Environmental Pollution 261 (2020) 114198

Contents lists available at ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

Sewage sludge application as a vehicle for microplastics in eastern Spanish agricultural soils☆

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ARTICLE INFO

Article history:
Received 24 November 2019
Received in revised form
31 January 2020
Accepted 14 February 2020
Available online 17 February 2020

Keywords:
Microplastics
Agricultural soils
Sewage sludge

ABSTRACT

Microplastic pollution is becoming a major challenge with the growing use of plastic. In recent years, research about microplastic pollution in the environment has become a field of study with increased interest, with ever expanding findings on sources, sinks and pathways of microplastics. Wastewater treatment plants effectively remove microplastics from wastewater and concentrate them in sewage sludge which is often used to fertilise agricultural fields. Despite this, quantification of microplastic pollution in agricultural fields through the application of sewage sludge is largely unknown. In light of this issue, four wastewater treatment plants and 16 agricultural fields (0–8 sewage sludge applications of 20–22 tons ha^{-1} per application), located in the east of Spain, were sampled. Microplastics were extracted using a floatation and filtration method, making a distinction between light density microplastics ($\rho < 1$ g cm^{-3}) and heavy density microplastics ($\rho > 1$ g cm^{-3}). Sewage sludge, on average, had a light density plastic load of 18,000 ± 15,940 microplastics kg^{-1} and a heavy density plastic load of 32,070 ± 19,080 microplastics kg^{-1}. Soils without addition of sewage sludge had an average light density plastic load of 930 ± 740 microplastics kg^{-1} and a heavy density plastic load of 1100 ± 570 microplastics kg^{-1}. Soils with addition of sewage sludge had an average light density plastic load of 2130 ± 950 microplastics kg^{-1} and a heavy density plastic load of 3060 ± 1680 microplastics kg^{-1}. On average, soils' plastic loads increased by 280 light density microplastics kg^{-1} and 430 heavy density microplastics kg^{-1} with each successive application of sewage sludge, indicating that sewage sludge application results in accumulation of microplastics in agricultural soils.

1. Introduction

Microplastics are commonly defined as plastic particles with a diameter ranging from a few µm to 5 mm. Microplastic (MP) pollution has been detected in numerous environmental compartments, such as the marine, terrestrial and atmospheric environment (Allen et al., 2019; Andrady, 2011; De Souza Machado et al., 2018). Research has primarily been focussed on the marine environment and quantification of sources of MP pollution in the terrestrial environment has largely been lacking (Bläsing and Amelung, 2018; Nizzetto et al., 2016; Weithmann et al., 2018). Microplastics can have a negative impact on soil biota by increasing mortality rate and reducing growth and reproduction rates of soil life (Huerta Lwanga et al., 2016; Zhu et al., 2018).

Wastewater treatment plants (WWTPs) are receptors of microplastics derived from industries, domestic wastewater and stormwater (Mahon et al., 2016). Studies have shown that wastewater treatment plants (WWTPs) effectively remove microplastics from wastewater, with a removal rate of up to 99% (Bläsing and Amelung, 2018; Magnusson and Norén, 2014; Murphy et al., 2016; Sun et al., 2019), concentrating microplastics in sewage sludge. Sewage sludge is widely used as a fertilizer because of its richness in organic and inorganic nutrients, its soil conditioning effects and economic advantageousness (Nizzetto et al., 2016; Singh and Agrawal, 2008). However, the sewage sludge's microplastics remain in the soil much longer than the nutrients, posing a threat to individual soil life and potentially even soil ecosystems

☆ This paper has been recommended for acceptance by Maria Cristina Fossi.

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https://doi.org/10.1016/j.envpol.2020.114198

(De Souza Machado et al., 2018).

Spain has an extensive use of sewage sludge in the agricultural sector with 65% of its country's sewage sludge production being recycled through agricultural soils, while Europe's average lies at 40% (Roig et al., 2012).

First studies of plastic pollution in agricultural soils by application of sewage sludge were performed almost 15 years ago (Zubris and Richards, 2005). However, quantification of microplastic pollution in agricultural soils by application of sewage sludge remains largely absent, with perhaps a few exceptions (e.g. Corradini et al., 2019; Xu et al., 2020). To address this knowledge gap, this study has evaluated the microplastic content of 16 agricultural fields, the number of sewage sludge applications to these fields, the amount of sewage sludge applied and the microplastic content of sewage sludge of 4 WWTPs in the east of Spain.

Our hypothesis was that receiving sewage sludge leads to a significant increase in accumulation of microplastics in agricultural soils.

2. Materials and methods

2.1. Study site

The study area for this research is located in the vicinity of Valencia, in the east of Spain (Fig. 1). A total of 16 fields were sampled for soil of which 11 had a history of sewage sludge application. Cereals were cultivated on most fields, with the exception of two fields which were olive orchards (Table 1). Number of sewage sludge applications ranged from 1 to 8 applications and the application load was 20–22 dry tons ha^{-1} per application. 4 WWTPs were selected and sampled for sewage sludge, 3 of which were the sewage sludge source of the last application of some of the sampled fields. Information on the origin of previous sewage sludge applications could not be obtained. The WWTPs were chosen to obtain sewage sludge samples from WWTPs with a diversity of size, treatment types and socio-geographical location. The WWTPs were located near Albaida, Canet d'En Berenguer, Ontinyent and Sagunto. A more detailed description of the WWTPs is included in Table S1 in the supplementary material.

2.2. Sampling

Per field, soil samples were collected at 4 randomised points at a depth of 0–10 cm and 10–30 cm using a soil auger. This sampling depth was chosen to cover the active soil layer as soil life is concentrated in the top layer, and the rooting depth of cereals and ploughing depth do not exceed 30 cm. Roughly 100 g of soil was sampled and collected into sealable polypropylene bags. Interviews were held with landowners to rule-out land management practices that could influence the microplastic content of the soil e.g. application of plastic mulch or compost (Rillig, 2012; Van Schothorst, 2018). Per WWTP, 4 sewage sludge samples of roughly 250 g were taken and collected into sealable polypropylene bags.

2.3. Extraction of microplastics

Extraction of microplastics was performed by using a modified floatation method of Zhang et al. (2018) and identification was based on the circularity, transparency and shininess under heating. During all the laboratory work cotton lab coats were worn to limit the contamination of the samples from synthetic clothing. All the samples were dried for 72 h at a temperature of 40 °C. It was decided to use 3 ± 0.005 g per sample to limit the obscureness effect of organic materials in further analysis while still obtaining reliable data. Two extraction steps were performed on the same sample with different liquids, one with distilled water and another with a sodium iodide mixture. The former led to a floatation of plastic particles with $\rho < 1$ and the latter led to a floatation of plastic particles with $1 > \rho < 1.7$. From here on the different densities of plastic will be referred to as light density plastic and heavy density plastic respectively.

Fig. 1. Study area located in the east of Spain. Dots indicate soil sampling points.

Table 1
Details on number of sewage sludge applications, sewage sludge application rates and sources, crop cultivation and ploughing strategy of sampled fields.

Field number	Number of sewage sludge applications	Origin of last sludge application	Crop	Ploughing	Application rate (t ha-1)
1	0	–	Cereal	yes	–
2	0	–	Cereal	yes	–
3	0	–	Olive	no	–
4	0	–	Cereal	no	–
5	0	–	Olive	no	–
6	1	Sagunto	Cereal	no	22
7	3	Sagunto	Cereal	yes	22
8	3	Sagunto	Cereal	yes (conservation tillage)	20
9	3	Sagunto	Cereal	yes	20
10	4	Ontinyent	Cereal	yes	20
11	4	Ontinyent	Cereal	yes	20.5
12	5	Ontinyent	Cereal	yes	22
13	5	Ontinyent	Cereal	yes	22
14	6	Albaida	Cereal	yes	21
15	8	Sagunto	Cereal	yes (conservation tillage)	20
16	8	Sagunto	Cereal	yes	21

The subsample was put in a 50 ml laboratory tube, 40 ml of distilled water was added and then mixed using a Gerhardt Laboshake at 120 RPM for 2 h. The sides and cap of the tube were rinsed off, topped up to 50 ml and centrifuged for 10 min at 3000 RPM using a Heraeus Varifuge 3.0 to force sediments to settle at the bottom. Supernatant material was filtered using a Whatman 91 filter with an 11 μm pore size. Filters were taken aside and replaced when clogged, and stored in sealed petri dishes. These steps were repeated until no more floating material was observed with a minimum of 3 repetitions. After extraction of light density microplastics (MPs), the subsample was dried at 40 °C and heavy density MPs were extracted using the same procedure, but by using a 600 g l^{-1} NaI liquid ($\rho \sim 1.7$ g cm^{-3}) instead of distilled water.

2.4. Observation of microplastics

Filter residues were placed on a glass plate and 2 pictures were taken using a ZEISS Stemi 508 microscope, with a heating period of 8–10 s at 120–130 °C in between the picture taking. The pictures were compared and microplastics were identified, based on an increase in circularity, transparency and shininess after heating (Fig. 2, Zhang et al., 2018). The plastic particles' count, maximum diameter and shape (fibre, fragment or film) of the preheating picture were analysed using ImageJ photoshop software (Schneider et al., 2012). To get further insight on the type of plastic present in sewage sludge and soils a microscope coupled with a spectrometer was used to perform a micro Fourier transform infrared (μFTIR) analysis (Agilent Technologies Cary 600 series FTIR spectrometer). Five frequently found particles in the samples were scanned at 1000–3500 cm^{-1} wavelength with a 4 cm^{-1} resolution, measuring transmittance to record their absorbance spectra.

2.5. Data analysis

A Shapiro-Wilk tests showed a non-normal distribution of data ($p < 0.05$). Differences in quantities and size of MPs of sample groups were determined with Mann-Whitney U tests. Correlations in quantities and size of MPs of sample groups were determined with Spearman tests. Descriptive statistics were used to characterise previous test results. All the statistical analyses were performed in SPSS and a significance level of 0.05 was chosen for this study.

3. Results

3.1. Application vs. no application

Microplastics were found in 97% of the analysed samples. Soils without application of sewage sludge contained, on average, 930 ± 740 light density MPs kg^{-1} and 1100 ± 570 heavy density MPs kg^{-1}. Soils with application of sewage sludge contained, on average, 2130 ± 950 light density MPs kg^{-1} and 3060 ± 1680 heavy density MPs kg^{-1}. A significant difference in MP content was found between soils with a history of sewage sludge application and soils without sewage sludge application for both densities of plastic ($p < 0.05$). The size distribution of microplastics showed that the majority of the MPs are present in the lower spectrum of the microplastic size range (Fig. 3), with the largest number of MP

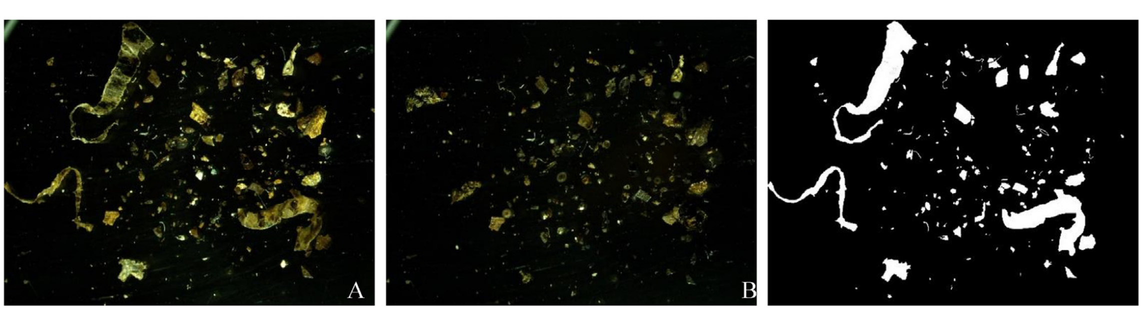

Fig. 2. Microscopic pictures of a sample before heating (A), after heating (B) and the processed image with plastic particles singled out (C).

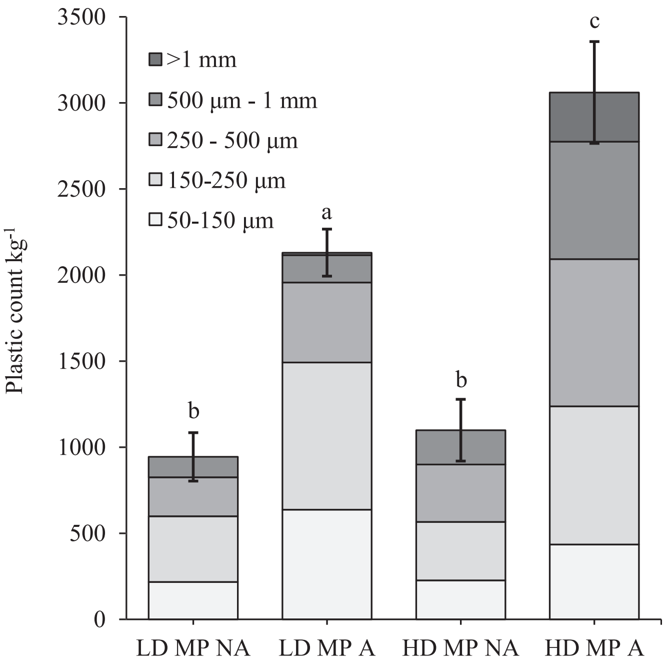

Fig. 3. Light density (LD) and heavy density (HD) microplastic (MP) counts per kilogram of dried soil of fields with application of sewage sludge (A, n = 88) and fields with no application of sewage sludge (NA, n = 40) and their respective standard error. Significant differences between sample groups (p < 0.05) are indicated with lowercase letters. No differentiation in depth was made in this analysis.

present in the size bin 150–250 μm.

Sewage sludge contained an average of 18,000 ± 15,940 light density MPs kg^{-1} and 32,070 ± 19,080 heavy density MPs kg^{-1}. The microplastic content of sewage sludge would result in a total average of 3.78×10^8 light density MPs ha^{-1} and 6.74×10^8 heavy density MPs ha^{-1} entering agricultural fields per application. Some differences in sewage sludge's MP content between WWTPs was observed, but no conclusive cause could be found (Table S2, supplementary material).

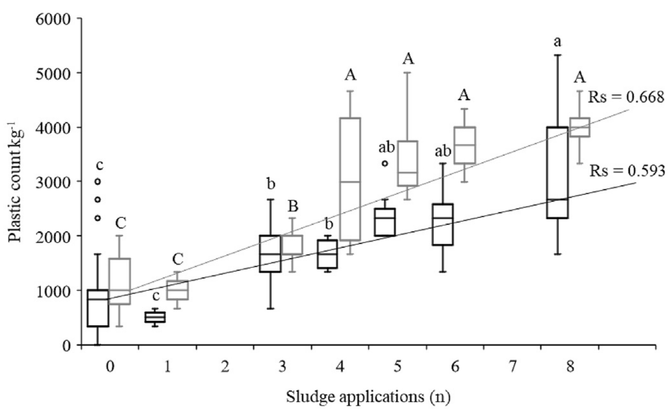

Fig. 4. Microplastic counts per kg dried soil by number of sewage sludge applications for light density plastic (black) and heavy density plastic (grey) n = 128. Boxplots showing median, 25%–75% range, range without outliers and outliers. Different letters indicate significant difference among sample groups (p < 0.05). Differences in light density plastic content are indicated with a lower case letter and difference in heavy density plastic content are indicated with capital letters. Lines are showing Spearman correlation coefficient.

3.2. Number of applications

On average, an increase in microplastic content was observed with each successive sludge application (Fig. 4). Spearman tests showed a significant positive linear correlation between the number of applications and MP content (Rs = 0.593 for light density plastic and Rs = 0.668 for heavy density plastic, $p < 0.05$). On average, soil microplastic content would increase with 280 light density MPs kg^{-1} and 430 heavy density MPs kg^{-1} dried soil with each successive sewage sludge application.

3.3. Differences in depth

No significant depth-dependent differences in MP load for both densities of plastic were observed within each sample group (Fig. 5). When only looking at fields were no ploughing was done, samples taken at fields with application of sludge at a depth of 0–10 cm had a light density MP load of 1730 ± 920 kg^{-1} and a heavy density MP load of 3410 ± 2330 kg^{-1}. Respective samples taken at a depth of 10–30 cm contained, on average, 1610 ± 920 light density MPs kg^{-1} and 3180 ± 2430 heavy density MPs kg^{-1}. For samples taken at a depth of 0–10 cm at fields were no ploughing was done and no sewage sludge was applied, a light density MP load of 960 ± 420 kg^{-1} and a heavy density MP load of 1140 ± 450 kg^{-1} was found. Respective samples taken at a depth of 10–30 cm contained, on average, 920 ± 480 light density MP kg^{-1} and 1070 ± 570 heavy density MP kg^{-1}. The slight difference in plastic content between depths for fields which were not being ploughed remained statistically not significant ($p > 0.05$).

3.4. Plastic types

For all sample groups, the majority of the present MPs were fragments (Fig. 6). In general, light density plastic had more fibres and less films than heavy density plastic. Plastics in soils that had not received sludge had the smallest share in fibres and films and

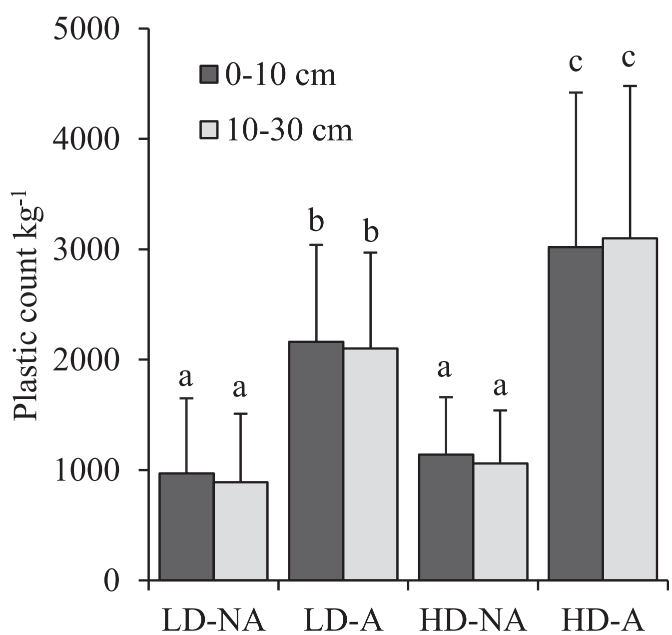

Fig. 5. A comparison of depth dependent light density (LD) microplastic count kg^{-1} and heavy density (HD) microplastic count kg^{-1} found in soils which had received no application of sewage sludge (NA) and soils which had received sewage sludge application (A). Significant differences ($p < 0.05$) between sample groups are indicated with lower case letters.

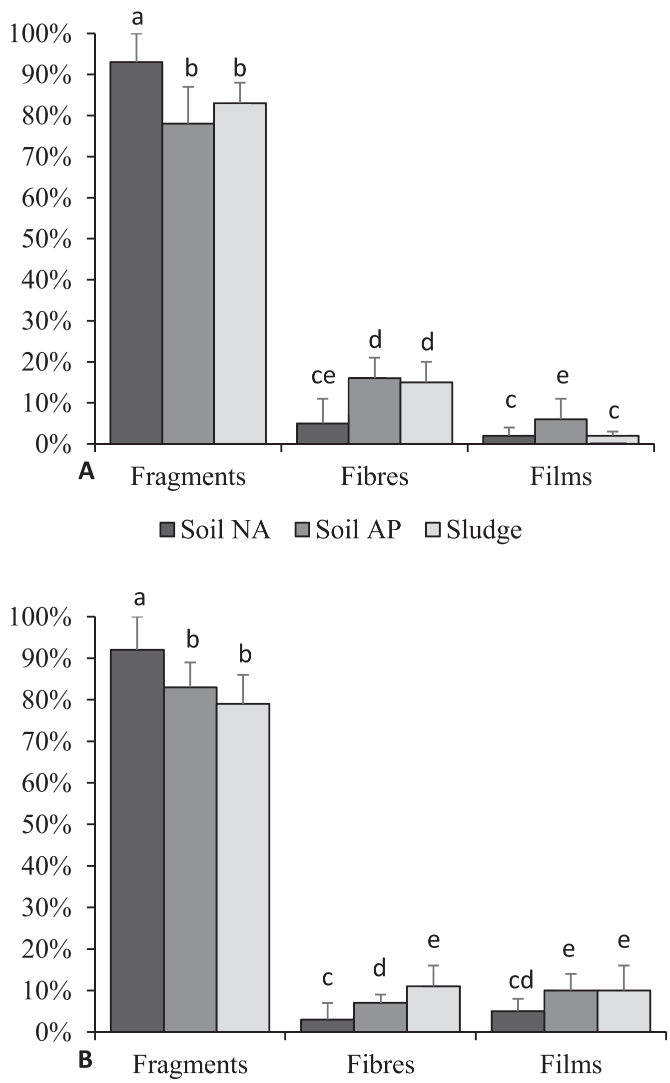

Fig. 6. A comparison of types of plastic found in soils which had received no application of sewage sludge (NA), soils which had received sewage sludge application (AP) and sewage sludge, for light density plastic (A) and heavy density plastic (B). Significant differences ($p < 0.05$) between sample groups are indicated with lower case letters.

the largest share in fragments out of all the sample groups. The μFTIR analysis showed that out of 5 particles, 3 were polypropylene (PP) and two were polyvinylchloride (PVC). The μFTIR results are shown in Fig. S1 in the supplementary material.

4. Discussion

4.1. Accumulation of microplastics in soils

Our findings strengthen the hypothesis that microplastics accumulate in the soil with each successive sewage sludge application. Soils with a history of sewage sludge application had, on average, a 256% higher microplastic content than soils without application of sewage sludge. Sewage sludge was found to be, by far, the most polluted entity of this study. The majority of the plastic particles found had a density >1 g cm^{-3}, underlining the importance of extracting heavy density MPs to fully grasp plastic pollution.

Other studies reporting microplastic content in sewage sludge showed slightly lower particle loads compared to the results found in this study. A study about numerous WWTPs in China reported an average of 22,700 ± 12,100 particles kg^{-1} sewage sludge (Li et al., 2018). Particle loads of studied sewage sludge in Ireland ranged from 4196 to 15,385 MP kg^{-1} (Mahon et al., 2016). In Chile, a median particle load of 34,000 particles kg^{-1} sewage sludge was found (Corradini et al., 2019). Seasonal variability, different socio-geographical locations and different treatment types may explain some of the variation found (Li et al., 2018; Mahon et al., 2016).

Comparing the plastic load of soils without application of sewage sludge with other studies proved difficult due to numerous uncontrolled sources of microplastics that are difficult to quantify. An example of the previous mentioned is the atmospheric transport and deposition of microplastics. Allen et al. (2019) reported atmospheric deposition rates of 365 MP m^{-2} d^{-1} in the Spanish Pyrenees. Furthermore, water and wind erosion are suspected to be transporters of microplastics (Hurley and Nizzetto, 2018; Rezaei et al., 2019), both acting as possible sources or sinks. Somewhat comparable studies were those of Van Schothorst (2018) and Huerta Lwanga et al. (2017). Van Schothorst (2018) found an average of 2341 ± 1248 light density MPs kg^{-1} for fields in Cartagena, Spain that applied biodegradable plastic mulch while this study found a much lower particle load. A more comparable particle load of 870 ± 1900 MP kg^{-1} was found in home gardens' soils in Mexico by Huerta Lwanga et al. (2017).

Corradini et al. (2019) reported a median between 1100 and 3500 MP kg^{-1} dried soil for fields in Chile with 1–5 applications. Which is 2 times lower than the range of medians found in this study (2000–7600 MP kg^{-1}) while the application rate was 2 times higher (40 tons ha^{-1}). It has to be noted that a denser extraction liquid was used in our study ($\rho = 1.7$ g cm^{-3} versus $\rho = 1.55$ g cm^{-3}) which could extract a wider range of plastic types. Furthermore, the authors reported an average of 34,000 MP kg^{-1} dried sludge while this study found an average of 50,000 MP kg^{-1} sludge.

No difference in MP load was found between the sampled depths. The majority of the sampled fields were being ploughed, mixing the top soil. Earthworms could transport and mix microplastics in the top soil even further (Huerta Lwanga et al., 2016). Furthermore, it may be speculated that a downward water flow may also wash out MPs to deeper soil layers or even the groundwater (Hurley and Nizzetto, 2018; Yu et al., 2019).

The majority of the MPs found were fragments for all sample groups. A significant difference in fibre content was observed between soils that had received sewage sludge and soils that had not received sewage sludge, reaffirming that synthetic fibres can be used as an indicator for past sewage sludge application (Zubris and Richards, 2005).

The found results show that microplastics accumulate in the terrestrial environment. This raises the question to what extent microplastics are being remobilised and what pathways they could take. Studies indicate that micro sized plastics are too big to be taken up by plants, but when microplastics degrade into smaller pieces and certain size thresholds are reached, plants can take up and transport these particles (He et al., 2018; Ng et al., 2018). Plant species vary in their uptake, translocation and accumulation of contaminants due to a range of anatomical and physiological differences (Ng et al., 2018). However, to the best of our knowledge, no study has been undertaken to look at uptake of microplastics by cereals specifically. In Spain, sludge is widely used on cereal fields and therefore looking at the uptake and transport of nano sized plastic particles within cereal plants would be an interesting topic for future research.

4.2. Method limitations

One of the main benefits of the chosen method was that it provided a simple and low-cost technique to extract and identify microplastics. However, the identification of microplastics remains an arbitrary procedure in which human error could have resulted in false positives. Previous studies have shown that presence of organic materials hamper the analysis of taken pictures (Van Schothorst, 2018; Zhang et al., 2018). In this study some pictures were clouded by organic materials, complicating the analysis. We would propose the use of an extra extraction step with a liquid with a density in between most organic materials and plastics. Ethanol is deemed a suitable liquid, however the efficiency of removing organic materials and the effect on plastic particles stands unstudied. Furthermore, the use of a centrifuge (3000 RPM) resulted in a speed up settling process of sediments and turned out to be a considerable timesaving component for this methodology.

There is no generalised protocol to report microplastic content. Microplastics are being reported per surface area, per weight, per volume or as a weight ratio (Cole et al., 2011; Corradini et al., 2019; Horton et al., 2017) hampering comparison between studies. It is crucial to introduce standardised units to promote data exchange and comparison. It was therefore decided to report our results in particles per weight as proposed by Horton et al. (2017).

No method validation could be performed due to the lack of natural blank samples. As previously discussed, even soils without application of sewage sludge contained plastic particles, making them unsuitable as blank. Zhang et al. (2018) reported 90% recovery rates and it is expected that our method harvested similar recovery rates. Some contamination of samples was observed during the picture analysis. This is a renowned issue across different microplastic studies (Corradini et al., 2019; Horton et al., 2017; Zhang and Liu, 2018) and taking a larger amount of soil/sludge would limit the influence of contamination on the results.

5. Conclusion

Application of sewage sludge causes increased presence of microplastics in agricultural soils. By evaluating fields with varying numbers of sewage sludge applications, evidence was found of accumulation of microplastics with each successive application. Quantifying transport of microplastic within fields and remobilisation to other environmental compartments pose a difficult yet essential task to understand the fate of plastic in the overall environment. Currently, there are no threshold values for microplastics in soils leading to a negative impact on soil quality.

CRediT authorship contribution statement

Pim van den Berg: Data curation, Formal analysis, Investigation, Software, Writing - original draft, Writing - review & editing. **Esperanza Huerta-Lwanga:** Conceptualization, Supervision, Methodology, Writing - review & editing. **Fabio Corradini:** Data curation, Formal analysis, Investigation, Software, Writing - original draft, Writing - review & editing. **Violette Geissen:** Writing - review & editing.

Acknowledgements

We would like to thank EPSAR and Global Omnium for granting access to several wastewater treatment plants and allow sampling of sewage sludge. Furthermore, we would like to thank landowners of the sampled fields for allowing sampling at their fields and providing information about management practices. Lastly, we would like to thank everyone else who contributed in some way.

This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2020.114198.

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