

ORIGINAL ARTICLE

Imaging of microplastic distribution–related unsaturated water flow in sand

A. Cramer^{1,2}  | P. Benard^{1,3}  | A. Kaestner⁴  | M. Zarebanadkouki⁵  |
P. Lehmann¹  | A. Carminati¹ 

¹Physics of Soils and Terrestrial Ecosystems, ETH Zurich, Zurich, Switzerland

²Institute of Soil Physics and Rural Water Management, BOKU University, Vienna, Austria

³Soil Mechanics and Soil Management, SLU Uppsala, Uppsala, Sweden

⁴Instrument Scientist ICON, PSI Villigen, Villigen, Switzerland

⁵Soil Biophysics and Environmental Systems, TUM Munich, Freising, Germany

Correspondence

A. Carminati, Physics of Soils and Terrestrial Ecosystems, ETH Zurich, Universitätsstrasse 16, 8092 Zurich, Switzerland.
Email: andrea.carminati@usys.ethz.ch

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Abstract

Soil is the largest sink of microplastic (MP) in terrestrial ecosystems, yet little is known about the transport of MP in soils and its effect on water flow. Since pristine MP can generally be considered water repellent, the transport of MP and water flow are subject to feedback processes. To investigate MP transport in porous media and the interactions with water dynamics, we applied neutron and X-ray imaging methods to columns filled with mixtures of sand (700–1200 µm) and MP (polyethylene terephthalate, 20–75 µm). Simultaneous neutron and X-ray CT were used to image MP distribution in 3D before and after each experiment, while time-series neutron radiography was applied to image water distribution during repeated wetting and drying cycles. The imaging revealed that MP impacted water infiltration. High local MP content caused an initial delay in infiltration, plausibly due to water repellency, but was eventually bypassed by water flow. Rapid and preferential propagation of the wetting front as well as reduced local water saturation above the wetting front were the outcome. The discrepancy between the initial delay and rapid stage infiltration was increased with an increase in total MP content. No significant vertical transport of MP could be observed during two wetting and drying cycles, probably since water bypassed regions containing MP. In conclusion, the interactions between hydrophobic MP and water impact the infiltration of water, with implications for the retention and fate of MP in soils.

Plain Language Summary

A lot of microplastic is deposited in soil, but we do not fully understand how it moves in soil or how it affects water flow. Microplastic is usually water repellent. This can create an interaction that changes how water and microplastic move in soil. To study this, we filled columns with sand-microplastic mixtures and used imaging

Abbreviations: A, anisotropy; AWI, air-water interface; CT, computed tomography; FA, fractional anisotropy; L, linearity; MP, microplastic; PET, polyethylene terephthalate; PS, polystyrene.

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tools. These tools allowed us to see where microplastic was before and after wetting. We also tracked how water spread in the columns.

Microplastic affected how water soaked into the sand. Areas with a lot of microplastic slowed down water flow at first because it repelled water. But over time, water found ways to flow around these areas. The microplastic did not move vertically through the sand during wetting, likely because water avoided the areas with microplastic. Microplastic can change how water flows through soil, and this affects where microplastic ends up over time. This is important to assess the risk of groundwater contamination by microplastic.

1 | INTRODUCTION

Microplastic (MP), defined as plastic particles of diameters <5 mm, contaminates our environment worldwide (Geyer et al., 2017; Liu et al., 2023). Besides aquatic environments, terrestrial ecosystems are reported to contain large amounts of MP of different origin, polymer type, shape, size, and state of degradation (Geyer et al., 2017; Xu et al., 2020). Soils are large sinks of MP with anthropogenically influenced areas containing the highest number of particles (Lam et al., 2018; Osman et al., 2023; Xu et al., 2020). Additionally, rural and remote areas like Antarctica have also been shown to be affected by MP contamination (Kelly et al., 2020; Lam et al., 2018; Scheurer & Bigalke, 2018). Polyethylene-group materials used for packaging constitute the bulk of plastic waste (Lam et al., 2018; Osman et al., 2023). Sources of MP contamination are identified as mismanagement of polymer-containing waste, for example, sewage sludge application on agricultural fields, as well as the neglected life cycle of polymer-containing consumer products, for example, cosmetics, clothing, and packaging materials (Anagnosti et al., 2021; Lam et al., 2018; Nizzetto et al., 2016; Osman et al., 2023). Besides long-range transport of airborne MP and its subsequent deposition on terrestrial surfaces, degradation of larger polymer fragments poses a potential source of MP input into soils (Bläsing & Amelung, 2018; Khalid et al., 2023; Xu et al., 2020). Mechanism of MP incorporation into soil are numerous, including plowing of agricultural fields, mulch film residue degradation, and bioturbation (Huerta Lwanga et al., 2017; Khalid et al., 2023; Rillig, Ingraffia, et al., 2017; Shafea et al., 2023; Xu et al., 2020).

While most studies focus on developing methods to detect and quantify MP in soils (e.g., Nabi et al., 2022; Tirkey & Upadhyay, 2021; Tophinke et al., 2022), less attention is dedicated to the effects of MP on soil properties, processes, and related spatial distribution of MP. However, there are plenty of studies showing a soil depth-dependent occurrence of MP and MP enrichments in the top soil (Heinze et al., 2024; Vinoth & Merline, 2021; Yi et al., 2020). Though, when a certain vol-

ume of soil is analyzed for its MP content, it is done based on a highly intensive procedure (density separation), destroying the structure of the soil and providing no information on the spatial distribution of MP within the soil volume. Following the deposition pathways of polymers to the environment, it can be assumed that MP, like all soil constituents, are unlikely to be homogeneously distributed. Furthermore, MP has the potential of affecting the physicochemical properties of soils, and, vice versa, the fate of MP can be influenced by soil characteristics. Various effects, negative or positive, on plant-soil systems, water dynamics, soil respiration, soil aggregation, etc., have been reported depending on polymer type, shape, and state of degradation (Cramer et al., 2022, 2023; Ju et al., 2023; Lozano & Rillig, 2020; Lozano et al., 2023; Rillig et al., 2021; Xie et al., 2023). Furthermore, MP may be retained in soil (Ju et al., 2023; Rillig et al., 2021; Xu et al., 2020), which could be related to its low wettability. The low wettability of MP acts to reduce the contact between MP and water, especially under unsaturated conditions (Wan & Wilson, 1994). This would cause water to bypass soil pores with high MP content. On the other hand, the water repellency of MP would drive it onto the air-water interface (AWI), and fluctuations of the AWIs could play a role in the displacement of MP, at least on the pore scale.

Wettability and physicochemical properties of MP depend on the degradation state and polymer type (Cramer et al., 2023; Meides et al., 2021; Schefer et al., 2023; Shah et al., 2008). A number of MP, for example, pristine PET (polyethylene terephthalate) and polystyrene, are water repellent, although to a different extent (Cramer et al., 2023; Schefer et al., 2023). However, polymer surfaces can be functionalized by UV radiation when exposed to sunlight, increasing their wettability (Ju et al., 2023; Meides et al., 2021; Menzel et al., 2022; Song et al., 2017). Furthermore, over time, MP becomes part of the soil matrix, and eco-coronas can form on its surface, changing its wettability (Cramer et al., 2023; Rillig et al., 2021; Rillig, Ingraffia, et al., 2017; Schefer et al., 2023; Yao et al., 2023). Nevertheless, low wettability pristine MP particles in soil might prevent extensive contact with

soil water (Cramer et al., 2022, 2023), and thus, colonization, degradation, and functionalization processes like hydrolysis and metabolization by soil microorganisms may be prevented or delayed. The time necessary for these processes to take place in the environment and to overcome the MP inherent hydrophobicity remains unclear.

To simultaneously investigate the transport of MP and water in porous media and to explore the effects of MP heterogeneous distribution and water repellency on the infiltration and percolation of water, two wetting and drying cycles in columns consisting of sand (700–1200 μm) mixed with PET (20–75 μm) in three different contents were monitored. The homogenous structure of the sand allows one to directly relate observed effects on water infiltration to MP and to track the mobility of MP. The initial distribution of MP as well as the distribution after each wetting and drying was simultaneously imaged with neutron and X-ray tomography. Additionally, time-series neutron radiographs were captured during each wetting to capture water infiltration dynamics. We hypothesize that due to the low wettability of MP, infiltrating water bypasses sand pores containing MP, causing locally reduced water contents changes in infiltration dynamics and, in turn, affecting MP transport.

2 | MATERIALS AND METHODS

2.1 | Investigated media

We mixed MP with sand at three MP contents and added a control with pure sand (see Table 1). As MP, we used PET provided by Veolia Deutschland GmbH. The raw plastic was processed using a centrifugal mill (ZM200, Retsch GmbH) and then air-jet sieved into fractions with diameters ranging between 20 and 75 μm . Particle image analysis (Microtrac, Retsch GmbH) of the resulting MP fraction showed irregularly shaped particles, ranging from nearly spherical to fibrous, mimicking a realistic and environmentally relevant composition of MP with regard to shape and size distribution. MP was mixed with sand (Raneem, Sand-Schulz GmbH), which ranged in diameter from 700 to 1200 μm . The sand was treated with hydrogen peroxide and thoroughly washed with deionized water to remove organic matter residues to solely capture the effect of MP on the wettability of the sand matrix. Additionally, magnetic particles were removed with a magnet to prevent imaging artefacts from high X-ray attenuation from iron-containing particles. Both materials, sand and MP, were characterized in terms of wettability and grain size distributions in previous studies by Cramer et al. (2022, 2023). The assessment of static contact angles (CA) was conducted using the Sessile Drop Method, following the procedure outlined by Bachmann et al. (2000, 2003). The CA, determined by the relation of the three involved inter-

Core Ideas

- There is high awareness of microplastic (MP) contaminating environments, especially soil, raising concerns worldwide.
- Soil is the largest sink of MP in terrestrial ecosystems, yet we know little about the transport mechanisms of MP in soil and its effect on water dynamics.
- MP debris is usually more water repellent than most other soil constituents and represents a hydrophobic surface addition to soil, potentially increasing soil water repellency.
- We present an analysis of MP within porous media and show effects on water infiltration and percolation.
- Furthermore, we show that the transport of hydrophobic MPs does not adhere to the mechanisms that more wettable soil particles underlie.

facial energies (liquid-vapor, solid-vapor, and solid-liquid), is formed at the three-phase contact line. It controls whether water can spread across a surface, and it can be used as a measure for the wettability of a surface (Bachmann et al., 2000). In porous systems, a liquid may wet a pore at a CA < 90°, whereas a CA > 90° leads to beading of a liquid and promotes uneven wetting. The sand had a static CA of ca. 35° and the PET of ca. 123°. Therefore, the MP used in this study can be categorized as hydrophobic.

2.2 | Experimental setup

Cylindrical aluminum columns with a wall thickness of 1 mm, an outer diameter of 20 mm, and a height of 70 mm were filled with different sand and MP mixtures (see Table 1). The bottom of each column was covered with water-permeable tissue to prevent material loss and to provide outflow. In repetition of one, sample variants of control (pure sand), MP 0.35% (w/w), MP 1.05% (w/w), and MP 2.10% (w/w) were prepared and imaged with dual neutron and X-ray imaging techniques. The analyzed “region of interest” of the samples was the top 17 mm, where the peak MP content was detected (see Figures 1 and 2). The MP contents were chosen based on static CA extrapolations shown in Figure 3 in Cramer et al. (2022). These combinations of wettable and non-wettable surfaces (here sand and MP) theoretically reflect average static CA of 60°, 90°, and >90°.

MP and sand were mixed in glass beakers and subsequently poured into the sample containers. The dry mixing proce-

TABLE 1 Experimental setup of the samples for imaging.

Variant	Sand (g)	Sand volume fraction (%)	MP (g)	MP volume fraction (%)
Control	27.40	58.05	0.00	0.00
MP 0.35%	27.40	58.05	0.10	1.12
MP 1.05%	27.41	58.07	0.29	3.20
MP 2.10%	27.43	58.11	0.58	5.37

Abbreviation: MP, microplastic.

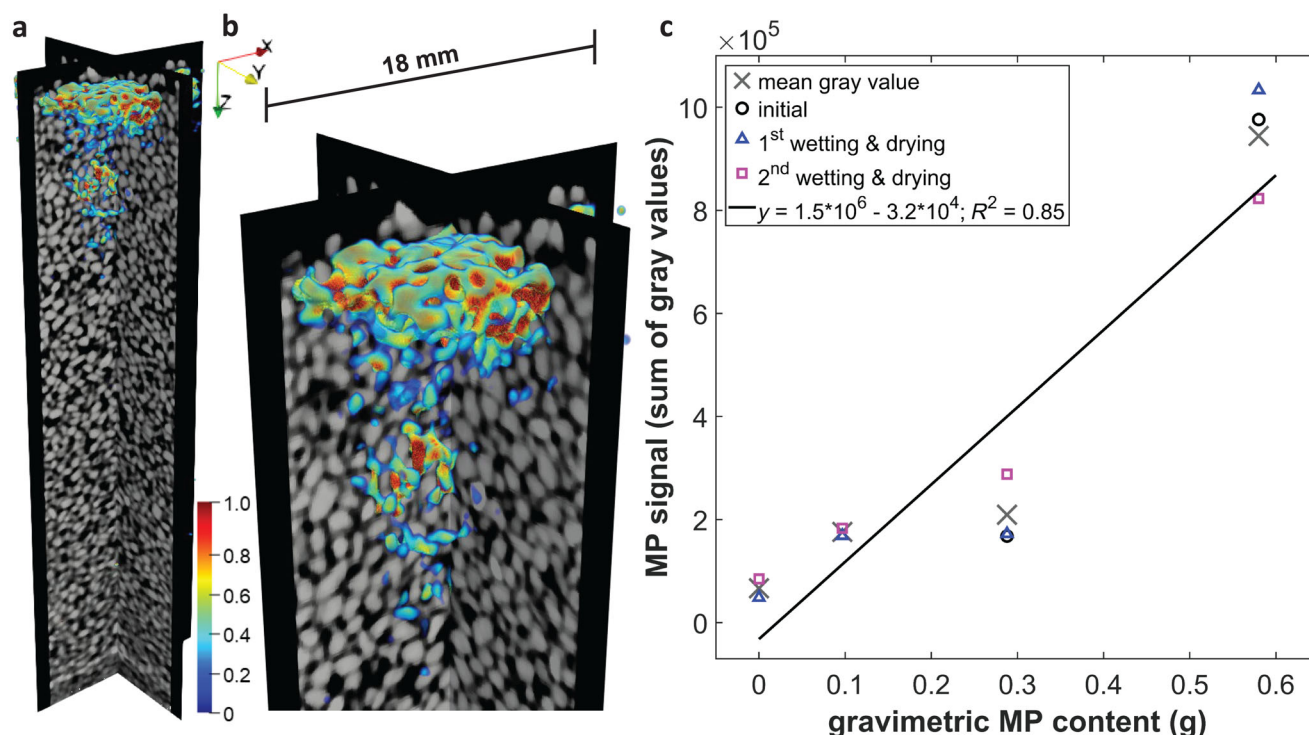


FIGURE 1 (a) 3D reconstruction of the combined neutron and X-ray computed tomography (CT) images. The sand particles retrieved from X-ray imaging are shown in gray, and the microplastic (MP) signal (gray values) within the pore space retrieved from dry neutron imaging is shown in vibrant colors ranging from blue (low content) to red (high content). The reconstructed and interpolated voxel size is 68 μm . The depicted sample is the MP 2.10% variant in its initial distribution before the first wetting and drying cycle. (b) Close-up of the analyzed region of interest of the peak MP content. (c) MP signal (sum of gray values caused by the presence of MP in each voxel) retrieved from dry neutron CTs after removing volumes assigned to sand (from X-ray CTs) over the corresponding gravimetric MP content of the treatments. The mean MP signal of each variant is depicted by a gray “X,” while the individual signals from each wetting and drying cycle are shown as color-coded circles, triangles, and squares (see legend). The black line indicates the result of a linear regression.

ture resulted in large MP remnants in the glass beaker, which were translocated on top of the sample. Each sample was concussed 10 times to ensure the intrusion of MP into the sample matrix. This led to a heterogeneous distribution of MP within the column and, when following the deposition pathways of polymers to the environment, mimicked locally MP-enriched areas of soil.

Constant water infiltration rates were imposed using a peristaltic pump (Ismatec-ISM937C, Cole-Parmer Instrument Company Ltd.). To establish an unsaturated water flow through the sample, the pump was set to a flow rate of 1 mL

min^{-1} (3.93 mm min^{-1}) for 25 s, resulting in ca. 0.42 mL (1.64 mm) of water applied to the samples from above via a syringe positioned over the center of the samples. The lower boundary condition is a seepage face boundary, so water and air were able to exit the column at the bottom under wet conditions. Afterward, water was drained from the bottom and then left to evaporate the remaining water for ca. 24 h at ambient temperature (25°C) prior to repeated irrigations. The infiltration experiments were recorded with time-series neutron radiography. Before and after each wetting and drying cycle, the 3D distribution of MP and sand was simultane-

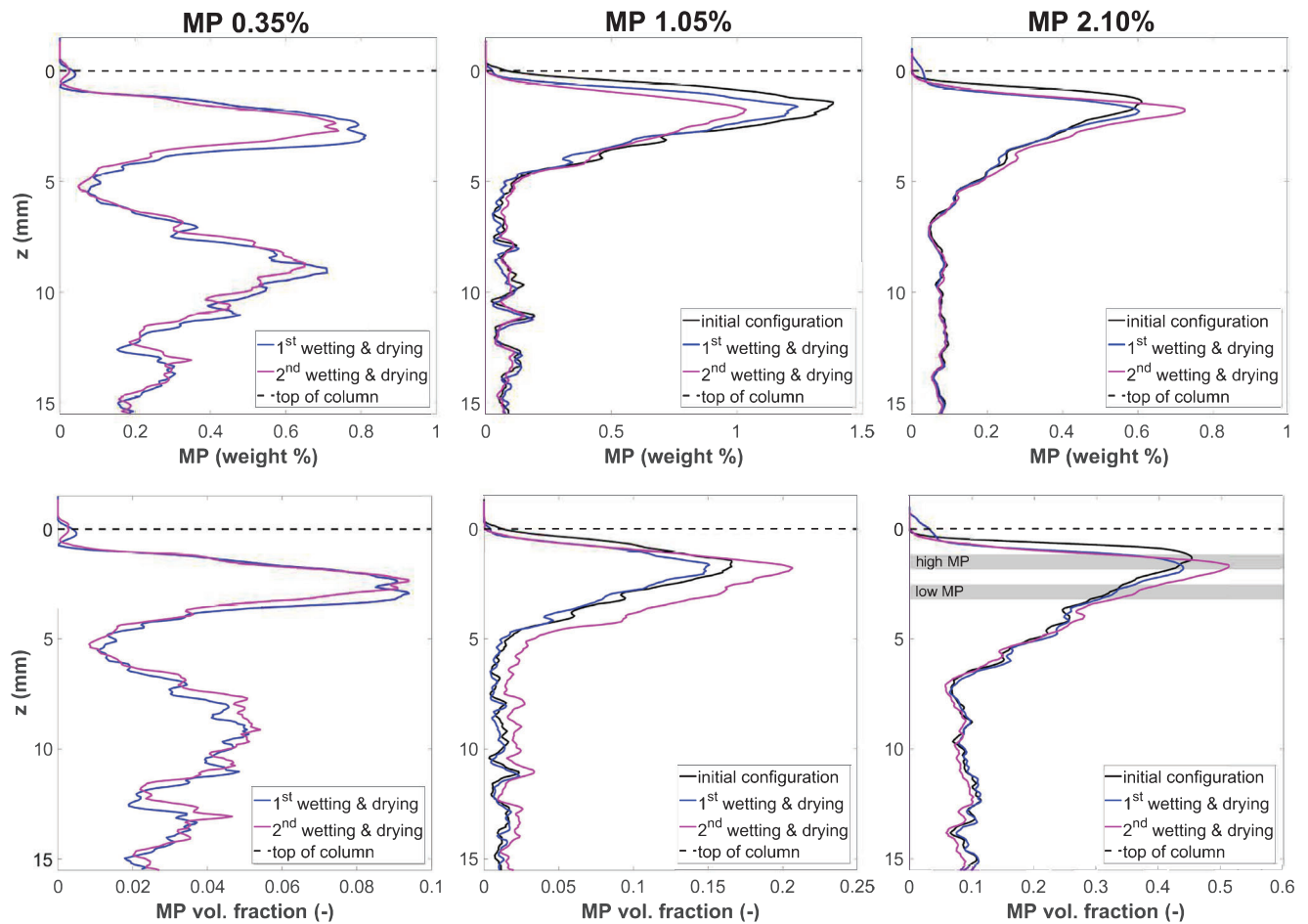


FIGURE 2 Top row: Microplastic (MP) weight percentage along the top 17 mm of the peak MP content of the profile (z). Bottom row: Volume fraction of microplastic along the top 17 mm. From left to right, the sample variants MP 0.35%, 1.05%, and 2.10% are depicted (note the different scale on x -axes). The volume fraction describes the MP occupied slice volume. Black lines represent the initial distribution of MP (missing for MP 0.35% due to beam failure), blue after the first wetting and drying cycle, and magenta after the second cycle. The dashed gray lines indicate the top of the sample cylinder. The gray areas in the right graph reflect the analyzed regions in Figure 3. Though small deviations in the vertical MP distribution are visible after each wetting and drying cycle, the bulk of the amended MP vertically stayed in place.

ously imaged with neutron and X-ray computed tomography (CT).

2.3 | Combined neutron and X-ray imaging

Neutron and X-ray imaging of cylindrical aluminum columns filled with mixtures of sand and MP was conducted at the ICON beamline at the Paul-Scherrer Institute (Kaestner et al., 2011). The objective was to simultaneously image the 3D distribution of MP via neutron CT imaging and the sand structure via X-ray CT imaging. Additionally, time-series neutron radiography was conducted during water infiltration to characterize the impact of MP on water flow. The exposure time of Neutron CT images was 3 s per projection angle. The exposure time of X-ray CT images was 2 s per projection angle. The total scanning time per sample was 31 min 20 s per sample (376 projections). One frame per projection angle was used

for the neutron CTs and the average of two frames per angle for X-ray CTs. The exposure time for the neutron radiographs was 0.5 s, resulting in the analysis of 12 s or 24 images of the captured the wetting process. Neutron radiographs and CTs were captured at a pixel size of 68 μm and X-ray CTs at 50 μm .

The combination of X-ray and neutron imaging provides fundamental advantages. Neutrons are sensitive to hydrogen-containing materials, while X-rays are sensitive to density. PET particles have a density comparable to water (ca. 1.38 g cm^{-3}) and are not easily detected with X-ray imaging techniques at the required spatial resolution. The combination of both imaging methods was shown to be optimal for visualizing and quantifying pore geometry and water distribution (A. Carminati et al., 2007; Kaestner & Schulz, 2015; Pleinert & Lehmann, 1997; Rogasik et al., 2003). Furthermore, not only water but also hydrogen-containing polymers like PET can be detected with neutrons, which allows for their quantification (Cramer et al., 2022, 2023; Tötze et al., 2021, 2024).

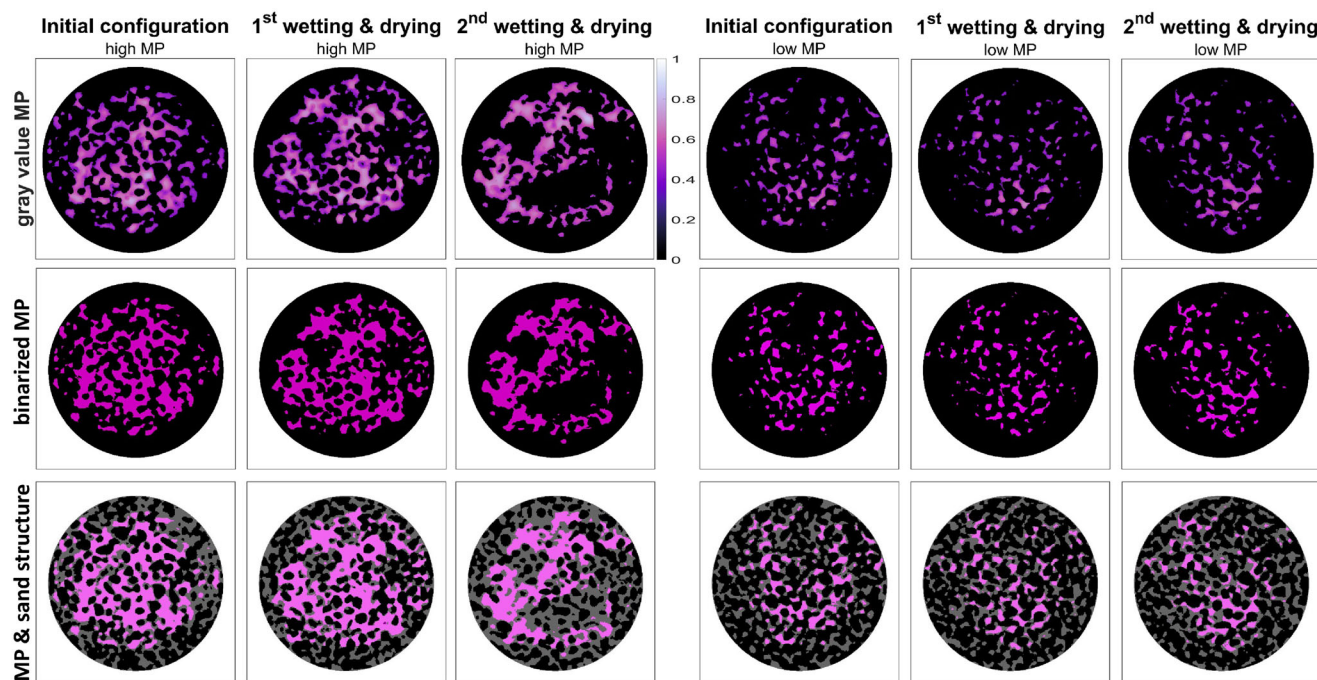


FIGURE 3 Averaged microplastic (MP) signal of slices within the top 17 mm exemplary depicted for the MP 2.10% variant. Images are separated by high (depth = 1.16–1.84 mm; left panels) and low MP content (depth = 2.52–3.20 mm; right panels), initial distribution of MP, and distribution of MP after the first and after second wetting and drying (see titles). The top row relates to the gray values caused by the presence of MP. The brighter the color, the higher the gray value, and accordingly, the more MP is present within a voxel (see color bar). The mid row represents the binarized MP gray values from the top row with magenta corresponding to the presence of MP and black showing no presence of MP within a voxel. The bottom row shows the binarized MP gray values from the mid row (magenta) overlaid onto the segmented sand (gray relates to the pore space and black to the sand grains). While the low MP range shows less deviations, cavities in the high MP range appear to be enlarged (i.e., MP is more clustered after wetting and drying).

The projections were reconstructed to volumes, normalized to the open beam signal, spot cleaned, noise reduced, and median filtered utilizing the software MuhRec, KipTool, and MATLAB (R2021a) (Boillat et al., 2018; Burger et al., 2006; C. Carminati et al., 2019; Kaestner, 2011; Kaestner & Schulz, 2015). The X-ray projections were reconstructed using a cone beam reconstruction algorithm due to the divergent X-ray beam. Volumes of the initial distribution of the MP 0.35% variant could not be reconstructed due to X-ray beam failure. The reconstructed volumes were rotated to align X-ray and neutron CTs, cropped to remove edge effects, and interpolated for the same image resolution (68 μm). The position referencing was conducted with guidance from reference copper beads attached to the outside of the sample cylinders.

To quantify the distribution of MP within the sand matrix, the respective volumes of sand were removed from dry neutron CT volumes (sand and MP). This allowed for the detection of MP distribution based on the remaining gray values of each voxel, later referred to as MP signal. Furthermore, the individual MP signal of each voxel is used to estimate the MP occupied volume of each horizontal slice volume, later referred to as MP volume fraction.

X-ray CT volumes were segmented by separating the sand grains and pore space using the stack histogram and applying the default thresholding mode of the software ImageJ (Version 1.54 g). The procedure was tested and validated against the porosity of the sand packing estimated via pycnometer experiments (mean porosity = 41.96%, $n = 3$). Water distribution in two dimensions was quantified by means of time-series neutron radiography by subtracting the respective signal of dry samples from wetted samples. (A. Carminati et al., 2007). The water signal was corrected according to the cylindrical shape of the sample container and the resultant varying depths in direction of the neutron beam.

Advancing wetting front depths were measured by binarizing the neutron time-series images (radiography) into wetted and dry zones (Shokri et al., 2008). The wetting front was defined as the interface between the water-saturated zone and the unsaturated zone (Shokri et al., 2008). The wetting front velocity of each time step corresponds to the vertical distance between the deepest pixel of the wetting front and the deepest pixel of the prior time step. A piece-wise linear regression was performed to identify breaking points in the infiltration behavior.

To characterize the shape and transformation of the MP phase with wetting and drying, a tensor-based morphometric analysis were performed, in which eigenvalues of the second-moment (covariance) tensor of the concentration distribution were used (see Table S1) (Boušek et al., 2025).

The shape analysis and statistical analysis was performed using MATLAB (R2021a). A non-parametric Kruskal–Wallis test was applied to explore potential significant differences, and the post hoc test of Bonferroni was employed to identify significant differences of treatments ($p < 0.01$). The capital letters depicted in the figures in the results section indicate significant differences where the same letter shows similarities between those groups.

3 | RESULTS

3.1 | MP distribution

The dry mixing procedure of sand and MP resulted in a heterogeneous distribution of MP over depth. Figure 1a and b show a three-dimensional reconstruction of the sample with the highest total MP content (2.10% w/w). Visible are the sand particles (in gray) and the regions of high MP content (vibrant colors). Due to the voxel size of 68 μm for neutron CTs, individual MP particles (20–75 μm) could not be resolved. However, the spatial distribution of MP was visible and quantifiable.

Figure 1c shows the MP signal (sum of gray values) as a function of the total amount of MP added to each sample variant. The MP signal of the samples was obtained from the neutron CTs after excluding the volume of sand particles retrieved from segmentation of X-ray CTs. Since the samples were dried prior to CT imaging, the remaining gray values relate to the MP content. Indeed, the MP signal correlated well with the independently measured gravimetric MP content. Errors as visible from this correlation are likely the result of noise in the neutron CTs and uncertainties in the segmentation of sand particles from X-ray CTs. Manifestations of these uncertainties can be seen in Figure 2 that depicts the MP profiles over depth.

Due to these uncertainties in segmentation, in Figure 2 the MP volume fraction of the for example, MP 1.05% variant of the second wetting and drying cycle is increased compared to the first cycle. The figure shows the volume fraction of MP in its initial distribution and after two wetting and drying cycles over the depth of 17 mm, the region of interest. The volume fraction describes the MP occupied volume of a total slice volume (slice thickness = 68 μm). Though small deviations in the MP distribution are visible after each wetting and drying cycle, the bulk of the MP remained vertically immobile, indicating that the transport of MP by unsaturated water flow is negligible in these conditions.

Analysis of MP distributions on the pore scale reveals a predominantly horizontal rearrangement of MP. Figure 3 shows the distribution of MP in two cross sections of the sample with highest total MP content (MP 2.10%), with one cross section chosen at the highest local MP content (depth: 2.2–2.9 mm) and the other cross section located deeper in the column corresponding to a lower local MP content (depth: 3.5–4.2 mm). The MP distribution partly changed during the wetting and drying cycles. This is particularly visible in the location of the highest MP content. In regions of locally high MP content, MP seems to cluster. MP distributions in the low local MP content region appeared more persistent.

The morphometric analysis of MP distributions in the first 17 mm of the samples shows a partly inconsistent picture across the variants (see Table S1). Generally, the MP 2.10% and MP 1.05% have similar and more spherical shapes compared to MP 0.35%, which is a prolate structure (elongated along the flow direction). Also, the change in shape by wetting and drying is different for the three MP concentrations. For the 0.35% variant (the initial shape is missing), the second wetting and drying acted as a refining force, increasing both anisotropy (A, +1.72%) and fractional anisotropy (FA, +2.79%) and enhancing linearity (L, +2.60%). The combination of increasing volume (V, +1.31%) and decreasing MP cluster count (−1.15%) is an indicator of slight structural coalescence constituting a more unified shape.

The MP phase of the MP 1.05% variant underwent the most drastic transformations. It follows a “collapse and expand” trajectory. The first wetting and drying stretches the MP-distribution plume, narrowing the cross section and causing a reduction in V (FA +24.37%, L +26.98%, and V −13.64%). The second wetting and drying causes a loss of this directionality and represents an isotropic expansion of the MP phase, becoming even more spherical and voluminous than its original state (S +19.13%, FA −26.53%, V +58.67%, and cluster number +22.13%).

The MP phase of the MP 2.10% variant follows a “stretch then fuse” evolution. It responds to the first wetting and drying by stretching and fragmentation (L +9.5%, FA +9.5%, cluster number +14.12%). The second wetting and drying acts as a stabilizer, inducing a consolidatory effect (cluster number −34.09%, V −1.7%), slightly reducing the internal fragmentation (FA −2.07%). The MP phase underwent a transition characterized by a minor initial elongation and fragmentation, followed by spatial reorientation and structural consolidation.

3.2 | MP influence on water infiltration

Figure 4 shows the percolating water during water infiltration imaged with neutron radiography. The temporal pattern is highly sensitive to the amount of MP.

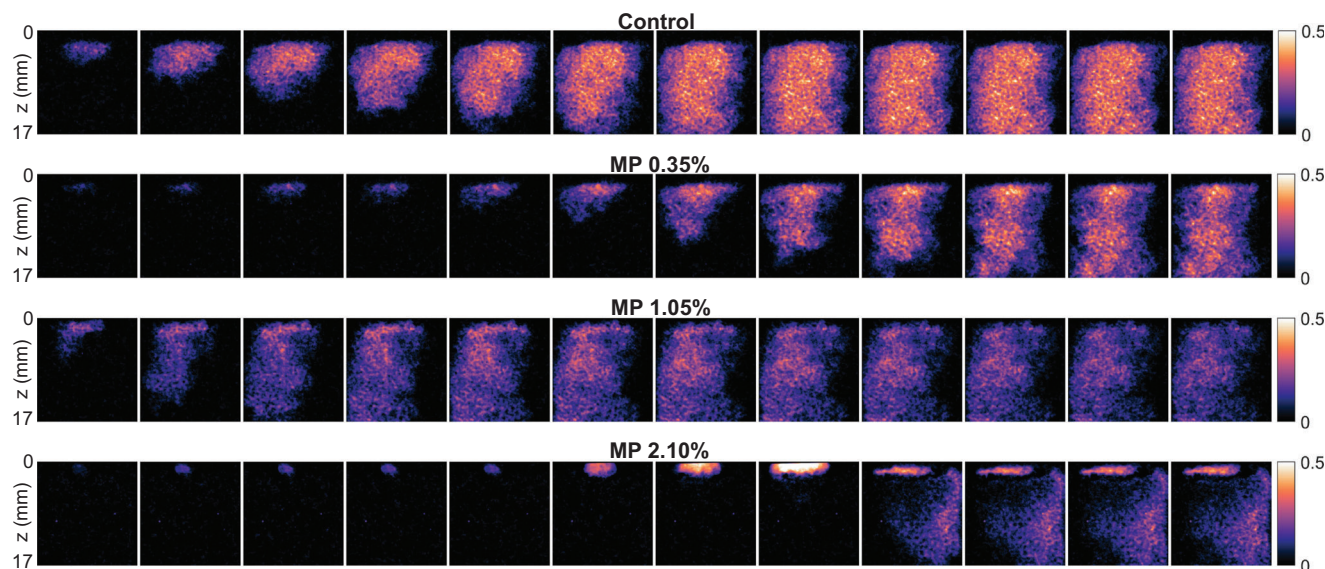


FIGURE 4 Time-series neutron radiographies of infiltrating water (gray values) during the first wetting in the top 17 mm. The brighter the color, the higher the gray value and the more water is present in a pixel (see color bar). From top to bottom: control sample, followed by the microplastic (MP) 0.35%, 1.05%, and 2.10% variants (see titles). From left to right, the time steps are in seconds of infiltration, with the last image representing the water status after 12 s of infiltration (see thick blue lines in Figure 5). Increasing MP contents results in less even infiltration, increased fingering, and reduced water signal above the wetting front, indicating a local MP-content-dependent infiltration of water. Note that the corresponding figure for the second wetting is shown in Figure S1.

In the control treatment, homogeneous water infiltration and even infiltration fronts were observed. More irregular patterns and fingering appeared with an increase in total MP content. The most pronounced irregularities were observed in the sample with 2.10% MP. Here, ponding water above the region with the highest MP content occurred. The establishment of a hydrostatic pressure of approx. 3 mm was necessary to overcome the hydrophobic patch of MP. Subsequently, water bypassed this region and rapidly infiltrated into deeper layers.

Figure 5 depicts water saturation derived from neutron radiographs during the first wetting. The individual figures also show the corresponding MP volume fraction. The thick blue lines refer to the water saturation after 12 s of infiltration that is, after 0.20 mL (0.79 mm) of applied water. At this time step, water had not reached the bottom of the samples. The thin gray lines depict water saturation over time at a resolution of 1 s, where the increase in gray level corresponds to the increase in time. In the control variant, a front with high water saturation moves downward. In the MP variants, the frontal region has a low water content, spreading quickly across the entire region of interest with water saturation increasing over time. The steeper the lines appear, the less even and more preferential infiltration took place covering only parts of the available pore space, resulting in reduced final water saturation (thick blue line) (see Figure 4; Table S2).

Similarly, Figure 6 (left) highlights that the average water saturation values in the wetted area were lower in samples with MP compared to those in the control sample. Here, MP restricts the volume available for the percolation of water (see Table S2 for porosity). All MP treatments resulted in significantly reduced water saturation compared to the corresponding wetting cycle of the control sample. There is a partly significant trend toward a decrease in water saturation with increasing MP content, especially visible during the first wetting (see Table S2).

Though being significantly lower than the control, during the second wetting, differences between the treatments are less clearly distinguishable and show larger similarities, arguably due to the establishment of preferential flow paths. Additionally, the second wetting partly resulted in increased water saturation values and a larger range in standard deviations (Table S2) when compared to the first wetting. This is especially pronounced in the control sample and thus cannot be solely associated with the redistribution and presence of MP. The redistribution of MP during the first wetting with mainly horizontal rearrangements and clustering of MP particles might have contributed to larger pore regions available for water flow (see Figure 3). Furthermore, minor adhesion water left from the first wetting cannot be excluded due to the accuracy of the used balance (two decimal places) and might have increased the overall wettability of the samples. Though considered to be minor, another influencing factor

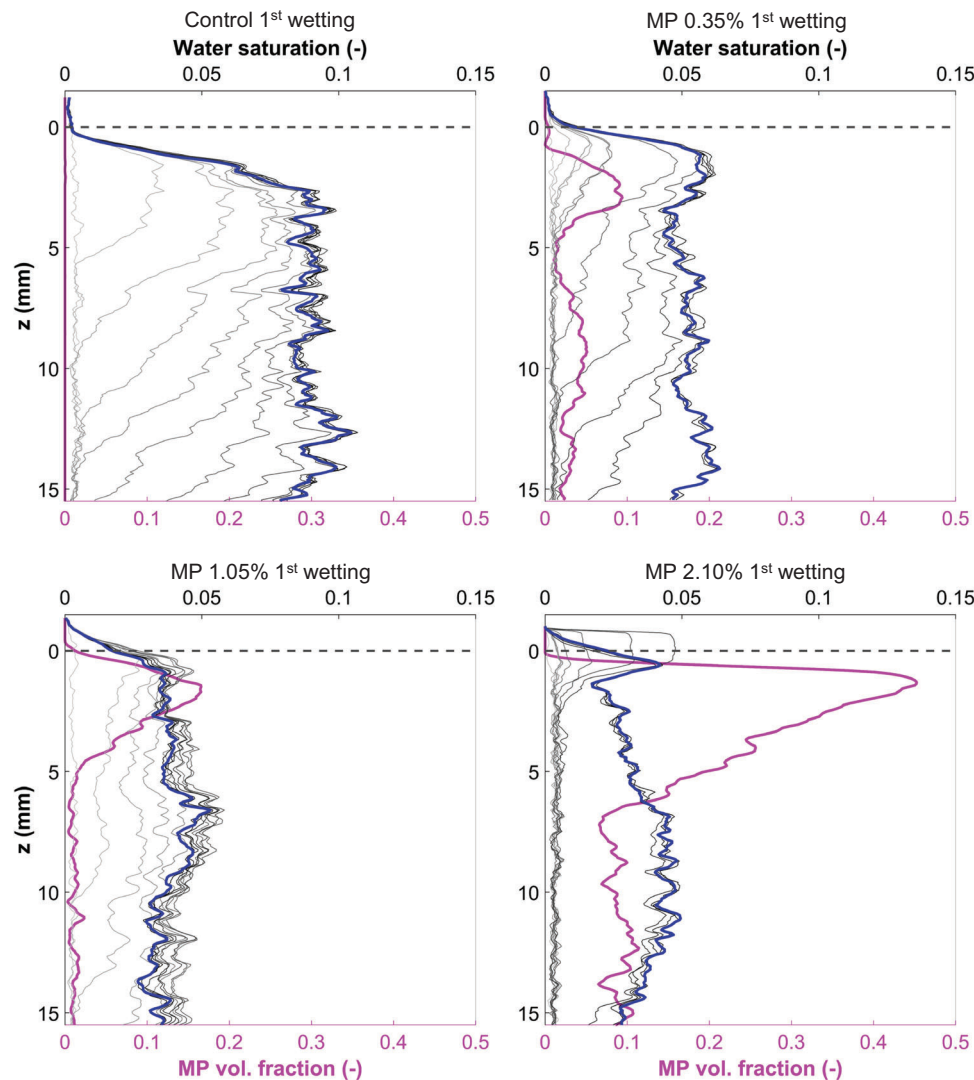


FIGURE 5 Profiles of water saturation (gray to black thin lines) in the first 17 mm during water infiltration and local microplastic (MP) volume fraction (magenta lines) before the first wetting (for second wetting, see Figure S2). The top of the sample cylinder is indicated by the dotted lines. Sample variants of control, MP 0.35%, 1.05%, and 2.10% are depicted (see titles). The thick blue line in each plot represents water saturation after 12 s of infiltration at 1 mL/min. The thin gray to black lines depict the water saturation at different times where the increase in gray level corresponds to the increase in time (resolution = 1 s). Water infiltration is increasingly impeded with increasing MP content, leading to ponding of the water above high local MP contents. Water saturation profiles after 12 s of all MP treatments are significantly lower than the respective control treatment (see Figure 6).

would be a decrease in contact angle of the sand by the first wetting.

The sharp increase in slope in the rapid phase further illustrates MP as cause for a two-stage infiltration pattern: (1) MP-occupied pores block water flow, followed by (2) re-direction of delayed water leading to rapid and preferential infiltration. In the case of lower local MP content and according to the concept of Wan and Wilson (1994), MP particles might be displaced by fluctuations of the AWI. But the acquired spatial image resolution is not sufficient to resolve this mechanism of MP translocation. The presence of MP influenced water saturation by restricting the pore space for water flow. This is illustrated by lower mean water satura-

tion values in the wetted regions after 12 s of infiltration. At very high MP contents, infiltration is prevented until a positive pressure is built (Figures 4 and 5). In the case of the MP 2.10% variant, ponding is indicated by a high water saturation near the surface.

4 | DISCUSSION

The experiments confirmed the hypothesis that the presence of hydrophobic MP within the pore space can restrict unsaturated water infiltration, resulting in delayed infiltration followed by rapid wetting front advancement and preferen-

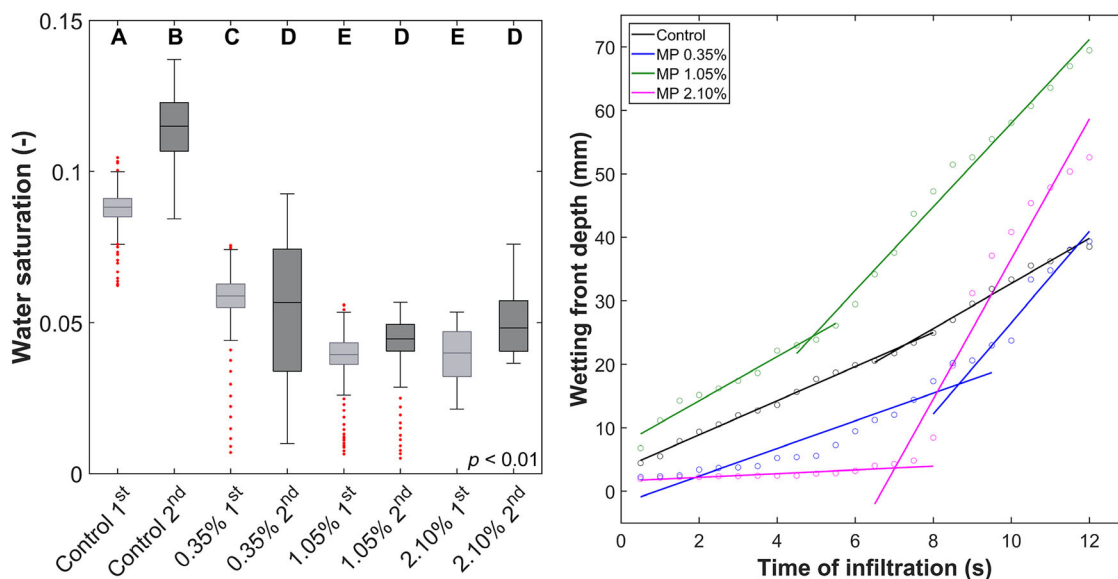


FIGURE 6 Right: The depth of wetting fronts over time during the first wetting (see Figure S3 for the second wetting). The infiltration in the control shows a relatively constant velocity of the wetting front. In contrast, the microplastic (MP) samples show a different dynamic, with an initial slower front movement followed by a faster front rate once the flow is initiated. A piecewise linear regression (see Table S3) was conducted to identify breaking points in this infiltration behavior, and two stages of infiltration in the MP amended samples can be separated: an initial delay stage, followed by a rapid stage where water quickly reaches greater depths. While the delay stage shows a gradual decrease in the slopes with an increase in MP volume fraction, there is a sharp increase in slopes of the rapid stage. This can be explained by water at first not being able to flow through the pore space, which is blocked by high local MP contents during the delay stage, followed by the breakthrough of water. Breakthrough of water happens by either (1) bypassing if enough wettable flow paths are available, (2) creating a positive pressure by ponding to overcome the water-entry value of pores with MP, or (3) redistribution of the MP to create wettable flow paths. This redistribution of MP can be caused by the buildup of hydrostatic pressures above the hydrophobic patches until local MP is pushed aside to lower the water-entry value (compare Figures 3 and 4). Left: Boxplots showing the range in water saturation above the wetting front after 12 s of infiltration (see Figure 5, thick blue lines) within the top 17 mm. From left to right, the sample variants of control, MP 0.35%, 1.05%, and 2.10% at their corresponding wetting cycles are depicted. Significant differences in water saturation are indicated by capital letters at the top of the plots ($p < 0.01$). All MP treatments resulted in significantly reduced water saturation compared to the corresponding wetting cycle of the control sample. During the first wetting, there is a partly significant trend toward a decrease in water saturation with an increase in total MP content. During the second wetting, differences in the MP treatments are less clearly distinguishable. Right: Wetting front depth over time of infiltration during the first wetting (see Figure S3 for the second wetting). A piecewise linear regression (lines) was conducted to identify breaking points in the infiltration behavior. Single time steps are indicated by circles. While the infiltration in the control shows a relatively constant wetting front velocity during the first wetting, two stages of infiltration in the MP-amended samples can be clearly separated, with an initial “delay” followed by a “rapid” stage.

tial flow. During infiltration, water bypasses regions with high local MP content (i.e., low wettability), leading to irregular wetting fronts and reduced water saturation behind the wetting front. The magnitude of these effects increases with an increase in local MP content. The sample variant with the highest total MP content investigated (MP 2.10%) and the resulting MP volume fractions in this treatment represent an extreme case in terms of MP-occupied pore space. Many soils will never reach a contamination level to this degree. Though, when considering the common deposition pathways of MP to soil (i.e., degrading plastic litter and aerial deposition), heterogeneously distributed and high local MP contents can be expected. As we hypothetically increase the scale of this experiment and replace the relatively homogeneous sand used in this study with a more heterogeneous soil, the distribution

of MP, and the resulting local water flow patterns, would still be heterogeneous.

Further, we observed no significant vertical movement of MP within the resolution, boundary conditions, and duration of the experiment. Pore regions with high MP content are less likely to be crossed by water due to their water repellency. We argue that low local wettability prevents effective MP transport. Nevertheless, the shape of the MP phase within the pore space is not static but instead undergoes transformations during the wetting and drying. The shape analysis shows that the first wetting and drying caused a minor stretching of the MP phase. The second wetting and drying caused the MP 2.10% variant to consolidate, while the other variants experienced an expansion. Arguably, the largely occupied pore space in the MP 2.10% variant caused consolidations, whereas

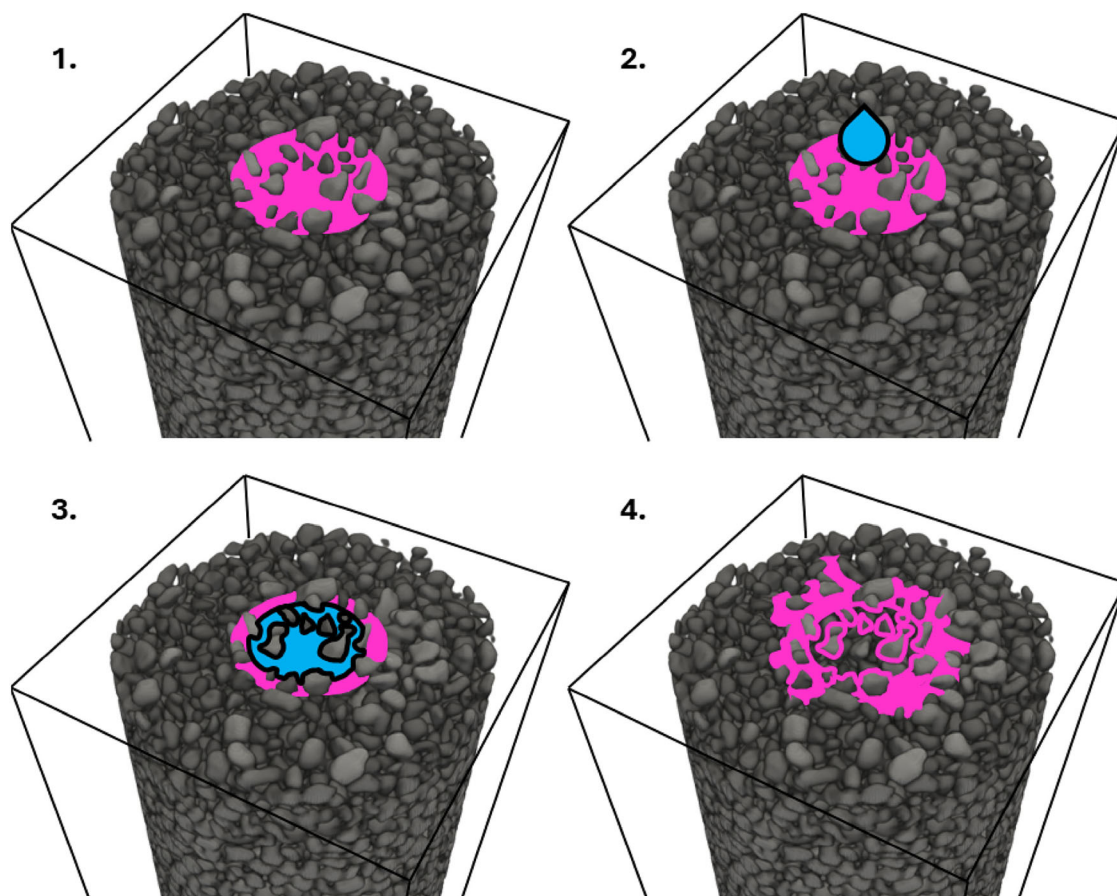


FIGURE 7 Conceptual illustration of hypothetical pore-scale mechanisms on microplastic (MP) mobility in porous media. (1) Hydrophobic MP (magenta) is deposited in the pore space (gray grains). (2) Water (blue droplet) arrives at the surface. The black border of the droplet represents the air-water interface (AWI). (3) The MP is collected at the AWI. As the AWI advances, the accumulating MP increasingly restricts the infiltration of water. Water and the MP-occupied AWI continue to spread until pores are rendered hydrophobic by the accumulated MP. (4) Water diverts into more wettable pores and pushes low-content MP outward. Water, driven by gravity and capillary forces, proceeds to infiltrate deeper and deposits the AWI-collected MP in pore throats (image adopted from Cramer [2024]).

in the lower MP variants, sufficient pore space is available for migration.

These findings are comparable to the results by Wan and Wilson (1994), who analyzed different saturation conditions in relation to the transport of hydrophobic and hydrophilic colloidal particles through porous media. In their study, they show that the retention of both kinds of particles is elevated with increased air content in the sample. Particles generally sorb onto the AWI, with the degree of sorption increasing for hydrophobic particles, resulting in a strong retention. It is suggested that the preferential sorption to the AWI is an important retention mechanism, especially with a stationary liquid and gas phase in the sample. This mechanism could also be applied to this study in relation to the observation that MP is not translocated vertically, although the applied wetting and drying cycles were expected to result in fluctuations of the AWIs. Possibly, such fluctuations resulted in a small scale (i.e., pore scale) redistribution to MP. Such pore-scale

redistributions of MP were partly visible during wetting and drying cycles, resulting in the clustering of particles at high MP content.

Water as well as MP strive to minimize their contact area with each other due to hydrophobic interactions. This might result in dynamic effects, formation of preferential flow path, and local accumulation of MP. Pore regions with high MP content, where MP might aggregate, are bypassed by water. Although in regions with low MP content the AWI might mobilize MP particles and push them aside, creating wettable pores and facilitating preferential flow (see Figure 7). Additional experiments conducted with varying infiltration rates would be required to test this concept.

Considering MP in natural soil environments, it is evident that its surface properties change over time. MP surfaces can become more functionalized and degraded, and with this, more wettable. This would in turn reduce hydrophobic interactions, solely leaving the occupation of available

pore space as an influencing factor on infiltration and would possibly facilitate the transport of MP with advection. However, the temporal dynamics of overcoming MPs inherent hydrophobicity in natural soil environments remain unclear and are most likely related to its wettability since degradation and functionalization processes in soils largely require the presence of water. Given these findings, we suggest that future research should focus on the temporal dynamics of surface combined wetting kinetics of MP as a factor to predict the transport of environmentally altered MP with advection.

AUTHOR CONTRIBUTIONS

A. Cramer: Conceptualization; data curation; formal analysis; investigation; methodology; software; validation; visualization; writing—original draft; writing—review and editing.

P. Benard: Conceptualization; investigation; methodology; software; supervision; validation; writing—review and editing.

A. Kaestner: Conceptualization; data curation; investigation; methodology; resources; software; supervision; validation; writing—review and editing.

M. Zarebanadkouki: Conceptualization; investigation; resources; supervision; validation; writing—review and editing.

P. Lehmann: Formal analysis; investigation; methodology; resources; supervision; validation; writing—review and editing.

A. Carminati: Conceptualization; formal analysis; funding acquisition; investigation; methodology; project administration; resources; supervision; validation; writing—review and editing.

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CONFLICT OF INTEREST STATEMENT

The authors declare no conflicts of interest.

ORCID

A. Cramer  <https://orcid.org/0000-0002-3853-7714>

P. Benard  <https://orcid.org/0000-0002-0922-5527>

A. Kaestner  <https://orcid.org/0000-0003-4054-4726>

M. Zarebanadkouki  <https://orcid.org/0000-0001-6342-5792>

P. Lehmann  <https://orcid.org/0000-0001-6315-7441>

A. Carminati  <https://orcid.org/0000-0001-7415-0480>

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