

TECHNICAL REPORT

Emerging Contaminants

Fate and transport of fragmented and spherical microplastics in saturated gravel and quartz sand

Ahmad Ameen^{1,2}  | Margaret E. Stevenson^{1,2}  | Alexander K. T. Kirschner^{2,3,4}  |
Stefan Jakwerth^{2,3}  | Julia Derx^{1,2}  | Alfred P. Blaschke^{1,2} 

¹Institute of Hydraulic Engineering and Water Resources Management E222/2, TU Wien, Vienna, Austria

²Interuniversity Cooperation Centre (ICC) Water and Health, Vienna, Austria

³Institute for Hygiene and Applied Immunology, Water Microbiology, Medical University of Vienna, Vienna, Austria

⁴Division Water Quality & Health, Karl Landsteiner University of Health Sciences, Krems an der Donau, Austria

Correspondence

Margaret E. Stevenson, Institute of Hydraulic Engineering and Water Resources Management E222/2, TU Wien, Karlsplatz 13, A-1040 Vienna, Austria.
Email: stevenson@waterresources.at

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Abstract

Microplastics in urban runoff undergo rapid fragmentation and accumulate in the soil, potentially endangering shallow groundwater. To improve the understanding of microplastic transport in groundwater, column experiments were performed to compare the transport behavior of fragmented microplastics (FMPs ~1- μm diameter) and spherical microplastics (SMPs ~1-, 10-, and 20- μm diameter) in natural gravel (medium and fine) and quartz sand (coarse and medium). Polystyrene microspheres were physically abraded with glass beads to mimic the rapid fragmentation process. The experiments were conducted at a constant flow rate of 1.50 m day⁻¹ by injecting two pore volumes of SMPs and FMPs. Key findings indicate that SMPs showed higher breakthrough, compared to FMPs in natural gravel, possibly due to size exclusion of the larger SMPs. Interestingly, FMPs exhibited higher breakthrough in quartz sand, likely due to tumbling and their tendency to align with flow paths, while both sizes (larger and smaller relative to FMPs) of SMPs exhibited higher removal in quartz sand. Therefore, an effect due to shape and size was observed.

Plain Language Summary

Microplastic pollution has emerged as a global concern, particularly in the terrestrial environment, where they undergo rapid fragmentation due to abrasion with debris. The effect of microplastic shape and size on transport in saturated porous media was investigated using lab-scale column experiments. Different-sized fragmented and spherical microplastics were tested in natural gravel and quartz sand. Findings revealed that fragmented microplastics of similar size displayed a higher breakthrough compared to spherical microplastics in saturated quartz sand. In contrast, larger spherical microplastics exhibited greater mobility than smaller fragmented microplastics in saturated gravel material. Under natural conditions, microplastics continuously break down into fragmented and irregular-shaped particles,

Abbreviations: CFT, colloid filtration theory; FMP, fragmented microplastic; SMP, spherical microplastic; SPC, solid-phase cytometer.

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increasing the risk of entering deeper soils and being transported in groundwater. This study provides information about this mobility and its potential environmental consequences.

1 | INTRODUCTION

Plastics are undoubtedly a marvel of the modern era, which have benefited society across many sectors and outpaced almost any other material with a global annual production of 368 million metric tonnes (da Costa et al., 2020). Improper handling and disposal of plastics in the environment lead to various degradation processes, including mechanical abrasion, in situ weathering, ultraviolet-induced degradation, photo-degradation, bio-degradation, oxidative degradation, and hydrolysis (Andrady, 2011; Cai et al., 2018; Fournier et al., 2021; Mendoza & Balcer, 2019; Ren et al., 2020; Song et al., 2017), ultimately resulting in the formation of microplastic particles smaller than 5 mm (Thompson et al., 2004). The European Commission defines microplastics as ranging in size from 0.1 to 5 mm and can be found in various forms such as filaments, fragments, spheres, and foams (Mendoza & Balcer, 2019). Microplastics are also released directly into the environment due to anthropogenic activities (Rochman, 2018); the main sources being cosmetic products (Habib et al., 2020), wastewater effluent and sludge (Edo et al., 2020; Kosuth et al., 2018), apparel and home textiles (Belzagui et al., 2019; Henry et al., 2019), abrasion of vehicle tires (Knight et al., 2020; Luo et al., 2021) and paint fragments generated from boats and ships, road markings, and buildings (Gaylarde et al., 2021; Turner, 2021). Over the past few years, there has been increasing concern regarding the widespread presence and distribution of microplastics (Kershaw & Rochman, 2015; Marsden et al., 2019) across various environments, including marine habitats, rivers, surface runoff, lakes, groundwater, sediments, soil, glaciers, and in such isolated places as Arctic and Antarctic regions (Aves et al., 2022; Cha et al., 2023; Chia et al., 2021; Koutnik et al., 2021; Miranda et al., 2020; Wu et al., 2019).

Microplastics, often referred to as colloidal contaminants, can quickly migrate in the environment, posing ecological and health risks (Flury & Aramrak, 2017; Liu et al., 2021; Molnar et al., 2015; Senathirajah et al., 2021). Due to their small size and altered surface properties, they can act as carriers for contaminants (Cortés-Arriagada, 2021; Liu et al., 2019). As microplastics age, their physical and chemical properties can change, making their fate more unpredictable (Luo et al., 2020). Physical fragmentation, the breaking down of microplastics into smaller pieces, is an important aging mechanism that is often overlooked. Microplastics that accumulate in urban stormwater runoff experience physical aging

due to mechanical abrasion, as stormwater runoff contains a mixture of precipitation, suspended sediment, natural and anthropogenic debris, and chemical pollutants (Gilbreath & McKee, 2015; McKee & Gilbreath, 2015; Werbowski et al., 2021).

Microplastics that accumulate from surface and urban stormwater runoff have the potential to move downward into the subsurface through infiltration enhanced by rainfall and natural freeze-thaw cycles (Koutnik et al., 2022a; Koutnik et al., 2022b; Li et al., 2023). Although groundwater is a critical resource for many regions of the world, the presence of microplastics in groundwater has been neglected as a potential risk due to the assumption that soil serves as an effective barrier against microplastic contamination (Alimi et al., 2018; Goeppert & Goldscheider, 2021; Panno et al., 2019; Re, 2019; Samandra et al., 2022). On the contrary, O'Connor et al. (2019) argued that soil is not only a microplastic sink but a viable route to deeper soils and groundwater. Goeppert and Goldscheider (2021) have experimentally demonstrated the transport of microplastics up to a distance of 200 m (in the groundwater flow direction) in an alluvial aquifer. Samandra et al. (2022) also identified the presence of eight most commonly found microplastics, ranging in size from 18 to 491 μm , within an unconfined alluvial aquifer in Australia. Mintenig et al. (2019) analyzed groundwater and drinking water samples for the presence of microplastics (50–150 μm) and found polyethylene, polyamide, polyester, polyvinylchloride, and epoxy resin. Susceptible to pollution from surface runoff due to fracture networks and preferential flow, karst groundwater systems are especially at risk (Panno et al., 2019).

Several factors, such as microplastic shape and size, porous media properties, and groundwater chemistry, influence the transport of microplastics in the subsurface environment (Hou et al., 2020). Koutnik et al. (2021) found that only 20% of global research on microplastics considers particles smaller than 20 μm , which are more likely to be transported to groundwater. Nonetheless, the potential impact of these smaller particles (diameter < 20 μm) is likely underestimated. A limited number of studies have investigated microplastics at the micron level even though most fragmented particles found in soil environments are micron sized (Hou et al., 2020). It is generally agreed upon that larger microplastics have less mobility in saturated porous environments, as they are likely to get filtered out by the pore structure in aquifer material (Dong et al., 2018). The most recent research demonstrates that smaller microplastics have increased vertical transport

in porous media, and also the morphology of microplastics can play a critical role in attachment and detachment within the subsurface environment (Dong et al., 2021; Dong et al., 2022; Hou et al., 2020; O'Connor et al., 2019; Rillig et al., 2017; Waldschläger & Schüttrumpf, 2020). The aquifer material characteristics also play a significant role in the transport of microplastics. Coarse-grained porous media promote the transport of microplastics due to their larger pores and preferential flow paths (Hou et al., 2020). Recently, the focus has been directed toward studying the transport of microplastics in groundwater, but limited to investigating uniform-shaped microspheres, while in nature, most microplastics found are fragmented and irregular in shape (He et al., 2020; He et al., 2019; Hitchcock, 2020; Piñon-Colin et al., 2020). Despite this, the impact of various parameters (e.g., fragmentation, size, and shape) on the transport of microplastics in different aquifer materials has not yet been extensively studied.

The present study aims to understand and investigate the transport behavior of spherical microplastics (SMPs) and fragmented microplastics (FMPs) in two different sizes of gravel and quartz sand. Polystyrene microspheres were chosen to replicate the impact of fragmentation on microplastics within an urban environment. The reason for using polystyrene is due to its abundance in the natural environment and high resistance to weathering and heat (Alimi et al., 2018). These microspheres have a density ranging from 1.04 to 1.11 g cm⁻³ and can exist as submerged or floating in the aquatic environment (Duis & Coors, 2016). The present research focused on the rapid physical breakdown of 10 and 20 µm microplastics through mechanical abrasion, resulting in a random size distribution (average diameter ~1 µm). In gravel, 20 µm SMPs are compared to 1.60 and 1.80 µm FMPs, whereas in quartz sand 10 µm SMPs are compared to 1.20 and 1.40 µm FMPs. Also, 1.40 µm FMPs were compared to their corresponding similar-sized SMPs (1 µm) in coarse quartz sand for a better understanding of the role of shape and size during subsurface transport.

2 | MATERIALS AND METHODS

2.1 | Preparation of microplastics

Commercially available plain polystyrene yellow-green fluorescent microspheres were used in the column transport experiments: 1, 10, and 20 µm (Fluoresbrite YG Microspheres, Polysciences Inc.). The major benefit of using these microspheres for laboratory experiments is the ease with which they are detected by solid-phase cytometry and epifluorescence applications due to their strong fluorescent intensity and emission/excitation spectra (Stevenson et al., 2014). To mimic irregular-shaped microplastics present in the terrestrial environment, 10 and 20 µm polystyrene microspheres

Core Ideas

- Shape and size influence microplastic transport in saturated porous media.
- Fragmented microplastics exhibit higher breakthrough than spherical, of the same size, in saturated quartz sand.
- Larger spherical microplastics are more mobile than smaller fragmented microplastics in saturated gravel material.

were physically fragmented through a mechanical abrasion process, based on the method developed by Ranhand (1974). To achieve rapid and reproducible fragmentation, 1 mL of the microsphere stock solution was mixed with 1 g of glass beads (1-mm diameter) in a FastPrep-24 Classic homogenizer (MP Biomedicals). To ensure consistent fragmented particle size, a controlled constant mixing speed (impact velocity) of 20 revolutions per minute was applied to provide consistent impact force between the microspheres and the glass beads. The abrasion process was conducted for a duration of 10–15 min. This time duration was chosen based on trials to achieve the target FMP size range of approximately 1 µm. The abrasion time was varied depending on the microsphere size: 10 min for 10 µm and 15 min for 20 µm microspheres. The mixing time for 20 µm microspheres was extended due to the Zetasizer measurement limit of 10 µm. The fragmented particles were extracted from the solution, using a pipette, after the heavier glass beads were allowed to settle. For the characterization of microplastics, the zeta potential and mean diameter (Table S1) were measured with a Zetasizer P ZSU3200 (Malvern). Each measurement was repeated using at least three different samples.

2.2 | Properties of influent water

The influent water used for the column experiments was standard Viennese tap water, sourced from an Alpine karstic spring. In this study, tap water offered several advantages over local groundwater, exhibiting lower electrical conductivity, reduced levels of iron and sulfate, and being free from emerging contaminants (Oudega et al., 2021). Also, tap water has a consistent quality (Table S2) and is readily available in large volumes, making it an ideal choice for maintaining controlled conditions during experiments. Furthermore, the use of tap water provided a more realistic simulation of microplastic fate in the natural environment, enabling accurate determination of critical transport parameters such as attachment, detachment, and dispersion coefficients within different porous media.

Throughout the column experiments, from injection to effluent collection, the pH, temperature, and electrical conductivity remained stable. Zhao et al. (2024) found that many water quality parameters are poorly correlated with the microplastics in an urban drinking water source. Also, studies in freshwater ecosystems suggest total dissolved solids and electrical conductivity have a minimal influence on microplastic aggregation. While pH, temperature, total suspended solids, and dissolved oxygen have a significant impact (Buwono et al., 2021; Khoironi et al., 2023; Rakib et al., 2023; Zhao et al., 2024).

2.3 | Tracer preparation and analysis

A solution of sodium bromide (1 mM L^{-1} , 102.89 mg L^{-1}) prepared in tap water served as a conservative tracer to investigate the transport properties of the packed columns. The bromide concentration and electrical conductivity (EC) of the injected tracer were measured by a flow-through cell (WTW TetraCon 325) and a hand-held portable EC meter (WTW ProfiLine Cond 3310, Xylem Analytics Weilheim).

2.4 | Porous media

Four different soil materials were used as porous media to study the transport behavior of microplastics. Natural aquifer material was extracted from a borehole depth of 5–6 m below the ground surface at a site southeast of Vienna, Austria, and the portions held back on the 8 and 4 mm sieves were used for the “medium gravel” and “fine gravel” columns, respectively (both with a bulk density of 1.52 g cm^{-3}). The median grain diameters (d_{50}) values of 4 and 8 mm for medium and fine gravel, respectively. Similarly, two types of quartz sand (grain size: 0.6–1.3 mm and 0.4–0.8 mm, bulk density: 2.65 g cm^{-3}) were purchased for the “coarse quartz” and “medium quartz” sand column experiments (Carl Roth GmbH + Co. KG). The median grain diameters (d_{50}) of 0.95 and 0.60 mm for coarse and medium quartz, respectively. Based on the standard gravimetric method, the porosity of the porous media was calculated by measuring the volume of water needed to saturate the dry soil used to pack the column, divided by the total column volume (Table S1).

2.5 | Column experiments

Transport experiments were conducted under saturated flow conditions using Plexiglas columns (70 mm inner diameter and 300 mm length). For quartz sand experiments, the column diameters met the minimum d_{col}/d_{50} ratio (the ratio of

the column diameter to the media effective particle diameter) that was much higher than the recommended value of 50 to ensure minimal potential wall effects in the column (Knappe et al., 2008). Although the d_{col}/d_{50} ratio in the gravel experiments was below 50, the symmetrical shapes of both the tracer and microplastic breakthrough curves resemble those observed with quartz sand, suggesting negligible wall effects (Figure S1). Stainless steel-mesh screens (Spectrum Labs) were placed on both ends of the column to prevent the loss of porous media and the caps were threaded to tighten against the outside of the column. A rubber O-ring was embedded in the column end to create a tight seal. All tubings were made of Teflon. To ensure homogeneous packing and minimize air entrapment, the column was filled with the porous media in 2 cm increments while gently stirring with a small steel rod. This careful filling procedure ensured uniform flow throughout the column, as evidenced by the good recovery rates of the tracer (NaBr) and the nearly identical, symmetrical breakthrough curves with minimal tailing (Figure S1), as expected for a homogeneously packed soil column (Sobotkova & Snehota, 2014). The pore volume (PV) for the influent solution was calculated by the product of porosity and the volume of the column. Before the experiment, the columns were flushed for a minimum of 20 PV with tap water. The influent microplastics concentration (Table 1) was prepared by mixing 1 mL of microplastics stock solution (spherical or fragmented) and two PVs of tap water. To preclude the possibility of ripening or blocking, relatively low microplastic concentrations (approx. 10^4 particles mL^{-1}) were introduced in the influent suspension solution, which was pumped in an upward direction at a constant flow rate of 4 mL min^{-1} (Darcy velocity = 1.5 m day^{-1}). Afterward, the columns were flushed with 5 PV of tap water at the same flow rate. A magnetic stirrer was used to stir the suspension constantly during the experiments to ensure that microplastics were evenly dispersed in the influent. A fraction collector (CF2 Fraction Collector, Spectrum Chromatography) automatically collected column effluent sample in a 15-mL test tube every 5 min. All column tests were done with at least two replicates (each column freshly packed).

2.6 | Microplastic enumeration

The enumeration of FMPs and SMPs in the effluent solution was carried out by solid-phase cytometry (SPC) using the ChemScan RDI (bioMérieux). The SPC system has been used in the past mainly for the detection of colloids in environmental samples (Baudart et al., 2002; Mignion-Godefroy et al., 1997; Schauer et al., 2012). Stevenson et al. (2014) used this simple and efficient method for enumerating microspheres in natural water samples. Details on the SPC

TABLE 1 Summary of experimental and modeling results using HYDRUS-1D in porous media: Mean microplastic diameter (d_{MP}), dispersivity (D), initial influent concentration (C_0), maximum effluent concentration (C_{max}), attachment rate coefficient (k_{att}), detachment rate (k_{det}), coefficient, and coefficient of determination (R^2).

Porous media	Column test	Type	d_{MP} (μm)	D (cm)	Flow Rate (cm min^{-1})	C_0 (particles mL^{-1})	C_{max} (particles mL^{-1})	C_{max}/C_0	k_{att} (min^{-1})	k_{det} (min^{-1})	R^2
Medium gravel (8 mm)	A1	FMPs	1.60	0.8	0.110	1.65×10^4	1.26×10^4	0.76	0.0028	0.00015	0.99
	A2	FMPs	1.60	0.8	0.100	1.68×10^4	1.25×10^4	0.74	0.0028	0.00015	0.96
	B1	SMPs	20	0.8	0.110	6.45×10^3	5.74×10^3	0.88	0.0012	0.00015	0.99
	B2	SMPs	20	0.8	0.110	6.45×10^3	5.79×10^3	0.90	0.0012	0.00015	0.98
Fine gravel (4 mm)	C1	FMPs	1.80	0.8	0.110	5.75×10^4	3.40×10^4	0.59	0.0060	0.00015	0.98
	C2	FMPs	1.80	0.8	0.110	5.79×10^4	3.60×10^4	0.62	0.0060	0.00015	0.95
	D1	SMPs	20	0.8	0.100	6.76×10^3	5.00×10^3	0.74	0.0032	0.00015	0.98
	D2	SMPs	20	0.8	0.110	6.76×10^3	4.88×10^3	0.72	0.0035	0.00015	0.97
Coarse quartz sand (0.6–1.3 mm)	E1	FMPs	1.40	0.4	0.084	2.32×10^4	7.25×10^3	0.31	0.0090	0.00002	0.98
	E2	FMPs	1.40	0.4	0.084	2.32×10^4	7.05×10^3	0.30	0.0097	0.00002	0.97
	F1	SMPs	10	0.4	0.086	4.84×10^4	5.77×10^3	0.12	0.0182	0.00002	0.94
	F2	SMPs	10	0.4	0.091	4.84×10^4	6.78×10^3	0.14	0.0178	0.00002	0.97
	G1	SMPs	1	0.4	0.091	2.55×10^4	6.27×10^3	0.24	0.0120	0.00002	0.98
	G2	SMPs	1	0.4	0.096	2.55×10^4	5.62×10^3	0.22	0.0145	0.00002	0.94
Medium quartz sand (0.4–0.8 mm)	H1	FMPs	1.20	0.4	0.088	2.30×10^4	4.30×10^3	0.19	0.0136	0.00002	0.98
	H2	FMPs	1.20	0.4	0.090	2.30×10^4	5.25×10^3	0.23	0.0126	0.00002	0.97
	I1	SMPs	10	0.4	0.085	4.64×10^4	2.18×10^3	0.04	0.0245	0.00002	0.92
	I2	SMPs	10	0.4	0.094	4.64×10^4	2.38×10^3	0.05	0.0265	0.00002	0.94

Abbreviations: FMPs, fragmented microplastics; SMPs, spherical microplastics.

system and the enumeration process are described by Mignon-Godefroy et al. (1997). The following section provides a comprehensive explanation of the microplastic enumeration procedure.

- a. *Filtration*: 1 mL of effluent solution was filtered onto a 25 mm black polyester 0.4 μm pore size filters (AES Chemunex, bioMérieux) using a Multifold Vacuum Filtration Device (Pall). The black polyester filter was then carefully placed on a support pad (AES Chemunex, bioMérieux), which was already saturated with 100 μL of phosphate-buffered saline to hold the filter in place.
- b. *Automated enumeration with SPC*: The filter was placed on the ChemScan RDI sample holder, and the scan was initiated. An argon laser (488 nm emission wavelength) scanned the entire 25 mm filter area in 3–5 min. Microplastic particles (referred to as fluorescent events) were distinguished from other background interferences based on their fluorescence intensity (500–530 nm wavelength with the green channel) and then counted. Following a complete scan, the system generates raw data and result maps based on discriminant settings, highlighting fluorescent events. These maps mark the exact location of each microplastic on the filter. Subsequently, these microplastics were visually examined and confirmed using a microscope equipped with a motorized stage. The most important discriminant parameters are the number of “lines” and the number of “samples” for the shape and the peak intensity of the signal. The ratio of the fluorescent light detected in the secondary channel (fluorescence from microplastics) to the fluorescent light detected in the primary channel (fluorescence from the probe) is another important discriminant parameter and is referred to as the S/P ratio. The discriminant settings were set as follows: number of “lines,” 1–50; number of “samples,” 1–250; peak intensity, 250–65536; and S/P ratio, 0–0.898 (Table S3).
- c. *Validation of counted microplastics*: The verification of microplastics was achieved through a visual inspection by a Nikon Eclipse 80i epifluorescence microscope directly connected to the ChemScan RDI system. The microscope employs a 100x magnification objective, resulting in a final magnification of 1000x for detailed analysis of each fluorescent particle identified by the laser scan. The microscope is equipped with an automatic stage that can be driven by the user such that the whole filtration area can be scanned. Up to 250 events were validated per filter, with a ratio being used if above, and all events were validated when enumeration results were less than 250. For each effluent sample, a minimum of three replicates were analyzed.

3 | THEORETICAL CONSIDERATIONS

3.1 | Filtration efficiency

The transport and deposition behavior of the microplastics (SMPs and FMPs) in the saturated porous media was quantified using colloid filtration theory (CFT), which calculates the attachment (collision) efficiency (α). The attachment efficiency represents the fraction of collisions (contacts) between suspended microplastic particles and collector sand grains that result in successful attachment (Elimelech & O’Melia, 1990). It is common to use column experiments to determine the attachment efficiency for given physicochemical conditions. The collision efficiency was computed using the following equation by Tufenkji and Elimelech (2004):

$$\alpha = -\frac{2}{3} \frac{d}{(1-\theta)x\eta_0} \ln \frac{C}{C_0} \quad (1)$$

where d is the mean soil grain size (mm), θ is the porosity, x is the column length (cm), C/C_0 is the normalized steady-state breakthrough microplastic concentration (particles mL^{-1}), and η_0 is the predicted single-collector contact efficiency and can be computed using the following equation (Tufenkji & Elimelech, 2004):

$$\eta_0 = \eta_D + \eta_I + \eta_G \quad (2)$$

where η_0 is the overall contact efficiency between microplastics and soil grain and it is dependent on hydrodynamic interactions, van der Waals, and gravitational forces. η_D is the contact due to Brownian force (diffusion), η_I is the contact due to interception, and η_G is the contact due to gravity (settling). Equation (2) relies on additional equations and parameters outlined in Table S4. The respective contribution of different filtration mechanisms was computed by dividing the individual contact efficiency by the total contact efficiency (i.e., diffusion: η_D/η_0 , interception: η_I/η_0 , and gravitation: η_G/η_0). The calculations governing microplastic filtration and their respective contributions in different soils are provided in Table S5. The removal efficiency (η) is computed using the following equation (Tufenkji & Elimelech, 2004):

$$\eta = \eta_0 \times \alpha \quad (3)$$

3.2 | Transport modeling

The breakthrough curves (BTCs) of the microplastics (SMPs and FMPs) were modeled using the HYDRUS-1D computer software package (Simunek et al., 2013). HYDRUS-1D is a

widely used finite-element model that can simulate the movement of water and particle-like solutes in both saturated and unsaturated porous media (Simunek et al., 2016). Several studies have already simulated the transport of microplastics in saturated gravel and quartz sand using HYDRUS-1D (Gui et al., 2022; Li et al., 2024; Ren et al., 2021). In this study, the advection-dispersion equations were implemented in a numerical model using an attachment–detachment model composed of the following equations:

$$\frac{\partial C}{\partial t} + \frac{\rho_b}{\theta} \frac{\partial S}{\partial t} = \lambda \nu \frac{\partial^2 C}{\partial x^2} - \nu \frac{\partial C}{\partial x} \quad (4)$$

$$\frac{\rho_b}{\theta} \frac{\partial S}{\partial t} = k_{\text{att}} C - \frac{\rho_b}{\theta} k_{\text{det}} S \quad (5)$$

where C is the microplastics concentration (particles mL^{-1}), t is the time (min), ρ_b is the dry bulk density (g cm^{-3}), S is the microplastic concentration (particles mL^{-1}), D is the dispersivity (cm), x is the distance along the flow path (cm), ν is the pore water velocity (cm min^{-1}), k_{att} is the attachment rate coefficient (min^{-1}), and k_{det} is the detachment rate coefficient (min^{-1}).

4 | RESULTS

4.1 | Microplastic fragmentation

The fragmentation rate of a material is influenced by the number, mass, and impact velocity of colliding particles (Arnold & Hutchings, 1989). During the abrasion process, the impact velocity remained constant, but the abrasion time was varied (high for 20 μm SMPs), which increased the number of particle strikes and reduced the SMP diameter. The hardness of the particles also impacts the fragmentation rate. The glass beads used in this study were harder (>5000 HV) than the sand (1200 HV) and polystyrene (20 HV) (Barlet et al., 2015), resulting in high abrasion and significant particle size reduction (Figure 1).

Another key factor regarding the effectiveness of the fragmentation process is the microplastic shape. Visual observation showed the plowing impact of the glass beads on the smooth and regular surface of the microspheres by displacing them to the side and causing a frontal impact. Perfect spherical particles (SMPs) now become rough and irregular in shape, after abrasion, and are broken down into smaller particles and debris (FMPs). Walley et al. (1987) found that at equal impact velocities, angular particles such as sand caused more damage to polyethylene surfaces due to increased contact points. Although spherical glass beads were used in this study to simulate abrasion of microplastics, particles in the environment are often angular, which can significantly increase fragmen-

tation and wear rates, potentially by a factor of 10 or more (Hutchings, 1992).

4.2 | Microplastic transport in natural gravel and quartz sand

The transport behavior of SMPs and FMPs was analyzed by comparing BTCs in two different grain size ranges and two textural classes each (natural gravel and quartz sand). In gravel, a higher removal of FMPs was observed, whereas in quartz sand, there was more removal of SMPs (Figure 2). In coarse gravel (8 mm), the percent mass recovery was 88%–90% for SMPs and 74%–76% for FMPs (Figure 2a). In fine gravel (4 mm), there was a decrease in mass recovery ranging from 72% to 74% for SMPs and 59% to 62% for FMPs (Figure 2b), indicating more FMP retention in gravel than for SMPs. The percent mass recovery of microplastics was further reduced in both quartz sands but, in contrast to the findings in the gravel experiment, more retention of SMPs compared to FMPs was observed (Figure 2c,d). In coarse quartz sand (0.6–1.3 mm), the mass recovery ranged from 12% to 14% for SMPs, whereas it was 30% to 31% for FMPs. A similar trend was observed in the medium quartz sand (0.4–0.8 mm), where the percent mass recovery was 5%–6% for SMPs and 19%–23% for FMPs.

Irrespective of the porous media, larger-sized SMPs (10- and 20- μm diameter) experienced straining (Table S1), defined by $d_p/d_{50} > 0.0017$ (Bradford et al., 2002). To characterize the SMPs and FMPs behavior in the porous media, BTCs were modeled using HYDRUS-1D (Figure 2). For gravel media, high values of dispersivity (D) and the detachment rate (k_{det}) highlight the importance of grain size and type of porous media (Table 1). The attachment rates (k_{att}) in medium and fine gravel were 0.0028 and 0.0060 cm^{-1} for FMPs and 0.0012 and 0.0032 cm^{-1} for SMPs, respectively, indicating higher attachment of FMPs compared to SMPs, in gravel, which was also evident from the percent recovery results (mentioned above). In contrast, in coarse and medium quartz sand, the attachment rates were 0.0090 and 0.0136 cm^{-1} for FMPs and 0.0182 and 0.0245 cm^{-1} for SMPs, respectively, indicating higher attachment of SMPs compared to FMPs, which is also in accordance with the percent recovery results.

4.3 | Effect of particle shape on microplastic transport

To investigate the role of particle shape during microplastic transport in the porous medium, column experiments were conducted using FMPs and SMPs with a similar average diameter of approximately 1 μm in saturated coarse

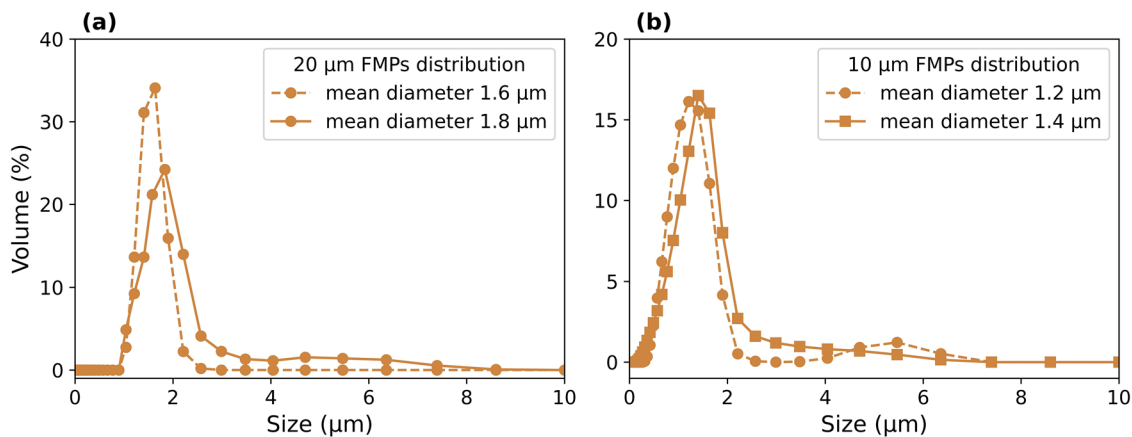


FIGURE 1 Particle size distribution of microplastics after going through an abrasion process. Fragmented microplastics (FMPs) generated from (a) 20 µm and (b) 10 µm. The mean diameter values are the volume-based distribution median.

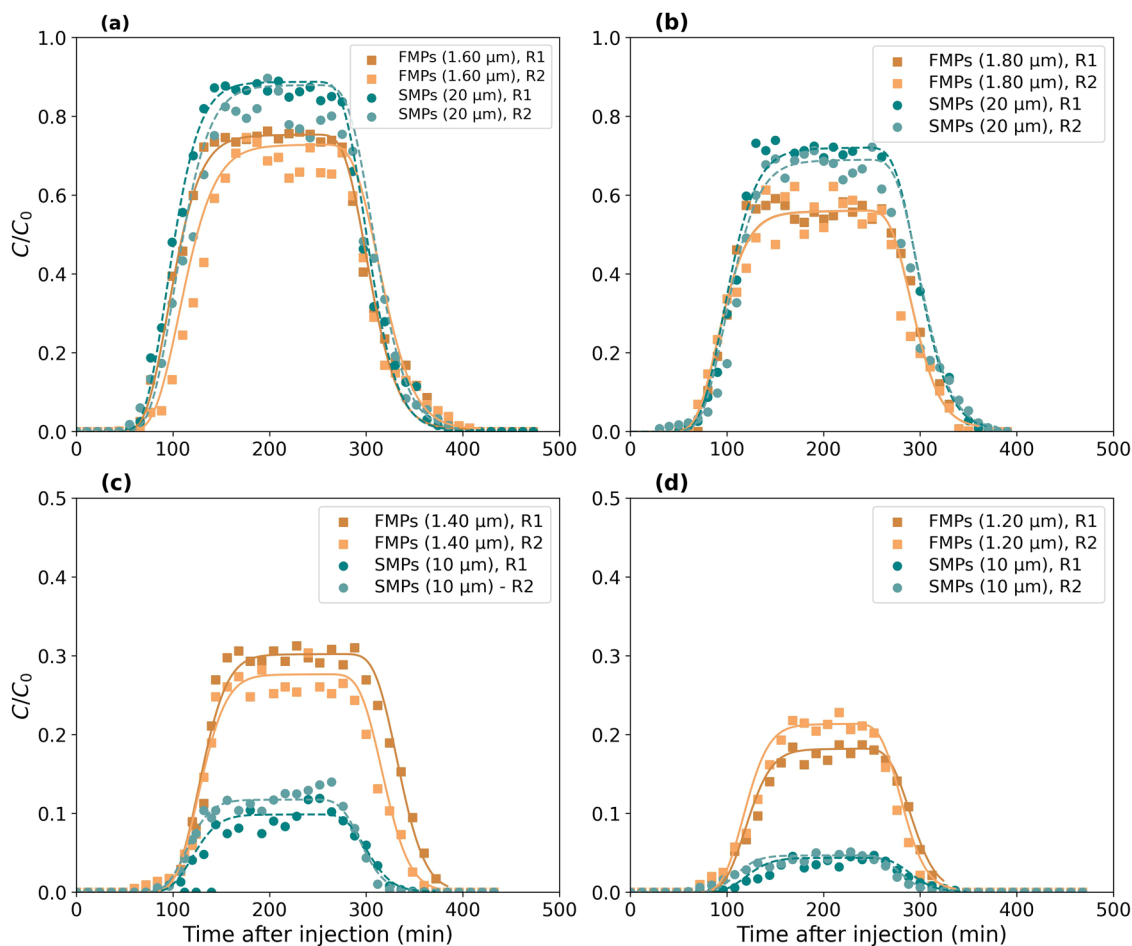


FIGURE 2 Observed and simulated breakthrough curves (BTCs) for SMPs and FMPs in four different materials (a) medium gravel 8 mm, (b) fine gravel 4 mm, (c) coarse quartz sand 0.6–1.3 mm, and (d) medium quartz sand 0.4–0.8 mm. Symbols represent experimental data, and the lines are the fitted breakthrough curves using HYDRUS-1D. SMPs, spherical microplastics (10 and 20 µm only); FMPs, fragmented microplastics. R1 and R2 are experimental runs 1 and 2, respectively. The column was packed with fresh material for each run. BTCs were plotted in the form of the normalized effluent concentration (C/C_0).

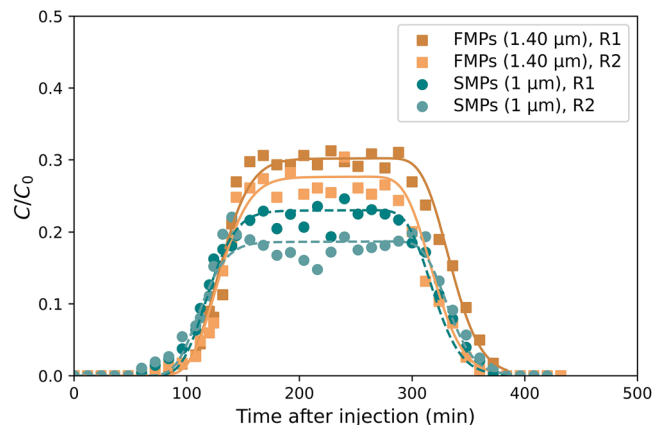


FIGURE 3 Average values of observed breakthrough curves (BTCs) of fragmented microplastics (FMPs) and spherical microplastics (SMPs) (both $\sim 1 \mu\text{m}$) in coarse quartz sand (0.6–1.3 mm). Symbols represent experimental data, and the lines are the fitted breakthrough curves using HYDRUS-1D. R1 and R2 are experimental runs 1 and 2, respectively. The column was packed with fresh material for each run. BTCs were plotted in the form of the normalized effluent concentration (C/C_0).

quartz sand (experiments E1, E2, G1, and G2; Table 1). The FMPs were visually classified as asymmetrical with uneven and fractured surfaces. The average diameter of injected FMPs was $1.40 \mu\text{m}$. The FMPs showed higher values of C_{max}/C_0 (Table 1) and higher values of percent mass recovery (31%) compared to SMPs (24.5%), indicating higher mobility (Figure 3).

4.4 | Filtration efficiency of microplastics

CFT was applied to better understand the filtration, retention, and transport of microplastics in different saturated aquifer materials. Figure 4 shows the values based on averages of column test runs 1 and 2, each run being done in a freshly packed column. As shown in Figure 4a, the single-collector contact efficiency “ η_0 ” of SMPs ranged from 0.58 (medium gravel) to 0.49 (fine gravel), while FMPs exhibited lower values of 0.0051–0.0063, respectively. However, in the case of coarse and medium quartz sand, η_0 ranged from 0.092 to 0.087 for SMPs and 0.0078 to 0.0096 for FMPs, respectively. High values of η_0 indicate that SMPs are expected to experience more collisions with the collector surface (Figure 4a).

As illustrated in Figure 4b, the attachment efficiency “ α ” of FMPs ranged from 1.62 (medium gravel) to 1.11 (fine gravel), whereas SMPs exhibited lower values of 0.006 and 0.009, respectively. This indicates a high removal rate of FMPs compared to SMPs through attachment in gravel (Figure 4d). However, the computed α values for FMPs are reduced to 0.54 (coarse quartz sand) and 0.38 (medium quartz sand), whereas SMPs exhibited increased values of 0.079 and 0.084,

respectively (Figure 4b). Compared to gravel, the significantly higher α values of SMPs in both quartz sands result in a higher removal rate of SMPs by attachment (Figure 4d). Moreover, SMPs also experience straining phenomena in quartz sand (Table S1), leading to high removal rates despite having lower α values compared to FMPs. Since the CFT does not apply to gravel porous media ($\alpha > 1$) and irregular colloids, a comparison of attachment rate coefficient k_{att} (min^{-1}) clearly showed high attachment of FMPs in gravel and high attachment of SMPs in quartz sand (Figure 4c). Furthermore, a comparison was made between the attachment efficiency of $1\text{-}\mu\text{m}$ -sized SMPs ($\alpha = 0.60$) to similar-sized FMPs ($\alpha = 0.54$) in coarse quartz sand (experiments E and G), which clearly showed high attachment of $1\text{-}\mu\text{m}$ -sized SMPs, which is consistent with the experimental data (Figure 3).

Figure 4e,f presents a summary of the contribution of different filtration mechanisms (gravity settling, interception, and Brownian diffusion) to microplastic retention and transport in porous media. In medium and fine gravel, the retention of FMPs (1.60- and $1.80\text{-}\mu\text{m}$ diameter) was mostly influenced by settling (0.71 and 0.65, respectively) and, to a lesser degree, diffusion (0.29 and 0.34), while the behavior of the SMPs (20- μm diameter) is entirely controlled by settling. On the contrary, in coarse and medium sand, respectively, the filtration of FMPs (1.40- and $1.20\text{-}\mu\text{m}$ diameter) is influenced mostly by diffusion (0.73 and 0.85) and, to a lesser degree, settling (0.24 and 0.12), while the behavior of the SMPs (10- μm diameter) is almost entirely controlled by settling (0.95 and 0.90) and interception (0.04 and 0.08).

5 | DISCUSSION

5.1 | Role of different aquifer materials in microplastic transport

Both gravel types (medium and fine) had a similar impact on the transport of FMPs under saturated conditions, resulting in a high retention of FMPs ($\sim 1 \mu\text{m}$) when compared to the larger-sized SMPs (20 μm). In such coarse material, it is surprising that the smaller particles were retained more. One potential reason for this phenomenon is that in natural gravel, the presence of clay particles may fill up the concave locations within the pore spaces and decrease the availability of these locations to retain larger SMPs (Liang et al., 2022). Another important explanation involves the size exclusion of the SMPs from small pore spaces. In comparison to the conservative tracer BTCs, earlier peak times of SMPs BTCs were observed in gravel (Figure S1). This suggests that SMPs can only be transported through the larger pores of gravel, resulting in an earlier detection peak. Similar observations have been reported regarding MPs and colloidal particles, which are compelled to navigate through the broader preferential

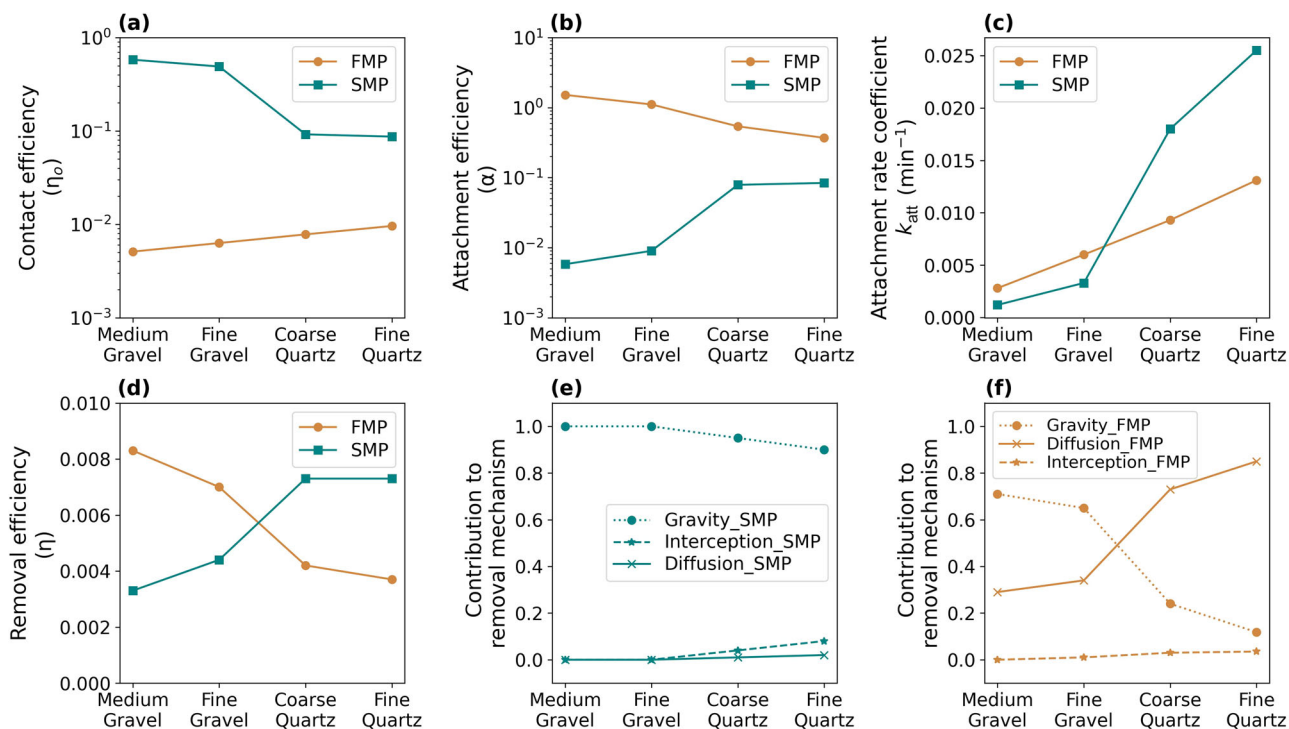


FIGURE 4 Filtration efficiencies of fragmented microplastics (FMPs) and spherical microplastics (SMPs) in various porous media. Panels (a–d) show single-collector contact efficiency, attachment efficiency, attachment rate coefficient (k_{att}), and removal efficiency, respectively. Panels (e, f) depict the contribution of SMPs and FMPs to different transport mechanisms. Data represent the average of the column test runs 1 and 2 with fresh soil material.

flow paths to facilitate their transport (Bradford et al., 2003; Li et al., 2024). In this context, Sirivithayapakorn and Keller (2003) experimentally demonstrated that the observed preferential flow paths are less evident in the case of smaller colloids ($0.05 \mu\text{m}$) compared to the larger colloids ($1\text{--}3 \mu\text{m}$). On the contrary, size exclusion phenomena were not observed in the BTCs for either type of quartz sand.

In the coarse and medium sands, FMPs showed higher mobility, compared to the $10 \mu\text{m}$ SMPs, due to the effect of straining of the SMPs (Supplemental Table S1), which is the removal of colloids due to small pore spaces acting as a sieve and is dependent on median grain size. Minimum values of d_p/d_{50} (d_p is the colloid diameter and d_{50} is the median grain size), where straining has been observed in angular silica sand, range from 0.0017 (Bradford et al., 2002) to 0.02 (Tufenkji et al., 2004). Since some straining values in the experiments are in the above-mentioned range (Table S1), straining could provide a possible explanation for the increased retention of SMPs in both quartz sands; however, none are above the 0.02 threshold from Tufenkji et al. (2004).

The zeta potential represents surface charge and serves as a key factor for microplastic stability in solutions (Dong et al., 2018). Variations in the zeta potential reflect size-dependent dispersion and aggregation of the microplastics. The zeta-potential of FMPs ranged from -21.75 ± 3.79 to

-22.17 ± 2.05 mV, while the zeta potential of the SMPs ranged from -18.22 ± 1.51 to -19.55 ± 3.61 mV (Table S1). These values indicate strong repulsive forces between the microplastics, causing them to disperse more easily from each other. The zeta potential influences the electrostatic interactions (Mei & Lu, 2014), which are crucial for microplastic attachment to porous media. A more negative zeta potential is more conducive to adsorb to less negatively charged particles (Shen et al., 2020).

Since no tailing was observed in any of the experiments (Figure 2), both FMPs and SMPs were likely deposited in the primary minimum, with no energy barrier hindering their interaction with the porous media (Knappett et al., 2008). Moreover, the calculation of single-collector contact efficiency took into account hydrodynamic interactions and van der Waals forces (Table S5). The primary interaction mechanism between SMPs and porous media across all soils was gravitational force (Figure 4e), whereas, for FMPs, Brownian diffusion was the key mechanism in quartz sand (Figure 4f), as reflected by aspect ratio and van der Waals number (Table S5). Consistent with previous experimental observations (Elimlech, 1994; Knappett et al., 2008; Zhuang et al., 2005) and theoretical computation (Huber et al., 2000), microplastics-collector interaction changes nonlinearly with microplastic size. Retention and transport of smaller colloids are more subjected to the influence of diffusion, while behaviors of

larger colloids are mainly controlled by interception and sedimentation (Close et al., 2006; Knappett et al., 2008).

5.2 | Microplastic shape affects its environmental fate and transport

The experiments (tests E1, E2, G1, and G2) examining the influence of microplastic shape on transport were performed with a similar size of microplastic in coarse quartz sand, for which straining due to large particle sizes is not a removal mechanism ($d_p/d_{50} < 0.0017$), and zeta potential is similar for both particle types (Table S1). It was found that the FMPs ($\sim 1.40 \mu\text{m}$) exhibit a higher degree of mobility compared to SMPs ($\sim 1 \mu\text{m}$). This may be due to the unique ability of FMPs to align themselves with the direction of flow, thereby significantly enhancing their overall transport efficiency. Previous studies have found that microplastics with a smaller diameter had higher mobility in the quartz sand (O'Connor et al., 2019; Pradel et al., 2020), which contradicts the findings of tests E and G where the $1 \mu\text{m}$ SMPs are transported less than the slightly larger FMPs (Figure 3). Therefore, the observed enhanced mobility of FMPs may be due to shape instead of size; an explanation for this could be that the higher aspect ratio of FMPs (Table S5) exposes them to larger surface tension forces, causing them to re-orient or re-align with the flow direction to counteract the dynamic fluid forces (Liu et al., 2010; Xu et al., 2008). This could also be described as a tumbling (rotational) movement leading to greater mobility within saturated quartz sand (Aramrak et al., 2013). Comparable to this study, Tumwet et al. (2022) also reported high mobility of FMPs in quartz sand, due to velocity fluctuations because of the asymmetrical shapes, and experimentally demonstrated that the shape of microplastics is one of the critical factors during transport. The current study found that even though FMPs had a larger average particle diameter and would be anticipated to have higher removal, the irregular shape of the particles emerged as the primary factor influencing mobility within saturated coarse quartz sand.

5.3 | Microplastic response to filtration mechanisms

Considering the transport of FMPs in natural gravel, high values of attachment efficiencies predicted that more removal of FMPs would occur (Figure 4b), which was in agreement with the experimental breakthrough data (Figure 2a,b). However, there were inconsistencies in the case of quartz sand. Despite higher values of the attachment efficiency for FMPs, compared to the larger $10 \mu\text{m}$ SMPs (which would imply more removal), experimental breakthrough data showed less removal of FMPs (Figure 2c,d). Several factors can poten-

tially contribute to these inconsistencies, including the size, shape, and roughness of the FMPs. One explanation is that since the majority of the FMPs used in the current study were approximately $1.20\text{--}1.80 \mu\text{m}$ in diameter (Table 1), this diameter range falls in the transition zone where all three filtration mechanisms can act simultaneously (i.e., Brownian diffusion, interception, and gravity settling). Moreover, straining is also likely playing a role, as Bradford et al. (2002) advised using caution when applying CFT when the d_p/d_{50} ratio is above the 0.0017 threshold, which is the case for the 10 and $20 \mu\text{m}$ SMPs, but especially (high d_p/d_{50}) in the coarse and medium quartz sand. Additionally, the particle shape and surface roughness of FMPs may influence the kinetic interactions with collector surfaces during the filtration process leading to more breakthrough. Several researchers suggest that grain surface area, angularity, and roughness can all play a role in physicochemical filtration (Bhattacharjee et al., 1998; Saiers & Ryan, 2005; Shellenberger & Logan, 2002). These inconsistencies indicate that additional factors beyond those accounted for by CFT should be taken into account, such as the shape of microplastics.

5.4 | Implications for groundwater management

Understanding microplastic retention and transport behavior in saturated porous media is imperative to be able to evaluate the potential risks of microplastics in groundwater. The results of this study suggest that the size and shape of microplastics were important factors influencing groundwater transport in various aquifer materials, and fragmented particles showed greater mobility in quartz sand (Figure 2c,d). Concerning microplastic shape and size, Pivokonsky et al. (2018) reported fragments as the most abundant morphotype (size range: $1\text{--}10 \mu\text{m}$), whereas spherical particles were the least prevalent in raw drinking water. Despite this fact, there is a lack of comprehensive data concerning the size, shape, and rates at which microplastics undergo fragmentation and how they are transported in various aquifer materials. The findings of the current study showed that these fragmented particles could pose a more significant threat to sandy aquifers, as they can potentially be transported further than spherical particles of the same size and could act as vectors for long-distance transport due to their co-existence with pathogens in wastewater effluents and sludge (Edo et al., 2020; Pham et al., 2021).

The experiments in the current study are focused on saturated conditions, whereas in the terrestrial environment, microplastic transport to groundwater via infiltrating water primarily occurs through the unsaturated (vadose) zone. The transport processes in the vadose zone are highly complex, characterized by intricate air–water interfaces within pore spaces (Bradford & Torkzaban, 2008). This

complexity results in the formation of liquid film entrainment, which contributes to the fate of microplastics influenced by flow velocity, volume fraction, and pore sizes (He et al., 2023; Wu et al., 2021). In natural environments, soil saturation conditions fluctuate, underscoring the importance of studying both conditions together to improve predictions of microplastic mobility and potential contamination risks.

6 | CONCLUSIONS

This study aimed to understand the transport of microplastics in saturated porous media based on their size and shape. The transport behavior of FMPs and SMPs was compared in different aquifer materials, such as natural gravel and quartz sand. The experiments showed that the soil type and the size and shape of the microplastics influence the transport of microplastics. In gravel, the larger SMPs experienced higher breakthrough than FMPs, possibly due to size exclusion. The irregular shape and rough surface of FMPs may increase their attachment to impurities (e.g., clay particles) present on the natural gravel surface. Due to straining, the larger SMPs showed higher retention in quartz sand. It was found that FMPs were more mobile than similar-sized SMPs in the coarse quartz sand, possibly due to the high aspect ratio forcing them to re-align with the flow direction. The results indicate that settling was the primary retention mechanism for SMPs in gravel, while diffusion was dominant for FMPs in quartz sand. The outcome of this study suggests that future research should use irregularly shaped plastic materials to closely replicate microplastics in the natural environment and to better assess the risks of microplastic transport in soil and groundwater.

AUTHOR CONTRIBUTIONS

Ahmad Ameen: Conceptualization; methodology; data curation; formal analysis; software; investigation; visualization; writing—original draft; writing—review and editing. **Margaret E. Stevenson:** Conceptualization; methodology; formal analysis; software; validation; supervision; visualization; writing—review and editing. **Alexander K. T. Kirschner:** Funding acquisition; resources; supervision; writing—review and editing. **Stefan Jakwerth:** Methodology; resources. **Julia Derx:** Funding acquisition; writing—review and editing. **Alfred P. Blaschke:** Conceptualization; funding acquisition; supervision; writing—review and editing.

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CONFLICT OF INTEREST STATEMENT

The authors declare no conflicts of interest.

ORCID

Ahmad Ameen  <https://orcid.org/0009-0005-6123-9159>

Margaret E. Stevenson  <https://orcid.org/0000-0002-4259-4033>

Alexander K. T. Kirschner  <https://orcid.org/0000-0002-9797-3073>

Julia Derx  <https://orcid.org/0000-0002-9931-088X>

Alfred P. Blaschke  <https://orcid.org/0000-0001-8617-5802>

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