



Transport of micron-sized polyethylene particles in confined aquifer: Effects of size, aging, and confining pressure

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ABSTRACT

Contamination of groundwater by microplastics (MPs) is increasingly reported and attracts a growing attention due to their potential risks. To understand how MPs migrate into groundwater, many previous works have investigated their transport using man-made microspheres of few microns or smaller as models. However, MPs observed in the environment are more diverse in size, shape, and type, which may have different migration behaviors. In this work, the transports of irregularly shaped polyethylene (PE) particles in the sand packed column were studied. Small MPs (22–37 μm) generally have high mobility than large MPs (44–74 μm) but can also be affected by aging. Aging decreased the hydrophobicity of the MPs and increased their surface negative charge, which can facilitate the transport of MPs. However, the physical barrier of space in the porous media might have a greater influence on their transport. The retention of the MPs was enhanced with the increase of pressure due to compression that decreased pore size. Results from this study showed that MPs of environmental features can also be transported in the groundwater but the processes could be governed by different factors, such as physical interception and steric hindrance.

1. Introduction

Mankind is now in the plastic age with the world's plastic production increasing from 1.7 million tons in 1950 to 367 million tons in 2020 [1,2]. The improper disposal of waste plastics has caused serious environmental pollution [3]. The plastic debris can break down into microplastics (MPs, <5 mm in size) through abiotic and biotic processes, leading to MPs pollution being a more serious problem [4,5].

Studies showed that MPs are ubiquitous in all environmental compartments [6–8]. Most of the previous works on microplastics focused on the aquatic environment while recent research indicated that microplastic pollution in terrestrial environments is even more serious [9]. It's reported that the annual release of plastic wastes into the terrestrial system was 4–23 times more than that enters the ocean [10]. Recent evidence demonstrated that MPs in the terrestrial environment can be transported into groundwater [7,11–13], which aroused potential concern on drinking water safety. Microplastics have been identified in drinking water and food products for human consumption. Thus, it may directly or indirectly impact human health by acting as physical stressors or vectors of other

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contaminants [14].

Several works have investigated the vertical transport of microplastics in porous media. However, most of the works use spherical polystyrene (PS) as a model, while MPs in the environment have various shapes that may affect their transport, and polyethylene (PE) is the main type of microplastic pollution [2,15]. Transports of PE MPs were found to be influenced by internal factors like the aging of plastic [16]. In addition, many external factors, such as input concentration, flow rate, size of the MPs, and surfactant concentration can also have an effect [17,18]. However, the transport of MPs in external environmental conditions, such as confined water, remains unclear.

The specific aims of this study are: (1) to determine the effect of size and aging on the transport of polyethylene microplastics in confined water. (2) apply and test numerical models for polyethylene microplastics transport under tested conditions. Results from this work can provide a better understanding of the transport of MPs in the groundwater environment and the controlling factors.

2. Materials and methods

2.1. PE MPs characterization and preparation

PE MPs was purchased from Huachuang Plastic Company (Dongguan, China). According to the supplier, the MPs were prepared by pulverizing large plastics in a mill with liquid nitrogen and passed through a 6.5 μm sieve in a frozen state. However, the measured size distribution of the purchased MPs was 22–74 μm at room temperature, using S3500 Particle Size Analyzer (Microtrac, USA) (Fig. S1). Due to a wide size range, the MPs was further passed through a 45 μm sieve in the laboratory, and two different size ranges after sieving were 22–37 μm and 44–74 μm , respectively.

MPs of two different size ranges were aged in a UV aging chamber (30 W, 365 nm irradiation) for 40 days to simulate photo weathering. MPs were placed in a Petri dish with ultrapure water 15 cm from the UV lamps. Water loss was replenished every two days.

The stability of the PE MPs was determined by counting with a fluorescence microscope in 2 h. The suspension of PE MPs with a concentration of 20 mg/L (equivalent to about 1.5 billion particles/L) was chosen. Nile red (NR) stain was used for the quantification of MPs referring to Shruti [19], Perez-Guevara (19). Concentrations were calculated by counting under a fluorescence microscope (S1). To ensure a uniform distribution of PE MPs during counting, we took samples in batches and mixed the solution with a vortex mixer before each sample, and counted 50 fields. The infrared spectra of the MPs were analyzed using a Nicolet iN10 attenuated total reflection Fourier-transform infrared spectrometer (ATR-FTIR) (Thermo, USA) (Fig. S2). The morphology of the MPs was examined using a JSM-IT 300HR Scanning Electron Microscope (SEM) (JEOL, Japan) (Fig. S3). The contact angle of unaged and aged PE MPs was measured by an OCA Contact Angle Tester (Dataphysics, Germany) (Fig. S4).

2.2. Preparation of river sand columns

The river sand (density: 2.75 g/cm³) purchased from Weiteng Company (Yueyang City, Hunan, China) was used for the experiment. The sand was air-dried and sieved through a 2 mm sieve to remove large particles and impurities, and then baked at 400 °C for 3 h to remove organic materials that may interfere with the quantification of MPs [20]. The prepared sand was stored in a desiccator before being used in the experiment. The composition of the river sand was analyzed using a SmartLab SE X-Ray Diffractometer (Rigaku, USA) (Fig. S5). For measurement of zeta potentials, the sand was grinded and passed through 200 mesh sieve and the PE MPs suspension was the same as in experiments (20 mg/L, Milli-Q water as the background solution). The zeta potentials of PE MPs and river sand were determined by Zetasizer Nano ZEN2600 (Malvern Instruments, UK). The properties of the tested materials are presented in Table 1.

2.3. Column experiment

A flexible wall permeameter (FWP) was employed for the column experiment [21]. Confining pressure was adjusted to simulate different head pressures of a confined aquifer. The latex membrane was embedded in a hollow drilled polyvinyl chloride (PVC) pipe (inner diameter: 5 cm, height: 12 cm) to form a three-dimensional pressure and filled with 10 cm sand. Once the initialization phase was completed, the background solution started to inflow. The integral device is depicted in Fig. 1.

We selected three confining pressures (0.1, 0.3, 0.5 MPa), two sizes (22–37, 44–74 μm), and non-aging and aging processing as variables to generate 12 treatment groups. Details on the three sets of matching variables are described in Table 1. The sand was packed into the column with a porosity of 0.46 under atmospheric pressure. Changes in the pore volume of the sand column were determined using a Model WG Single Lever Consolidmeter by measuring the amount of compression deformation and the compression

Table 1
Properties of the tested materials.

Materials	Aging	Size (μm)	Contact angle (°)	Zeta Potential (mV)	pH
PE	Unaged	22–37	141.37	−10.00	7.73
		44–74	141.37	−11.31	7.76
	Aged	22–37	138.28	−15.53	7.53
		44–74	130.77	−17.23	7.35
River sand	-	704–2000	-	−23.68	-

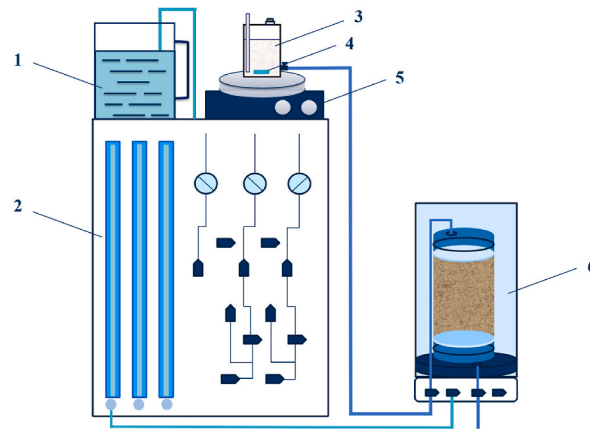


Fig. 1. The schematic diagram of the experimental device. 1: Water supply tank, 2: Logic panel, 3: Mariotte bottle, 4: Magnet, 5: Magnetic agitator, 6: Permeameter cell.

process. Details are provided in the Supplementary Materials.

A Mariotte bottle served as an inlet device to deliver a constant flux for the permeameter [22]. A magnetic agitator was placed under the Mariotte bottle to mix the MPs with water. At the beginning of the experiment, 5 pore volumes (PVs) of deionized water were loaded into the column for pre-equilibration, followed by 3 PVs of MPs suspension. Then, the column was eluted with 3 PVs of deionized water. The effluent samples were collected in glass bottles from the outlet every 0.5 PVs. In the end, the sand column was sliced at an increment of 1 cm and MPs that remained in each layer were analyzed. The extracted sand samples were transferred into glass bottles and weighed. MPs were extracted following a previous method [23].

Subsequently, nonreactive tracer tests were carried out with 1 mM KBr to determine the hydrodynamic properties of the sand columns with no retardation, and details are provided in the Supplementary Materials (S5). All experimental groups were conducted in duplicate.

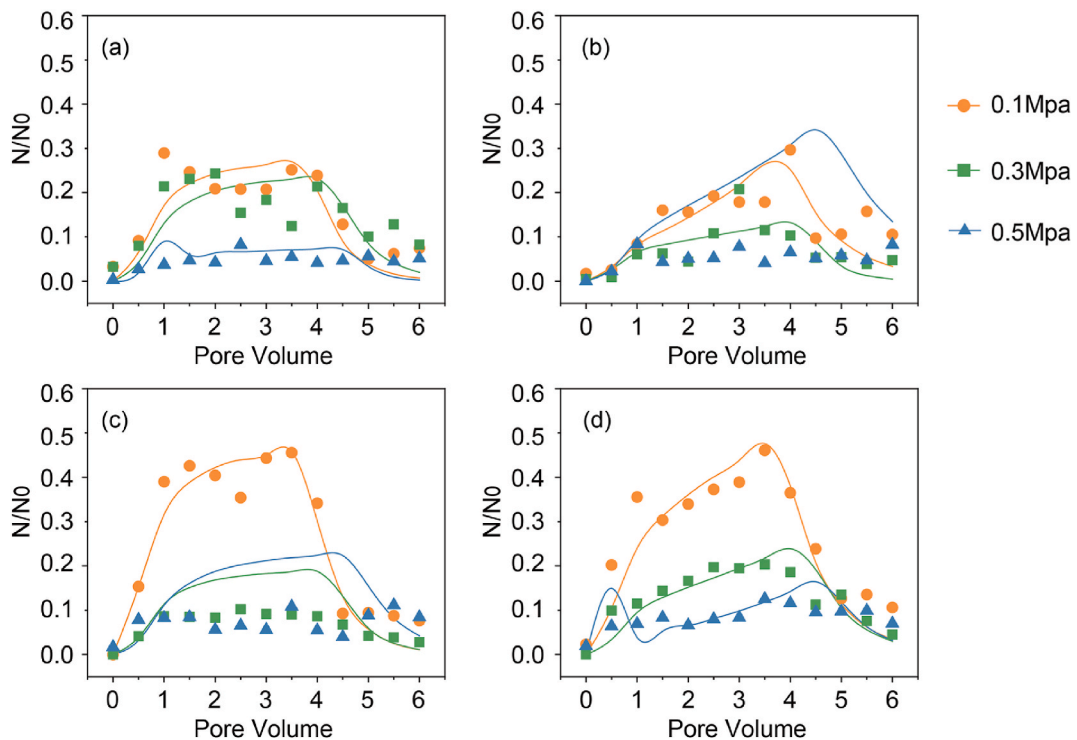


Fig. 2. Breakthrough curves of unaged 22–37 μm MP (a), unaged 44–74 μm MP (b), aged 22–37 μm MP (c), and aged 44–74 μm MP (d) under different confining pressures. Symbols represent experimental data, and the solid lines are the fitted BTCs. (Symbols represent the mean value of experiments performed in duplicate).

2.4. Modeling

The advection-dispersion equation (provided in S6 in the supporting information) was used to describe the transport of MPs in the sand column. This model was used to simulate the breakthrough curves of the tracer test (Br^-) and obtain the best-fit values of D (dispersion coefficient, $\text{cm}^2\text{min}^{-1}$). The breakthrough curves of PE MPs were simulated inversely fitted using HYDRUS-1D to obtain the best-fit values of K (first-order retention coefficient, min^{-1}) and S_{max} (the maximum solid phase particle concentration), used as the fitting parameters.

3. Results and discussions

3.1. Transport and retention of MPs

The stability of the suspensions PE MPs with and without treatment in 1 h were displayed in Fig. S6. Changes of normalized concentration (i.e., C/C_0) of PE MPs was negligible within the time period of the transport study, indicating a good stability.

Breakthrough curves (BTCs) of the MPs at different test conditions are presented in Fig. 2. MPs were detected in the effluent after loading of 0.5 PV MP suspension and reached a plateau after loading of 1 PV suspension. After 3 PV suspension, MPs in the effluent began to decrease and reached equilibrium after 2 PVs of deionized water. Our results demonstrated that irregularly shaped MPs could be transported in the confined aquifer. Our results agree with previous observations, which showed that spherical MPs can be transported in natural soils and sand [24–26].

Retained profiles of MPs in the column at the end of the experiment were presented in Fig. 3. In all treatments, a large portion of the MPs was retained in the column after elution with 3 PVs of deionized water, which could be attributed to the presence of small pores in the column that blocked MPs physically. The retained profiles (RPs) scatter considerably within a small dynamic range. This can be attributed to the non-uniform size of sand (Fig. S1a), which provides the different deposition sites in each section of the column. The incomplete symmetrical shape of the tracer BTC (Fig. S7) indicates that the non-homogeneous sand column may also cause scattering. In addition, it is observed that when the ratio of particle-to-media size exceeds the theoretical value of 3×10^{-3} , the straining effects of particles become more prominent [27]. This phenomenon is caused by the velocity gradient generated by the fluid passing through a porous medium, which creates a pressure drop across the medium and lead the fluid to strain and deform as it passed through small pores. Additionally, straining can occur via bridging mechanism, allowing multiple particles to simultaneously enter a pore and accumulate in a constriction [28]. Such behaviors resulted in irregular fluctuations in the concentration of retained particles within a segment.

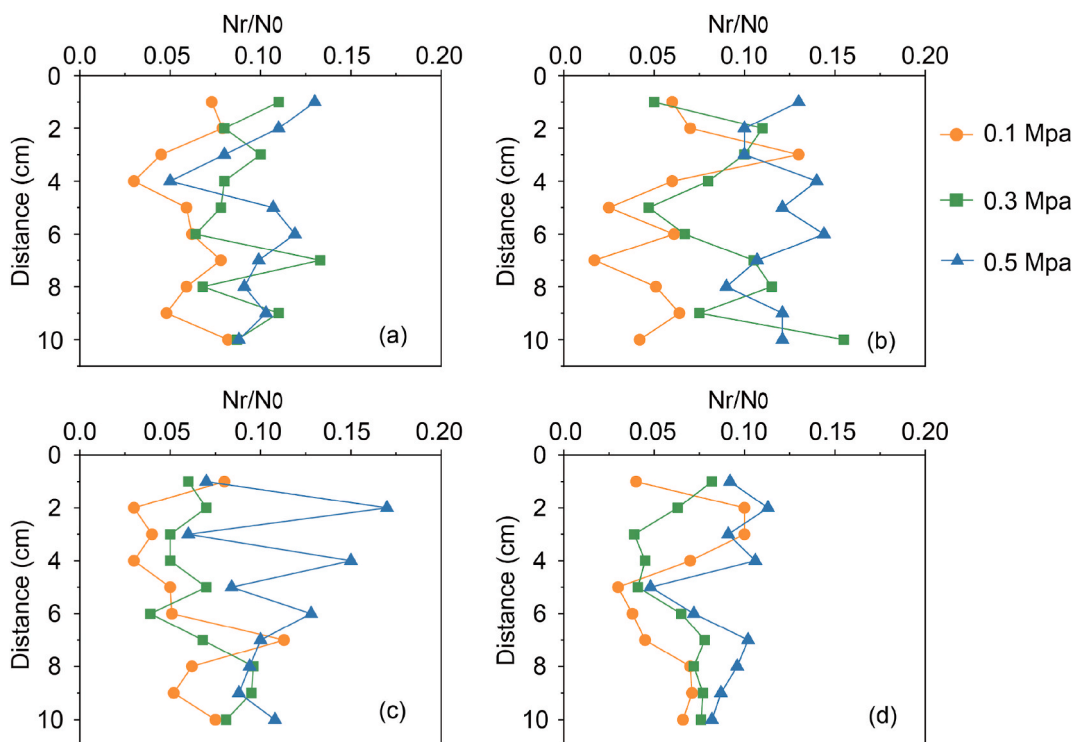


Fig. 3. Retention profiles of unaged 22–37 μm MP (a), unaged 44–74 μm MP (b), aged 22–37 μm MP (c), aged 44–74 μm MP (d) at the end of the experiment. (Symbols represent the mean value of experiments performed in duplicate).

The mass balance and parameters of the BTCs of MP transport in a confined aquifer are presented in Table 2. The percentage of MPs retained in the column varied from 58 to 117%, which are much higher than the results from previous works with smaller MPs [24,29]. The fitness of the BTCs varied from 0.007 to 0.870, which is relatively low compared to those works with MP spheres [16,29]. According to Bradford, Yates [30], the numerical simulations demonstrate that traditional colloid attachment theory is not well suited to describe colloid transport in systems that exhibit straining, which caused deviation from the ideal situation. Especially for high confining pressure the pores are even smaller so that the straining is more serious and leads to particularly poor fitted values.

3.2. Effect of size on the transport of MPs

Comparing the transport of MPs of two size ranges, more 22–37 μm unaged MPs were eluted out of the column in the effluent than 44–74 μm unaged MPs (Table 1). However, 22–37 μm aged MPs in the effluent were comparable to even less than 44–74 μm aged MPs at certain sites. The infiltration of MPs was found to depend on the ratio between the diameter of MPs and the diameter of the porous media $d_{\text{MP}}/d_{\text{PM}}$ [31]. Our results suggested that the transport of MPs in a confined aquifer is related to the size of the MPs but can also be affected by aging.

Dong, Qiu (28) studied the transport and retention of MP spheres with diameters ranging from 0.1 to 2.0 μm , complete breakthroughs of all MPs were observed in deionized water while retention of MPs was enhanced in simulated seawater, and an increase of MP size from 0.8 to 2.0 μm resulted in reduced MP transport. Cai, He (31) showed that the breakthrough of polystyrene (PS) MPs with a diameter of 0.2, 1, and 2 μm in quartz sand decreased with the increase of MP diameter and the presence of TiO_2 nanoparticles enhanced the retention of MPs.

In those previous works, MPs investigated are a few microns to hundreds of nanometers in size. Those MPs can exhibit colloidal properties during transport in porous media. Aggregation of MPs was observed and has a great influence on their transport in porous media [24,29]. In this work, the sizes of the MPs are much larger and are closer to MPs actually detected in the environment, and more MPs are retained.

Another work studied the transport of irregularly shaped polyvinyl chloride (PVC) and low-density polyethylene (LDPE) (125–300 μm) in saturated quartz sand and found that small-sized MPs and fragmentation promoted migration while larger MPs experienced obstruction [32]. It was suggested that secondary microplastics with irregular shapes are more likely to experience fragmentation and infiltrate deeper than primary microplastics when transported through porous media [7].

3.3. Effect of aging on the transport of MPs

Aging decreased the contact angle of the MPs and the zeta potential became more negative (Table 1), indicating that aged MPs became more hydrophilic and the double layer repulsive force between particles became greater. Also, the increased contents of oxygen functional groups on aged PE enhanced the role of hydrogen bonding in the process [33]. Thus, aging enhanced the transport of MPs under all conditions (Table 2).

Previously, both chemical and photoaging were found to be able to increase the transport of MPs in saturated soils [16,34]. Similar to the result of this work, the aging of MPs caused the increase in surface negative charge and hydrophobicity. The extended Derjaguin-Landau-Verwey-Overbeek (XDLVO) model calculation suggested that greater surface hydrophilicity was mainly responsible for the higher mobility of the aged MPs, whereas the contribution from increased surface charge negativity was relatively small [34].

In this work, the contact angle of both aged and unaged PE MPs was larger than 90° . Although aging decreased the contact angle of the PE MPs, which contributes to a decrease in hydrophobicity, their surface was still hydrophobic. However, the contact angle

Table 2
Mass balance and parameters of the BTCs of MP transport in the experiment.

Size (μm)	Pressure (MPa)	Mass Balance			Parameters		
		$M_{\text{eff}}^{\text{a}}$	$M_{\text{ret}}^{\text{b}}$	$M_{\text{tot}}^{\text{c}}$	S_{max} (mg/kg)	K (min^{-1})	R^2
Unaged MPs 22–37	0.1	0.28	0.62	0.90	0.497	0.189	0.633
	0.3	0.27	0.91	1.18	0.107	0.131	0.328
	0.5	0.08	0.98	1.06	0.601	0.152	0.147
44–74	0.1	0.23	0.58	0.81	0.289	0.167	0.472
	0.3	0.13	0.90	1.03	0.216	0.811	0.671
	0.5	0.09	1.17	1.26	0.102	0.172	0.007
Aged MPs 22–37	0.1	0.45	0.58	1.03	0.396	0.154	0.868
	0.3	0.12	0.68	0.80	0.982	0.216	0.858
	0.5	0.12	1.05	1.17	0.601	0.152	0.092
44–74	0.1	0.46	0.63	1.09	0.166	0.168	0.828
	0.3	0.23	0.64	0.87	0.194	0.259	0.870
	0.5	0.15	0.89	1.03	1.022	0.172	0.500

^a Percentage of MPs eluted out of the column in the effluent.

^b Percentage of MPs retained in columns.

^c Total percentage of MPs recovered from each treatment.

decreased from $>90^\circ$ to $<90^\circ$ for polystyrene (PS) nanoplastics after aging [34], which changed from hydrophobic to hydrophilic. The zeta potential of MPs became more negative after aging in our study and previous works due to the introduction of hydrophilic functional groups as a result of oxidation [35]. Both the MPs and the river sand are negatively charged, and the electrostatic repulsion between the aged MPs and between the aged MPs and the river sand increased.

For small MPs, the retention of MPs could be related to the deposition or electrostatic attraction. As demonstrated by Wu, Lyu (26), the retention of PS nanoparticles in soils was positively correlated with Fe/Al oxides contents due to electrostatic attraction. In another study, it was found that enhanced deposition of PS latex microspheres (diameter of 0.2, 1, and 2 μm) with carboxylic functional groups in the presence of nano-TiO₂ at pH 5, which resulted in increased retention of the MPs [36]. However, MPs used in this work are larger and have an irregular shape, the physical barrier ($d_{\text{MP}}/d_{\text{PM}}$) of pores in the river sand column may be more important in the retention of the MPs.

3.4. Effect of confining pressure on the transport of MPs

Groundwater is buried underground and could be under a certain pressure in the confined aquifer. In this work, the effect of confining pressure on the transport of MPs was assessed up to 0.5 MPa, which corresponds to a 50 m water head difference. The porosity of the sand column after pressurization of 0.1, 0.3, 0.5 Mpa was 0.40, 0.37, and 0.33, respectively. For both aged and unaged MPs, an increase in confining pressure enhanced the retention of MPs. The retention of unaged MPs increased from 62 to 98% and from 58 to 117% for 22–37 and 44–74 μm MP, respectively. The retention of aged MPs increased from 58.3 to 105% and from 63 to 89% for 22–37 and 44–74 μm MP, respectively.

The influence of pressure could be related to the deformation of the sand column, which resulted in a decrease in pore size due to compression. The result of the consolidated test is presented in Fig. 4, deformation of the sand columns under pressure reached equilibrium in a few minutes and increased with the increase of pressure. At higher pressure, sand particles are more densely packed, and the space between the particles becomes smaller. Therefore, MPs are more likely to be retained. Enhanced retention under higher pressure suggests that MPs could be more difficult to transport in deep groundwater in general. However, characteristics of the porous media, such as particle size, mineral composition, and hydro-chemical characteristics, could also have an impact on the migration of MPs [37].

4. Conclusion

Transport of PE MPs in a confined aquifer was studied using a flexible wall permeameter with the packed sand column in a simulation experiment. Effects of size, aging, and confining pressure on the MPs were assessed. Results indicated that MPs of two different size ranges (22–37 and 44–74 μm) can be transported in the sand column. Small size MPs have high mobility but their transport was affected by aging. Aging decreased the hydrophobicity of the MPs and increased surface negative charge, which could facilitate the transport of MPs. However, the physical obstacle of space in the porous media may have a greater influence on the transport of large MPs compared with those of few microns or less. The retention of the MPs was enhanced with the increase in pressure due to compression that can decrease pore size. Therefore, deep groundwater might be less vulnerable to MPs. Different processes may be involved for MPs of different sized. In the future, more attention should be paid to the transport of MPs with environmental features other than man-made plastic spheres.

Author contribution statement

Xin Chen: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Wrote the paper. Yong Wan: Conceived and designed the experiments; Performed the experiments; Contributed reagents, materials, analysis tools or data. Jia Jia: Analyzed and interpreted the data. Qiang Xue: Contributed reagents, materials, analysis tools or data. Chenxi Wu: Conceived and designed the experiments; Wrote the paper. </p>

Data availability statement

Data will be made available on request.

Additional information

Supplementary content related to this article has been published online at [URL].

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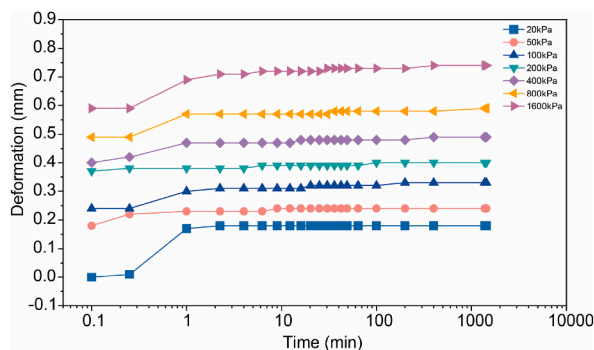


Fig. 4. Deformation of the sand columns under different pressure.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary data

Supplementary data related to this article can be found at <https://doi.org/10.1016/j.heliyon.2023.e18464>.

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