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Transfer and transport of microplastics from biosolids to agricultural soils and the wider environment

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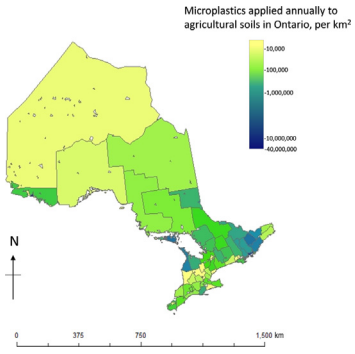
HIGHLIGHTS

- Microplastic (MP) concentrations of up to 1.4×10^4 MP kg^{-1} were found in biosolids.
- Storage of biosolids should be explored as a method for reducing biosolids' MP content.
- Some MP fibers are retained in soils, while fragments are predominantly exported.
- >99% of MPs applied from biosolids were likely exported to the aquatic environment.
- Biosolids application legislation may need revisions to incorporate MP management.

GRAPHICAL ABSTRACT



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ABSTRACT

Between April to November of 2017, microplastics (MPs) were analysed in biosolids from two separate suppliers, and in the soils of three agricultural fields to which they were applied, in Ontario, Canada. Soils of a control site with no history of biosolid application were also examined. High MP concentrations of between 8.7×10^3 MP kg^{-1} and 1.4×10^4 MP kg^{-1} were found in biosolids samples. Lower MP concentrations observed in Provider 2 biosolids may be due to storage, settling and supernatant removal prior to applications. Annual MP additions to agricultural soils across Ontario were estimated at between 4.1×10^{11} and 1.3×10^{12} particles. All fields receiving biosolids had higher soil pre-treatment MP concentrations than the control. The field with the greatest number of previous biosolid treatments had the highest pre-treatment soil MP concentrations; suggesting some MP retention in soils between applications. Immediately following biosolids applications, two fields demonstrated significant increases in soil MP concentrations, with preferential retention of MP fibers over fragments observed, while a reduction in soil MP concentrations were observed in the third. Surprisingly, only one field demonstrated a net gain in soil MPs over the course of the study. At all three fields, >99% of MPs applied in biosolids in 2017 were unaccounted for. The study suggests that despite adhering to applicable legislation, biosolids applications at all sites likely result in high rates of MP export. This study is the first to track MP transport through soils following their application in biosolids, and contributes to filling current knowledge gaps regarding export of MPs to aquatic systems from the terrestrial environment.

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1. Introduction

Plastic items with a longest dimension smaller than 5 mm are defined as microplastics (MPs) and include both intentionally manufactured particles (primary MPs) and those originating from the breakdown of larger macroplastics (secondary MPs). Although secondary MPs are more common in the environment, primary MPs have gained the greatest public attention, and been the target of most legislation and management (Rochman et al., 2015; Xanthos and Walker, 2017). Due to their ubiquitous presence in environmental matrices and potential interaction with biota, MPs have emerged as a serious global concern.

While the generation of MPs is largely terrestrial, their transmission to the aquatic environment is believed to be driven primarily by urban surface runoff and wastewater discharge (Galafassi et al., 2019). Large volumes of MPs can be removed, however, when stormwater runoff, industrial effluent and household drainage pass through wastewater treatment plants (WWTP) (Carr et al., 2016). Despite this, MP concentrations in lakes and oceans continue to rise; there are an estimated 2.69×10^5 tons of MPs in the ocean (Xanthos and Walker, 2017), and an average of 4.30×10^4 MP km^{-2} in the surface waters of the Laurentian Great Lakes (Eriksen et al., 2013).

Once removed from wastewater, MPs culminate in biosolids; a nutrient-rich semi-solid waste product, created during the wastewater treatment process. In many countries, biosolids are applied to agricultural lands as a soil amending agent and fertilizer (Nizzetto et al., 2016a). It has been calculated that each year farmed soils in North America may receive to up to 300,000 tonnes of MPs through biosolid applications (Nizzetto et al., 2016a), however very little is known about their fate and transport through the terrestrial environment (Nizzetto et al., 2016a; Rocha-Santos and Duarte, 2015; Rillig, 2012; Ng et al., 2018; de Souza Machado et al., 2018). Specifically, no empirical information is available on whether subsequent to biosolids application, the soils act as an MP source to rivers and lakes, with MPs moving laterally towards watercourses; or whether soils act as a sink, with downward vertical movement dominating. Under repeated biosolids applications, some MPs may be retained in soils, which could result in significant MP accumulation over time (Brodhagen et al., 2017; Corradini et al., 2019), although MP retention has not yet been quantified. While the notion of a soil MP 'store' might be desirable for reducing transfer of MPs to aquatic systems, the permanence of such a store and its impacts on plant growth, soil biota and overall soil health are largely unknown.

Through the analysis of 1300 soil samples collected before, during and after biosolids applications from three cultivated fields and from one control site in 2017 (Fig. 1), in addition to samples of the biosolids applied, we aimed to quantify the spatial and temporal variability in MP fate and transport through agricultural soils in Ontario, Canada. Key objectives were to 1) identify MPs in biosolids applied to agricultural soils; 2) quantify impacts of biosolid applications on the MP content of agricultural soils; and 3) to identify biosolids management strategies to reduce MP inputs to agricultural soils.

2. Methods

2.1. Site description

Four agricultural fields in Ontario, Canada, were studied. Three active treatment sites were sampled (Field 1, Field 2 and Field 3) where biosolids had been previously applied between 2012 and 2016, and where applications were planned for 2017. A fourth control field with no history of biosolids use was selected within the same region. Proximity was important for minimizing between-site variability in exposure to airborne MP contamination. All sites were located proximal to tributaries which drained into large economically important waterbodies (Lake Simcoe and Lake Scugog) and were situated in sandy loam soils (Fig. 1).

Field 1 (10.24 ha) and Field 3 (34.4 ha) were treated with biosolids from the same supplier (Provider 1) whereas biosolids applied to Field 2 (26.1 ha) were supplied by a separate haulage company (Provider 2). Although the two providers obtained biosolids from different WWTPs, all treatment plants had separate stormwater and sanitary systems and served similar sized cities (≈140,000 people). There are, however, notable differences in biosolid treatment and storage methods between WWTPs. The WWTP supplying Provider 1 anaerobically digests solids in mixing tanks, whereas the WWTP supplying Provider 2 uses an additional aeration pre-treatment process to promote bacterial breakdown. Provider 1 transports biosolids directly from the WWTP to the fields on the day of application and incinerates waste from fall to spring when biosolids spreading is prohibited. Provider 2 collects biosolids from a centralized facility, used year-round by the WWTP for biosolid storage and settling (Albert, 2013), where the top liquid supernatant is syphoned off and returned to the WWTP for additional treatment. Thus, Provider 2 uses only the thicker portion of the settled biosolids for land applications.

In Ontario, biosolids may be either surface applied or injected into the soils, although restrictions exist as to time and conditions of use (Ontario Nutrient Management Act, 2002). Permitted application rates vary depending on soil saturation levels, season, proximity to watercourse and slope. Opportunities to apply biosolids were limited in early 2017 by heavy and frequent rainfall, resulting in soil saturation. Soil conditions improved sufficiently by April 27th for treatment to proceed at Field 3, where Provider 1 applied biosolids to the surface using precision methods with a tanker and hose, simultaneously applying and ploughing biosolids into the top soil layer. At the other two treatment sites, conditions were unfavorable for surface application until May 23rd (Field 2, Provider 2) and June 13th (Field 1, Provider 1), by which time crops had already been planted, precluding post-application ploughing. At all sites, heavy rainfall events combined with high runoff volumes were observed shortly after application. Application rates and total amounts of biosolids applied are summarized in Table 1.

2.2. Sample collection

A cylindrical stainless steel corer, 5x8cm was used to extract soil samples from three depths; 0–5 cm, 5–10 cm and 10–15 cm. Soil samples were not taken below 15 cm due to heavy soil compaction at all sites at this depth, making MP transport below this level unlikely. The 5 cm corer was chosen to minimize soil compaction during sampling. Samples from each profile depth were separately wrapped in aluminum foil on site, and frozen until analysis. Soil samples were taken before, during and after biosolid application (Table S1). Following biosolids application, soil samples were taken monthly and following heavy rainfall events. Coring locations were selected according to observed field characteristics. Fields 2 and 3 had relatively steep slopes and a clear ridge and furrow pattern. At these fields, on each collection date 11 cores were taken across a transect at each profile depth (between two ridges and furrows), and four additional cores collected at each profile depth across ridges and furrows at the highest and lowest points of the field. Field 1 and the control site were predominantly flat, with less variable microtopography (i.e., no ridge or furrow patterns). In these fields, 14 soil cores were taken at each depth on each collection date. Locations of soil cores in these fields were randomly selected, although the existence of dense alfalfa crops in Field 1 restricted sampling to the southern field perimeter. Biosolid samples were also collected directly from the trucks and hoses during application. Three to five liters of biosolids were collected during each event and stored in a fridge prior to analysis.

Due to highly localized precipitation patterns and biosolids application times, sampling dates did not necessarily coincide at all sites. Precipitation was monitored using electronic tipping bucket rain gauges

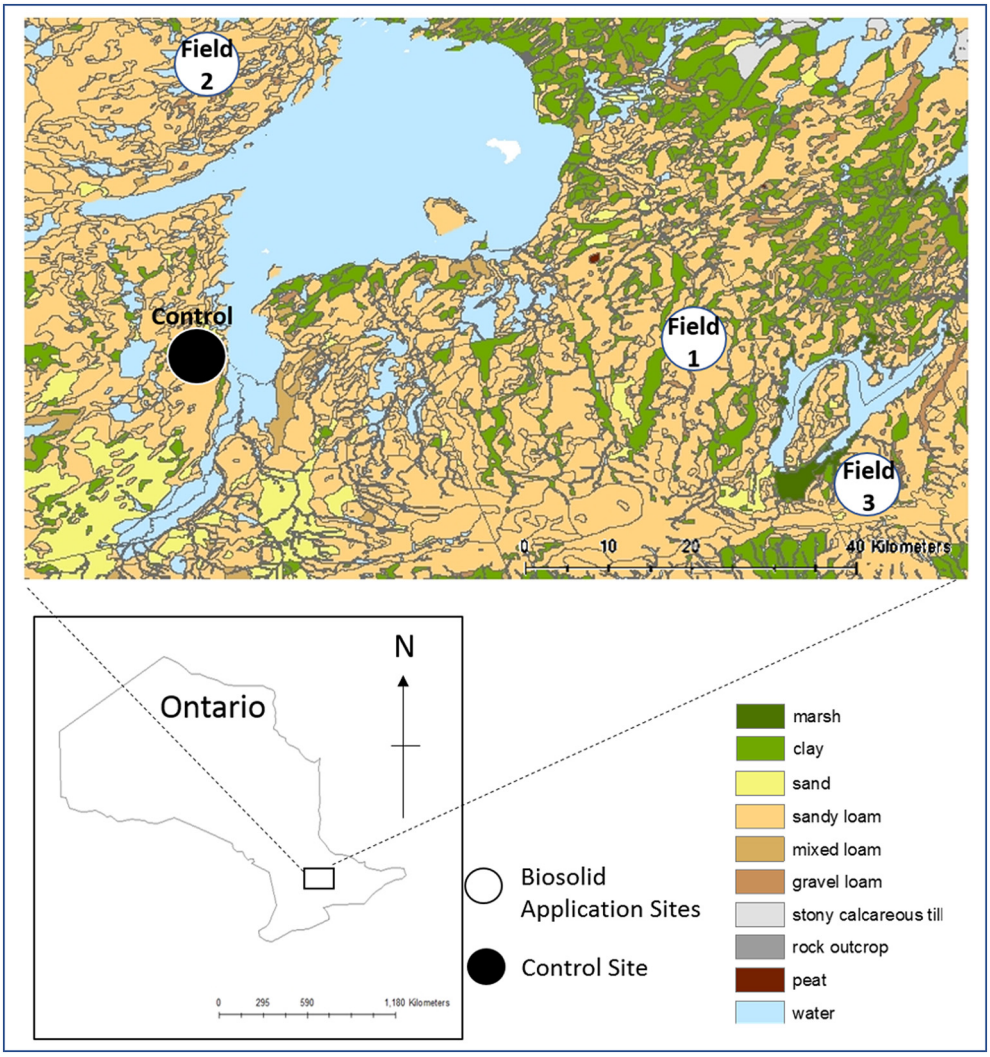

Fig. 1. Study sites, Ontario, Canada. Soils data derived from Soil Landscape of Canada Working Group (2010). Only approximate field locations are provided to ensure landowner anonymity.

installed in Fields 1 and 2. At the Control site and Field 3, established meteorological stations were used which were located within close proximity.

As with any field study, there remain limitations to the numbers of samples which can be collected. For example, fewer samples were taken from Field 3 due to limited site access. Every effort was made however to obtain a representative sample of the soil landscape, with over 1300 cores collected across the four fields. Through taking such a large number of samples, we incorporated variance in soil MP content across a range of soil depths, slope, ridges, furrows and general patchiness in soil matrices. This is the first MP study conducted within the soil environment at such a large spatial and temporal scale.

2.3. Analytical methods

At the University of Windsor, Ontario, soil samples were defrosted and 10 cm^3 selected for analysis from each core. This volume was weighed, and then placed in an oven at 65 °C until dry (with a minimum drying duration of 24 h). During drying, samples were covered with foil lids, with small holes inserted, to minimize likelihood of contamination.

Table 1
Biosolid application rates, and total amounts of MPs applied.

Site	Distance to water	Area (ha)	Biosolids provider	Biosolids application rate[a] (m^3 ha^{-1})	Application method	Total biosolids applied (m^3)	Biosolids dry weight MP concentration (MP kg^{-1})	Biosolids dry weight MP mass (mg kg^{-1})	Biosolids wet weight MP content (MP L^{-1})	Total number of MPs applied to field
Field 1	<150 m	10.24	1	78	Surface	799	11,469.4	Min: 1180 Max: 1276	686	5.48×10^8
	>150 m	N/A	1	N/A						
Field 2	<150 m	3.79	2	75	Surface	3183.2	8678	Min: 626 Max: 654	275	8.74×10^8
	>150 m	22.39	2	130						
Field 3	<150 m	3.67	1	74	Surface with post-application ploughing	4258.7	14,407	Min: 471 Max: 536	884	3.77×10^9
	>150 m	30.67	1	130						

[a] Biosolid application rates for Field 1 and 3 were supplied by Provider 1. Application rates for Field 2 were derived from regulatory guidelines (Ontario Nutrient Management Act, 2002).

Soil dry weights were calculated, and a composite sample for each field and sampling event created by combining dry samples from the same depth; resulting in 3 dried soil samples for each site sample date; vis, a 0–5 cm, 5–10 cm and 10–15 cm sample. Total volumes of biosolids sampled at each field were measured in a volumetric flask. Samples were poured into aluminum trays, covered, oven dried at 65 °C and then weighed. Temperatures were maintained ≤65 °C at all times to minimize plastic degradation. Finally, all dried samples were wrapped in aluminum foil, inserted in paper sampling bags, and shipped to the Norwegian Institute for Water Research (NIVA) for separation and identification of MPs.

As MPs in this study are encased in organic matrices (soils and biosolids), several processing steps are required for extraction including organic matter digestion, density separation, microscopic selection of MPs and identification of polymer types. All samples were first treated to remove organic matter using Fenton's reagent for digestion, following Hurley et al. (2018). MPs were then isolated from the sediment matrix through a series of density separations (SI). Following extraction, MPs were first visually identified using a Nikon SMZ 745T stereomicroscope at 20–40× magnification. Larger microplastics >300 μm, excluding fibers, were analysed on an Agilent Cary 630 single bounce ATR-FTIR. All smaller particles and fibers were analysed on a PerkinElmer Spotlight 400 μFT-IR. Spectra were compared to several libraries: including the Agilent Polymer and Elastomer, Oring and Seal ATR, PerkinElmer ATR Polymer, and BASEMAN libraries (Primpke et al., 2018). The a, b and c axis of each identified particle was recorded and used to establish particle volume, to facilitate a mass-based conversion between particle counts and particle masses. A detailed methodological description is provided in SI, including quality assurance and control (QA/QC) measures. Raw MP data are reported in Tables S5 and S6.

2.4. Contamination control

Throughout the study, care was taken to minimize MP contamination. During sample collection and drying, only glass or metal containers and instruments were used. Containers and instruments were prewashed with millipore water. Samples were covered where possible using aluminum foil or glass. At NIVA, sample processing and analysis was performed using a series of QA/QC measures, including measures to limit contamination, and the use of controls (SI), both positive (spiked matrix samples) and negative (blanks). The positive controls confirmed high extraction efficiencies for different particle types (PE beads: 97.3% ± 4.67%; PVC fragments: 92.7% ± 6.17%; Car tyre particles: 88.2% ± 5.75%; PET fibers: 80.0% ± 9.53%). For negative controls, only cellulosic particles were observed in biosolid samples, although viscose fibers were identified in soil samples. As viscose fibers represent <3% of MP types found within the soil samples, they were excluded from the dataset to account for a possible lab contamination source.

2.5. Data analysis

Three replicates of each composite soil sample were analysed, with dry weights between 19.3 and 30.8 g. The size, type (fragment/fiber), and polymer of each MP was recorded. Concentrations of MPs kg^{-1} in dried biosolid or soil were calculated by dividing the number of MPs in the replicate by the dried weight of the sample analysed. MP concentration L^{-1} of biosolid was calculated by dividing the number of MPs in the sample by the volume of sample analysed. Average MP concentrations and standard errors were calculated for the three replicates. Changes in MP distribution were calculated as the difference in MP soil concentrations at each soil depth between the baseline (before biosolid application) and date of sampling. MP accumulation was calculated as the sum of observed changes in MP distribution (positive and negative) over all soil depths, across the study period.

3. Results and discussion

3.1. Microplastics in biosolids

Microplastics were found in all biosolid samples (Fig. 2, Table 1). The highest dry and wet weight concentrations were found in biosolids supplied by Provider 1, applied to Field 1 (11,469 MP kg^{-1} or 686 MP l^{-1}) and Field 3 (14,407 MP kg^{-1} or 884 MP L^{-1}). The lowest concentrations were found in biosolids supplied by Provider 2, and applied to Field 2 (8678 MP kg-1 or 275 MP L^{-1}), and were significantly lower than those applied to Field 3 ($p < 0.01$). This range of concentrations is similar to those reported elsewhere (Primpke et al., 2018). Total numbers of microplastics applied to the fields ranged between 5.5×10^8 and 3.8×10^9 (Table 1).

The lower MP concentrations in biosolids from Provider 2 may be a result of several factors. First is the possibility of a difference in initial MP inputs to WWTPs; although treatment plants serve similar population sizes and have separate stormwater and sanitary systems. Second is the potential for differences in MP removal efficiency, i.e., the WWTP supplying Provider 2 may release more MPs in effluent and retain less in biosolids. High spatial and temporal variability in removal efficiency has been reported previously, e.g. between 54 and 91% MP removal efficiency was reported over 5 days in a Turkish WWTP (Gündoğdu et al., 2018) compared to a much more consistent 97–99% efficiency observed over a 13 day period in Canada (Gies et al., 2018). While this could partially explain the two-fold differences in biosolid MP concentrations measured in this study, short term variability in removal efficiency would likely have a minimal impact on biosolid MP concentrations, as biosolids are created from cumulative contributions from effluent flow over time. The third possibility is the difference in storage mechanisms; whereby Provider 2 used biosolids only after long-term storage and settling, and Provider 1 used materials directly from the WWTP. This settling process, in which the liquid fraction is syphoned off for re-treatment, equates to a form of density separation. As many plastic polymers are less dense than water (Nizzetto et al., 2016b), it is likely that MPs would be removed from biosolids during this process. The results indicate that long-term storage could be a possible mechanism for biosolid MP concentration reduction, and further research is therefore required to isolate the mechanisms causing variability in biosolids' MP concentrations between sources.

Several of the polymers identified in biosolids were found to be common between sources (Fig. 3, Table S2); polyethylene was found in similarly high concentrations in all samples from both providers, ranging from 4772 MP kg^{-1} (Provider 1) to 3926 MP kg^{-1} (Provider 2). Polypropylene, acrylics and polyesters were also found in high concentrations in all biosolids samples. Concentrations of other plastics common to all samples e.g. silicone, polyurethane, and ethyl-vinyl acetate were significantly lower in biosolids from Provider 2, at less than 30MP kg^{-1}, compared to up to 870MP kg^{-1} in biosolids from Provider 1. Furthermore, some plastic polymers found within Provider 1 biosolids samples at over 100MP kg^{-1} were entirely absent in biosolids from

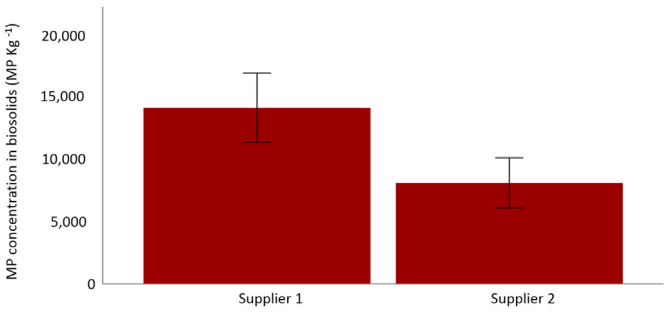

Fig. 2. MP concentrations in biosolids observed during 2017. Total samples analysed = 11. Error bars represent ± 1 Standard Error.

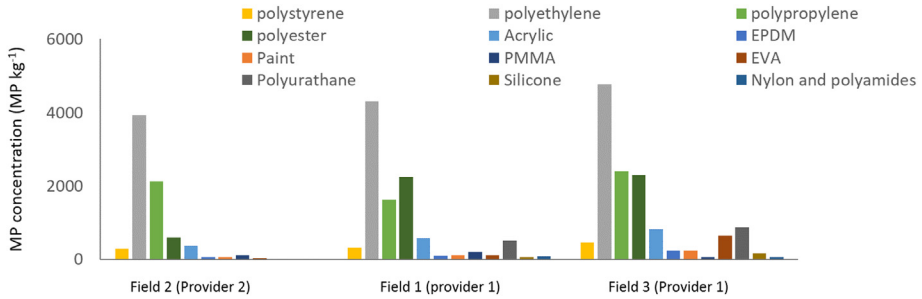

Fig. 3. Common types of plastics found in all biosolids from both providers applied to different fields.

Provider 2, including polycarbonate, polybutylene terephthalate, ethylene propylene rubber (EPR) and fluoroelastomers. These differences may again be due to the storage and density separation methods of Provider 2; since several of these polymers have a density less than or close to that of water, they are likely to have been preferentially removed during the settling process prior to application. Differences could also result from contrasting industrial inputs to the WWTPs, with Provider 1 receiving inputs from the automotive industry, and manufacturers of mining and railway equipment, steel fabrication and rubber products; and Provider 2 receiving inputs from manufacturers of motor vehicle parts, plastics, cement and concrete products.

All biosolids applied to Fields 1, 2 and 3 contained a similar dominance of fragments over fibers, with fragment content ranging from between 63% (Field 1) and 73% (Fields 2 and 3). This supports existing observations of preferential release of fibers in WWTP effluent waters (Sutton et al., 2016) and retention of fragments in biosolids (Weithmann et al., 2018).

3.2. Microplastics in soils

Soil MP concentrations prior to biosolid application were significantly different, both between depths and between sites (Fig. 4). Soils at Field 3 contained the highest number of MPs before application (average 541 MP kg^{-1}), with the control having the lowest (average 4 MP kg^{-1}). Biosolids were previously applied in 2013 and 2015 to Field 3, but only on one prior occasion on Fields 1 and 2; and there was no record of applications at the control site. High residual MP levels at Field 3, both in terms of MP numbers and MP mass (Table S3) may therefore represent accumulation of MPs from previous applications. Such accumulations have been hypothesized through laboratory experiments (Zubris and Richards, 2005) although not previously empirically observed in the environment.

Similar to the biosolids, high proportions and high concentrations of polypropylene, polyethylene, polyesters and acrylic were found in soils at all treated sites (Table S4). Soils also contained some of the polymers which were unique to individual biosolids providers. For example, poly butylene terephthalate (PBT) was found only in biosolids and soils from

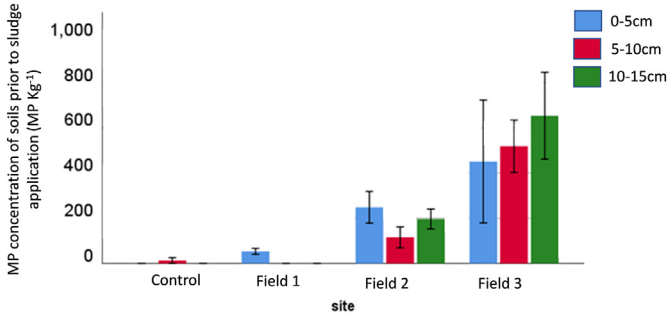

Fig. 4. Total MP concentrations in soils prior to 2017 biosolid application. Error Bars = ±1 standard error.

Provider 1, and polyvinyl chloride acetate (PVCA) was found only in biosolids and soils from Provider 2. The similarity in MP polymer types between individual biosolid sources and the soils to which they were applied indicates direct transfer and retention of plastic polymers between biosolids and agricultural soils. Much higher proportions of polyester MP fibers were found in soils (41–45%) (Fig. S1) than in original biosolid applications (8–21%). The higher dominance of polyester MP fibers in all soil sites, including the control, compared to biosolids, indicates that polyester MPs from an alternate source, e.g. atmospheric deposition, are preferentially retained in the soil profile.

Following biosolids applications, an immediate increase in soil MP concentration was observed at both Field 1 and Field 2 (Fig. 5), whereas no increase was seen in Field 3, despite the higher MP concentration and quantity of biosolids applied at this site. At Field 1, elevated soil MP concentrations persisted throughout the study period, amounting to a net increase of 41% compared to pre-treatment concentrations (Table 2); although after 2 months the majority of the increase was limited to the bottom 10–15 cm, in contrast to the upper soil layer (0–5 cm) that displayed a partial loss of MPs during this period. Biosolids were applied to this field via surface spreading on established crops, with no subsequent ploughing; and the initial MP accumulation in upper soil layers, followed by its decline and sustained accumulation in deeper layers is consistent with a downward movement of MPs. Unique polymers found both in the biosolids and soil profile indicate that MPs found in Field 1 originate from Provider 1 (Table S4).

In Field 2 biosolids applications were also surface spread without ploughing, subsequent to crop planting. Initial MP enrichment in upper soil layers was also observed here for seven weeks. Beyond this timeframe, a net loss of MPs compared to the pre-application phase was observed from the top layer. Unlike Field 1, however, this loss was not matched by concomitant MP increases in lower layers, suggesting limited MP infiltration, followed by removal during runoff events. Throughout the study period, a net loss of 30% MPs was observed throughout the soil profile at this site (Table 2). In Field 3 the sustained MP losses of 45%, and absence of unique MPs matching Provider 1 biosolids, suggests that the majority of MPs from biosolids were removed during early surface flushing events following application. Field 3 was the only field where biosolids were applied with ploughing and prior to crop establishment. High rainfall following biosolids application may account for the lack of MP accumulation during 2017. Considering the high historical MP accumulation in soils in Field 3, however, it appears that such flushing is not a common occurrence at this site.

As between 5.5×10^8 and 3.8×10^9 MPs were applied through biosolids to each site during the study period, this small net increase, and large net reductions in soil MP concentrations indicate that between 99% (Field 1) and >102% (Fields 2 and 3) of the MPs applied in biosolids were unaccounted for during soil sampling, and were likely ultimately exported from the soils, where >100% indicates that both biosolids and existing baseline MPs were removed. Surprisingly, net soil MP accumulation was lowest where the highest biosolid MP concentrations and volumes were applied (Field 3; Table 2), indicating that the response of soil MP concentrations to applications through biosolids is not solely

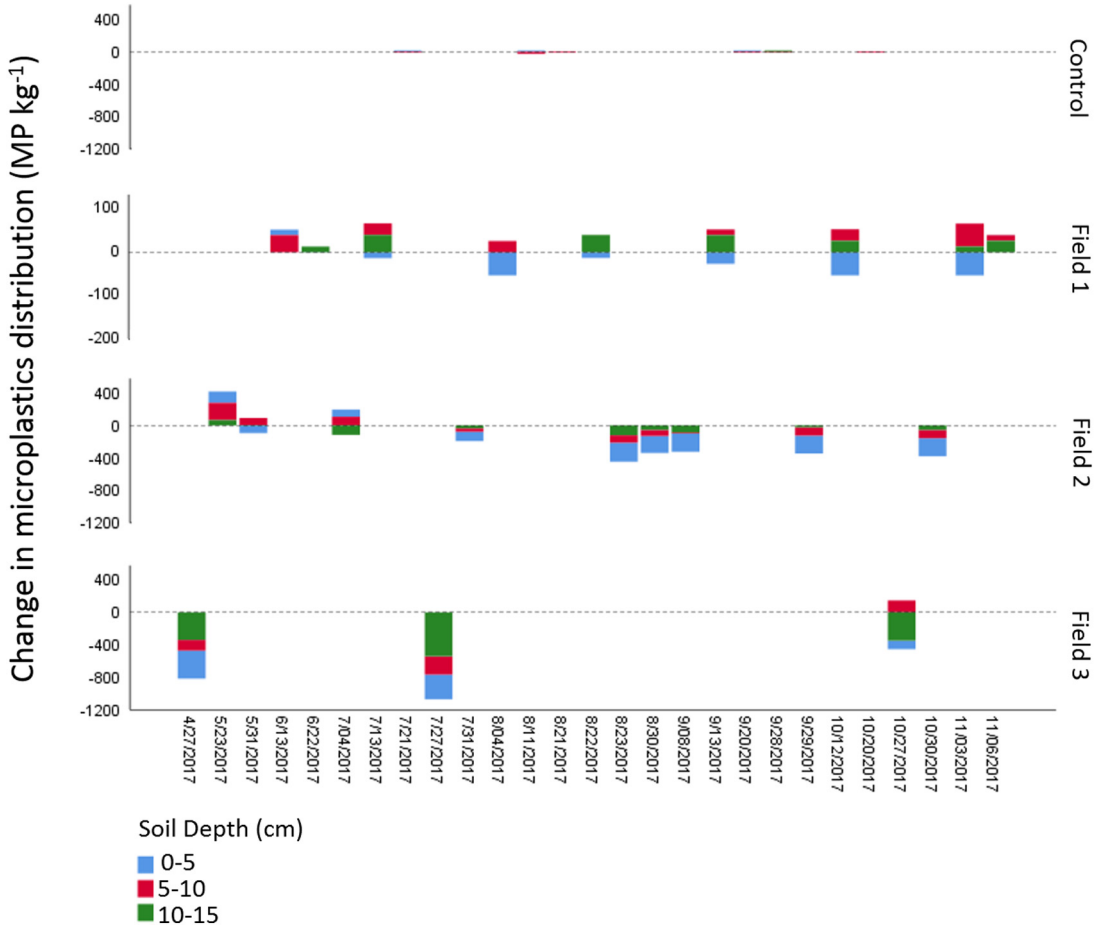

Fig. 5. Change in distribution of microplastics over time by soil depth. The change in distribution is calculated as MP numbers counted in soils prior to biosolids applications, subtracted from those identified after its application. Negative values indicate a reduction in MPs compared to the baseline; positive values indicate an increase. Note the larger scale axis for Field 1.

driven by the quantity of material used, but is influenced by a number of factors which affect subsequent MP mobilization within the soil. For instance, there is significant negative correlation between soil MP retention and soil wet density (Fig. 6). Fields 1 and 2 which retained MPs at deeper layers (Fig. 5), and which lost fewer MPs throughout the study period (Table 2) had significantly lower soil density than Field 3; which demonstrated no retention of MPs. The soil density is associated with its hydraulic properties. With higher density, and by association, fewer void spaces, MPs in Field 3 may have been subject to greater losses from surface runoff (Zemke et al., 2019) particularly during the unusually high rainfall events of 2017, with accumulation demonstrated in much drier years (Fig. 7). Applications of biosolids at Field 3 also occurred much earlier in the year (April) which likely increased the MPs exposure to these high rainfall amounts and saturated soils. Precipitation in April to June was more than double that in July; thus in Fields 1 and 2 where biosolids were applied later in the season, they were exposed to less rainfall, lower soil saturation, and were more easily able to move vertically through the soil profile. Pre-establishment of crops (Fields 1 and 2) could also facilitate vertical transport through bioturbation (Horton and Dixon, 2017) and preferential flow pathways enhanced by root growth; as it has been suggested that MP movement through soils could be impacted by soil macropores, strength of the soil matrix, accumulation within organic matter, and movement by soil biota (Rillig et al., 2017).

In all fields an increase in the proportion of MP fragments, a key characteristic of biosolids used, was noted either during or shortly following applications; although in Field 2 the increase was not observed until a full month after treatment (Fig. 8). Increases in fragments ranged

Table 2
Net change in average soil MP concentrations calculated between samples taken before and after spring applications of biosolids in 2017, including % relative error calculated across sample replicates and depth, measured as site average MP concentration divided by total range of MP concentrations.

Site	MP concentration (MP kg^{-1})		Net change	
	Before application	After	MP kg^{-1}	% increase
Field 1	18 ±22.2%	25 ±20.8%	7 ±43%	41 Min: 24% Max 59%
Field 2	187 ±53.1%	130 ±23.1%	−56 ±76%	−30 Min: −7% Max: −53%
Field 3	541 ±56.4%	298 ±39.1%	−244 ±95%	−45 Min: −2% Max: −88%

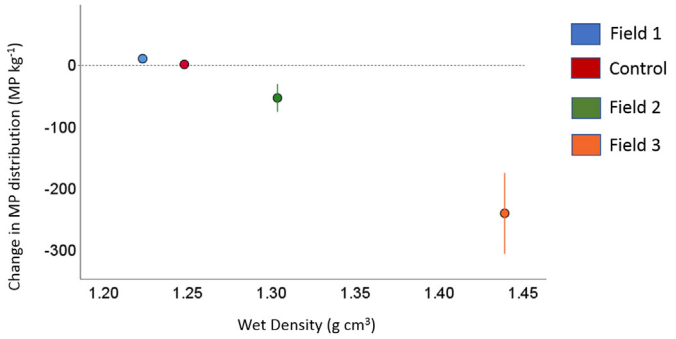

Fig. 6. Total change in microplastic distribution across all depths throughout the study period, by soil density. Error Bars = ±1 standard error.

from 4% (Fields 1 and 2) to 10% in Field 3. In Field 3 the increase was seen only during the application event itself, in comparison to Field 1 where the increase was sustained for several months. The eventual decline in proportion of fragments and increase in fibers in two of the three application sites might indicate that fragments from biosolids are preferentially transported out of the soil matrix, while fibers are retained. Previous studies on agricultural soils observed a high proportion of fibers, and suggested possible retention (e.g., Corradini et al., 2019; Zubris and Richards, 2005) due to entanglement within the matrix (Horton and Dixon, 2017).This study is the first empirical assessment of MP fragment and fiber transfer between biosolids and soils which is able to support this theory.

3.3. Wider implications

Total annual biosolid applications to agricultural land in Ontario, Canada (Central Lake Ontario Conservation Authority, 2007) have been estimated at 1.5×10^6 m^3. Using upper and lower biosolids MP concentrations from this study (Table 1), annual MP additions to agricultural soils in Ontario through biosolids use are between 4.1×10^{11} and 1.3×10^{12} particles (Fig. 9). Such high MP application rates, combined with the minimal capacity of soils to store MPs, as observed in this study, demonstrate the need for a targeted program for the protection of agricultural soils and the wider environment. For instance, results indicate that the long-term storage of biosolids and syphoning of supernatant liquid may be a relatively simple step which could reduce biosolid MP content and the quantities of MPs introduced to the environment.

4. Conclusion

This study reinforces the hypothesis that biosolids are a significant source of MPs to agrosystems, since up to 3.8×10^9 of MPs were applied to a single field in 2017. Different measures may need to be conceived to reduce MP inputs to agricultural soils treated with biosolids. Recently announced policies in many parts of the world, including Canada, to restrict single use plastics, along with existing regulations banning use of polymeric microbeads in cosmetics and personal care products will likely indirectly reduce the loads of MPs entering WWTPs, and thus reduce concentrations in sludge and biosolids. It is clear, however, that fibers, originating from laundry of synthetic clothes, will be unaffected by such legislation. In addition, revision of existing legislation surrounding biosolids management will be required to incorporate MP management, as results show that large numbers of MPs were lost from some sites following biosolid applications, despite adhering to all applicable regulations (e.g. use of Nutrient Management Plans). Meteorological conditions, soil characteristics, vegetative cover and timing of applications all impacted MP soil retention, and new legislation should be implemented to incorporate these factors and minimize transfer of MPs to watercourses.

Due to the current paucity of data on MP removal during different wastewater treatment steps and the near complete lack of MP measurements in agricultural soils, additional research is clearly needed to answer pressing questions. For instance, might heavy rainfall during spring alter agricultural soils from acting as an MP store, to an MP source? Can biosolid application subsequent to crop establishment reduce transmission of MPs into watercourses? What proportion of MPs are removed from biosolids through storage and filtering of surfactant

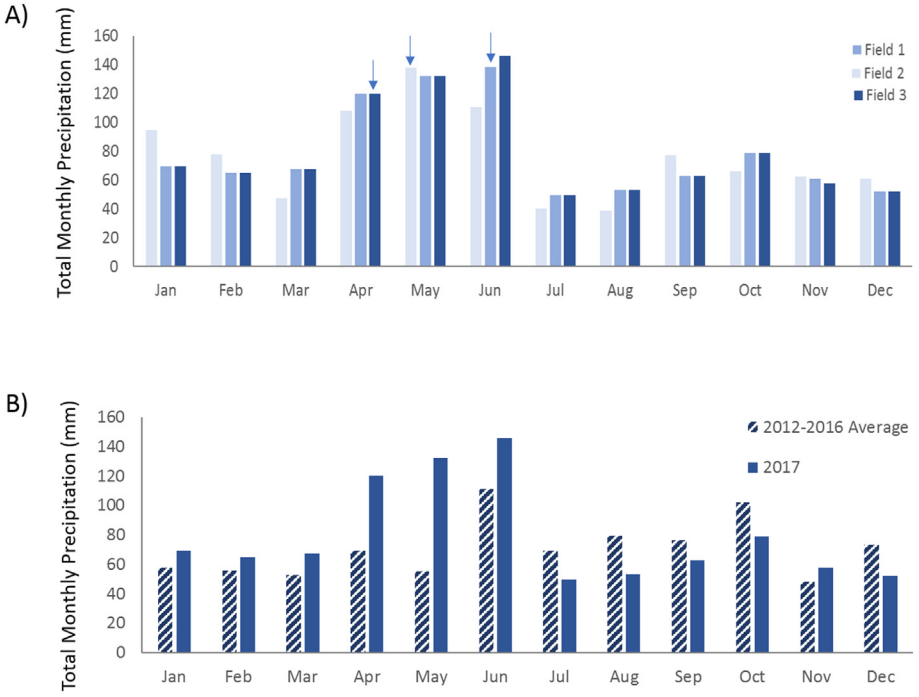

Fig. 7. A) total monthly precipitation (mm) at each field site during study period; arrows indicate locations and dates of biosolids application. B) Comparison of monthly precipitation amounts received at Site 3 between 2017 study period and average conditions over preceding 5 years (2012–2016) (ECCC, 2019).

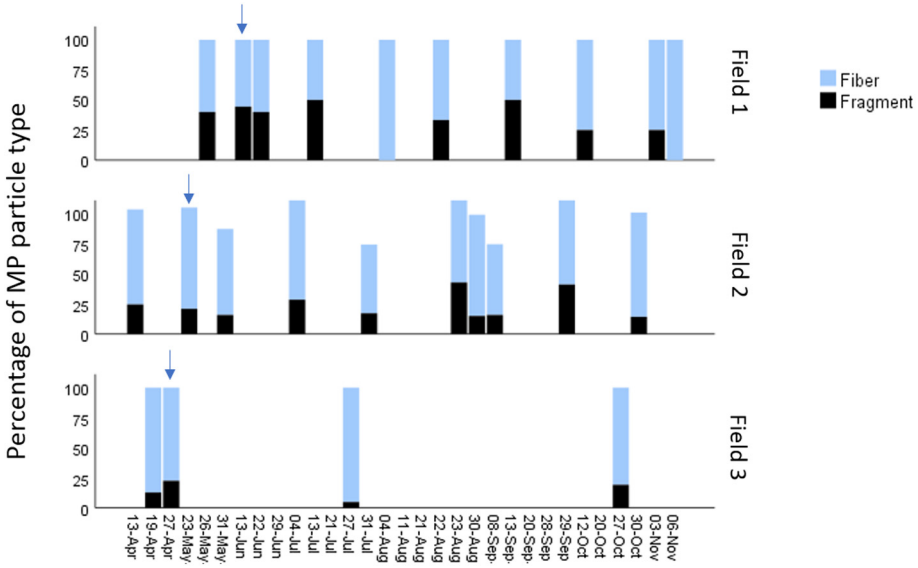

Fig. 8. Percentage of soil MP particles identified as fragments throughout the soil profile at active biosolid treatment sites (control site not included due to low numbers of particles). Arrows indicate dates of biosolid application.

liquids? Importantly, thresholds for ecological impact of MPs on soil biota have not yet been identified, meaning that currently an ecological risk assessment cannot be conducted on the results from this study. This study indicates that establishing a framework of estimating exposure to MPs in soil, a crucial step in any risk assessment, will require dealing with highly dynamic and complex processes. Additional research is therefore required as to whether the observed long-term accumulation of MP fibers in soils on plant growth and soil biota could pose a risk to

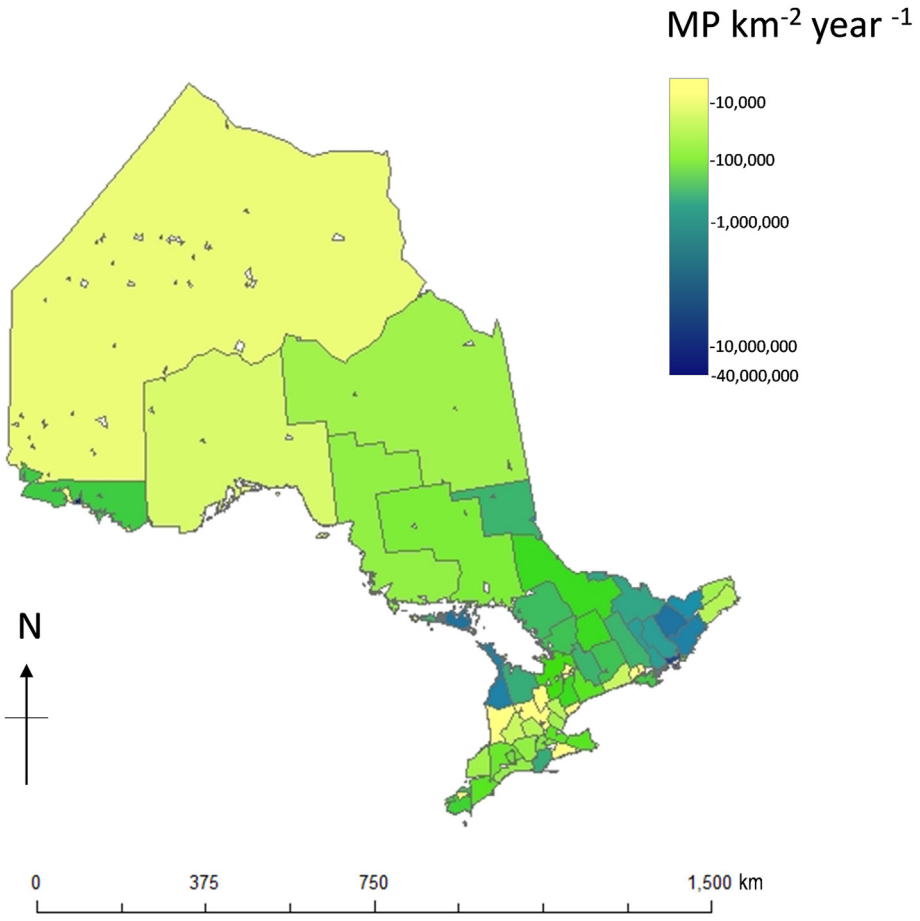

Fig. 9. Total number of microplastics applied to Ontario agricultural soils, calculated using annual biosolid application volumes (Central Lake Ontario Conservation Authority, 2007), farmland area (Ontario Ministry of Natural Resources, 2002) and observed average biosolid MP concentrations as quantified within this study.

agricultural sustainability. Researchers urgently need to address these uncertainties in order to enable effective management of terrestrial and aquatic MP pollution.

CRediT authorship contribution statement

Jill Crossman: Conceptualization, Funding acquisition, Methodology, Investigation, Formal analysis, Data curation, Visualization, Writing – original draft. **Rachel R. Hurley:** Methodology, Formal analysis, Data curation, Writing – original draft. **Martyn Futter:** Conceptualization, Funding acquisition, Investigation, Writing – original draft. **Luca Nizzetto:** Conceptualization, Funding acquisition, Methodology, Writing - review & editing.

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.2020.138334.

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