Polyester Microplastic Fibers affect Soil Physical Properties and Erosion as a Function of Soil Type

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10 Abstract. Microplastic is recognized as a factor of global change <u>contaminating</u>affecting many environmental compartments.

11 Agricultural soils are very likely to receive microplastic contamination and are of particular concern due to their role in food

12 production. Microplastic has already been shown to be able to affect soil properties, but its effect on different soil types is

13 poorly understood. Moreover, limited information is available on how the presence of this pollutant can affect soil water

14 erosion processes, which are extremely important issues in many environments. In the light of this, we performed two

15 experiments (carried out on a microscale) to investigate how the presence of polyester microplastic fibers affects soil physical

- 16 and hydrological parameters and processes such as aggregate formation and soil erosion in three different agricultural soil
- 17 types (a Vertisol, an Entisol, and an Alfisol).

Our data show that the effects of polyester microplastic fibers on soil physical parameters and erosion are strongly dependent on soil type. We found that microplastic fiber contamination can affect soil bulk density, capacitive indicators of soil physical quality, and decrease the formation of new aggregates (labile in the incubation period applied in our experiments) but did not affect their stability in water. However, we found that polyester MP fibers reduced soil loss and sediment concentration, especially in the most erodible soils. In this paper we provide some hypotheses but certainly future data are still needed to confirm or not our hypotheses.

Overall, our results highlight the importance of broadly exploring soil properties such as texture, mineralogy, organic carbon content, etc. to better understand how the various soil types respond to microplastic fiber contamination.

26 1 Introduction

Although the production of plastic is relatively recent (~ 1950), its versatility and low cost have made it one of the most used daily materials today and it is hard to think of a world without plastic. World production of plastics has exceeded 350 million tons per year. Only a modest fraction of it is recycled (6-26%; Alimi et al., 2018) while the vast majority generates plastic waste. Mishandling of plastic waste can lead to environmental contamination; and indeed, small plastic particles have been

- documented even in the most remote areas of the globe (Bergami et al., 2020; Napper et al., 2020). In the last decade, research
- 32 on environmental impacts of plastic has received growing attention especially through the study of microplastic (defined as 33 plastic particles smaller than 5 mm; Hartmann et al., 2019).
- 34 Most studies on plastic pollution concern aquatic environments, although most plastics are likely present in the terrestrial 35 environment (estimated in quantities 4 to 23 times greater than those in the ocean; Horton et al., 2017). Given their role in food 36 production, agricultural ecosystems are of particular concern. Microplastics (MPs) can reach agricultural soils through different 37 routes: (i) incorporation of compost, sewage sludge, coatings of seeds and fertilizers, and other organic compounds often 38 contaminated with MPs (Weithmann et al., 2018; Zubris & Richards, 2005; Zhang et al., 2020); (ii) fragmentation of plastic 39 films used for mulching (Qi et al., 2020) or (iii) atmospheric fallout (Dris et al., 2018). Therefore, it seems clear that agricultural 40 land contains MPs and, considering that the degradation of plastic in the soil is generally very slow (O'Kelly et al., 2021), the current hypothesis is that the MP content in agricultural land will slowly increase over time. 41
- 42 Once the MP particles reach the soil surface, they can be easily incorporated into the soil profile through terrestrial fauna 43 (earthworms: Rillig et al., 2017; Collembola: Maaß et al., 2017), biopores, mechanical operations (e.g., ploughing), soil 44 cracking (Rillig, Ingraffia, et al., 2017), cryoturbation, etc. Incorporated MPs into soil can interact with soil bio-physical 45 processes. Indeed, several studies have shown that the presence of MP can affect soil physical characteristics with effects widely differentiated in relation to the soil characteristics and MP type, shape, and size, and concentration (Boots et al., 2019; 46 47 Lozano et al., 2021; Machado et al., 2018; Zhang et al., 2019). Among the various microplastic types, polyester MP fibers are 48 one of the most detected in agroecosystems (Büks and Kaupenjohann, 2020; Crossman et al., 2020) and have been shown to 49 be able to modify markedly (often stronger than other MP types) several soil physical properties. For instance, Machado et al. (2018) found that polyester MP fibers at a concentration of 0.4% decreased soil bulk density and increased soil water holding 50 51 capacity in an Albic Luvisol, while Lehmann et al. (2019), in the same soil type, found that this contaminant tends to decrease 52 soil aggregate water-stability when present at a concentration of 0.1 %.
- As soil physical characteristics are related to its susceptibility to erosion (Bradford et al., 1987; Lowery et al., 1995; Mamedov and Levy, 2019), it is reasonable to expect that MPs in the soil can influence water erosion processes, which are extremely important issues in many environments (Bagarello et al., 2018). However, to the best of our knowledge, limited research has been conducted to evaluate if and to what extent MP contamination of soil affects water erosion processes.
- In light of the above, plastic likely represents a risk for the functionality and resilience of ecosystems. Moreover, due to erosion processes, microplastic particles embedded in the upper soil layer can be transported, eventually reaching water bodies such as rivers, lakes or the sea (Rehm et al., 2021). To better manage this situation, it is necessary to understand the sources, movements, fate, and impacts of plastic when it reaches the different environmental compartments.
- 61 The main objective of this research is to expand the knowledge on the effects of MP contamination on the physical properties 62 of three different soil types and evaluate the impact of polyester MP fibers on the water erosion processes. The underlying 63 hypotheses are: i) polyester MP fiber contamination negatively affects the structural state of the soil and increases soil erosion; 64 ii) the adverse impact of polyester MP fiber contamination on physical soil properties and hydrological processes differs with

- soil type, with more marked effects on soils that are characterised by a low aggregation ability. In these soils the presence of
- 66 MP fibers could substantially increase the susceptibility to the erosion processes. Collecting this information is essential for
- 67 identifying the damages and developing solutions that can reduce the impact of microplastics on ecosystems and, at the same
- 68 time, concentrate efforts in terms of resources and interventions in the most vulnerable areas.

69 2 Material and Methods

70 2.1 Soils and polyester microplastic fiber treatment

- 71 For our experiment we chose the following three different soils, widely spread in the Mediterranean area:
- i) Typic Haploxerert (Vertisol). This soil type is widespread in areas with a flat or slightly sloping morphology. The sampled
 soil is well-structured, with clay texture, a good water and nutrient accessibility, sub-alkaline reaction, fair or high presence of
- organic matter and other elements of fertility (P, K, N, etc). Smectite (montmorillonite) is the dominant clay mineral. This soil
- is characterized by large and deep cracks along the profile during the dry season. It has a medium-high production potential.
- ii) Typic Xerorthent (Entisol). It is a light brown type of soil with a sub-alkaline reaction, widespread in areas with steep and
 uneven morphologies, frequently affected by erosion phenomena; it generally has a limited thickness with an abundant skeleton
 presence. It contains illite as the dominant clay mineral followed by kaolinite. The structure is not very stable, the organic
- 79 matter content is modest, and the production potential is medium-low.
- 80 iii) Typic Rhodoxeralfs (Alfisol). It is a typical soil widespread on the carbonate platforms of many Mediterranean 81 environments; it can also be found in mountain contexts that are carbonated and when located in high slopes they are 82 susceptible to severe erosion. The colour tends to be red due to the considerable presence of iron oxides linked to the leached 83 clays. Kaolinite is the dominant clay mineral. This soil is characterized by strong pedological aridity, due to its calcareous 84 nature, and low amounts of organic matter and fertility elements.
- The soils were sampled at the end of October 2019 from the upper 30 cm of agricultural fields. After sampling, the soil was air dried, sieved at 600 µm and stored at 4 °C until the beginning of the experiments in December 2019. This was to minimize
- 87 changes in the natural microbial community. At the sampling time, we checked and ensured that the three soil types were not
- 88 contaminated with meso- and/or macroplastic particles; however, we did not carry out analytical procedures to assay the
- 89 contamination of smaller plastic particles and therefore we cannot exclude that the control treatments might already contain a
- 90 detectable amount of micro- and/or nanoplastic particles.
- 91 Soils were characterised as follows: pParticle size distribution was determined using conventional methods and soil texture
- 92 was classified according to USDA (Gee and Bauder, 1986);- tTotal nitrogen (TN; Kjeldhal), total organic carbon (TOC;
- 93 Walkley–Black procedure according to Nelson & Sommers, 1996), pH, saturated electrical conductivity at 25 °C (EC), and
- 94 cation exchange capacity (CEC). S-of the considered soils properties are listed in Table 1.
- 95 **Table 1.** Physical and chemical properties of the three different soils used in the experiment.

Soil	Site coordinates	Clay	Silt	Sand	TN	TOC	pН	EC	CEC
		(g kg ⁻¹)		(dS m ⁻¹)	(cmol kg ⁻¹)				
Vertisol	37.556140°N,	415	357	228	1.54	15.78	7.74	1.89	30.0
v crusor	13.515400°E,	415	557	220	1.54	15.78	/./4	1.09	50.0
Entire1	37.561368°N,	209	461	330	1.20	9.25	7.84	1.88	18.4
Entisol	13.512904°E								
A10 1	37.643511°N,	152	431	417	0.77	11.20	7.58	2.01	13.8
Alfisol	12.628327°E								

Clay, Silt, and Sand were classified according to USDA (Clay < 2μm, Silt 2-50 μm, and Sand 50-2000 μm); TN, Total Nitrogen; TOC, Total
 Organic Carbon; EC, Electrical Conductivity; CEC, Cation Exchange Capacity.

98 For microplastic contamination, a 100% polyester white rope (Marlow, Marlowbraid classic rope) was manually cut to produce 99 secondary microplastic fibers. These fibers were characterized by scanning at least 200 fibers on PVC travs 10 times (Epson 00 Perfection Scan V800, 8-bit grayscale, 800 dpi) and then analyzing the scans with WinRhizo (WinRHIZO Prov. 2007d, Regent 01 Instrument Inc., Quebec, Canada). The mean and the standard deviation, SD, of fiber length were 2.87 mm and 0.31 mm, respectively, and the mean and SD of fiber diameter were 87 um and 3 um, respectively. The polyester fibers were incorporated 02 03 into the soil at a concentration of 0.5% w/w of the dry soil weight. This microplastic level was similar to that used in previous 04 studies, which reported noticeable changes in the soil biophysical environment and plant response (Lehmann et al., 2020; 05 Lozano et al., 2021; Machado et al., 2018, 2019; Ingraffia et al., 2022). To contaminate the soils, the fibers were added into a 06 blender (Waring® WSG30, Waring Commercial, Torrington, Connecticut) as a band sandwiched between two layers of soil. 07 We chose to incorporate the fibers into the soil using a blender to provide a more homogeneous distribution of the fibers in the 08 soil. We tested the impact of mixing time to establish a protocol which ensured a homogeneous distribution of the polyester 09 MP fibers into the soil, and that preserved the integrity of the MP fibers (which was evaluated through visual inspection using 10 a stereo microscope Zeiss Stemi 2000-C; Fig. 1a and b). The mixture of soil and fibers was mixed five times for 5 s each. The 11 same disturbance was also applied to the soil of the control treatment. 12 The three soil types, both treated and untreated, were used to fill 36 cylinders in total (height = 0.05 m, diameter = 0.05 m; 80

13 g of soil; Fig. 1c) to evaluate the impact of microplastic on soil properties (experiment I) and 36 soil trays of $0.30 \times 0.15 \times$

14 0.10 m³ perforated at the bottom (Fig. 1d) for the rainfall simulation tests (experiment II).



5 Figure 1. a) and b) contaminated soils; c) experimental units Experiment I; d) experimental units Experiment II.

For both treatments, that is the control (non-treated, Ctr) and the soils contaminated with polyester MP fibers, the soil samples were watered with distilled water to near field capacity by capillarity and then incubated in a growth chamber in the dark at 23 ± 2 °C and 60 ± 5 % relative humidity for nearly 6 months. During the incubation period, the soil samples were watered once a week with distilled water to field capacity by capillarity. The experimental units were re-randomized at each irrigation event.

21 2.2 Experiment I. Impact of polyester microplastic fiber on soil properties

For each soil type, the Ctr treatment was set up in 8 replicates while the MP treatment consisted of 4 replicates (for a total of 36 cylindrical samples).

The soil water retention curve was determined after incubation using the hanging water column apparatus (Dane et al., 2002a) for pressure head, h, values ranging from -0.03 to -1 m and the pressure plate extractor (Dane et al., 2002b) for h values ranging from -3.3 to -150 m.

Briefly, the soil samples were placed on the porous plate of a glass funnel and saturated from the bottom by progressively raising the water level in a graduated burette (height was adjustable). Equilibration at *h* values of -0.2, -0.1, and -0.05 m was obtained successively in steps of 24 hours each. Finally, the samples were fully saturated by submersion for 2 hours. From saturation, soil samples were desorbed by imposing a sequence of eight *h* values (h = -0.03, -0.05, -0.10, -0.20, -0.30, -0.50, -

31 0.70, and -1.0 m). At each h level, the volume of drained water into the burette was recorded. The volumetric water content, θ

32 (m³m⁻³), at each equilibrium stage was calculated by adding the drained volumes to the final θ value corresponding to h = -133 m that was determined by oven-drying the samples at 105°C. The dry soil bulk density, *BD* (g cm⁻³), was calculated by the 34 measured volume at the end of the experiment (i.e., h = -1 m) and using the oven-dried weight of the soil sample. Given that 35 polyester can withstand temperatures as high as 150°C and the added amount of fibers (0.5%) was low, the drying process was 36 not expected to have a significant effect on the measured *BD* values.

Then, the oven-dried soil samples were gently crushed and split into two sub-samples: one was used to determine the soil water content down to -150 m, while the other was used to determine the soil structure and the soil water-stable aggregates.

To determine the soil water content at lower pressure head values, the soil was packed to the same *BD* value of the larger samples in rings with a diameter of 0.05 m and a height of 0.01 m. These repacked soil samples were then used to determine the soil water content corresponding to h = -3.3, -10, -33 and -150 m using the pressure plate apparatus (Dane et al., 2002b).

The water retention model proposed by van Genuchten (1980) was then fitted to experimental data obtained for each soil sample. The water content values corresponding to saturation, θ_s , and to the pressure heads h = -0.10, -1.0, and -150 m ($\theta_{-0.1}, \theta_{-1.0}, \theta_{-1.0}, \theta_{-1.50}$) were then estimated from the model. The following "capacitive" indicators of soil physical quality were considered that are directly linked to the soil water retention properties (Iovino et al., 2016; Reynolds et al., 2009) :

46	Macroporosity	$P_{mac} = \theta_s - \theta_{-0.1}$	(1)
47	Air capacity	$AC = \theta_s - \theta_{-1.0}$	(2)
48	Plant available water capacity	$PAWC = \theta_{-1.0} - \theta_{-150}$	(3)

The P_{mac} indicator gives the volume of large (macro) pores (i.e. > 0.3 mm equivalent pore diameter), which indicates the soil's ability to quickly drain excess water and facilitate root proliferation (Reynolds et al., 2009). The *AC* index represents the ability of soil to store and provide essential soil air (Topp et al., 1997). The *PAWC* indicator is a measure of the ability of the soil to store and provide soil water that is available to crop roots.

To investigate soil aggregation and soil water-stable aggregates, we used a wet sieving apparatus (Retsch AS 200) with a set of stacked sieves mesh of 2, 1, 0.6, 0.5, 0.25, and 0.106 mm. For the soil aggregation, we closed the water valve and we performed a dry sieving using a 40 g subsample of air-dried soil from each treatment; the sieves were moved vertically at rate of 30 cycles per minute over a period of 3 minutes, and after recording the weight of each sieved fraction, the whole sample was reconstructed and used to assess water-stable aggregates by using the same procedure with the opened water valve. All the analyses were conducted in a laboratory under controlled conditions of temperature (mean T = 22° C).

59 Data obtained from dry and wet sieving were used to calculate:

60 1) percentage of newly formed aggregates (> 600 μ m; *NFA*) according to Lehmann et al. (2021):

61
$$NFA(\%) = \frac{W_{>600}}{W_{sample}} \times 100$$
 (4)

62 where $W_{>600}$ is the weight of the >600 µm fraction after air dried or wet sieving process, W_{sample} is the total weight of the 63 sample.

As the soil at the beginning of the experiment was sieved at 600 μ m, this metric represents the formation of new macroaggregates (> 600 μ m) certainly built from smaller size aggregates representing a shift in aggregate size distribution.

Mean weight diameter (*MWD*; mm) calculated on data derived from dry (*MWD_{dry}*) and wet (*MWD_{wet}*) sieving using
 the following equation (Deviren Saygin et al., 2012):

$$MWD = \sum_{i=1}^{7} \quad \frac{W_i}{W_{sample}} \times d_i \tag{5}$$

where *i* is the fraction size (> 2; 1-2; 1-0.6; 0.6-0.5; 0.5-0.25; 0.25-0.106; < 0.106 mm), W_i is the oven-dry weight of each fraction after sieving process (dry and wet); d_i is the mean diameter of each size fraction (i.e. the mean inter-sieve size; for the fraction > 2.0 mm, that was extremely small and always less than 1%, it was considered 2.2 mm).

72 2.3 Experiment II. Rainfall simulation

Soil with (MP) and without (Ctr) polyester MP fibers was used to perform the experiments with the rainfall simulator. For a given treatment, air dried soil was used to fill the soil tray after placing a cotton guard cloth at its bottom to avoid soil loss through the draining holes. The soil was gradually poured into the tray and it was compacted manually by repeatedly dropping the tray from a height of approximately 0.05-0.10 m until soil compaction ceased. Each soil sample had an initial bulk volume of $0.30 \times 0.15 \times 0.10 = 4.50 \times 10^{-3}$ m³. For a given soil and a given treatment (MP and Ctr), six soil trays were prepared (total sample size, N = 3 soils × 2 treatments × 6 soil trays = 36).

The rainfall simulator is a nozzle type rainfall simulator, very similar to those described by Iserloh et al. (2012) and Ries et al. (2009). The major parts of the rainfall simulator (Fig. 2a) are: a square metal frame (120 cm \times 120 cm) with a nozzle located at its centre and four telescopic steel legs in order to position the nozzle at different heights above the ground surface. A tank ensures the supply of water which is pressurized by a low pressure 12 V bilge pump (model Shurflo 2088 713-515) (Fig. 2b). The water supply to the tank assures a steady flow. The flow rate is regulated by the control valve located on a recirculation circuit and it is checked by a digital manometer (model PCE- DMM 10) characterized by an accuracy equal to \pm 0.5%. In order to check the pressure measure, a further analog manometer is installed.

Rainfall intensity and its uniformity over the wetted area were checked before performing the runoff and soil loss measurement experiment. Initially, a total of 55 plastic cups, each with an upper surface area of 35.3 cm², were placed on a circular metal frame, with a diameter of 60 cm, that was placed at a distance from the nozzle equal to the average distance between the nozzle and the soil trays and centred exactly below the nozzle of the rainfall simulator (Fig. 2c). Simulated rainfall was collected for 30 min and a rainfall intensity value was then determined for each cup. Taking into account that four trays were planned to simultaneously be subjected to a given rainfall event, rainfall uniformity among these trays was verified. In particular, the data obtained on the cups corresponding to each tray were averaged to obtain four mean rainfall intensity values, that is a value to be associated with each soil tray. Rainfall uniformity was assessed by calculating the following uniformity coefficient, CU (Christiansen, 1942):

(6)

$$CU = 100 \left(1 - rac{\sum_{i}^{n} |R_i - R|}{nR}
ight)$$

being n = 55 the number of cups, R_i = the rainfall intensity recorded by each cup and R = the mean rainfall intensity.



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Figure 2. a) Scheme of the rainfall simulator and b) the hydraulic setup; c) arrangement of the plastic cups to evaluate rainfall intensity uniformity and overlap between the plastic cups and the soil trays (in red); d) view of the experimental soil trays during a rainfall simulation event.

01 Each individual rainfall simulation experiment was carried out by placing four soil trays on a frame imposing a steepness of 02 15% to each try (Fig. 2d). This steepness value was chosen to consider a potentially favourable condition to appreciable soil 03 erosion processes and also in the perspective to replicate the experiment with natural rainfall events and larger plots in the 04 future. This new experiment could be performed at the Sicilian Sparacia experimental area, where micro-plots and plots were 05 established on a 14.9% slope (Bagarello & Ferro, 2004). For each soil try, surface runoff and the associated sediment load was 06 conveyed through a gutter to a plastic container and the same was done with reference to deep percolation. Lids and plastic 07 sheets were used in order to only expose soil surface to rainfall during the experiment. The soil tray with the soil was weighed before and immediately after the rainfall event to obtain the change in the stored water in the soil volume. The nozzle was 08 09 placed at a mean height of 1.70 m (measured from the middle of each tray). The whole experiment was carried out with a mean rainfall intensity of 33.4 mm h⁻¹ (CV = 3.0%) for 2 hours. The rainfall simulation experiment was completed in nine days. At 10 11 the beginning of each working day, rainfall intensity was measured by collecting water for ten minutes in a cylindrical bucket 12 with a diameter of 30 cm, and at a distance from the nozzle equal to the average distance between the nozzle and the soil travs. 13 At the end of the experiment, all containers were transported to the laboratory and dried at 105 °C to constant weight to 14 determine the total surface runoff volume, the total drained water volume and the associated load of solids. For the drained 15 water volume, this last quantity was always small and practically under the detectable quantity. Therefore, drained water was 16 essentially clear in all cases. The data were then used to calculate, for each soil tray, total runoff per unit area, V_e (mm), total percolation per unit area, P_e (mm), total soil loss per unit area, A_e (g/m²), and sediment concentration in the surface runoff 17 18 volume, C_e (g/m³). A check of the reliability of the V_e and P_e data was made, taking into account that the sum of total runoff, 19 total percolation and change in the stored water in the soil volume, denoted as S_{e} (mm), was expected to nearly coincide with 20 the rainfall depth for the simulated event, h_{e} .

21 2.4 Statistical analysis

The data of both experiments were analyzed in R (R Core Team, 2020) using a generalized least square models in the "nlme" package (Pinheiro et al., 2021) with implemented varIdDent() function to account for heterogeneity of variance. Model residuals were checked for heteroscedasticity and normal distribution.

All response variables within each soil type were compared between the two groups (MP minus Ctr) using the package "dabestr" (Ho et al., 2019) to calculate the effect sizes as unpaired mean differences and generate a bias-corrected and accelerated bootstrapped 95% Confidence Intervals (CIs). We used this combined approach based on the expanding recognition of the limitation of using only "P-value statistic" approach and avoid dichotomous cutoffs (Ho et al., 2019; Wasserstein and Lazar, 2020).

30 3 Results

31 3.1 Experiment I

The physical and hydrological effects of the treatment with polyester MP fibers differed with the soil type (Table S1). In particular, for the Vertisol, polyester MP fibers caused a decrease of bulk density (-9%), and an increase in air capacity (+ 34%; Fig. 3) whereas no effect was found on soil macroporosity and plant available water content (Fig. 4). In the Entisol, the contamination with polyester MP fibers did not cause any evident effect on the physical and hydrological characteristics. In the Alfisol, polyester MP fibers did not exert effects on *BD* but induced pronounced decreases in both *AC* (- 26%) and *P_{mac}* (-85%) and an increase in *PAWC* (+ 19%).



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Figure 3. Soil bulk density (BD) and air capacity (AC): raw data of Control (Ctr, grey dots) and polyester MP fibers added treatment (MP, colorued dots) are shown in the plot for each soil. The filled curve indicates the resampled distribution of mean differences (Δ), given the observed data. Horizontally aligned with the mean of the test group, Δ is indicated by the black circle. The 95% confidence interval of Δ is illustrated by the black vertical line.



Figure 4. Soil macroporosity index (P_{mac}) and plant available water content (PAWC): raw data of Control (Ctr, grey dots) and polyester MP fibers added treatment (MP, coloured dots) are shown in the plot for each soil. The filled curve indicates the resampled distribution of mean differences (Δ), given the observed data. Horizontally aligned with the mean of the test group, Δ is indicated by the black circle. The 95% confidence interval of Δ is illustrated by the black vertical line.

The data obtained from dry sieving show how the contamination with polyester MP fibers negatively affected the percentage of newly formed aggregates (> 600 μ m) (-32% in Vertisol; -47% in Entisol; -33% in Alfisol; Fig. 5). Only a small fraction of the newly formed aggregates was stable to the subsequent wet sieving (Fig. 5). In addition, MP had a minimal positive effect on NFA_{wet} for the Vertisol but it was uninfluential for the Entisol and the Alfisol (Fig. 5). Polyester MP fibers contamination determined a general decrease of MWD_{dry} (-21% in Vertisol and Alfisol; -13% in Entisol; Fig. 6), while no effects due to polyester MP fibers were observed on MWD when soil samples were wet sieved (Fig. 6; Tables S2 and S3).

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Figure 5. Newly formed aggregates (> 600 µm) calculated from dry (NFA_{dry}) and wet (NFA_{wel}) sieving: raw data of Control (Ctr, grey dots) and polyester MP fibers added treatment (MP, coloured dots) are shown in the plot for each soil. The filled curve indicates the resampled distribution of mean differences (Δ), given the observed data. Horizontally aligned with the mean of the test group, Δ is indicated by the 59 black circle. The 95% confidence interval of Δ is illustrated by the black vertical line.



60minus Ctrminus Ctr61Figure 6. Mean weight diameter (MWD; mm) calculated from dry (MWD_dry) and wet (MWD_wet) sieving: raw data of Control (Ctr, grey dots)62and polyester MP fibers added treatment (MP add, coloured dots) are shown in the plot for each soil. The filled curve indicates the resampled63distribution of mean differences (Δ), given the observed data. Horizontally aligned with the mean of the test group, Δ is indicated by the64black circle. The 95% confidence interval of Δ is illustrated by the black vertical line.

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66 **3.2 Experiment II**

67 **3.2.1 Preliminary check of the simulated rainfall**

Rainfall intensities, *RI*, measured by the 55 cups ranged from 31.3 to 37.4 mm h⁻¹, with a mean of 33.8 mm h⁻¹, a coefficient of variation, CV = 4.7% and a uniformity coefficient, *CU*, equal to 96.1%. Considering only the cups corresponding to the area of the soil trays, RI = 33.1 mm h⁻¹ and CU = 97.1% were obtained. At the individual soil tray scale, the mean rainfall intensity varied from 32.5 to 34.0 mm h⁻¹, depending on the tray, and the *CU* values were in the range 96.8-98.2%. As compared with the performances of other rainfall simulators (Iserloh et al., 2013), these results suggested that rainfall intensities were

reasonably uniform among the four simultaneously tested soil trays and also on each individual tray.

- 74 Mean rainfall intensities were overall stable in the nine days of the rainfall simulator experiment (from 32.1 to 34.9 mm h⁻¹).
- The whole experiment was carried out with a mean rainfall intensity of 33.4 mm h⁻¹ (CV = 3.0%) and it had a duration of 2
- hours. Therefore, the mean rainfall depth, h_e , was 66.8 mm.
- On average, $h_e (S_e + V_e + P_e)$ was equal to 5.3 mm. However, neglecting the seven cases characterized by values of $h_e (S_e + P_e)$
- $V_e + P_e$) greater than 20 mm, for which some anomaly occurred in the experiment, the mean of the remaining 29 values of h_e -
- 79 $(S_e + V_e + P_e)$ is very close to zero (0.14 mm). Considering the consistency between h_e and $(S_e + V_e + P_e)$, the seven suspect
- 80 runs were excluded from the considered dataset to check soil treatment effect on V_e , P_e , A_e , and C_e

81 **3.2.2** Runoff, percolation, soil erosion and sediment concentration

- 82 Soil polyester MP fibers contamination did not induce changes in surface runoff (Fig. 7) in all soil types, although a slight
- V_e decrease of V_e was perceived for both the Entisol and the Alfisol (unpaired mean difference -4.19 and -7.65 for Entisol and
- 84 Alfisol respectively; 95% CIs -10 to 3.74 for the Entisol and -16.7 to 0.69 for the Alfisol; Table S1). However, it affected
- 85 percolation, which varied with the soil type. In particular, the analysis carried out separately by soil type allowed to highlight
- 86 some different responses of the soils under evaluation; in the Alfisol, the contamination with polyester MP fibers increased
- 87 percolation by 144% (Fig. 7) as compared with the Ctr treatment. No effects were observed in the Vertisol and in the Entisol.



88minus Ctrminus Ctr89Figure 7. Surface runoff and percolation: raw data of Control (Ctr, grey dots) and polyester MP fibers added treatment (MP, coloured dots)90are shown in the plot for each soil. The filled curve indicates the resampled distribution of mean differences (Δ), given the observed data.91Horizontally aligned with the mean of the test group, Δ is indicated by the black circle. The 95% confidence interval of Δ is illustrated by92the black vertical line.

Soil erosion varied widely among the soils under evaluation, resulting in a decidedly limited amount in Vertisol and in a particularly high one in Alfisol (Fig. 8; Table S1). Contamination with polyester MP fibers did not cause variations in the amount of eroded soil in the Vertisol (unpaired mean difference -0.01; 95% CIs -0.06 to 0.06), while it resulted in substantial reductions in both Entisol (-75%; unpaired mean difference -0.46; 95% CIs -0.63 to -0.07) and Alfisol (-80%; unpaired mean difference -1.43; 95% CIs -2.11 to -1.05). Similar results were observed for sediment concentration, as shown in Figure 8 and in Table S1.



99 ÓÓ Figure 8. Soil erosion and sediment concentration: raw data of Control (Ctr, grey dots) and polyester MP fibers added treatment (MP, 01 coloured dots) are shown in the plot for each soil. The filled curve indicates the resampled distribution of mean differences (Δ), given the observed data. Horizontally aligned with the mean of the test group, Δ is indicated by the black circle. The 95% confidence interval of Δ is

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03 illustrated by the black vertical line.

04 **4** Discussion

05 As hypothesized, the contamination with polyester MP fibers generally resulted in differentiated effects on soil aggregation 06 and hydrological characteristics among the tested soils. In particular, the presence of polyester MP fibers determined a decrease 07 of the soil bulk density on the Vertisol, a slight decrease on the Entisol, and no effects on the Alfisol. The data available in the 08 literature about the impact of polyester MP fibers on soil bulk density are limited and not always concordant. For instance, 09 Machado et al. (2018 and 2019) observed a decrease in bulk density of a loamy sand soil at increasing concentration of 10 polyester MP fibers while Zhang et al. (2019) in a study conducted on a clay loam soil (in field and in greenhouse conditions) 11 found no differences in soil bulk density.

12 Polyester MP fiber contamination negatively affected air capacity and macroporosity index of the Alfisol, maybe as a 13 consequence of the smaller mean diameter of the newly formed aggregates. Given that the BD of this soil was unaffected by

14 polyester MP fiber contamination, it can be supposed that total porosity was redistributed from macropores to meso/micropores 15 as confirmed by the increase of PAWC. No modification of the capacitive indicators of soil physical quality was observed on 16 the Entisol. For the Vertisol, the increase of both total porosity (decrease of *BD*) and *AC* suggested that MP addition favoured 17 macroporosity. Meso/micropores did not change appreciably, as indicated by the similar *PAWC* results for the Ctr and MP 18 treatments.

19 Our data obtained from dry sieving show that the presence of polyester MP fibers changed the cohesion between aggregate-20 forming particles with a general reduction (although with different effect size on the three soil types) of the formation of new 21 macro-aggregates (> 600 µm). Similar results were reported by Boots et al. (2019) and Lozano et al. (2021). Zhang & Liu 22 (2018) in a survey study of microplastic contamination in agricultural soils found that the abundance of aggregate-associated 23 plastic fibers is greater in the micro-aggregates than in the macro-aggregates and this, according to the same authors, would 24 suggest that the presence of fibers in the micro-aggregate limits their possibility to be combined into macro-aggregate. 25 Machado et al. (2018) found that contamination of a loamy sand soil with polyester MP fibers decreased the fraction of soil 26 forming dry aggregates larger than 1 mm but at the same time increased the formation of large soil clumps and therefore 27 potentially provided additional macro-structures, absent in the non-contaminated soil. In contrast, Zhang et al. (2019) found 28 that polyester MP fibers improve soil aggregation helping to entangle soil particles more efficiently to form aggregates. 29 Interestingly, in this experiment, the negative effects induced by the addition of polyester MP fibers on the aggregation capacity 30 of the soils were, to a certain extent, of a decreasing magnitude as TOC increased (Vertisol>Alfisol>Entisol; according to the 31 results of the dry sieving; Fig. 5, NFAdry; Table 1). Therefore, it would seem that the addition of polyester MP fibers interfered 32 with the formation of macro-aggregates by altering the binding mechanism in the soil. In particular, high organic matter levels 33 made the MP contamination effects smaller. Similar results were also obtained by Liang et al. (2021) who found that the effects 34 of polyester MP fibers on soil aggregation were organic matter dependent.

35 The presence of the contaminant, although determining a general decrease in newly formed aggregates, did not affect the 36 formation of water stable aggregates, since we generally did not observe any appreciable effect of the polyester MP fibers on 37 the MWD and NFA values obtained with the wet sieving. In contrast, Zhang et al. (2019) observed a significant increase of 38 stability of the macro-aggregates (> 2 mm) in water and in the volume of the macro-pores (> 30 μ m) after addition of polyester 39 MP fibers in a clay soil. On the other hand, Machado et al. (2018) found a significant decrease in water-stable aggregates with 40 increasing polyester MP fiber concentration in a sandy loam soil. Evidently, soil properties play an important role in guiding 41 the effects of polyester MP fibers in the formation of macro-aggregates and their water stability. The interactions that can 42 occur between polyester MP fibers and fine soil particles are still poorly studied. Our results show that the effects of polyester 43 MP fibers on soil structure and hydrological characteristics are most probably related to the different characteristics of the soil 44 used in the experiments, as already hypothesized by Xu et al. (2020) and Zhang et al. (2019). Our three soil types differed for 45 various characteristics (e.g. texture, clay mineralogy, TN, TOC, CEC) which could have played a role in the observed responses 46 to the contamination. Certainly, it is very difficult to identify how and on which factor/s polyester MP fibers influenced our 47 results, also considering the several biotic and abiotic factors that interplay in the process of soil aggregation. For instance, our 48 three soil types widely differ for clay mineralogy: montmorillonite in the Vertisols, illite in the Entisols, and kaolinite in the

Alfisols. These three clay types largely differ for size, shape, specific surface area, and structure, and, as a consequence, for their ability to form aggregates (Lal and Shukla, 2004). Therefore, it is possible to presume that polyester MP fibers could have differentially affected the ability of the above-mentioned clay types to form aggregates during the incubation period. Those aspects deserve further investigation to clarify the mechanisms causing the different responses observed.

53 Polyester MP fiber contamination did not affect surface runoff and drainage in the Vertisol and in the Entisol although a slight 54 decrease of V_e was perceived for this last soil. However, it caused a slight decrease of surface runoff and an increase of 55 percolation in the Alfisol, which has the coarsest texture among the three tested soils (Table 1). Moreover, the three studied 56 soils differed widely in their susceptibility to erosion (Vertisol < Entisol < Alfisol). This result was partly expected, as many 57 authors have highlighted that more silt and less organic matter in the soil enhance erodibility(Bonilla and Johnson, 2012; 58 Chaney and Swift, 1984; Meyer and Harmon, 1984; Wischmeier and Smith, 1978). Adding polyester MP fibers had no effect 59 on soil erosion in the least erodible soil (Vertisol), whereas it resulted in a pronounced decrease of soil erosion in the more 60 erodible soils (Entisol, Alfisol). In other words, the tendency of microplastics to decrease soil erosion increased as the soil became intrinsically more erodible. The data therefore showed that contamination with polyester MP fibers modified only a 61 62 little and not for all soils the soil hydrological behaviour (rainfall partition into surface runoff and percolation) but it affected 63 the soil erosion phenomena. Another way to summarise these results is that a decrease of soil erosion (Fig. 8) was linked with 64 a slight decrease of the total runoff (Fig. 7). The MP influence on soil erosion was moderate or even negligible when the soil 65 was inherently little erodible. Instead, it became appreciable in those cases in which the erosion phenomena were noticeable.

At the end of the rainfall experiment, we observed differences between the Ctr and MP trays at the soil surface. In the trays of the Ctr treatment, micro-rills oriented parallel to the slope appeared during the rainfall-runoff event, especially in the contact zone between the soil and the box walls, or small cracks developed soon after rainfall, depending on the soil type (Fig. 9). Instead, neither micro-rills nor cracks were ever detected in the soil trays of the MP treatment. Therefore, it seems that the polyester MP fibers performed a soil particle binding action, possibly microbially-mediated, that likely induced a decrease of soil erodibility. This decrease was not suggested by the sieving experiments, probably because mechanical impact of sieving



Figure 9. Experimental units of Experiment 2 (Vertisol left hand side 3andAlfisol right hand side) after the rainfall simulation.383

cannot be considered equivalent to the impact of the raindrops (Fox and Le Bissonnais, 1998; Loch, 1994).

Moreover, after rainfall, the soil surface of the Ctr trays generally appeared smoother than the ones of the MP trays and, especially in the darker soil, the polyester MP fibers were noted to form a diffuse fluff on the soil surface by the end of the experiment (Fig.9). Therefore, polyester MP fibers appeared to generally induce a greater resistance of the soil to flow tractive forces. An additional possible reason for the observed results was that these fibers were exposed to some degree as a consequence of erosion

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- during the early stage of the experiment but they remained entrapped by the subsoil. Therefore, the soil surface of the MP trays appeared to have an additional micro-roughness compared to that of the Ctr travs. Consequently, flow velocity and sediment
- appeared to have an additional micro-roughness compared to that of the Ctr trays. Consequently, flow velocity and sediment
 transport capacity were likely greater in the Ctr trays than in the MP ones (Zhang et al., 2011). Perhaps, the fluff formed by
 the exposed fibers also contributed to limit rainfall detachment, acting as a mulch.
- 89 The active erosion processes vary with the measurement scale (Cammeraat, 2002). At the soil travs scale adopted in this 90 investigation, erosion is expected to be due to rain splash and sheet flowrainfall impact and interrill flow (Bagarello and Ferro, 91 2004) and it should be a transport-limited process as a consequence of the reduced rain impacted flow and the limitation of 92 flow velocity (Boix-Fayos et al., 2006; Chaplot and Le Bissonnais, 2000). Therefore, the collected data provide information 93 on particle detachment and the early stage of their transport that can be expected to occur in upland agricultural soils during 94 intense rainfall events. However, in agricultural fields, rill erosion can dominate total soil erosion due to the simultaneous 95 occurrence of long slopes (dozens of meters or more) and the exposure of bare soil surfaces to rainfall in some periods of the 96 year (Reiman and Brodowski, 2005). The effects of polyester MP fibers on rill erosion require specific testing but, also 97 according to this investigation, it could be expected that rill development is hindered since the presence of the contaminant 98 makes the soil intrinsically less erodible.
- 99 Additional investigations should also be carried out by varying the steepness of the soil trays to verify if a linear or less than
- 00 linear relationship between interrill erosion rate and slope (Fox & Bryan, 2000) remains detectable after MP fibers addition
- 01 into the soil.

02 5 Conclusion

03 Although the current MP contamination level in agroecosystems is some orders of magnitude below the concentration applied 04 in our experiment, in some areas, it is steadily increasing (Büks and Kaupenjohann, 2020); therefore it is of key importance to 05 investigate such contamination levels which may represent future scenarios, as is common practice in global change biology. -06 The results showed that the presence of polyester MP fibers limits the neoformation of soil aggregates (labile in incubation 07 period applied in our experiments). Likely, polyester MP fibers hinder the natural aggregation processes since they interpose 08 between the fine soil particles and hence reduce the possibility of establishing bonds between these. Such results seem in 09 contrast with what we observed in our rainfall simulation experiment where polyester MP fibers reduced soil loss and sediment 10 concentration, especially in the most erodible soils, which would suggest that the polyester MP fibers had a soil particle binding 11 effect that reduced erodibility of the porous medium. In particular, the lack of rills in the MP treated trays suggested that 12 polyester MP fibers induced a greater resistance of the soil to flow tractive forces of runoff. Other factors that could have 13 played a role on the observed results include: i) mechanical impact of sieving cannot be considered equivalent to raindrop 14 impact; ii) the presence of polyester MP fibers favoured a higher micro-roughness at the soil surface as compared with the 15 non-treated soil and consequently the flow velocity and sediment transport capacity decreased with the addition of polyester 16 MP fibers; and iii) the contaminant, acting as a mulch, could have exerted a physical protective action by intercepting the

- 17 raindrops, allowing the dissipation of their kinetic energy, and limiting the splash effect that leads to the disintegration of the
- 18 aggregates.
- In conclusion, our experiments showed that the contamination of soils with polyester MP fibers exerts an impact on the soil structure, susceptibility to erosion and other hydrological characteristics as a function of soil type. This aspect is of great interest and underlines how a complete understanding of the potential impacts of polyester MP fibers on terrestrial ecosystems requires special attention to the processes that occur in the soil and to the knowledge of the mechanisms underlying the different responses. The effects we observed are short-term (the incubation period applied in our experiments was ~6 months) and we
- 24 currently do not know whether such effects would be maintained in the long term. Moreover, in this study, we used fresh MP
- 25 particles and the contamination with aged MP particles may lead to different results (Waldman and Rillig, 2020). However,
- 26 there is insufficient information about aged MP particles at this time.
- 27 The applied experimental methodologies in this investigation provide much information on the effect of polyester MP fibers
- addition on several physical soil properties and hydrological processes. These methodologies should be applied in the future
- 29 to different MPs concentrations and types to better understand the effects of those contaminants on different soils and to help
- 30 cover the potentially very large parameter space that, as suggested by Rillig & Lehmann (2020), represents a major challenge
- 31 of MPs research in terrestrial ecosystems.

32 Author contributions

- 33 All authors (RI, GA, VB, FGC, DG, MI, AL, MCR, and ASF) conceptualized the study. GA, DG, and ASF acquired the funds
- 34 to conduct the experiment. RI, FGC, DG, MI, and ASF carried out the experiments. RI, DG, and ASF analyzed the data. RI,
- 35 DG, GA, ASF, MI, and VB wrote the first draft of the manuscript. AL and MCR collaborated on the ideas and contributed
- 36 critically to the drafts. All authors gave the final approval for publication.

37 Competing interests

38 The authors declare that they have no conflict of interest.

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