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Microplastics removal from a primary settler tank in a wastewater treatment plant and estimations of contamination onto European agricultural land via sewage sludge recycling☆

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ABSTRACT

Wastewater treatment plants (WwTPs) remove microplastics (MPs) from municipal sewage flow, with the resulting bulk of MPs being concentrated within generated sewage sludge which is frequently recycled back onto agricultural land as accepted practice in many European countries as a sustainable fertiliser resource. This circular process means that MPs successfully removed from WwTPs are deposited into the soil and able to return into the natural watercourse by means of run-off or infiltration to groundwater. This study quantifies the removal efficiency of MPs with size ranging between 1000 and 5000 μm in a primary settlement tank (PST) at a WwTP serving a population equivalent of 300,000 and provides MP concentrations in the generated sewage sludge. Our study revealed that the proportion of MPs partitioning in a PST to settled sludge, floating scum and effluent was 96%, 4% and 0% respectively, implying 100% removal of MPs of 1000–5000 μm in size. The generated sewage sludge was estimated to contain concentrations of approximately 0.01 g of MPs or 24.7 MP particles per g of dry sewage sludge solid, equivalent to ~1% of the sewage sludge weight. Using these figures and data from the European Commission and Eurostat, the potential yearly MP contamination onto soils throughout European nations is estimated to be equivalent to a mass of MPs ranging between 31,000 and 42,000 tonnes (considering MPs 1000–5000 μm in size) or 8.6×10^{13}–7.1×10^{14} MP particles (considering MPs 25–5000 μm in size). An estimated maximum application rate of 4.8 g of MP/m^2/yr or 11,489 MP particles/m^2/yr, suggests that the practice of spreading sludge on agricultural land could potentially make them one of the largest global reservoirs of MP pollution. Hence, recycling raw sewage sludge onto agricultural soils should be reviewed to avoid introducing extreme MP pollution into the environment.

1. Introduction

Microplastic (MP) pollution, defined as plastic particles smaller than 5000 μm in size, is a well-documented threat to aquatic and terrestrial ecosystems worldwide (Eerkes-Medrano et al., 2015; Hamid et al., 2018; de Souza Machado et al., 2018). MPs which have the capacity to absorb organic contaminants onto their surface, leach toxic chemical additives throughout the process of degradation, and can serve as attachment media for hazardous bacterial pathogens, are readily ingested by a range of organisms, owing to their small size (Galloway et al., 2017; Hermabessiere et al., 2017; de Souza Machado et al., 2018). The ingestion of MPs can cause negative health effects to organisms and the trophic transfer of MPs from lower trophic organisms to top predators means that the impacts of MP exposure and ingestion not only effects organisms at an individual level but potentially impacts the whole food chain (Haegerbaeumer et al., 2019; D'Souza et al., 2020).

The pathways through which these emerging environmental contaminants enter the aquatic and terrestrial environments are currently not fully understood (Hardesty et al., 2017; Cera et al., 2020). One significant source of MPs entering the aquatic environment are the effluents from wastewater treatment plants (WwTPs) due to inefficient removal of MPs from incoming municipal sewage (Murphy et al., 2016; Talvitie et al., 2017). Whilst the quality and quantity of techniques used during the water treatment process determines the capacity of WwTPs to

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remove MPs, they can still release substantial amounts of MPs despite having high MP removal rates of 72–98% from inlet to effluent, exacerbated if large amounts of municipal sewages are treated every day (Iyare et al., 2020). For instance, a WwTP serving a 1.2 million population that treats 400 million litres of sewage per day, with a MP removal efficiency of 84%, could be responsible for a daily release of 1.6×10^8 MPs per day into the aquatic environment (Magni et al., 2019).

Although WwTPs are currently not purposefully designed to remove MPs, it is important to understand the prevalent MP removal processes in order to develop a suitable technology to curb the concentrations of MPs exiting these infrastructures and entering the environment. The primary stages of sewage treatment, that usually comprise of a primary settling tank (PST), are responsible for up to 68–98.4% of MP removal according to seven recent studies (Talvitie et al., 2015; Michielssen et al., 2016; Murphy et al., 2016; Gies et al., 2018; Blair et al., 2019; Sun et al., 2019; Yang et al., 2019). The purpose of the PST is to promote solid settling to minimise the size of suspended particles prior to the biological treatment (Riffat, 2013). The incoming sewage, with its MPs content, are separated by density in the PST; dense materials such as grit and organic solids settle to the bottom of the tank as 'sludge', where more than 90% of the MPs from the incoming sewage feed are deposited, while less dense fats, oils and grease (often known as FOGs) stay to the upper water column as 'scum' (Carr et al., 2016). Both the sludge and scum, along with the MP load from the incoming sewage, are combined to produce sewage sludge, which is then subjected to additional treatment such as thickening, aerobic digestion, and de-watering (Kelessidis and Stasinakis, 2012; Mintenig et al., 2017; Alavian Petroody et al., 2021). MPs have been found in sewage sludge and vary in abundance depending on the treatment processes used at the WwTP and the population equivalent it serves (Mahon et al., 2017; Edo et al., 2020; Rolsky et al., 2020). For example, at a Chinese WwTP serving a population equivalent of 100,000, 2.92 MP particles per g of dry sewage sludge was measured, corresponding to 1.14×10^{11} particles per year deposited in the sewage sludge generated, while from an Italian WwTP serving a population equivalent of 1.2 million, 113 ± 57 MP particles per g of dry sewage sludge was observed, corresponding to over 1.24×10^{12} MP particles per year deposited in the sewage sludge generated (Magni et al., 2019; Ren et al., 2020).

Sewage sludge is commonly recycled to agricultural land as a sustainable and renewable source of fertiliser throughout European countries, owing to the European Union's directives that promote diverting sewage sludge away from landfill and incineration, and towards energy production and agriculture, contributing to goals that lead to net-zero waste and sustainable economic growth (e.g. European Landfill (1999/31/EC(30)) and Renewable Energy (2009/28/EC(31)) (EU Commission, 2009a, 2009b; 1999; Mininni et al., 2015). The spreading of sewage sludge onto agricultural land has been acceptable practice until the ever-increasing MP presence appeared as a new environmental threat to terrestrial ecosystems through MP deposition onto agricultural lands. Concentrations as high as 541 MP particles per kg of soil were found in agricultural soils that had sewage sludge applied to them in Ontario (Canada), compared to 4 MP particles per kg in control non-sewage sludge applied soils, meaning that sewage sludge land application is contaminating agricultural soil with MPs (Crossman et al., 2020). Based on quantitative data from national MP inputs to WwTPs from Denmark, Sweden and Norway, it was projected that 63,000–430, 000 tonnes of MPs are applied onto European agricultural land each year, with average and maximum loadings per-capita of 2 and 80 g of MP/m^2/yr, respectively (Nizzetto et al., 2016). In addition, Mohajerani and Karabatak (2020) estimated that between 26,000 and 151,000 tonnes of MPs are disposed onto European agricultural soils using figures from a review of three papers which report concentrations of MPs in generated biosolids, produced from WwTP sewage sludge. As a result, agricultural land may represent one of the largest potential reservoirs of MP pollution worldwide, mirroring MP concentrations in global ocean surface waters (Sebille et al., 2015).

These recent findings have highlighted a need for understanding the transport of MPs throughout the environment, processes involved in their removal at WwTPs, the MP concentration within generated sewage sludge, and to provide further insights into the MP budget and contamination on agricultural soils. This study provides a better understanding of the partitioning of removed MPs in the size range of 1000–5000 μm in a PST into settled sludge, surface scum, and effluent by mass and abundance, obtained at a WwTP in Newport (Wales, UK). These field sampling data were used to estimate the MP concentration within the generated sewage sludge and to better estimate the potential magnitude of MP pollution on European agricultural soils for most nations, adopting data from yearly sewage sludge production and application rates from individual European nations (EU Commission, 2015, 2018; Eurostat, 2019b). These data will aid to improve the management of the WwTP sewage sludge process in order to minimise or remove the number of MPs contained, as well as to aid in the development of policies that regulate the MPs input limits for sewage sludge spreading on agricultural soils.

2. Methodology

2.1. Nash WwTP background and production of sewage sludge

The Nash WwTP in Newport (South Wales, the United Kingdom) is operated by Dwr Cymru Welsh Water (DCWW) and treats the combined sewage from a total population equivalent of 300,000 from Newport and Chepstow, with a pass forward flow limit of 1,415 L per second. Fig. 1 shows a diagram flow depicting the Nash WwTP consisting of an initial 6 mm screening and grit removal, a primary settling tank (PST), biological activated sludge treatment in an aerated basin, and secondary settlement, before discharging the final effluent to Julian's Pill, a tributary of the River Severn. Waste sludge from the secondary settler is returned to the PST stage for co-settlement. The PST separates incoming municipal sewage into scum and sludge, combined to create sewage sludge, which is then thickened on-site with sludge imported from smaller rural WwTPs also managed by DCWW and transported for advanced anaerobic digestion (AAD) treatment at Cardiff WwTP before being recycled back to agricultural land. From all DCWW's sludge centres (35 WwTPs), around 97,854 tonnes of dry solid (T_{ds}) sewage sludge was produced in 2019–2020, of which 5,311 T_{ds} (5.43%) was contributed by the Nash WwTP (DCWW, personal communication).

2.2. Sampling locations and procedure

Four locations at the Nash WwTP (shown in Fig. 2) were sampled to quantify the abundance of MPs in a PST and assess the removal efficiency of the PST, and to calculate the concentration of MPs in generated sewage sludge. These include: (1) the screened incoming sewage before the PST (PST feed), (2) the effluent discharged from the PST (PST effluent), (3) settled sludge removed from the base of the PST tank (sludge), and (4) surface FOG's and debris collected from the surface scum traps (scum). A 'flow and load' survey was conducted, with 24-h monitoring throughout a seven-day dry weather period, which is consistent with a standardised sampling programme to understand typical treatment works loading for other environmental pollutants (Environmental Agency, 2017; Bertanza et al., 2022), providing a total of seven samples per location (28 samples in total).

PST feed and effluent samples were collected in 15-min intervals using an automatic sampling system over 24-h periods (Aquamatic Aquacell P2 Coolbox). PST feed sample pipework was placed within the central baffle boards where the tank inlet pipework was located, while the PST effluent sample pipework was installed at the tank's outer edge where clarified effluent weirs over at the surface. On collection, the bulk samples containers were mixed and decanted into a 1 L glass sampling bottles once per day (Fig. 3). Spot samples of the PST sludge and PST scum were also taken each day. Sludge is removed from the PST tank by

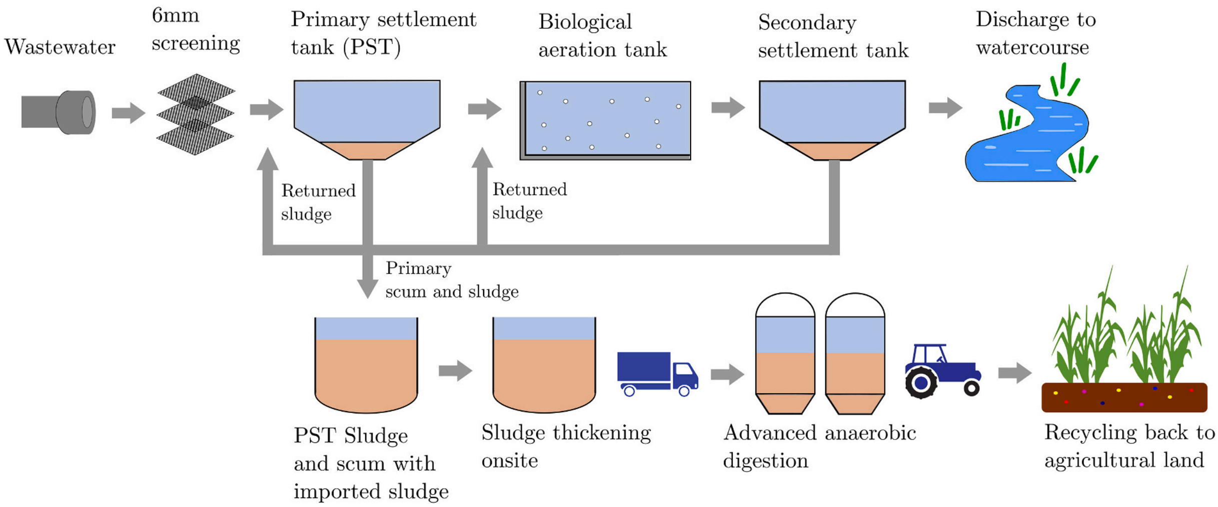

Fig. 1. Schematic diagram of the wastewater treatment flow process, sewage sludge production and environmental discharge at Nash WwTP (South Wales, UK).

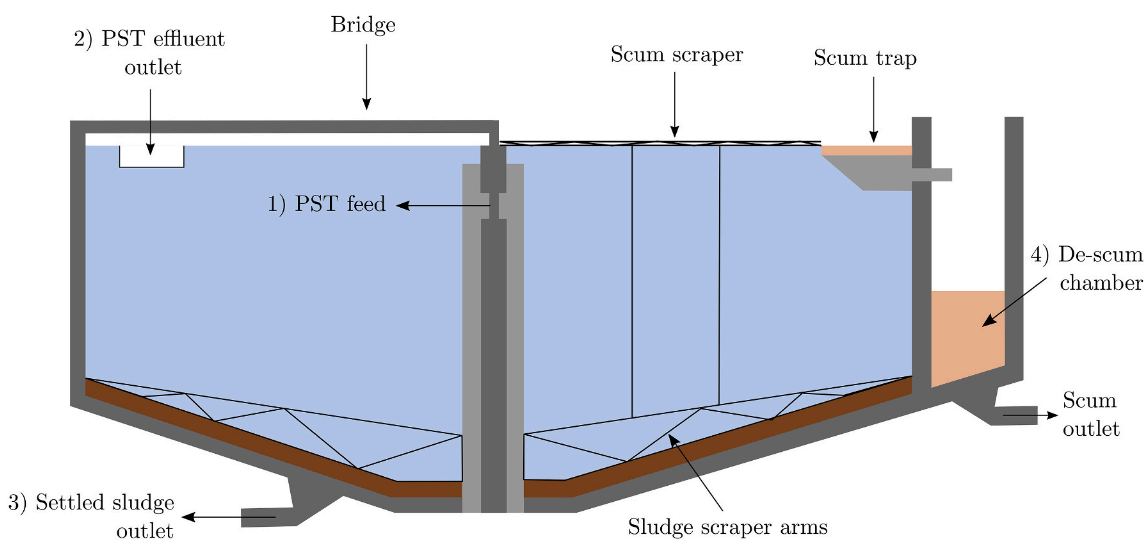

Fig. 2. A side-view cross-section of the PST and de-sludge chamber at the Nash WwTP showing the four sampling locations: 1) PST feed, 2) PST effluent, 3) settled sludge outlet, and 4) de-scum chamber.

Fig. 3. Photograph of collected samples stored in 1 L glass bottles. From left to right: PST feed, PST effluent, Sludge and Scum.

dedicated de-sludge pumps from the base of the PST tank once per hour for a maximum of 15 min or until the density of the sludge falls below 3.5% dry solids content. One litre of sludge sample was taken once per day using a tap located on the de-sludge pipework. For the scum samples, surface scrapers that revolve around the centre of the PST force the scum towards the outside of the tank into a scum trap (see Fig. 2) where the scum drains into a de-scum chamber once per revolution (approximately once per hour). A collection bucket was secured below the pipe connecting the scum trap to the de-scum chamber and decanted into 1 L glass sampling bottle (Koelmans et al., 2019). In addition, the total volume of incoming feed and outgoing effluent was recorded in litres, while the volume of scum and sludge generated by the PST was recorded in both litres and as grams of dry solid (g_{ds}) over a seven-day sampling period.

2.3. Extraction and identification of MPs from samples

MPs were recovered and identified from the samples collected at the four sampling locations (shown in Fig. 2) in four steps: (1) wet sieving, (2) wet peroxide oxidation, (3) density separation using zinc chloride ($ZnCl_2$), and (4) rose bengal staining and microscopic examination. Firstly, debris and organic material were removed from the samples using wet vacuum filtering with two custom graded wire meshes that divided the samples into 250–1000 μm and 1000–5000 μm size ranges; with only the latter fraction analysed in this study, as equipment to analyse the smaller size fraction was not available at the WwTP and would also require Water Companies to have specialised technicians and equipment. A volume of 200 mL from the 1 L glass bottles collected per day was used from the feed and effluent samples, while 10 mL of sludge and scum samples was used from the 1 L glass bottles collected per day, totalling 1.4 L of feed and effluent samples and 70 mL of sludge and scum samples, following recommendations by Koelmans et al. (2019) for feed samples. Due to the large amounts of FOGs that remained on the meshes during trial scum samples, an additional sample preparation step was added prior to wet sieving. This consisted of warming a mixture of 10 mL of scum sample, 100 mL of filtered water, and 10 mL of filtered washing-up liquid detergent in the oven at 50 °C for 1 h, which was then stirred for 10 min with a magnetic stirrer before sieving to remove the FOGs as they melted and disaggregated. The meshes were taken from the vacuum filtration equipment and dried in the oven overnight at 50 °C in accordance with recommendations made in Koelmans et al. (2019).

To digest the organic material in the samples, an iron catalyst and 30% hydrogen peroxide (H_2O_2) (commonly known as Fenton's Reagent) was used (Masura et al., 2015; Liu et al., 2019). The meshes containing the samples were placed into an 800 mL glass beaker, 10 mL ferrous sulphate was added, followed by 23 mL 30% hydrogen peroxide, all stirred using a glass stirring rod and a foil cover placed over the beaker. This was left overnight to allow the reaction to complete. The meshes were then removed from the beaker and washed using filtered water into the beaker. The solids were recovered by passing the samples through the mesh using the vacuum filtration unit, from which the meshes were removed and the samples dried in the oven overnight at 50 °C.

The samples were then transferred from the meshes to a 400 mL glass beaker and a 250 mL solution of 1.7 g/mol $ZnCl_2$ was added for density separation (Prata et al., 2019). The solution was stirred vigorously with a glass stirring rod and left for at least 20 h. To retrieve the MPs particles, the top quarter of the zinc chloride was sieved through the mesh. The process was repeated by adding up to 250 mL $ZnCl_2$, stirring vigorously to agitate the samples and release any flocculated MPs, and then passing the top quarter liquid through the meshes to recover the MPs.

Finally, the meshes containing the MP samples were placed in a foil tray and 0.2 mg/mL rose bengal solution was added until the samples were submerged and left to stain for 5 min (Lares et al., 2019). The waste rose bengal solution was passed through the meshes to recover any MPs. The foil tray and meshes were rinsed with filtered water using the vacuum filtration unit to recover any MPs and remove the rose bengal residue. The meshes were removed from the foil tray and dried in the oven overnight at 50 °C. The remaining MP particles were then analysed and counted at 40× magnification using a digital microscope. Particles that took up the stain were considered to be MPs and the remaining un-stained particles were discarded (Lares et al., 2019). Three rules for visual identification of MPs larger than 1000 μm proposed in Hidalgo-Ruz et al. (2012) were also used where: (1) no cellular or organic structures are visible, (2) fibres should be uniform in thickness, and (3) length and colour should be clear or uniform. MP particles were then placed on a weighed foil tray and re-weighed. MP concentrations over the seven-day sampling period were calculated as g of MPs (g_{mp}/L) and as MP particle abundance per litre (MP_p/L).

It is acknowledged that analysing MPs 1000–5000 μm in size over a seven-day sampling routine with a sample size of 200 mL per day may not fully represent the seasonally variability or total spectrum of MPs from a WwTP, while approaches such as using Fourier-transform infrared spectroscopy (FTIR) would have reduced uncertainty in the MP identification. However, the methods used in this present study are successful techniques for MP isolation and identification in WwTP sludge samples for MPs 1000–5000 μm in size using readily available equipment and reagents accessible at an on-site WwTP laboratory (Ziajahromi et al., 2017; Campo et al., 2019; Lares et al., 2019). Furthermore, the methods are designed in such a way that the approach could be routinely carried out by a Water Company together with the regular monitoring of other environmental pollutants, thus providing a standardised monitoring and estimation framework of MP concentrations in PST feed, effluent and sewage sludge.

2.4. Control methods

As MPs are ubiquitous and can be found in tap water, on clothing, and in the air, a number of control measures were implemented to minimise contamination of the samples (Dris et al., 2017; Brander et al., 2020). A negative control of filtered water was used to check for any MP contamination from the automatic sampling systems and throughout laboratory procedures. Filtered water was used for all washing and rinsing of laboratory equipment, sample collection buckets and bottles, and glass and metal equipment was used wherever possible. Glassware was rinsed twice with filtered water between sample transfers from one procedure to another. Furthermore, sample containers were covered with aluminium foil during waiting periods to avoid airborne MP contamination.

2.5. Data interpretation and upscaling methods

The mean MP concentration (*C*) from the seven-day sample was used together with the mean daily volume (*Vol*) processed at each of the four sampling locations to calculate the daily incoming feed of MP to the PST, and MPs leaving the PST as effluent, scum, or sludge, reported as a daily MP mass flux (g_{mp}/day, Eq. (1)) and a daily MP particle flux (MP_p/day, Eq. (2)).

$$\begin{aligned} &\textit{Daily MP mass flux}\ \left(\mathrm{g_{mp}}/\mathrm{day}\right) \\ &= \textit{Vol}\ (\mathrm{L}/\mathrm{day}) \times C\ \left(\mathrm{g_{mp}}/\mathrm{L}\right) \end{aligned} \tag{1}$$

$$\begin{aligned} &\textit{Daily MP particles flux}\ \left(\mathrm{MP_p}/\mathrm{day}\right) \\ &= \textit{Vol}\ (\mathrm{L}/\mathrm{day})\ \times C\ \left(\mathrm{MP_p}/\mathrm{L}\right) \end{aligned} \tag{2}$$

The total daily MP flux and the mean daily sewage sludge production (i.e. combined scum and sludge) at the WwTP were used to calculate the concentration of MPs per g of dry pre-thickened and pre-AAD treated sewage sludge (g_{ds}), as mass (g_{mp}/g_{ds}, Eq. (3)) and particle abundance (MP_p/g_{ds}, Eq. (4)).

$$\textit{MP mass per g of dry sewage sludge}\ \left(\mathrm{g_{mp}}\,/\,\mathrm{g_{ds}}\right) = \frac{\textit{Daily mass flux of MPs}\ \left(\mathrm{g_{mp}}/\mathrm{day}\right)}{\textit{Daily production of sewage sludge}\ \left(\mathrm{g_{ds}}/\mathrm{day}\right)} \tag{3}$$

$$\textit{MP particles per g of dry sewage sludge}\ (\mathrm{MP_p}/\mathrm{g_{ds}}) = \frac{\textit{Daily MP particles flux}\ (\mathrm{MP_p}/\mathrm{day})}{\textit{Daily production of sewage sludge}\ (\mathrm{g_{ds}}/\mathrm{day})} \quad (4)$$

Eurostat (2019b) is the main source of data about sewage sludge management in Europe and publishes yearly datasets, collected biennially by means of questionnaires, on the production and disposal of sewage sludge (between 2009 and 2018) for each European nation. Figures from Eurostat (2019b) are collected by the European National Statistical Institutes, by which no specific data collection method is imposed by Eurostat, from a variety of data sources, including regional or local authorities, environmental administrations and industry. An additional source of data on yearly sewage sludge management in Europe is from reports published by the EU Commission (2015, 2018) that summarise and analyse the implementation of Sewage Sludge Directives (EU Commission, 1986, 1991, 1994) by each European nation, through means of a questionnaire. The EU Commission (2015, 2018) reports are a synopsis of the replies submitted by European nations for the period 2010–2015.

Based on both the EU Commission (2015, 2018) and Eurostat (2019b) datasets, between 8–10×10^6 tonnes of dry sewage sludge (T_{ds}) per year is produced from European WwTPs between 2009 and 2018. Using the amount of MPs per unit of sewage sludge from the Nash WwTP and the average sewage sludge production from WwTPs in individual European countries between the years 2009 and 2018, an estimate of the MP concentration in generated sewage sludge across Europe was calculated as yearly MP mass in tonnes (T_{mp}/yr, Eq. (5)) and particle abundance (MP_p/yr, Eq. (6)). As the concentration of MPs present in the generated sewage sludge may vary depending on sewage sludge production processes, WwTP capacity, and population served, the number of MPs per unit of sewage sludge from five other studies which sample European WwTP's are also included in calculations, as presented in Table 1 (Lusher et al., 2017; Mahon et al., 2017; Mintenig et al., 2017; Sujathan et al., 2017; Lares et al., 2018; Edo et al., 2020). The other European WwTPs were chosen to include different countries where the facilities are located, WwTP operations and range of population equivalents served by the WwTP. The five studies only report the abundance of MPs per unit of sewage sludge and isolated MPs down to 25 μm in size from sewage sludge, thus calculations consider MPs smaller than the current study (<1000 μm) for MP abundance.

Results for MP mass are displayed as the lower and upper limit is based on the standard error for the production of sewage sludge for each individual European country over the available years datasets (between 2009 and 2018) from EU Commission (2015, 2018) and Eurostat (2019b) datasets, while for MP abundance, lower and upper limits are based on the lowest and highest MP_p per unit of sewage sludge from the Nash WwTP and the five other European studies.

$$\begin{aligned}&\textit{MP mass load from sewage sludge production}\ (\mathrm{T_{mp}}/\mathrm{yr})\\ &\quad= \textit{MP mass per unit of sewage sludge}\ (\mathrm{T_{mp}}/\mathrm{T_{ds}})\\ &\qquad\times \textit{EU production of sewage sludge}\ (\mathrm{T_{ds}}/\mathrm{yr})\end{aligned} \quad (5)$$

$$\begin{aligned}&\textit{MP particles load from sewage sludge production}\ (\mathrm{MP_p}/\mathrm{yr})\\ &\quad= \textit{MP particles per unit of sewage sludge}\ (\mathrm{MP_p}/\mathrm{T_{ds}})\\ &\qquad\times \textit{EU production of sewage sludge}\ (\mathrm{T_{ds}}/\mathrm{yr})\end{aligned} \quad (6)$$

The EU Commission (2015, 2018) and Eurostat (2019b) datasets also provide the amount of sewage sludge that is disposed to agricultural land by each individual European country between 2009 and 2018. Based on the average yearly disposal of sewage sludge onto agricultural land, 35–44% of total generated sewage sludge from European WwTPs was recycled for agricultural use between the years 2009 and 2018, equating to 3.5–3.8 million T_{ds} of sewage sludge applied to European agricultural lands. Therefore, the amount of MP application per year to European agricultural soils, as well as the upper and lower limits, were calculated using the amount of MPs per unit of sewage sludge from the Nash WwTP and the five other European studies, assuming that 100% of sewage sludge directed to agricultural use is recycled back onto European soils (Eqs. (7) and (8)). Furthermore, the fraction of sewage sludge that is recycled back to European agricultural soils was also calculated for each individual European country.

$$\begin{aligned}&\textit{MP mass recycled to agricultural land}\ (\mathrm{T_{mp}}/\mathrm{yr})\\ &\quad= \textit{MPs mass per unit of sewage sludge}\ (\mathrm{T_{mp}}/\mathrm{T_{ds}})\\ &\times \textit{sewage sludge recycled to agricultural land}\ (\mathrm{T_{ds}}/\mathrm{yr})\end{aligned} \quad (7)$$

$$\begin{aligned}&\textit{MP particles recycled to agricultural land}\ (\mathrm{MP_p}/\mathrm{yr})\\ &\quad= \textit{MP particles per unit of sewage sludge}\ (\mathrm{MP_p}/\mathrm{T_{ds}})\\ &\times \textit{sewage sludge recycled to agricultural land}\ (\mathrm{T_{ds}}/\mathrm{yr})\end{aligned} \quad (8)$$

Finally, the application rate of MPs to agricultural land as MP mass (g_{mp}/m^2/yr, Eq. (9)) and MP particle abundance (MP_p/m^2/yr, Eq. (10)) was calculated considering the maximum amount of total nitrogen/ha/yr permitted for European agricultural soils (250 kg of total nitrogen/ha/yr), which typically represents the limiting factor determining the rate of application of sewage sludge to agricultural land (EU Commission, 1991a,b; DEFRA, 2018; Collivignarelli et al., 2019). For a typical digested sewage sludge cake, such as that produced at DCWW's AAD site in Cardiff, which is where the sewage sludge from the Nash WwTP was sent for processing before direct application to agricultural land, 250 kg of total nitrogen/ha/yr equates to a maximum of 18.7 T of wet sludge at

Table 1
Comparison of the concentration of MP particles per g of dry sewage sludge, the population served by WwTP, the lower size limit used in isolating the MPs from the sample, and section of the sewage sludge generation sampled from five other European studies, in order of decreasing lower MP size limit.

Reference	Population served by WwTP	Country	Lower size limit	MP concentration (MP_p/g_{ds} ± SE)	Type of sewage sludge
Mintenig et al. (2017)	7.0 × 10^3–2.1 × 10^5	Germany	500 μm	1–24	Combined PST surface scum and settled sludge
Lares et al. (2018)	N/A	Finland	250 μm	23 ± 4.2	Activated sludge
				170.9 ± 28.7	Digested sludge
				27.3 ± 4.7	Membrane bioreactor sludge
Lusher et al. (2017)	1.8 × 10^4–6.1 × 10^5	Norway	50 μm	1.7–19.8	Dewatered sludge and dried sludge
Mahon et al. (2017)	6.5 × 10^2–2.4 × 10^5	Ireland	45 μm	4.0–15.4	Aerobically digested sludge, thermal dried sludge, and lime stabilized sludge
Edo et al. (2020)	3.0 × 10^5	Spain	25 μm	183 ± 84	Combined primary and secondary settled sludge

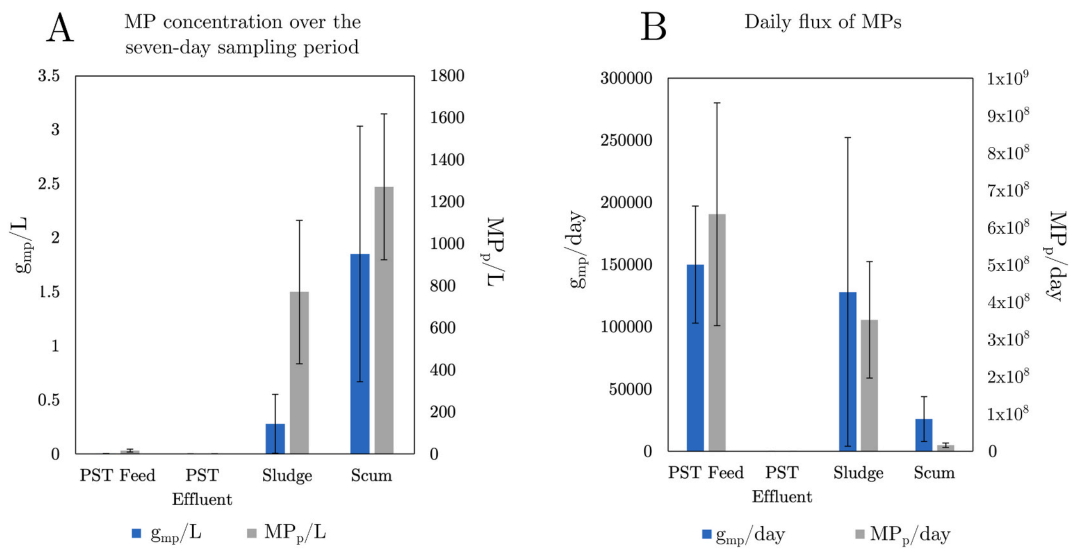

Fig. 4. **a**) Mean concentration of MPs in the size range 1000–5000 μm at each sampling location over the seven-day sampling period (n = 28) expressed as g_{mp}/L and MP_p/L. **b**) Mean daily flux of MP particles at each sampling location expressed as g_{mp}/day and MP_p/day.

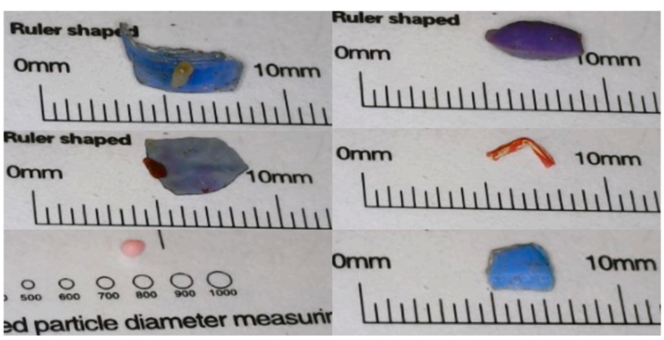

Fig. 5. MP particles of different shapes and sizes found at the final identification stages.

25% dry solid content/ha/yr, corresponding to 4.68 T_{ds}/ha/yr (DCWW, personal communication).

$$\begin{aligned}&\textit{App. rate of MPs mass to agricultural land}\ \left(g_{mp}/m^2/yr\right)\\&\quad=\left(\textit{MP mass per unit of sewage sludge}\ \left(g_{mp}/T_{ds}\right)\right.\\&\qquad\times\textit{app. rate of sewage sludge onto agricultural}\\&\qquad\qquad\left.\textit{land}\ \left(T_{ds}/ha/yr\right)\right)\cdot 10^{-4}\end{aligned} \tag{9}$$

$$\begin{aligned}&\textit{App. rate of MP particles to agricultural land}\ \left(MP_p/m^2/yr\right)\\&\quad=\left(\textit{MP particles per unit of sewage sludge}\ \left(MP_p/T_{ds}\right)\right.\\&\qquad\times\textit{app. rate of sewage sludge onto agricultural}\\&\qquad\qquad\left.\textit{land}\ \left(T_{ds}/ha/yr\right)\right)\cdot 10^{-4}\end{aligned} \tag{10}$$

3. Results and discussion

3.1. Quantification of daily MP flux in the PST at Nash WwTP

The mean concentration of MPs sized 1000–5000 μm over the seven-day sampling period, and the daily flux of MPs from each of the four sampling locations at the PST are presented in Fig. 4 in terms of MP mass and abundance. Examples of the MP particles found at the Nash WwTP PST are presented in Fig. 5 to illustrate the wide spectrum of MP size and shapes found. Mean concentrations of 0.004 ± 0.001 g_{mp}/L or 16 ± 7 MP_p/L were observed in the incoming PST feed. Despite these being relatively low over the seven-day sampling period, approximately 40,651 m^3/day of raw sewage was processed by the PST, which equates to 150,000 ± 47,000 g_{mp}/day or $6.34\times10^8 \pm 2.99\times10^8$ MP_p/day entering the PST. These data suggest that about 497 g_{mp}/day and 2106 MP_p/day are sent to the WwTP per 1000 residents of Newport and Chepstow (South Wales, UK). No MP particles were found in the negative control samples examined for contamination throughout sample preparation and analysis.

MPs 1000–5000 μm from the incoming sewage feed either settled at the base of the PST as sludge or floated to the surface of the PST as scum. The settled sludge represents the fraction MPs that have a higher density than water, have been aggregated with cohesive particles, such as silts clays and iron oxides, or become colonised by microorganisms, rendering them overall denser than water (Maliwan et al., 2021). Over the seven-day sampling period, concentrations of MPs contained in the sludge were 0.28 ± 0.27 g_{mp}/L and 771 ± 341 MP_p/L (Fig. 4a), whilst the daily flux was 128,000 ± 124,000 g_{mp}/day and $3.52\times10^8 \pm 1.56\times10^8$ MP_p/day (Fig. 4b). This amounts to 83% and 96% of the MPs from the incoming raw sewage, by mass and abundance respectively, illustrating that the majority of MPs from the raw sludge feed concentrate in the sludge. This is similar to results in previous studies (Magnusson and Norén, 2014; Carr et al., 2016), demonstrating that around 90% of MPs in PSTs are contained within the settled sewage sludge.

The MPs found within the scum have a lower overall density than water or have become flocculated with FOGs forcing them to float to the surface. Over the seven-day sampling period, the MP concentrations in the scum were the highest of the four sampling locations, with 1.85 ± 1.18 g_{mp}/L and 1271 ± 347 MP_p/L (Fig. 4a). However, only 13 m^3/day of scum is generated daily at this WwTP, meaning the daily flux of MPs in the scum is much lower than the sludge, with 26,000 ± 18,000 g_{mp}/day and $1.6\times10^7 \pm 6.1\times10^6$ MP_p/day (Fig. 4b). As a result, the scum only accounts for 17% and 4% of MPs in the incoming daily supply of wastewater in terms of mass and number of MP particles, respectively. Hence, combining the sludge and scum as the sewage sludge at the Nash WwTP, a daily concentration of 153,000 ± 125,299 g_{mp}/day, and abundance of $3.68\times10^8 \pm 1.56\times10^8$ MP_p/day, accumulate within the sewage sludge. This budget is summarised in Fig. 6.

During the seven-day sampling period the Nash WwTP produced a daily average of 14.98 T_{ds} of pre-thickened and pre-AAD treated sewage sludge. Using these values with the daily mass and number of MP particles exiting the PST as sewage sludge (Eqs. (3) and (4)), a concentration of 0.01 ± 0.008 g_{mp}/g_{ds} or 24.6 ± 10.4 MP_p/g_{ds} are estimated to be generated from Nash WwTP. Thus, around ~1% of the dried mass of sewage sludge processed at the Nash WwTP are MP particles.

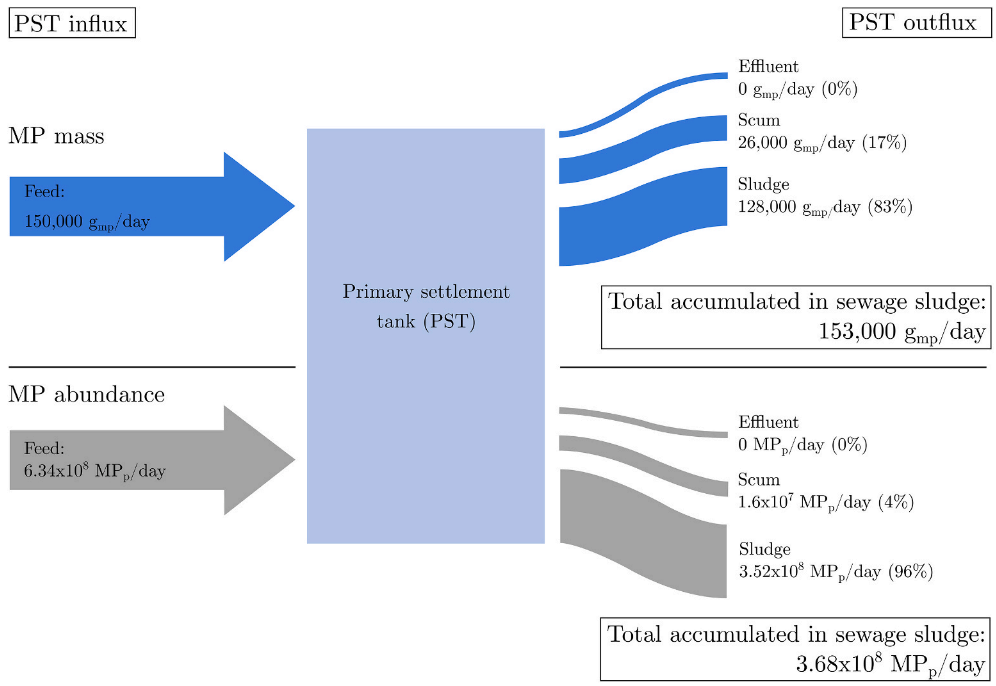

Fig. 6. Mass (g_{mp}/day) and number (MP_p/day) of MP particles 1000–5000 μm entering the PST as feed, and leaving the PST as Effluent, Scum and Sludge, with the percentual contribution of each sampling site.

The results show that 100% removal efficiency of MPs in the range of 1000–5000 μm was achieved at the PST, with 0 MPs of the sampled range identified in the PST effluent over the seven-day sampling period. Thus, a negligible quantity of MPs in the size range 1000–5000 μm is expected to enter the aquatic environment from the Nash WwTP outflow. It should be borne in mind that particles smaller than 1000 μm could be still present in the PST effluent and transported to secondary and tertiary stages of treatment. The removal rate observed by the Nash WwTP PST effluent is greater than those reported in previous studies, which could be attributable to the sampling approach employed to isolate the MPs from the collected PST samples, as most studies target MPs smaller than 1000 μm. For example, Tagg et al. (2020) found that the PST removed 100% of MPs larger than 600 μm, while MPs smaller than 600 μm were transported to secondary and tertiary treatment. Dris et al. (2015) observed that the PST reduced the proportion of MPs 1000–5000 μm from 45% in PST feed to 7% in PST effluent. Talvitie et al. (2017) suggested that the PST treatment may be efficient in removing MPs more than 300 μm, while smaller MPs (<300 μm) are transported to secondary or tertiary stages of treatment.

3.2. MPs concentrated in generated sewage sludge at nash WwTP

MP abundance held within generated sewage sludge can vary due to the methods used to isolate the MPs from the sample, as well as the section of sewage sludge production process sampled, as shown in Table 1. Mintenig et al. (2017) studied the MP budget from the combined PST surface scum and settled sludge from 12 German WwTPs serving a similar population-equivalent scale and sewage sludge production processes to the present study but with a lower size limit of MP retention of 500 μm and obtained a concentration of 1–24 MP_p/g_{ds} of dry pre-thickened and pre-AAD sewage sludge. The upper limit of their measurements agrees with the value of 24.6 ± 10.4 MP_p/g_{ds} from the present study. Edo et al. (2020) also sampled a similar treatment stage and population-equivalent scale at a Spanish WwTP but adopted a significantly lower size limit of MP retention of 25 μm. They observed concentrations of 183 ± 84 MP_p/g_{ds} in PST sewage sludge samples, i.e. around seven times higher than in the current study, implying that smaller sized MPs are prevalent in sewage sludge samples and that MP concentrations in sewage samples are dependent on sampling approach to separate the MP particles from the sample and lower size limit of MP retention.

The samples obtained in this study were collected prior to any sewage sludge processing. Previous studies have shown that the production steps of sewage sludge generation will change the abundance of MPs contained in the sewage sludge, however, from a mass balance point of view, the mass of MPs contained in sewage sludge should remain the same. For instance, Mahon et al. (2017) observed that thermal drying and lime stabilization increased MP concentrations due to the fragmentation of MP particles into a number of smaller MPs, while Alavian Petroody et al. (2021) observed that AAD increased concentrations potentially due to the degradation of flocs releasing MPs. Conversely, de-watering was found to reduce MP concentrations due to the breakdown of flocs in the digestive process and subsequent release of associated MP, which are reintroduced into the wastewater treatment process with the rejected water (Alavian Petroody et al., 2021).

3.3. Estimates of MP application onto european agricultural land via sewage sludge recycling

The concentration of MPs per unit volume of sewage sludge in European agricultural land is estimated from the present Nash WwTP measurements, that from the five European studies shown in Table 1, and datasets from the EU Commission (2015, 2018) and Eurostat (2019b). Based on this data, it can be estimated that in terms of MP mass between 72,000 and 110,000 T_{mp}/yr (based on MPs 1000–5000 μm in size) or in terms of MP abundance, 2.0^{14}–1.9×10^{15} MP_p/yr (based on MPs 25–5000 μm in size), lie in sewage sludge generated from European WwTPs. Considering the average application rates of sewage sludge onto European agricultural soils per European nation between 2009 and 2018, a conservative estimate of MP mass of between 31,000 and 42,000 T_{mp}/yr (based on MPs 1000–5000 μm in size), or in terms of MP abundance, 8.6×10^{13}–7.1×10^{14} MP_p/yr (based on MPs 25–5000 μm in size), is recycled back to European soils via sewage sludge application. Table 2 displays the lower and upper limits of MP mass (T_{mp}/yr) and MP

Table 2
The lower and upper limits of MPs recycled to agricultural land via sewage sludge disposal displayed as T_{mp}/yr (based on MPs 1000–5000 μm in size) and MP_p/yr (based on MPs 25–5000 μm in size) per European nation. Data is generated using the concentration of MPs per unit volume of sewage sludge from the Nash WwTP and five other European studies (Table 1), as well as from EU Commission (2015, 2018) and Eurostat (2019b) datasets.

	% of total produced sewage sludge recycled to agricultural land	T_{mp}/yr recycled to agricultural land via sewage sludge (MPs 1000–5000 μm)		MP_p/yr recycled to agricultural land via sewage sludge ($\times 10^{10}$) (25–5000 μm)	
Country	%	Lower	Upper	Lower	Upper
Albania	30–55	285	531	98	729
Austria	12–22	434	571	104	806
Belgium	11–12	0	480	49	429
Bulgaria	27–38	157	221	43	363
Croatia	3–5	6	12	2	20
Cyprus	31–50	22	36	7	52
Czechia	14–44	327	1,015	89	1,744
Denmark	52	758	1,620	182	1,448
Estonia	2–5	4	242	2	216
Finland	3–5	48	66	14	102
France	35–48	3,809	6,876	953	10,842
Germany	7–28	1,400	5,396	568	9,266
Greece	7–16	91	211	32	331
Hungary	8–15	156	277	43	399
Ireland	73–89	535	654	139	1,067
Italy	24–29	2,681	3,232	673	5,776
Latvia	25–33	59	76	16	123
Lithuania	22–33	101	161	28	281
Luxembourg	26–41	23	37	7	63
Netherlands	5–6	185	211	48	354
Norway	45–48	679	725	168	1,255
Poland	19–21	1,095	1,179	273	2,062
Portugal	7–61	168	1,384	50	1,712
Romania	5–10	92	188	34	250
Slovakia	1–2	4	9	1	14
Slovenia	1–2	1	5	1	7
Spain	71–77	8,918	9,717	2,154	17,024
Sweden	24–30	507	633	127	1,062
Switzerland	0.3	6	6	1	11
Turkey	4–6	129	187	38	282
United Kingdom	61–75	9,058	11,040	2,412	18,155

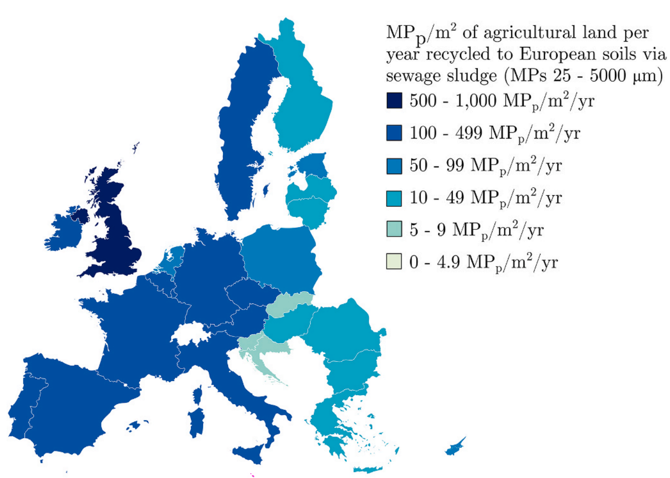

Fig. 7. The relative MP pressure on European agricultural soils, per nation, caused by recycling MP-laden sewage sludge, expressed as MP_p/m^2/yr. Data is generated from the Nash WwTP and five other European studies (Table 1), as well as from the EU Commission (2015, 2018) and Eurostat (2019a, 2019b) datasets.

abundance (MP_p/yr) of the fraction of sewage sludge that is recycled back to agricultural land, per European nation, from the EU Commission (2015, 2018) and Eurostat (2019b) datasets. Gaps in the data are related to the omission of European nations to respond to questionnaires on their sewage sludge production and disposal routes.

According to these estimates, agricultural soils may be one of the greatest environmental reservoirs of MP pollution, mirroring concentrations of MPs in ocean surface waters worldwide (Sebille et al., 2015). It should be noted that such estimates for MP mass are based on results at the Nash WwTP assuming that similar sewage sludge production processes are used across European WwTPs as those found at Nash WwTP, and that 100% of sewage sludge directed to agricultural use is applied to agricultural land. Estimates for MP mass can be deemed conservative as MPs less than 1000 μm are omitted, thus they are likely to underestimate of the true scale of the issue. As a result, our estimations are one order of magnitude lower than those from Nizzetto et al. (2016), that roughly projected that 63,000–430,000 T_{mp}/yr were applied to European agricultural land, including MPs smaller than 1000 μm, while the lower limit of Mohajerani and Karabatak (2020) estimations of 26,000–151,000 T_{mp}/yr, that included MPs 10 μm–5000 μm, agrees with the results of the present study.

Based on data from the EU Commission (2015, 2018) and Eurostat (2019a, 2019b) datasets, Fig. 7 depicts the relative MP pressure to European agricultural land, per nation, denoted as MP_p/m^2 of agricultural land per year. Fig. 7 and Table 2 indicate that MP-laden sewage sludge production and subsequent input onto European soils is not uniform across European nations, resulting in potentially greater MP pressures in some countries. The total amount of sewage sludge produced by each European country differs due to the percentage of the resident population served by WwTPs and the water treatment technologies used. Germany, the United Kingdom, Spain, France, and Italy account more than 76% of the total mass of MPs concentrated in sewage sludge in Europe. The percentage of generated sewage sludge that is recycled back to agricultural land is determined by local agricultural policy, population density or the availability of agricultural areas for sewage sludge spreading (Collivignarelli et al., 2019). Countries such as Spain (74–77%), Ireland (73–85%), the United Kingdom (61–75%) and Denmark (52%) recycle more than half of their WwTP sewage sludge and implement policies that encourage recycling sewage sludge to their land, which result in higher MP contamination in their agricultural soils. Similarly, countries such as Luxembourg have limited areas of agricultural land but yet recycle high amounts of their sewage sludge to agricultural soils (26–41%), resulting in higher MP pressure. On the other hand, Poland has large areas of agricultural land but recycle a relatively small amount of sewage sludge to those areas (19–21%), meaning in lower MP pressure in their agricultural soils. Slovenia (1–2%), Finland (3–5%), and Croatia (3–5%) also have a low availability of agricultural soils for the spreading of sewage sludge, suggesting MP pressure may also be lower in these countries. Conversely, countries such as the Netherlands recycle almost none of their sewage sludge to their agricultural land (5–6%) due to concerns over the heavy metal contents in the sewage sludge, favouring other disposal methods such as incineration (Mininni et al., 2015).

In most European nations, the nitrogen content of the sewage sludge represents the limiting factor determining the rate of application of sewage sludge to the land, with a maximum of 250 kg of total nitrogen/ha/yr permitted for agricultural land. Based on applying the concentration of MPs contained in sewage sludge from the Nash WwTP, an estimated application rate of 4.8 g_{mp}/m^2/yr or 11,489 MP_p/m^2/yr is applied to European agricultural land. This exceeds the estimates using data from the EU Commission (2015, 2018) and Eurostat (2019a, 2019b) datasets which range between 30 and 800 MP_p/m^2/yr, likely due to the use of the maximum permissible amount of total nitrogen in calculations, representing the upper limit of spreading MP-laden sewage sludge onto agricultural soils. It should be noted that these estimates are based on a sewage sludge treatment analogous to the one at the Nash WwTP, and different treatment processes may vary the nitrogen content within the sewage sludge, altering the maximum permissible amount of sewage sludge/m^2/yr (McCarty, 2018; Van der Hoek et al., 2018). The estimations using the maximum permissible amount of nitrogen for agricultural land are similar to recent estimates by Nizzetto et al. (2016), who projected an average and maximum areal per-capita loadings of 2 and 80 g_{mp}/m^2/yr input onto European agricultural land, respectively.

3.4. Wider implications and future direction

At present, there remains inadequate solutions for the explicit release and control of MP pollution into the aquatic and terrestrial environment from WwTPs. The majority of research focuses on the MP removal efficiency of WwTPs processes, with PSTs being highly effective in this function, and whether MPs are recirculated to natural watercourses. Unfortunately, the lack of a strategy of Water Companies to manage MP waste present in the sewage sludge, leads to a recycling management in which these contaminants are transported back into the soil that will eventually return to the aquatic environment. This questions whether MPs are being removed at WwTPs or effectively shifted around the environment. In Europe, the use of sewage sludge on agricultural land has been limited by the nitrogen levels and can be prohibited if it contains high levels of harmful chemical contaminants, such as heavy metals, and pathogens (Kelessidis and Stasinakis, 2012). However, MPs are an emerging contaminant with an insufficient amount of research on the effects that MP exposure may have on soils, plants, and biota (Rillig et al., 2019; de Ruijter et al., 2020). There is currently no European legislation that limits or controls the MP input into recycled sewage sludge based on the loads and toxicity of MP exposure, thus disposing MP-laden sewage sludge onto agricultural land is and has been an accepted strategy worldwide (Gianico et al., 2021). There are limited exceptions such as Germany, which has some of the strictest fertiliser contamination regulations, and has placed upper limits on impurities such as glass and plastics, allowing up to 0.1% of wet fertiliser weight of plastics larger than 2000 μm (Weithmann et al., 2018). The presented results from the Nash WwTP (South Wales, UK) show that 1% of dry sewage sludge weight is contributed by MPs of size larger than 1000 μm, thus direct application of the sewage sludge from the Nash WwTP may be prohibited should adequate legislation were in place like in Germany's agricultural policy.

One immediate action that could be taken to control MPs from the entering sewage sludge is to avoid the mixing of surface scum with the sewage sludge, in which the scum can be recovered for biofuel (Bi et al., 2015; Cobb et al., 2020) and biogas production (Long et al., 2012), preventing up to 26,000 ± 18,000 g_{mp}/day or $1.6\times10^7 \pm 6.1\times10^6$ MP_p/day from accumulating in the sewage sludge (Fig. 6). Other alternatives to land application for sewage sludge include disposal to landfill, which is preferred in European nations such as Italy and Greece (Collivignarelli et al., 2019). However, MPs can leachate from landfill sites, contaminating the surrounding terrestrial and aquatic environment (He et al., 2019). Incinerating sewage sludge for energy recovery is also an emerging strategy for MP end-of-life management, whilst it can be questioned whether it is a sustainable solution because more energy is required to break down the plastics than can be recovered (Eriksen et al., 2018; Rollinson and Oladejo, 2019).

Before regulations on the limits of MP concentrated within sewage sludge and alternative solutions are discussed, there remains multiple knowledge gaps on the magnitude of the problem of MP pollution stemming from sewage sludge application. A more in-depth and widespread examination of the MP concentration in produced sewage sludge, including the shape, size and chemical composition of MPs, would improve our understanding of the fate and behaviour of MPs in soils. Additionally, understanding the storage of MPs in agricultural lands and their transport to aquatic environments via run-off or infiltration to groundwater is critical for estimating MP budgets in both environments, as it has been shown that up to 99% of MPs are transported away from the originally applied soils (Crossman et al., 2020). The influence that different WwTP sewage sludge treatments and population equivalent served may have on the abundance and composition of MPs in the sewage sludge should also be compared (Mahon et al., 2017). It is also necessary to carry out studies related to MPs in each WwTP process unit, as the selection of different treatment processes will significantly affect the removal rate of MPs in WwTPs. Finally, efforts should be made to increase standardised monitoring of MP concentrations in sewage sludge and agricultural soils, which would provide a more accurate picture of MP output from WwTPs and contamination levels in soils across Europe (Carr et al., 2016).

4. Conclusion

This study presents the quantification MPs, in terms of mass and abundance, in a primary settler tank (PST) at the Nash Wastewater Treatment Plant (WwTP) located in South Wales (United Kingdom). MPs 1000–5000 μm from raw incoming sewage were completely removed from the PST and separated by density into settled sludge, which contained 83% and 93% of MPs by mass and abundance respectively (128,000 ± 124,000 g_{mp}/day and $3.52\times10^8 \pm 1.56\times10^8$ MP_p/day), and surface scum which provided 17% and 4% of MPs by mass and abundance, respectively (26,000 ± 18,000 g_{mp}/day and $1.6\times10^7 \pm 6.1\times10^6$ MP_p/day). The scum and sludge portions are combined to generate sewage sludge, which was estimated to contain 0.01 g_{mp} or 24.7 MP_p per g of dried sludge, equating to about 1% of the sewage sludge weight.

On an international scale, adopting the data from the Nash WwTP and five other European studies, as well from the EU Commission (2015, 2018) and Eurostat (2019b) datasets with a maximum application rate of 4.8 g_{mp}/m^2/yr or 11,489 MP_p/m^2/yr of recycled sewage sludge to European agricultural lands, an estimate of between 72,000 and 11,000 T_{mp}/yr (based on MPs 1000–5000 μm in size) or 8.6×10^{13}–7.1×10^{14} MP_p/yr (based on MPs 25–5000 μm in size) are applied to European agricultural soils. These figures, based on the presented measurements from the Nash WwTP and five European WwTPs with MP particles ranging in size from 25 to 5000 μm, are conservative and are likely to be an underestimate. These data highlight the magnitude of the environmental problem of MP pollution derived from directly recycling sewage sludge into organic fertiliser, suggesting that agricultural soils are likely to be one of the largest environmental reservoirs of MP pollution due to the transfer of MPs from WwTPs to agricultural land.

Credit author statement

Conceptualisation and methodology by all authors. Experimental investigation conducted by **JL** and supervised by **VM**, **PO** and **CW**. Data curation and formal analysis by **JL**, **VM** and **PO**. Validation and manuscript drafted by **PO**, **VM** and **JL**. Data interpretation and final manuscript editing and approved by all authors. Supervision and funding acquisition by **PO** and **CW**.

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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