Environmental Pollution 266 (2020) 115097

Contents lists available at ScienceDirect

Environmental Pollution

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

Impact of plastic mulch film debris on soil physicochemical and hydrological properties[☆]

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ARTICLE INFO

Article history:
Received 10 February 2020
Received in revised form
28 May 2020
Accepted 23 June 2020
Available online 30 June 2020

Keywords:
Plastic pollution
Microplastics
Biodegradable plastic
Soil quality
Agricultural soil

ABSTRACT

The plastic mulch films used in agriculture are considered to be a major source of the plastic residues found in soil. Mulching with low-density polyethylene (LDPE) is widely practiced and the resulting macro- and microscopic plastic residues in agricultural soil have aroused concerns for years. Over the past decades, a variety of biodegradable (Bio) plastics have been developed in the hope of reducing plastic contamination of the terrestrial ecosystem. However, the impact of these Bio plastics in agro-ecosystems have not been sufficiently studied. Therefore, we investigated the impact of macro (around 5 mm) and micro (<1 mm) sized plastic debris from LDPE and one type of starch-based Bio mulch film on soil physicochemical and hydrological properties. We used environmentally relevant concentrations of plastics, ranging from 0 to 2% (w/w), identified by field studies and literature review. We studied the effects of the plastic residue on a sandy soil for one month in a laboratory experiment. The bulk density, porosity, saturated hydraulic conductivity, field capacity and soil water repellency were altered significantly in the presence of the four kinds of plastic debris, while pH, electrical conductivity and aggregate stability were not substantially affected. Overall, our research provides clear experimental evidence that microplastics affect soil properties. The type, size and content of plastic debris as well as the interactions between these three factors played complex roles in the variations of the measured soil parameters. Living in a plastic era, it is crucial to conduct further interdisciplinary studies in order to have a comprehensive understanding of plastic debris in soil and agroecosystems.

1. Introduction

In recent years, researchers have seen soil as a major sink for microplastics (MPs, particles with diameter < 5 mm), which is a threat to sustainable agriculture and food security (de Souza Machado et al., 2018a; Ng et al., 2018; Nizzetto et al., 2016; Rillig et al., 2019; Rochman, 2018). Subsequent studies have filled certain knowledge gaps with regards to MPs in terrestrial ecosystems, particularly in agricultural soil. For instance, the effects of MPs on soil biota have been studied (Cao et al., 2017; Huerta Lwanga et al., 2016; Zhu et al., 2018b), as well as their effects on multiple trophic levels (Huerta Lwanga et al., 2017b; Zhu et al., 2018a), underground transport (Huerta Lwanga et al., 2017a; Maass et al., 2017; Yu et al., 2019), and their interactions with other soil pollutants (Hodson et al., 2017; Rodríguez-Seijo et al., 2019; Yang et al., 2018; Yang et al., 2019). Although these studies have answered many questions, the most fundamental questions concerning MPs in soil have gone unanswered. Several major problems remain unresolved: no sufficient methods to quantify diverse MPs (Blasing and Amelung, 2018; Corradini et al., 2019; Fuller and Gautam, 2016; Schwaferts et al., 2019; Shan et al., 2018; Zhang et al., 2018b), very limited field surveys measuring the status of

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[☆] This paper has been recommended for acceptance by Baoshan Xing.

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https://doi.org/10.1016/j.envpol.2020.115097

MPs in the soil (Huang et al., 2020; Liu et al., 2018; Scheurer and Bigalke, 2018; Zhang and Liu, 2018; Zhou et al., 2020; Zhou et al., 2018), and lack of information concerning the impacts of MPs on soil physical, chemical and biological properties (Liu et al., 2017; de Souza Machado et al., 2018b; Qi et al., 2020). Moreover, recent studies have shown that MPs affected soil structure, hydraulic conductivity, water holding capacity, etc. (de Souza Machado et al., 2018b; Zhang et al., 2019). Therefore, it is crucial to study the impacts of MPs on soil physicochemical and hydrological properties to gain a better understanding of this emerging contaminant in soil and the agroecosystems.

As one of the main sources contributing to MPs in agricultural soil, plastic mulching practices play a crucial role in modern agriculture (Gao et al., 2019; Steinmetz et al., 2016). The use of plastic mulch film (PMF) to increase water use efficiency has been going on for years and thus it is relevant to study the effects of residual PMF on parameters related to soil water holding capacity. The prevailing use of plastics in agronomy started in the early 1950s (Espi, 2006). Since then, PMF has brought multiple benefits to agriculture such as instantly improving the quality and quantity of the harvests (Steinmetz et al., 2016). After decades of application, residual PMF fragments have accumulated in the soil and have had detrimental effects on soil quality and crop yield (Liu et al., 2014; Yan et al., 2014). With the highest amount of PMF usage in the world, China was the first to notice the plastic residue pollution in agricultural soil and has conducted many studies since the 1980s (Dong et al., 2015; Xiang et al., 1992; Xu, 1985; Zhao et al., 1998). In recent years, plastic residue in the soil has aroused intensifying concerns that the macroscopic plastic debris will eventually fragment into MPs (Barnes et al., 2009). From previous studies about residual PMF, researchers raised universal concerns about its long-term effects on farmland (Gao et al., 2019).

Due to the increasing global concern surrounding plastic pollution, a huge variety of biodegradable plastic mulch film (Bio PMF) was designed as a promising substitute for polyethylene films (Brodhagen et al., 2017; Kasirajan and Ngouajio, 2012). In 2016, the European commission estimated that among the 100,000 tonnes of PMF applied in Europe, 3000 tonnes were Bio PMF (European Commission, 2016). Bio plastics are made of polymers and additives that should degrade into carbon dioxide and methane or form new biomass (van Ginkel, 2007). According to current standards (e.g. ISO 17556 and EN 13432), Bio plastic should reach at least 90% biodegradation in the soil within two years (Miles et al., 2017). However, studies warned that some polymers used in these films may not be biodegradable in soil conditions (Brodhagen et al., 2017; Thompson et al., 2019). The application of Bio PMF in agriculture has aroused fierce debate (Bandopadhyay et al., 2018; Sintim and Flury, 2017) and sparked controversies surrounding the fact that Bio plastics are not only used within agroecosystems (Haider et al., 2018; Ren, 2003). Furthermore, only scant studies have been performed to investigate the function and disintegration of Bio PMF (Anzalone et al., 2010; Kapanen et al., 2008; Li et al., 2014b; Miles et al., 2012; Moreno et al., 2017). Therefore, both fundamental and in-depth studies examining Bio PMF are urgently needed to ensure their safe and sustainable application in agroecosystems.

In this study, we conducted mesocosm experiments in the laboratory using both low-density polyethylene (LDPE) and Bio PMF. The LDPE and Bio PMF were made into macro- and micro-sized debris to investigate the impacts of the plastic debris on soil physical, hydrological and chemical properties with a plastic content gradient (0, 0.5%, 1% and 2% w/w). LDPE was chosen since it is the most common mulch material and Bio PMF was chosen because it has become increasingly popular in agricultural applications (Kasirajan and Ngouajio, 2012; Steinmetz et al., 2016). We hypothesized that (i) tested soil parameters would have predictable responses to the presence of plastic debris, e.g. a decrease of bulk density, increase of porosity, increase of water flow, increase of water repellence, and (ii) different types, sizes and content of the plastic debris may have distinct effects on soil physicochemical and hydrological properties.

2. Materials and methods

2.1. Experimental setup

The mesocosm experiments were performed at 20 °C and 35% humidity in the laboratory of the Soil Physics and Land Management Group, Wageningen University & Research (WUR). Our test soil was a sandy soil (4% Organic matter, pH = 6) with 87% sand, 12% silt and 1% clay. It was collected from farmland at Unifarm, WUR and has been used for our previous studies (Qi et al., 2018; Qi et al., 2020). More information about the soil properties can be found in Table S1.

LDPE and Bio PMF were bought from the plastic mulch producer. The company states that the Bio PMF is produced from a formulated compound consisting mainly of polybutylene adipate terephthalate, starch and about 5% polylactic acid, blended with a black carbon masterbatch using a copolyester as a carrier resin. The presence of polybutylene adipate terephthalate and starch was confirmed by Fourier transform infrared spectroscopy and Differential scanning calorimetry. Macro- and micro-sized debris from LDPE and Bio PMF were prepared as described in a previous study (Qi et al., 2018). Macro-sized pieces were made by cutting PMF into $5 \times 5\ \mathrm{mm}^2$ squares by hand and the micro-sized powder was made by freeze grinding the plastic with liquid nitrogen. The powder consisted 25% of particles between 50 and 250 μm, 62.5% of particles between 250 and 500 μm and 12.5% of particles between 500 and 1000 μm. The effects of two types and two sizes of plastic debris (i.e. LDPE-Mi, Bio-Mi, LDPE-Ma, Bio-Ma) were each tested in the experimental soil at three concentrations: 0.5%, 1% and 2% of soil dry weight (Table 1). This concentration gradient is environmentally relevant and was chosen based on previous studies (de Souza Machado et al., 2018b; Qi et al., 2020). Soil without additional plastic was used as the Control.

In total, 13 treatments were tested and each treatment was replicated in three mesocosms. The three contents were always tested together with the Control treatment, during three different months making three incomplete blocks due to logistic reasons (Table 1).

The plastic debris was mixed with 2 mm sieved dry soil and water was added to reach a soil gravimetric water content of 20%. Four kg of the mixture was then manually packed into each plastic

Table 1
Treatment settings for the mesocosm experiments.

Block	Treatment	Plastic type	Plastic size	Plastic content (w/w)
1st	Control	–	–	0.0%
	LDPE-Mi_0.5	LDPE	Micro	0.5%
	Bio-Mi_0.5	Bio	Micro	0.5%
	LDPE-Ma_0.5	LDPE	Macro	0.5%
	Bio-Ma_0.5	Bio	Macro	0.5%
2nd	Control	–	–	0.0%
	LDPE-Mi_1	LDPE	Micro	1.0%
	Bio-Mi_1	Bio	Micro	1.0%
	LDPE-Ma_1	LDPE	Macro	1.0%
	Bio-Ma_1	Bio	Macro	1.0%
3rd	Control	–	–	0.0%
	LDPE-Mi_2	LDPE	Micro	2.0%
	Bio-Mi_2	Bio	Micro	2.0%
	LDPE-Ma_2	LDPE	Macro	2.0%
	Bio-Ma_2	Bio	Macro	2.0%

pot (4 L, 16.5 cm high) with a wooden pressing tool (Fig. S1). The compaction consisted of a define pattern of 10 hits repeated every kg of soil added. Each pot was covered loosely with a plastic lid and stored at 20 °C for 30 days. Every week, the mesocosms were weighed and watered to compensate for evaporation (about 10 g per week).

At the end of the experiment, four ring samples (5 cm diameter) were taken at the 0–5 cm depth and four others at the 7–12 cm depth. All the ring samples were analysed for porosity, dry bulk density (ρ_b), saturated hydraulic conductivity (ks), field capacity (FC) and water drop penetration time (WDPT). The pH, electrical conductivity (EC) and aggregate stability index (ASI) were measured from 2 mm sieved, air dried soil samples at both soil depths (two samples at 0–5 cm and two others at 7–12 cm) for each pot.

2.2. Measurements of soil parameters

After sampling, ring samples were water saturated for 24 h and weighed. The ks was then measured on saturated ring samples using the flow induction with constant head method (Klute and Dirksen, 1986), described in Fig. S2. Ring samples were then placed in a sandbox to measure the FC (Klute and Dinauer, 1986; Topp and Zebchuk, 1979), described in Fig. S3. The suction was gradually increased to pF 2 and the ring samples were weighed to measure the gravimetric water content. FC is defined as the gravimetric water content at pF 2. Soil water repellency was assessed on the ring samples at pF 2 using the WDPT method (Ritsema et al., 2008). An arbitrary WDPT threshold of 5 s was used to distinguish between hydrophilic (wettable) and hydrophobic (water-repellent) soils (Dekker et al., 2009). The ring samples were finally dried at 105 °C for 48 h. The dry mass was used to calculate the water content at saturation and at pF 2. The porosity was estimated using the volume of water in a saturated sample divided by the total volume (Klute and Dinauer, 1986). The ρ_b was measured using the dry mass of the sample and the ring volume (Klute and Dinauer, 1986).

pH (H_2O) and EC were measured in a suspension (1:5) of 5 g of 2 mm sieved dry soil in 25 ml demineralized water with a SenTix meter and a conductivity cell TetraCon 325, separately (Čapka et al., 2009). ASI was determined using an Eijkelkamp wet sieving apparatus with 4 g of 2 mm sieved soil and NaOH 2 g/L as a dispersing solution (Almajmaie et al., 2017; Kemper and Rosenau, 1986).

2.3. Statistical analysis

The results of each parameter were analysed using a linear mixed effect model (Eq. S1) implemented in SAS® 9.4 (Littell et al., 2006). Measured variables (i.e. porosity, ρ_b, ks, FC, WDPT, pH, EC and ASI) were modelled while taking into account the content, type and size of the plastic debris applied to the soil and the soil depth of the sample. Random terms were included to correct for temporal (Block) and positional effects (Pot and Pot-Depth combination).

After fitting the mixed models, the distribution of standardized residuals was checked for approximate normality. Residuals for all parameters, except ks, loosely followed a normal distribution. The residuals for $\log_{10}$(ks) followed a normal distribution, so $\log_{10}$(ks) was used for the analysis of ks. For all parameters, the soil depth factor was relatively unimportant (Table 3). Therefore, we decided to present the results averaged over both soil depths. The contribution of the main effects of each factor and each factor's interaction with the fitted model was quantified, using F-values and p-values (Table 3). The variance components for the random terms (i.e. Block, Block × Pot, Block × Pot × Depth and Residual) were calculated. The random terms contributing to the total variance of the individual observation are shown in Table S2. Means and standard errors of means were estimated for all the parameters (Table S3). Estimated means and standard errors of means were plotted in R version 3.4.2 (Team, 2013).

For convenience, the model was reparametrized, aggregating factors Type, Size and Content into one single factor Comb (Eq. S2) with 13 levels (1 control and 12 factor level combinations). This reparametrized model allowed for simple comparisons of treatments with the Control treatment, as well as other pairwise comparisons, using t-tests (Table 2).

In addition, a principal component analysis was performed for the parameters with the most effects (porosity, ρb, ks, FC and WDPT) and the correlations between porosity, ρb and ks was further explored with linear regressions. Two equations were tested to fit the porosity and ρb data. These analyses are presented in supplementary materials. The raw data, the outcomes of the model and the R script used for the plots and calculation are available on the GitHub page (https://github.com/NGBeriot/Plastic_mulch-soil_properties).

3. Results

3.1. Soil structure parameters: porosity, dry bulk density (ρ_b) and aggregate stability index (ASI)

The estimated mean porosity for the Control was 0.43 ± 0.02 (Table S3). Porosity of the Control was not significantly different for plastic treatments with 0.5% content (Fig. 1A). Size-wise comparisons for treatments with Bio plastics at both 1% and 2% showed that the macro-sized pieces had higher porosity than micro-sized particles (Table 2). Type-wise comparisons showed that LDPE-Ma_2 had lower porosity than Bio-Ma_2. Content-wise comparisons for LDPE-Ma showed that porosity for 1% was higher than the Control, 0.5% and 2% contents. For LDPE-Mi, the porosity for 1% was higher than the Control and 0.5% content but not different from the 2% content. For Bio-Ma, the porosity at 1% and 2% were not significantly different but they were both higher than the Control.

ρ_b of the Control was not significantly different from any of the plastic treatment with 0.5% content (Fig. 1B). ρ_b decreased with increasing 1% and 2% plastic content for all plastic debris except Bio-Mi. Size-wise comparisons showed that for LDPE_1% and LDPE_2%, the macro-sized debris had lower ρ_b than the micro-sized ones (Table 2). Type-wise comparisons showed that for 2% content, LDPE had lower ρ_b than Bio for both macro- and micro-sizes. Content-wise comparisons showed that the addition of LDPE-Ma significantly decreased ρ_b as the increase of content went from 0.5% to 2%.

The estimated mean value of ASI over all the treatments ranged from 0.48 ± 0.045 to 0.68 ± 0.045, with the Control being 0.56 ± 0.045 (Table S3). Bio-Mi_0.5 showed significantly higher ASI compared to Bio-Ma_0.5 and no other significant differences in ASI were observed among the treatments (Table 2).

3.2. Water infiltration parameter: saturated hydraulic conductivity (ks)

ks of the Control was not significantly different from any of the plastic treatments with 0.5% content (Fig. 1C). Size-wise comparisons showed that for Bio_1% and Bio_2%, the macro-sized debris had higher ks than the micro-sized ones (Table 2). Type-wise comparison showed that treatments LDPE-Ma_0.5 had lower ks than Bio-Ma_0.5, but LDPE-Mi_2 had higher ks than Bio-Mi_2. Content-wise comparisons showed that the increase from 0.5% to 1% of plastic debris increased ks, but not all the differences were

Table 2
Estimates of differences between treatments associated with p-value < 0.001.

	Porosity [-]	ρb [kg/m^3]	log10(ks) [-]	FC [-]	WDPT [s]	pH [-]	EC [μS/cm]	ASI [-]
Comparison size-wise (Mi-Ma); same type, same content								
LDPE-Mi_0.5 - LDPE-Ma_0.5	.	.	.	.	2.0	.	.	.
LDPE-Mi_1 - LDPE-Ma_1	.	0.09	.	.	.	.	.	.
LDPE-Mi_2 - LDPE-Ma_2	.	0.19	−0.47	0.01	.	.	.	.
Bio-Mi_0.5 - Bio-Ma_0.5	.	.	.	.	.	.	.	0.16
Bio-Mi_1 - Bio-Ma_1	−0.034	.	−0.55	0.007	1.9	−0.10	.	.
Bio-Mi_2 - Bio-Ma_2	−0.067	0.17	−1.23	.	.	.	.	.
Comparison type-wise (LDPE-Bio); same size, same content								
LDPE-Mi_0.5 - Bio-Mi_0.5	.	.	.	−0.01	1.5	.	.	.
LDPE-Mi_1 - Bio-Mi_1	.	.	.	−0.02	−3.9	.	.	.
LDPE-Mi_2 - Bio-Mi_2	.	−0.08	0.85	−0.02	−1.9	.	.	.
LDPE-Ma_0.5 - Bio-Ma_0.5	.	.	−0.51	.	.	.	.	.
LDPE-Ma_1 - Bio-Ma_1	.	−0.08	.	−0.01	−1.9	.	.	.
LDPE-Ma_2 - Bio-Ma_2	−0.044	−0.11	.	−0.03	.	.	.	.
Comparison content-wise (0.5−1, 0.5−2, 1−2); same type, same size								
LDPE-Mi_0.5 - LDPE-Mi_1	−0.060	0.08	−0.73	.	.	.	.	.
Bio-Mi_0.5 - Bio-Mi_1	.	.	.	−0.01	−4.6	.	.	.
LDPE-Ma_0.5 - LDPE-Ma_1	−0.062	0.20	−1.03	.	.	.	.	.
Bio-Ma_0.5 - Bio-Ma_1	.	.	−0.70	−0.01	−2.3	.	.	.
LDPE-Mi_0.5 - LDPE-Mi_2	.	.	.	.	.	.	.	.
Bio-Mi_0.5 - Bio-Mi_2	.	.	.	.	.	.	.	.
LDPE-Ma_0.5 - LDPE-Ma_2	.	0.28	−1.01	0.015	.	.	.	.
Bio-Ma_0.5 - Bio-Ma_2	.	0.13	.	−0.01	.	.	.	.
LDPE-Mi_1 - LDPE-Mi_2	.	.	.	.	.	.	.	.
Bio-Mi_1 - Bio-Mi_2	.	.	0.98	0.01	3.3	.	.	.
LDPE-Ma_1 - LDPE-Ma_2	0.047	0.09	.	0.021	.	.	.	.
Bio-Ma_1 - Bio-Ma_2	.	.	.	.	.	.	.	.

Cells are empty (.) if the p-value > 0.001. All estimated differences and associated p-value were provided in Table S4.
ρb: dry bulk density; ks: saturated hydraulic conductivity; FC: field capacity; WDPT: water drop penetration time; EC: electrical conductivity; ASI: aggregates stability index.

Table 3
Tests of Fixed Effects for the four factors and the factor interactions F-value (p-value).

Factor and Interaction	DF num, DF den	Porosity	ρb	log$_{10}$ks	FC	WDPT	pH	EC	ASI
Content	2, 32	**11.33 (0.0002)**	**26.13 (<.0001)**	**15.67 (<.0001)**	**12.1 (0.0001)**	8.3 (0.0017)	0.48 (0.62)	0.56 (0.57)	0.55 (0.65)
Type	1, 31	1.51 (0.22)	**51.42 (<.0001)**	2.42 (0.12)	**506.95 (<.0001)**	**64.65 (<.0001)**	**18.59 (0.0002)**	3.56 (0.063)	0.19 (0.66)
Size	1, 31	**40.38 (<.0001)**	**164.62 (<.0001)**	**71.4 (<.0001)**	6.09 (0.019)	2.33 (0.13)	2.63 (0.12)	0.27 (0.60)	5.32 (0.027)
Depth	1, 31	0.17 (0.68)	1.77 (0.19)	4.63 (0.038)	6.72 (0.014)	11.16 (0.0021)	4.9 (0.034)	1.35 (0.25)	0.77 (0.39)
Content×Type	2, 31	7.04 (0.0030)	**27.89 (<.0001)**	**17.5 (<.0001)**	**38.44 (<.0001)**	**31.28 (<.0001)**	1.69 (0.20)	0.41 (0.66)	1.11 (0.34)
Content×Size	2, 31	1.76 (0.19)	**72.08 (<.0001)**	**18.82 (<.0001)**	0.49 (0.62)	**12.06 (0.0001)**	**10.03 (0.0004)**	2.51 (0.089)	1.47 (0.24)
Type×Size	1, 31	**25.26 (<.0001)**	0.17 (0.69)	**29.07 (<.0001)**	0.75 (0.39)	0.23 (0.63)	6.66 (0.015)	0.0 (0.98)	6.15 (0.018)
Content×Type×Size	2, 31	4.8 (0.015)	6.55 (0.0042)	1.05 (0.36)	**22.84 (<.0001)**	**16.3 (<.0001)**	1.45 (0.24)	0.93 (0.39)	2.42 (0.10)
Depth×Content	2, 33	3.46 (0.043)	2.55 (0.0.93)	1.94 (0.16)	3.83 (0.032)	1.1 (0.34)	3.53 (0.041)	1.07 (0.35)	6.49 (0.0042)
Depth×Type	1, 33	0.83 (0.37)	0.66 (0.42)	1.81 (0.19)	4.19 (0.049)	0.52 (0.48)	12.79 (0.0011)	1.74 (0.19)	1.47 (0.23)
Depth×Size	1, 33	2.1 (0.16)	**13.7 (0.0008)**	8.92 (0.0053)	**13.06 (0.001)**	6.89 (0.013)	0.07 (0.80)	0.02 (0.89)	0.19 (0.67)
Depth×Content×Type	2, 33	2.45 (0.10)	3.75 (0.034)	5.13 (0.012)	6.57 (0.004)	1.55 (0.23)	4.29 (0.022)	1.49 (0.23)	1.35 (0.27)
Depth×Content×Size	2, 33	1.45 (0.25)	0.45 (0.64)	1.12 (0.34)	2.32 (0.11)	0.1 (0.90)	1.11 (0.34)	0.94 (0.40)	1.21 (0.31)
Depth×Type×Size	1, 31	4.6 (0.040)	0.27 (0.60)	0.47 (0.50)	5.31 (0.028)	0.12 (0.73)	10.27 (0.003)	1.17 (0.28)	0.63 (0.43)
Depth×Content×Type×Size	2, 33	0.12 (0.88)	2.2 (0.13)	3.31 (0.049)	1.48 (0.24)	3.63 (0.038)	4.19 (0.0239)	1.6 (0.21)	1.73 (0.19)

DF num and DF den are the degrees of freedom for numerator and denominator for the F-tests, respectively.
ρb: dry bulk density; ks: saturated hydraulic conductivity; FC: field capacity; WDPT: water drop penetration time; EC: electrical conductivity; ASI: aggregates stability index.
Bold values have $p < 0.001$.
Underlined values are the highest per parameter when $p < 0.001$.

statistically significant. There was no further increase of ks with the increase from 1% to 2% plastic debris.

3.3. Soil water retention parameter: field capacity (FC)

FC of the Control was not significantly different from any plastic treatments with 0.5% content (Fig. 1D). However, Bio_1% and Bio_2% of both macro- and micro-sizes had higher FC than the Control and LDPE_2% had lower FC than the Control. Size-wise comparisons showed that for Bio_1% and LDPE_2%, the macro-sized had lower FC than micro-sized ones (Table 2). Type-wise comparisons showed that the treatments with LDPE macro- and micro-sizes had lower FC as compared to Bio. Content-wise comparisons showed that the FC of Bio-Mi at 1% was higher than the Control, 0.5% and 2%.

3.4. Soil water repellency parameter: water drop penetration time (WDPT)

The WDPT was higher for all of the treatments with plastic residues as compared to the Control (Fig. 1E). Size-wise comparisons for LDPE_0.5% and Bio_1% showed that WDPT was lower for the macro-sized plastics than for the micro-sized plastics (Table 2). Type-wise comparisons showed that most of the treatments with

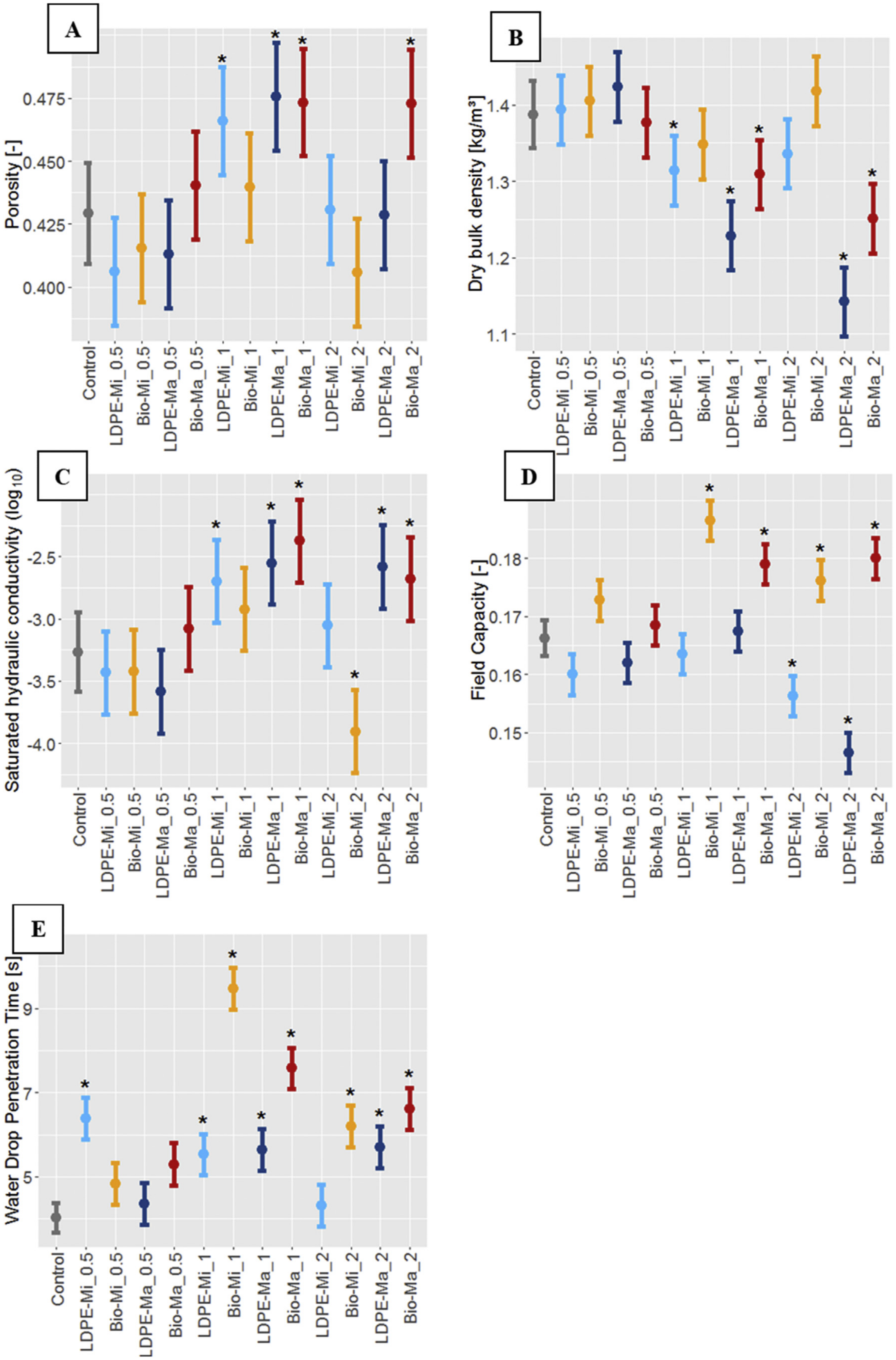

Fig. 1. Mean ± standard errors of means estimated for Porosity (A), Dry bulk density (B), Saturated hydraulic conductivity (C), Field capacity (D) and Water Drop Penetration Time (E) over the 13 treatments (Type × Size × Content). Treatments significantly different from the Control (p-value<0.001) are marked with a star.

LDPE had lower WDPT as compared to the treatments with Bio. Content-wise comparisons showed that the WDPT for Bio-Mi at 1% was higher than the Control, 0.5% and 2% contents. The WDPT for LDPE-Mi decreased with increasing content from 0.5% to 2%, but the differences were not statistically significant. All treatments, except for the Control, Bio-Mi_0.5, LDPE-Ma_0.5 and LDPE-Mi_2, were above the 5 s threshold defining water repellent soils.

3.5. Soil chemical properties: pH and electrical conductivity (EC)

The estimated mean value of pH over all of the treatments ranged from 6.28 ± 0.052 to 6.42 ± 0.052, with the Control at 6.33 ± 0.052 (Table S3). The estimated mean value of EC over all the treatments ranged from 431 ± 65 to 532 ± 65, with the Control at 492 ± 65 (Table S3). We did not observe important variation of pH and EC caused by the addition of the plastic debris (Table 2).

3.6. Main factors and interactions

The main factor affecting porosity, ρb and $\log_{10}(\mathrm{ks})$ was the size of the plastic whereas it was the type of the plastic for FC, WDPT and pH (Table 3). Both the type and the size of the plastic had important impacts on ρb. The type of plastic itself did not affect the porosity and the $\log_{10}(\mathrm{ks})$ very much but the Type × Size interaction was responsible for a lot of variation. The content of the plastic played a major role in the porosity, ρb, $\log_{10}(\mathrm{ks})$, FC and WDPT and always interacted with the Type factor. For each of these five parameters, more than one factor interaction had a significant impact and the 3-factor interaction Content × Type × Size was significant except for $\log_{10}(\mathrm{ks})$. The soil depth and its interactions with other factors was relatively unimportant. Overall, the studied factor had small effects on EC and ASI.

To further explore the correlation between parameters which were mostly affected by the main factors and their interactions, the principal component analysis for the parameters porosity, ρb, ks, FC and WDPT were conducted as additional information. The first, second and third principal components explained 89% of the variance (Table S5). The first and second principal components showed that porosity and ks were likely to be positively correlated, while both were likely to be negatively correlated to ρb (Fig. S4A). The correlations porosity/ρb, porosity/ks and ρb/ks had a coefficient of determination of 0.33, 0.54 and 0.65, respectively (Fig. S5). The correlation fit the data except for the treatment LDPE-Ma_2 which had values below the regression lines porosity/ρb and ρb/ks. Additionally, the equations Eq. S(6) and Eq. S(7) showed that the plastic content plays a minor role in explaining the correlation between porosity and ρb.

4. Discussion

The present study provides clear experimental evidence that incorporating PMF residues into the tested sandy soil aroused multiple effects on studied soil properties (Fig. 1). Differences were observed for physicochemical and hydrological parameters, when compared to the treatments with plastic additions and/or with the Control (Table 2). The size, type and content of plastic debris presented idiosyncratic effects on tested soil parameters. These soil parameters are closely related with soil type and we only used one sandy soil in this study. Nevertheless, our research aligns with previous studies, suggesting that further research is urgently needed to develop a comprehensive understanding of plastic pollution in agroecosystems.

4.1. Effects of the size, type and content of plastic debris on soil and agroecosystems

So far, only a few studies have been carried out that examine the effects of plastic residues on soil properties and the research was either focused on macro- or micro-sized debris (Dong et al., 2015; Jiang et al., 2017; de Souza Machado et al., 2018b). In this study, both macro- and micro-sized plastic residues had significant impacts on studied soil parameters. Significant differences between the Control and treatments were observed more frequently in treatments with macro-sized debris. In the research of de Souza Machado et al. (2018b), researchers found that MPs affected the ρ_b, water holding capacity, hydraulic conductivity and water stable aggregates. Dong et al. (2015) found that plastic film residues (0–100 cm^2) affected soil moisture content, porosity, pH, organic matter and worsened soil quality. Jiang et al. (2017) demonstrated that residual PMF fragments changed soil properties, e.g. soil water content, ρ_b, ks and porosity, and altered soil water distribution involved with plant roots. Since different conditions were used for these studies, it is hardly feasible to directly compare the results.

In a previous study, using the same kind of soil and plastic materials, the addition of 1% plastic residues had significantly negative effects on crop growth and micro-sized plastic residues showed more negative effects than macro-sized residues (Qi et al., 2018). In the current experiment, with the same plastic type and content, treatments with micro-sized residues showed significantly lower porosity and ks, and higher ρ_b, FC and WDPT, in some cases. Although the changes, even if statistically significant, were relatively small, we hypothesized that the changes in these soil properties brought about by the addition of plastic residues may negatively affect soil quality and plant growth. In the long run, plastic debris could be eventually degraded into micro- and nanoplastics due to various biotic and abiotic stressors (Barnes et al., 2009; Singh and Sharma, 2008). Studies about plastic debris in different sizes are needed to assess the long term effects of microplastics in soil.

In this study, we found that Bio and LDPE plastic debris showed significantly different effects on soil properties even with the same size and content. Correspondingly, with the same soil and plastic materials (at content 1%), Qi et al. (2018) concluded that Bio plastic debris had stronger negative effects on crop yield and growth than LDPE. Hence, the negative effects on plant growth could be partly explained by the effects of plastic debris on soil properties. Regarding different types of plastic debris, de Souza Machado et al. (2018b) tested four different types of MPs and found that polyester fibres showed the most noticeable impacts on the soil biophysical environment as compared with polyacrylic fibres, polyamide beads and polyethylene fragments. Unfortunately, previous studies of Bio mulch films mainly focused on their performance in agriculture (Anzalone et al., 2010; Kapanen et al., 2008; Miles et al., 2012) or their degradation and deterioration patterns (Li et al., 2014b; Moreno et al., 2017). Li et al. (2014a) buried two starch-based mulches, one polylactic acid mulch and one cellulose-based mulch, in a field for 18 months and suggested that the Bio mulch films had minor effects on the soil quality during the evaluation period. As Sintim and Flury discussed (2017), although Bio mulch films may be encouraging substitutes for traditional polyethylene films, in-depth and comprehensive studies, focussing on the potential release of micro- and nanoplastics during degradation processes among others, should be conducted before they are widely utilized. Overall, Bio plastics should not be considered as the panacea for plastic pollution in agroecosystems without in-depth research.

In our experiment, we set the same gradient for the four kinds of plastic debris tested, i.e. 0.5%, 1%, 2% and the Control at 0%. It is difficult to concisely summarize the content-wise effects of different plastic debris on various soil parameters since quite a few low-content plastic debris showed stronger effects than high-content debris even if the plastic sizes and types were all the same. Similarly, de Souza Machado et al. (2018b) added a series of concentrations for different MPs ranging from 0.05% to 2.00% to the soil and they found the apparent nonmonotonic dose responses of soil biophysical proxies. Although de Souza Machado et al. (2018b) suggested that it was unrealistic to assess this nonmonotonicity based on current experimental data, they intensively discussed the potential interactions among plastic particles and natural matter in the heterogeneous terrestrial ecosystem. The addition of plastic debris in the soil would affect multiple soil processes and the interactions between plastic particles and natural matter were unpredictable (de Souza Machado et al., 2018b).

Regrettably, to the best of our knowledge, there are no experiments that have been carried out on the effects of the MPs content gradients on crop growth so we could not estimate the dose responses of crops to MPs in the soil. Nevertheless, there are quite a few studies that have been conducted in China on the impacts of macroplastic residue gradients (from 0 to 1440 kg hm^{-2}) on crop growth and soil quality (Huang et al., 2019; Nan et al., 1996; Zhang et al., 2014; Zhao et al., 1998). For instance, Zhao et al. (1998) found monotonic responses of crop yield, ρ_b and porosity to the gradient of residual PMF weight (0, 37.5, 75, 150, 225, 300, 375 and 450 kg hm^{-2}). While Huang et al. (2019) also observed glaring adverse effects of plastic residues on the growth and yield of potato, they did not find any linear correlation between the yield and the residual amount of PMF (0, 90, 180, 360 and 720 kg hm^{-2}). Regardless, considering the undeniable nonmonotonicity in the responses of the soil matrix to plastic debris, further studies using a series of gradients are urgently needed to elucidate the mechanisms and dose responses.

4.2. Limitations and wider implications for ecological assessment of plastic debris in soil

We have asserted that the types, sizes and contents of the plastic debris has had distinct effects on selected soil properties in our study and interactions mattered in some cases. Parameters of soil structure, water infiltration, water retention and soil water repellency all responded vigorously during the experimental period, but not many effects were measured in the soil chemical parameters, i.e. pH and EC. On one hand, one month might be too short for plastic debris to initiate chemical alterations in the soil. On the other hand, other soil chemical parameters may react more swiftly than pH and EC. It is difficult to explain the variations of parameters in the presence of plastic debris in the soil. For instance, the effects of plastic debris on porosity and ρb cannot be explained by the lower density of plastic compared to the soil particles using Eq. S(6) and Eq. S(7). In this study, only the effective porosity was measured using the saturation method and only a sandy soil was tested. Therefore, more tests using different soil textures are required to understand how plastic debris may affect the soil's physical and hydrological parameters.

We did not expect the plastic to undergo significant degradation during this one-month experiment. PMF was designed to keep its integrity over the crop growing season (>1 month) and exposure to UV irradiation from the sun is a significant factor in plastic degradation (Napper and Thompson, 2019). The properties of plastic will change during degradation and therefore, we could expect they may have different interactions with the soil. Further studies should take into account the degradation of plastics in long-term experiments and aging plastic debris could be used to compare with virgin debris.

With regards to soil properties, a soil's biological trait is a vital aspect, along with soil physical and chemical parameters (Bünemann et al., 2018). With the same plastic materials and soil, Qi et al. (2020) found that the rhizosphere bacterial communities were significantly affected by the presence of Bio PMF residues. When Li et al. (2014a) investigated the effects of mulch film residues on soil quality, they calculated the soil quality index based on microbial biomass carbon, β-glucosidase, EC, total organic carbon and pH, so that the alterations of soil quality among treatments could be clearly presented by numerical comparisons. While scientists try to obtain an overall soil quality index for comparisons, as Bünemann et al. (2018) critically reviewed, an assessment framework based on a logical-sieve method would be useful for the assessment of targeted soil threats. Hence, establishing an assessment framework which can be applied universally for plastic debris in soil would be profoundly pragmatic for further studies.

5. Conclusion

Overall, we saw that both LDPE and Bio PMF debris in either macro- or micro-sizes had noticeable effects on soil physicochemical and hydrological parameters and these properties of tested sandy soil nonmonotonically responded to residual amounts of PMFs. For instance, the presence of LDPE debris decreased field capacity, while Bio plastic debris increased it. Macro-sized plastic debris presented more differences between the Control, compared to micro-sized ones. Special attention should be paid to the fundamental properties of soil in order to gain a comprehensive understanding of the potential effects of plastic residues on soils. Concerning their conspicuous mischief and long-term existence, we eagerly call on further interdisciplinary studies for various types, sizes and contents of plastic debris in soil and agroecosystems.

CRediT authorship contribution statement

Yueling Qi: Conceptualization, Investigation, Methodology, Validation, Writing - original draft, Writing - review & editing. **Nicolas Beriot:** Conceptualization, Investigation, Methodology, Formal analysis, Writing - original draft, Writing - review & editing, Visualization. **Gerrit Gort:** Formal analysis, Writing - review & editing, Visualization. **Esperanza Huerta Lwanga:** Conceptualization, Writing - review & editing. **Harm Gooren:** Conceptualization, Methodology. **Xiaomei Yang:** Conceptualization, Writing - review & editing. **Violette Geissen:** Conceptualization, Resources, Supervision.

Declaration of competing interest

The authors declare no conflict of interest.

Acknowledgements

This research was funded by the National Natural Science Foundation of China (41877072), the Natural Science Basic Research Plan in Shaanxi Province of China (2019JQ-639) and the EU Horizon 2020 project (ISQAPER: 635750). We are thankful for the financial support from the China Scholarship Council (CSC: 201604910510). We acknowledge Taede Stoker from Unifarm of Wageningen University & Research for providing the soil, the pots and helpful information. Many thanks to Klaas Oostindie and Piet Peters from the SLM group (WUR) for their expertise in water repellency and soil aggregate stability, respectively. Thanks to Henny Gertsen for helping prepare the plastic materials. We acknowledge Maarten van der Zee from the Agrotechnology & Food Science group of WUR for his expertise about plastics. We are very grateful to Robin Palmer for the language editing.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2020.115097.

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