Kim, J., Sherlock, C., Ho, A., Hung, C., 2019. Rethinking microplastics as a diverse contaminant suite. Environ. Toxicol. Chem. 38 (4), 703–711. https://doi.org/10.1002/etc.4371.

Rohatgi, A., 2020. WebPlotDigitizer. Version 4.6. [Online]WebPlotDigitizer, Pacifica Available at: https://automeris.io/WebPlotDigitizer/ [Accessed 14 December 2022].

Rolsky, C., Kelkar, V., Driver, E., Halden, R., 2020. Municipal sewage sludge as a source of microplastics in the environment. Curr. Opin. Environ. Sci. Health 14, 16–22. https://doi.org/10.1016/j.coesh.2019.12.001.

Roscher, L., Halbach, M., Nguyen, M., Hebeler, M., Luschtinetz, F., Scholz-Böttcher, B., Primpke, S., Gerdts, G., 2022. Microplastics in two German wastewater treatment plants: year-long effluent analysis with FTIR and Py-GC/MS. Sci. Total Environ. 817 (15), 152619.

Salmi, P., Ryymin, K., Karjalainen, A., Mikola, A., Uurasjärvi, E., Talvitie, J., 2021. Particle balance and return loops for microplastics in a tertiary-level wastewater treatment plant. Water Sci. Technol. 84 (1), 89–100. https://doi.org/10.2166/wst.2021.209.

Schell, T., Hurley, R., Buenaventura, N., Mauri, P., Nizzetto, L., Rico, A., Vighi, M., 2022. Fate of microplastics in agricultural soils amended with sewage sludge: is surface water runoff a relevant environmental pathway? Environ. Pollut. 293, 118520. https://doi.org/10.1016/j.envpol.2021.118520.

Scottish Water, 2021. Annual Report & Accounts 2020/21: Performance and Prospects. [Online]Scottish Water, Glasgow Available at https://www.scottishwater.co.uk/-/media/

ScottishWater/Document-Hub/Key-Publications/Annual-Reports/ScottishWaterAnnualReportAccounts202021PerformanceAndProspectsLRFINAL.pdf [Accessed 15 December 2022].

Severn Trent Water Limited, 2021. Annual Performance Report 2021. [Online]Severn Trent Water Limited, Coventry Available at https://www.stwater.co.uk/content/dam/stw/regulatory-library/ST.%20APR%202021-single-pages.pdf [Accessed 15 December 2022].

South West Water Limited, 2022. Annual Performance Report and Regulatory Reporting 2022. South West Water, Exeter.

Southern Water Services Limited, (2022) [Online] Worthing: Southern Water Services Limited. Available at: https://www.southernwater.co.uk/our-performance/reports/annual-reporting [Accessed 11 08 2022].

Sujathan, S., Kniggendorf, A., Kumar, A., Roth, B., Rosenwinkel, K.-H., Nogueira, R., 2017. Heat and bleach: a cost-efficient method for extracting microplastics from return activated sludge. Arch. Environ. Contam. Toxicol. 73, 641–648.. https://link.springer.com/article/10.1007/s00244-017-0415-8.

Sun, J., Dai, X., Wang, Q., van Loosdrecht, M., Ni, B.-J., 2019. Microplastics in wastewater treatment plants: detection, occurrence and removal. Water Res. 152, 21–37. https://doi.org/10.1016/j.watres.2018.12.050.

Tadsuwan, K., Babel, S., 2022a. Microplastic abundance and removal via an ultrafiltration system coupled to a conventional municipal wastewater treatment plant in Thailand. J. Environ. Chem. Eng. 10 (2), 107142. https://doi.org/10.1016/j.jece.2022.107142.

Tadsuwan, K., Babel, S., 2022b. Microplastic contamination in a conventional wastewater treatment plant in Thailand. Waste Manag. Res. 39 (5), 754–761. https://doi.org/10.1177/0734242X20982055.

Tagg, A., Brandes, E., Fischer, F., Fischer, D., Brandt, J., Labrenz, M., 2022. Agricultural application of microplastic-rich sewage sludge leads to further uncontrolled contamination. Sci. Total Environ. 806 (part 4), 150611. https://doi.org/10.1016/j.scitotenv.2021.150611.

Talvitie, J., Mikola, A., Setälä, O., Heinonen, M., Koistinen, A., 2017. How well is microlitter purified from wastewater? – a detailed study on the stepwise removal of microlitter in a tertiary level wastewater treatment plant. Water Res. 109, 164–172. https://doi.org/10.1016/j.watres.2016.11.046.

Tang, N., Liu, X., Xing, W., 2020. Microplastics in wastewater treatment plants of Wuhan, Central China: abundance, removal, and potential source in household wastewater. Sci. Total Environ. 745, 141026. https://doi.org/10.1016/j.scitotenv.2020.141026.

Teuten, E., Rowland, S., Galloway, T., Thompson, R., 2007. Potential for plastics to transport hydrophobic contaminants. Environ. Sci. Technol. 41 (22), 7759–7764. https://doi.org/10.1021/es071737s.

Thames Water Utilities Limited, 2022. Annual Performance Report 2021/22. [Online]Thames Water Utilities Limited, Reading Available at https://www.thameswater.co.uk/media-library/home/about-us/investors/our-results/current-reports/annual-performance-report-2021-2022.pdf [Accessed 11 November 2022].

The Company of Biologists Ltd, 2021. Leaving the Bar in Five Steps. [Online]The Company of Biologists Ltd, Cambridge Available at: https://thenode.biologists.com/leaving-bar-five-steps/research/ [Accessed 30 September 2021].

UK Water, 2010. Recycling of Biosolids to Agricultural Land. UK Water, London.

Uman, L., 2011. Systematic reviews and meta-analyses. J. Can. Acad. Child Adolesc. Psychiatry 20 (1), 57–59.. https://www.ncbi.nlm.nih.gov/pmc/articles/PMC3024725/.

United Utilities Group PLC, 2022. Annual Performance Report 2021/22. [Online]United Utilities Group PLC, Warrington Available at https://www.unitedutilities.com/globalassets/documents/pdf/united-utilities-annual-performance-report-2021-22 [Accessed 10 October 2022].

Van den Berg, P., Huerta-Lwanga, E., Corradini, F., Geissen, V., 2020. Sewage sludge application as a vehicle for microplastics in eastern Spanish agricultural soils. Environ. Pollut. 261, 114198. https://doi.org/10.1016/j.envpol.2020.114198.

Van Echelpoel, W., Vandegehuchte, M., Gueldre, G., Cauwenberghe, L., 2014. Microplastics in a biological wastewater treatment plant and the surrounding freshwater environment in Flanders: quantitative assessment. Advanced Master of Science in 'Technology for Integrated Water Management'. Universiteit Gent & Universiteit Antwerpen . https://libstore.ugent.be/fulltxt/RUG01/002/166/784/RUG01-002166784_2014_0001_AC.pdf.

Vardar, S., Onay, T., Demirel, B., Kideys, A., 2021. Evaluation of microplastics removal efficiency at a wastewater treatment plant discharging to the Sea of Marmara. Environ. Pollut. 289, 117862. https://doi.org/10.1016/j.envpol.2021.117862.

Vollertsen, J., Hansen, A., 2017. Microplastic in Danish Wastewater: Sources, Occurrences and Fate. [Online]The Danish Environmental Protection Agency, Copenhagen Available at https://vbn.aau.dk/en/publications/microplastic-in-danish-wastewater-sources-occurrences-and-fate [Accessed 19 December 2022].

Wagner, M., Lambert, S., 2018. Freshwater Microplastics: Emerging Environmental Contaminants? The Handbook of Environmental Chemistry. 58th series edition. Springer Nature, Switzerland.

Wei, F., Xu, C., Chen, C., Wang, Y., Lan, Y., Long, L., Xu, M., Uw, J., Shen, F., Zhang, Y., Xiao, Y., Yang, G., 2022. Distribution of microplastics in the sludge of wastewater treatment plants in Chengdu, China. Chemosphere 287, 132357. https://doi.org/10.1016/j.chemosphere.2021.132357.

Wessex Water Services Limited, 2022. Latest Performance. [Online]Wessex Water Services Limited, Bath Available at https://corporate.wessexwater.co.uk/our-performance/latest-performance [Accessed 19 December 2022].

WISE Freshwater, 2022. Overview: Urban Waste Water Production and Its Treatment. European Environment Agency, Italy. Copenhagen Available at: https://water.europa.eu/freshwater/countries/uwwt/italy [Accessed 11 August 2022].

Wiśniowska, E., Moraczewska-Majkut, K., Nocoń, W., 2018. Efficiency of microplastics removal in selected wastewater treatment. Desalin. Water Treat. 134, 316–323. https://doi.org/10.5004/dwt.2018.23418.

Wohlin, C., 2014. Guidelines for snowballing in systematic literature studies and a replication in software engineering. EASE '14: Proceedings of the 18th International Conference on Evaluation and Assessment in Software. Article No. 38, pp. 1–10 https://doi.org/10.1145/2601248.2601268.

Woodward, J., Rothwell, J., Hurley, R., 2021. Acute riverine microplastic contamination due to avoidable releases of untreated wastewater. Nat. Sustain. 4, 793–802. https://doi.org/10.1038/s41893-021-00718-2.

Xie, M., Huang, J.-L., Lin, Z., Chen, R., Tan, Q., 2021. Field to laboratory comparison of metal accumulation on aged microplastics in coastal waters. Sci. Total Environ. 797, 149108. https://doi.org/10.1016/j.scitotenv.2021.149108.

Xu, Q., Gao, Y., Xu, L., Shi, W., Wang, F., LeBlanc, G., Cui, S., Lihui, A., Lei, K., 2020. Investigation of the microplastics profile in sludge from China's largest water reclamation plant using a feasible isolation device. J. Hazard. Mater. 388, 122067. https://doi.org/10.1016/j.jhazmat.2020.122067.

Yang, G., Zhang, G., Wang, H., 2015. Current state of sludge production, management, treatment and disposal in China. Water Res. 78, 60–73. https://doi.org/10.1016/j.watres.2015.04.002.

Yang, Z., Li, S., Ma, A., Liu, P., Peng, D., Ouyang, Z., Gup, X., 2021a. Characteristics and removal efficiency of microplastics in sewage treatment plant of Xi'an City, Northwest China. Sci. Total Environ. 771, 145377. https://doi.org/10.1016/j.scitotenv.2021.145377.

Yang, J., Li, L., Li, R., Xu, L., Shen, Y., Li, S., Tu, C., Wu, L., Christie, P., Luo, Y., 2021b. Microplastics in an agricultural soil following repeated application of three types of sewage sludge: a field study. Environ. Pollut. 289, 117943. https://doi.org/10.1016/j.envpol.2021.117943.

Yorkshire Water Services Limited, 2022. Annual Performance Report 2021/11. [Online]Yorkshire Water Services Limited, Bradford Available at https://www.yorkshirewater.com/media/zuicmmat/annual-performance-report-2021-2022.pdf [Accessed 19 December 2022].

Yu, Q., Hu, X., Yang, B., Zhang, G., Wang, G., Ling, W., 2020. Distribution, abundance and risks of microplastics in the environment. Chemosphere 249, 126059. https://doi.org/10.1016/j.chemosphere.2020.126059.

Yuan, F., Zhao, H., Sun, H., Zhao, J., Xia, T., 2022. Investigation of microplastics in sludge from five wastewater treatment plants in Nanjing, China. J. Environ. Manag. 301, 113793. https://doi.org/10.1016/j.jenvman.2021.113793.

Zhang, L., Xie, Y., Liu, J., Zhong, S., Qian, Y., Gao, P., 2020. An overlooked entry pathway of microplastics into agricultural soils from application of sludge-based fertilizers. Environ. Sci. Technol. 54 (7), 4248–4255. https://doi.org/10.1021/acs.est.9b07905.

Zhang, Y., Wang, H., Xu, J., Su, X., Lu, M., Wang, Z., Zhang, Y., 2021a. Occurrence and characteristics of microplastics in a wastewater treatment plant. Bull. Environ. Contam. Toxicol. 107, 677–683.. https://link.springer.com/article/10.1007/s00128-021-03142-6.

Zhang, L., Liu, J., Xie, Y., Zhong, S., Gao, P., 2021b. Occurrence and removal of microplastics from wastewater treatment plants in a typical tourist city in China. J. Clean. Prod. 291, 125968. https://doi.org/10.1016/j.jclepro.2021.125968.

Ziajahromi, Shima, Neale, Peta, Silveira, Telles, Isabel, Chua, Andrew, Leusch, Frederic, 2021. An audit of microplastic abundance throughout three Australian wastewater treatment plants. Chemosphere 263, 128294. https://doi.org/10.1016/j.chemosphere.2020.128294.

Zubris, K., Richards, B., 2005. Synthetic fibers as an indicator of land application of sludge. Environ. Pollut. 138, 201–211. https://doi.org/10.1016/j.envpol.2005.04.013.

Nicholson, F., Gale, P., Hough, R., Jiang, Y., Kabir, M., Longhurst, P., Rollette, A., Taylor, M., Tompkins, D., Tyrell, S., Sakrabani, R., Williams, J., 2018. Biosolids to Market: A Strategic Proposal to Explore the Threats to Biosolids to Land-now and in the future. UK Water Industry Research Limited, London Available from https://ukwir.org/resume?object=b68d4460-53ba-4f58-a1b8-ceb59572d2a9. (Accessed 24 May 2023).

PlasticsEurope, 2018. Plastics - the Facts 2018; An analysis of European plastics production, demand and waste data. PlasticsEurope AISBL, Brussels Available from https://plasticseurope.org/wp-content/uploads/2021/10/2018-Plastics-the-facts.pdf. (Accessed 24 May 2023).