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Microplastic accumulation in groundwater: Data-scaled insights and future research

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ABSTRACT

Given that microplastics (MPs) in groundwater have been concerned for risks to humans and ecosystems with increased publications, a Contrasting Analysis of Scales (CAS) approach is developed by this study to synthesize all existing data into a hierarchical understanding of MP accumulation in groundwater. Within the full data of 386 compiled samples, the median abundance of MPs in Open Groundwater (OG) and Closed Groundwater (CG) were 4.4 and 2.5 items/L respectively, with OG exhibiting a greater diversity of MP colors and larger particle sizes. The different pathways of MP entry (i.e., surface runoff and rock interstices) into OG and CG led to this difference. At the regional scale, median MP abundance in nature reserves and landfills were 17.5 and 13.4 items/L, respectively, all the sampling points showed high pollution load risk. MPs in agricultural areas exhibited a high coefficient of variation (716.7%), and a median abundance of 1.0 items/L. Anthropogenic activities at the regional scale are the drivers behind the differentiation in the morphological characteristics of MPs, where groundwater in residential areas with highly toxic polymers (e.g., polyvinylchloride) deserves prolonged attention. At the local scale, the transport of MPs is controlled by groundwater flow paths, with a higher abundance of MP particles downstream than upstream, and MPs with regular surfaces and lower resistance (e.g., pellets) are more likely to be transported over long distances. From the data-scaled insight this study provides on the accumulation of MPs, future research should be directed towards network-based observation for groundwater-rich regions covered with landfills, residences, and agricultural land.

1. Introduction

Microplastics (MPs) with a particle size < 5 mm pose a significant challenge to the water security of freshwater systems and has implications for the World Health Organization's One Health Initiative (https://www.who.int/teams/one-health-initiative). MPs originate from the weathering and decomposition of plastic objects, car tires, clothing, etc., and can accumulate in organisms, thereby increasing morbidity and mortality (Vethaak and Leslie 2016; Vethaak and Legler 2021). Since the first detection of MPs in the oceans in the 1970s, MPs have been found in rivers, reservoirs, and lakes across all continents, including Antarctica (Cunningham et al., 2022; Rochman, 2018). Atmospheric deposition and surface runoff are the main pathways for the occurrence of MPs in surface water (Rochman and Hoellein, 2020; Wang et al., 2022). Higher abundances of MPs are found in densely populated, urbanized areas, and in waters with elevated deposition areas, long water-residence times, and high anthropogenic impacts (Nava et al., 2023). Remote areas without direct anthropogenic activity can also be polluted due to atmospheric transport and deposition (Allen et al., 2019; 2021; Sun et al., 2022). Surface water and groundwater can be inherently interconnected in riverbeds, floodplains, wetlands, and springs (Scanlon et al., 2023), which results in the storage and transfer of MP contaminants in groundwater.

Extensive investigation of MP contamination in groundwater, a significant drinking water source, highlights the widespread degree of contamination in these systems. Investigating the quality of

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groundwater is difficult due to its location below the ground surface, therefore it can only be sampled and measured by means of groundwater-fed springs, karst caves, or artificially dug wells. Although the particle size of MPs that can infiltrate into groundwater is limited by soil and rock pore sizes, the surfactants they carry can greatly enhance the mobility of MP particles in porous media (Jiang et al., 2021). In 2019, MPs were first detected in groundwater from karst aquifers, which provide 25% of global drinking water sources (Panno et al., 2019). Subsequently, several nations launched investigations into the occurrence of MPs in groundwater across diverse regions, leading to an increase in monitoring data on groundwater MPs (Chia et al., 2021; Viaroli et al., 2022). However, most existing studies have focused on the occurrence, distribution, and ecological risk of MPs in groundwater in a single or localized area, with the spatial and temporal heterogeneity from a large-scale perspective overlooked.

Employing a data integration methodology by mapping measurements from various scales to a uniform framework can provide insights into groundwater MP contamination on multiple scales. The analysis of datasets associated with other pollutants in groundwater, such as arsenic and fluoride, and application of machine learning techniques, have revealed statistical relationships with environmental parameters and pollutant concentrations. This has resulted in the generation of high-resolution regional/global distribution prediction maps (Podgorski and Berg, 2020; 2022; Araya et al., 2022; McDonough et al., 2020). However, to date, MP datasets are small and have not been able to support the creation of a global prediction map. Despite this, they have still provided some important ecological insights (Todman et al., 2023). For example, data-driven meta-analyses integrating over 400 MP data from reservoirs and bays and adopting the basic framework of "extraction, diversion and statistics" has yielded corresponding scientific findings in terms of spatial patterns, compositional patterns, and ecological risks (Guo et al., 2021; Liu et al., 2022). Here, we adopt and enhance these frameworks, according to the characteristics of land-use types that cannot be ignored during the migration of contaminants to groundwater (Ekpe et al., 2024).

In this study, we utilise a novel Contrasting Analysis of Scales (CAS) approach to systematically review and analyze data from numerous studies, expanding our understanding of groundwater MP distribution on three different spatial scales: overall, regional, and local. Through this, we aim to (i) identify the differences in MP characteristics and main pollution pathways between Open Groundwater (OG) and Closed Groundwater (CG) on the full data scale; (ii) elucidate the distribution characteristics and ecological risk of MPs covered by existing studies on a regional scale, and (iii) investigate the effects of groundwater flow pathway on MPs on a local scale. Our novel analysis explores the pollution characteristics, spatial distribution, and existing issues of groundwater MP contamination, which provides a scientific basis for formulating control and management measures for MP pollution.

2. Methods

2.1. Data extraction

In April 2024, a search in the Web of Science core collection database using the formula "Topic = "microplastic*" AND ("groundwater" OR "underground water" OR "aquifer")" to search the peer-reviewed papers on groundwater MPs, and 209 papers were obtained by specifying the retrieval types as articles and reviews. These papers were then manually screened according to the following criteria: (i) the studies conducted on untreated groundwater, and (ii) it contained at least one data on the characteristics of MPs, such as abundance or morphological characteristics (color, particle size, shape, and polymer type). 163 of these papers focused on targets other than the occurrence of groundwater MPs and 17 papers that did not contain MP data were therefore excluded. Based on the screening criteria, 29 papers providing information on groundwater MPs occurrence were identified and included in this study.

In the remaining 29 papers, information on sampling time, geographical location, land-use type, well depth, sample depth, sampling method, extraction and separation method, identification method, as well as MP abundance and morphological characterization data were extracted directly from the text or using a graphical digitization tool (Plot Digitizer 3.3). The abundance and morphological characteristics of MPs from different groundwater depths or sampling times at the same sampling point were considered different sample data. We constructed a groundwater MP dataset consisting of 386 records that included sampling site base information, MP abundance, and morphological characteristics (Tables S1-S6).

2.2. Data classification/grouping

Data classification and grouping followed dimensional scaling. Data grouping is a common method used in meta-analyses (Chaplin-Kramer et al., 2011), and the appropriate selection of data ranges produces more robust results (Desquilbet et al., 2021). Drawing upon the data regarding the distribution of MPs in groundwater, we undertook an exhaustive classification of groundwater type in full data scale, classifying data as originating from closed groundwater (CG) or open groundwater (OG) according to whether groundwater was sampled from the deep underground, or the surface. Among these, OG included springs and caves, with a total of 52 water samples, and the remaining 334 water samples were CG samples.

The regional scale analysis exclusively focused on MPs within CG, as most samples from OG lacked definitive descriptions of the surrounding environment. CG sampling points were classified into agricultural areas, residential areas, industrial areas, landfills, nature reserves, and unclassified areas of groundwater, with sample sizes of 162, 30, 50, 57, 16, and 30, respectively, using information from the source publication. Among them, nature reserves include estuaries, marshes, forests, and mudflats. Some sampling points are situated at the interface of two land-use types, and these sampling points undergo dual categorization, being assigned to both land-use types.

On a local scale, 6 study areas of 5 publications situated in caves, landfills, agricultural areas, residential areas, and nature reserves were chosen to examine variations in MP abundance with groundwater flow paths. These 6 areas encompassed both OG and CG, which represented a comprehensive range of land-use types discussed in this study. Upstream and downstream of the 6 study areas were delineated. Two of the study areas were around landfills. Considering the south-eastward flow of groundwater, the sampling points to the north-west of landfills were designated as upstream, while points to the south-east were designated as downstream. The remaining four study areas were characterized by non-point surface source contamination, for which the sampling points along the groundwater flow path were connected one by one, starting from the first sampling point through which the water flowed and reaching the last sampling point. Different groundwater flow directions resulted in distinct connections. Within the same connecting line, the connecting distance from the first sampling point to the last sampling point was calculated. Points located in the first 50% of this distance were designated as upstream sampling points, while those in the last 50% were designated as downstream sampling points.

2.3. Statistical analysis

We use a contrasting analysis approach here to calculate and analyze the available data on full data, regional, and local scales, focusing primarily on the abundance, color, shape, and polymer type of MPs in groundwater. Shapiro-Wilk tests indicated that neither the abundance nor the morphological characterization data were normally distributed (Shapiro and Wilk, 1965). Therefore, Mann-Whitney U tests and Kruskal-Wallis tests were used to compare the significance of differences in the abundance and morphological characteristics of MPs between different groups in SPSS 26 software (Kruskal and Wallis, 1952; Mann and Whitney, 1947). Meanwhile, the coefficient of variation (CV) of each groundwater group's abundance was calculated, through the ratio of the standard deviation to the average.

The pollution load index (*PLI*) and the polymer hazard index (*HI*) were employed to assess the risk of MPs in groundwater. *PLI* evaluates the pollution of MPs based on MP abundance in the environment, while *HI* considers the toxic effects of different polymers, calculated as following equations (Eq. 1-3) (Guo et al., 2021; Pico et al., 2021):

$$CF_i = \frac{C_i}{C_0} \tag{1}$$

$$PLI = \sqrt{CF_i} \tag{2}$$

$$HI = \sum_{n=1}^{n} P_n S_n \tag{3}$$

where CF_i represents the MP pollution index at sampling point *i*, which is the ratio of the observed MP abundance (C_i) to the background abundance (C_0). For this study, the background abundance is chosen as the lowest abundance of MPs detected in groundwater (0.001 items/L). Based on the values of *PLI*, the pollution levels in the sampling areas are categorized into low (< 10), medium (10-20), high (20-30), and extremely high (> 30). *HI* calculates the risk for different polymers. P_n denotes the proportion of each polymer in each sample, and S_n represents the hazard score of the corresponding polymers. Polyethylene (PE), polyamide (PA), polyacetal (PAT), polystyrene (PS), polyethylene terephthalate (PET), polyvinylchloride (PVC), polycarbonate (PC), polymethylmethacrylate (PMMA), polyester, polypropylene (PP), polyurethane (PU), and ethylene vinyl acetate/alcohol (EVA/EVAL) have hazard score values of 11, 47, 1500, 30, 4, 5001, 610, 1021, 1, 7384, and 9, respectively (Lithner et al., 2011). The values of *HI* are also divided into four categories: Category I (< 10, low risk), Category II (10-100, medium risk), Category III (100-1000, high risk), and Category IV (> 1000, extreme risk).

On a local scale, we assessed the impact of flow path using 6 study areas which were selected based on the availability of hydrogeological information in original papers. The spatial interpolation of 6 areas was conducted using the inverse distance weighted interpolation method within ArcGIS 10.7 software. Subsequently, MP abundance in the local vicinity of the sampling points were calculated and visualized using the following equations (Eq. 4 and 5) (Philip and Watson, 1982):

$$d_i = \sqrt[2]{(x - x_i)^2 + (y - y_i)^2} \tag{4}$$

$$z = \frac{\sum_{i=1}^{n}\left(z_i \frac{1}{d_i^k}\right)}{\sum_{i=1}^{n}\left(\frac{1}{d_i^k}\right)} \tag{5}$$

Where d_i represents the distance from the i^{th} known abundance point to the unknown point, with (*x*, *y*) and (x_i, y_i) denoting the coordinates of the unknown point and the known point in the two-dimensional plane, respectively. The variable *z* signifies the MP abundance value of the unknown point in the local area, *n* represents the number of known

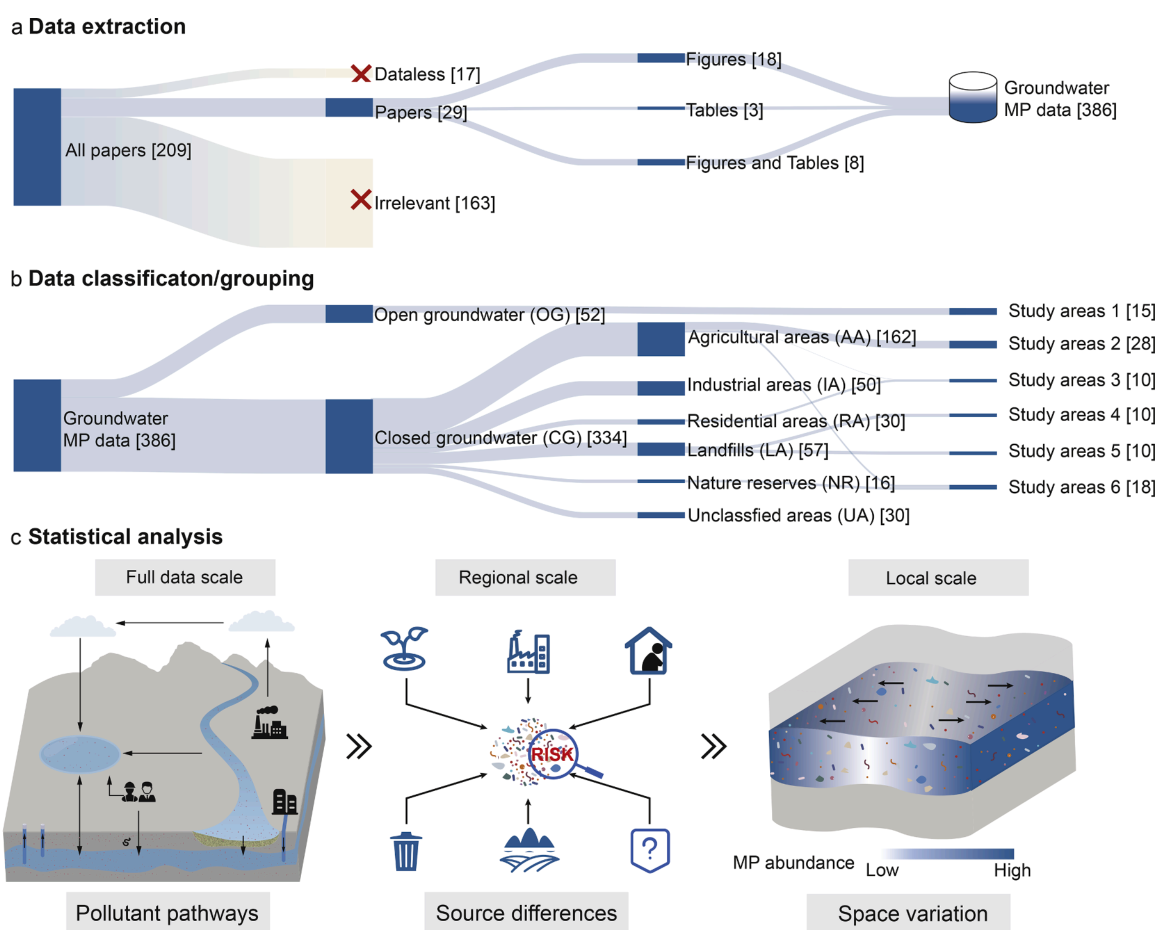

Fig. 1. A contrasting analysis of scales approach for (a) data extraction (values in brackets represent the number of papers with available data for use in this study), (b) data classification/grouping (values in brackets represent the number of samples), and (c) statistical analysis of microplastics (MPs) in groundwater at multiple scales.

points in the local area influencing the unknown point, z_i stands for the abundance value of the i^{th} known point, and k is the power of the Euclidean distance d_i, conventionally set to 2. All results were visualized through ArcGIS 10.7 software and Origin 2021 software. A summary of the framework for data extraction, classification/grouping and statistical analysis used in this study is presented in Fig. 1.

3. Results

3.1. Full data scale results: differences in MPs between OG and CG

The sampling points encompassed within the OG were predominantly situated in the karst landscape regions of North America, Europe, and East Asia (Fig. 2a). MP abundance ranged from 0 to 63.0 items/L with a median (Interquartile Range (IQR)) of 4.4 (0.2, 8.8) items/L (CV = 161.6%) (Fig. 2b). Transparent, multicolored, and blue MPs exhibited higher prevalence in the OG samples, constituting 34.5%, 26.2%, and 15.4%, respectively. Red, black, and grey MPs made up approximately 5% each, with the remaining colors in minimal proportions (Fig. 2c). Fibers (55.5%) were the shape that appeared most frequently in the OG, followed by fragments (23.2%) and films (16.4%) (Fig. 2d). A range of polymer types were identified, wherein PP and PE constituted the predominant types at 34.1% and 29.7%, respectively, followed by PS and PET, with each accounting for approximately 10%. The remaining polymer types collectively represented less than 5% (Fig. 2e). Approximately 30.6% of MP particles were smaller than 0.1 mm, with 69.4% of MPs being 0.1-5 mm (Fig. 2f).

The variation in MP abundance between OG and CG was not significant ($p > 0.05$), but the differences in morphological characteristics were notable. CG had a wider distribution of sampling points spanning North America, Europe, Asia, and Oceania (Fig. 2a). The abundance of MPs ranged from 0 to 6832.0 items/L with a median (IQR) of 2.5 (0.2, 16.0) items/L (CV = 883.5%) (Fig. 2b). Transparent (31.3%), black (22.8%), White (14.8%) and blue (11.5%) were more prevalent colors, followed by red and yellow (Fig. 2c). In terms of shape, fibers and fragments were two common shapes, accounted for 45.3% and 37.6% respectively (Fig. 2d). 15 polymer types were identified, with PP, PA, PET, and PE being the dominant polymers representing 84.0% of the total (Fig. 2e). Most sampling points in CG typically encompassed up to 80% of MPs smaller than 0.1 mm (Fig. 2f).

3.2. Regional scale results: distribution of MPs in CG from different areas

3.2.1. MP abundance and morphological characteristics

The variability in land-use types associated with CG led to an exceptionally high CV in abundance (883.5%). Groundwater around nature reserves and landfills had the highest median (IQR) abundance of MPs at 17.5 (11.8, 35.3) and 13.4 (8.0, 19.1) items/L, respectively, with CV being about 85%. The abundance of MPs in these areas was significantly different from that in agricultural areas ($p < 0.001$). MP abundance in agricultural areas had the widest range (CV = 716.7%), with the highest abundance reaching 6832.0 items/L, but the median (IQR) value was only 1.0 (0.1, 6.2) items/L. The extremely low MP abundance in sampling points located in South Korea greatly reduced the median abundance in agricultural areas. Furthermore, in industrial and residential areas, the median (IQR) abundance of MPs were 4.2 (1.0, 25.7) and 3.0 (2.0, 13.8) items/L, respectively. The CV of abundance in these two areas was 125.0% and 163.4%, respectively (Fig. 3a).

Differences in the morphological characteristics of MPs in groundwater were also detected across the various selected areas. In terms of color ($p < 0.001$), landfills exhibited the highest proportions of white, black, and red MPs at 16.6%, 36.2%, and 15.7%, respectively, with the remaining colors accounting for less than 10%. Nature reserves contained the highest proportions of white, black, and blue MPs at 24.9%,

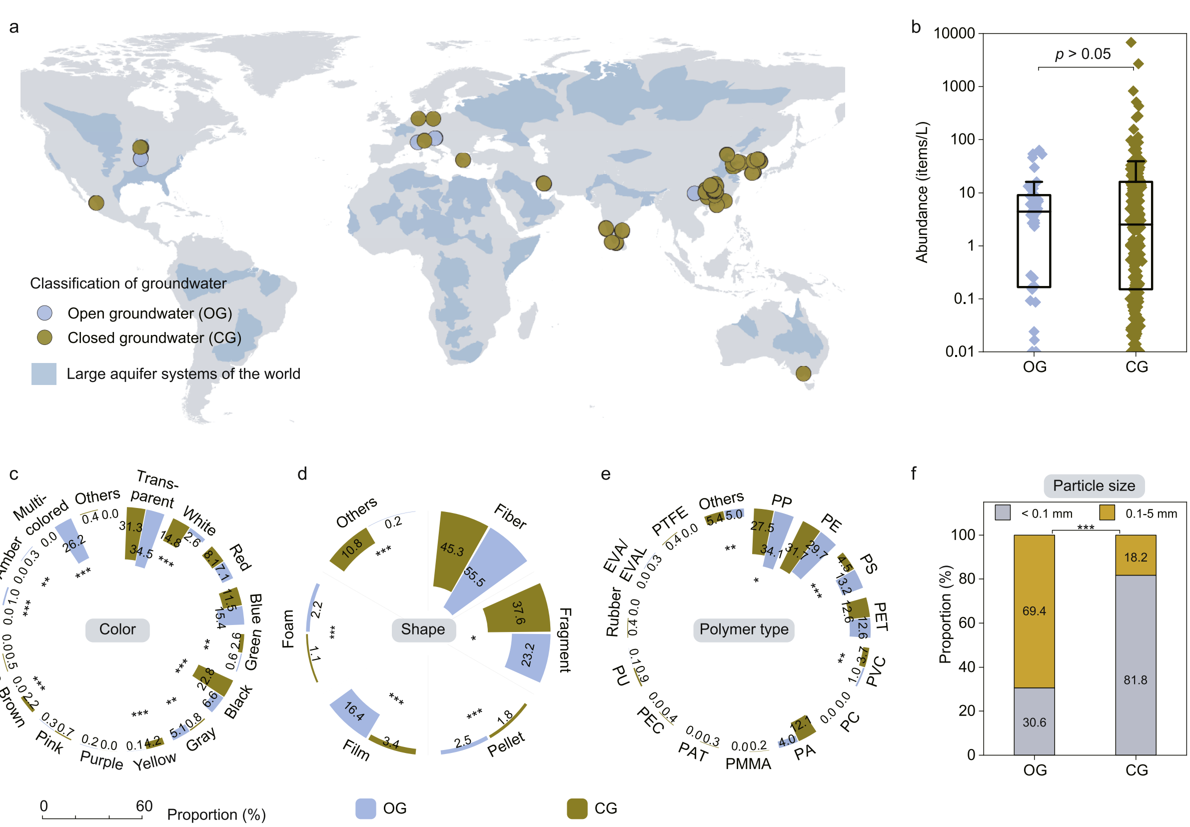

Fig. 2. Global distribution of sampling points (a), differences in MP abundance (b), color (c), shape (d), polymer type (e), and particle size (f) between open groundwater (OG) and closed groundwater (CG). *, ** and *** indicates $p < 0.05$, $p < 0.01$ and $p < 0.001$ respectively.

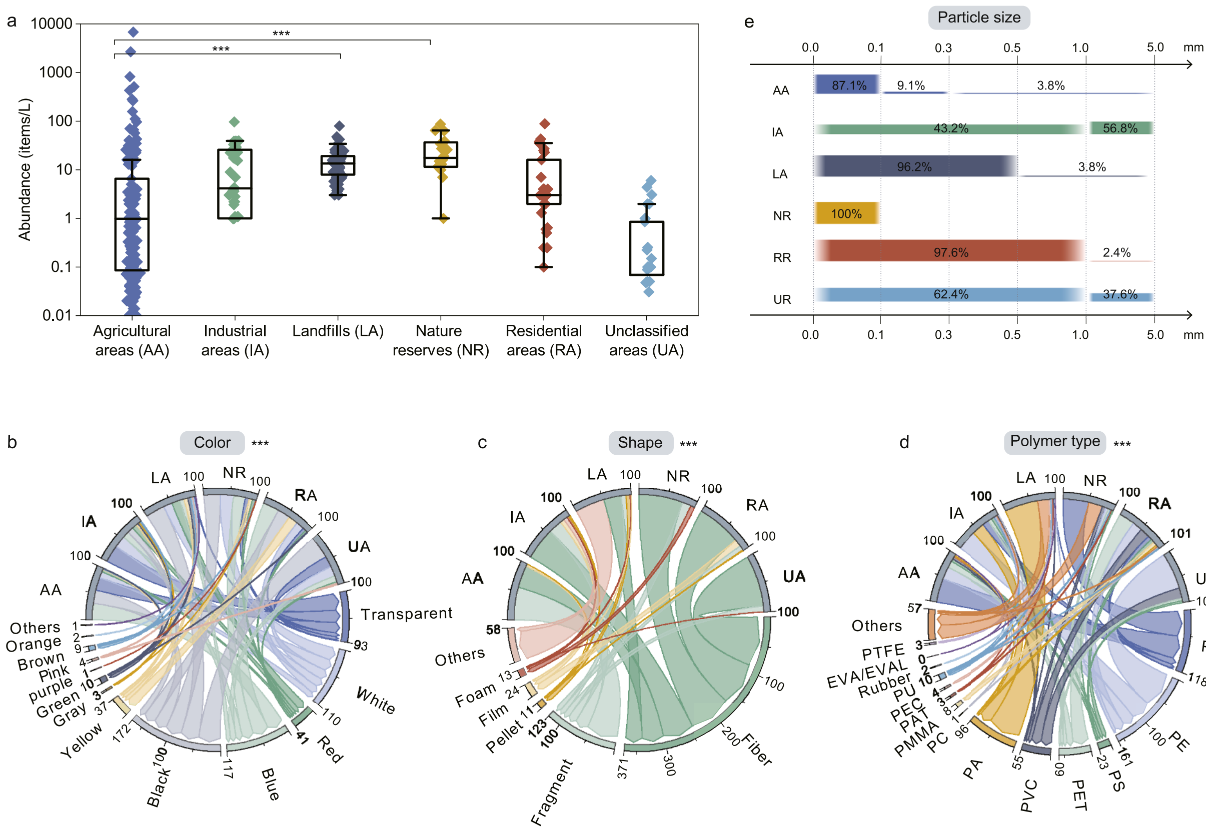

Fig. 3. Differences in MP abundance (a), color (b), shape (c), polymer type (d), and particle size (e) in agricultural areas (AA), residential areas (RA), industrial areas (IA), landfills (LA), nature reserves (NR) and unclassified areas (UA) of closed groundwater. *, ** and *** indicates $p < 0.05$, $p < 0.01$ and $p < 0.001$ respectively. (a) There are significant differences in the abundance of MPs between unclassified areas and other areas, which is not shown in the figure.

31.6%, and 22.7%, respectively. In industrial areas, there was a diverse range of MP colors, but more than half of the MPs identified as transparent and white. Residential areas predominantly featured blue and black MPs, accounting for 31.1% and 24.1%, respectively. Agricultural areas had an abundance of transparent, blue, and black MPs with 24.2%, 31.1%, and 25.6%, respectively (Fig. 3b, Fig. S1).

The differences in MP shapes were evident across distinct areas ($p < 0.001$). Landfills contained 30.2 % fibers, 8.8% fragments, 7.3% pellets, 1.9% foams, 0.2% films, and up to 51.6% other shapes. Only three types of shape, fibers, fragments, and foams, were identified in the nature reserve, accounting for 77.4%, 15.2%, and 7.5%, respectively. Industrial areas exhibited 56.2% fibers, 23.9% fragments, 7.3% films, 3.3% foams, 2.5% pellets, and 6.7% other shapes. Residential areas were dominated by fibers with a high percentage of 78.1%, followed by 12.1% films, 8.6% fragments, and 1.2% pellets. MPs in the agricultural areas were dominated by fragments at 56.7%, followed by 39.3% fibers, 3.8% films, and 0.1% pellets (Fig. 3c, Fig. S2).

The differences in polymer types were also significant ($p < 0.001$). The types of polymers found in landfills were unique compared to other areas, with PA making up the largest proportion (65%). Unique polymers such as polyethylene chlorinated (PEC), rubber, and EVA/EVAL were also identified in landfills. Nature reserves exhibited a relatively high abundance of PP at 50.4%, with 27.3% attributed to other polymers. PE (40.0%), PA (24.5%), and PP (12.5%) were more prevalent in groundwater from industrial areas, with other polymer types accounting for less than 10%. In residential areas, PE (19.8%), PVC (19.4%), and PET (31.0%) were the predominant polymers. In agricultural areas, PP and PE were the most common polymers at 46.7% and 24.3%, respectively, followed by PET at 13.1% (Fig. 3d, Fig. S3).

Different studies had varied categorizations for the particle sizes of MPs. In landfills, most MPs were less than 0.5 mm (96.2%). In nature reserves, the monitored MPs were even smaller, all less than 0.1 mm. In agricultural areas, MPs in the 0-0.1 mm range constituted nearly 90%. In industrial areas, about 43.2% of MPs fell within the 0-1.0 mm range, while in residential areas, MPs within this size range accounted for 97.6% (Fig. 3e).

3.2.2. MP risk in groundwater

According to the *PLI*, 65.0% of the groundwater was extremely polluted. 21.2 %, 9.6 %, and 69.2 % of the OG samples had low, medium, and extremely high pollution levels, respectively. In the CG, different land-use types had inconsistent levels of pollution loads. Sampling points at both landfills and nature reserves have reached extremely high levels of pollution. In agricultural areas, the proportions of groundwater with low, medium, high, and extremely high pollution were 26.2%, 15.2%, 6.7%, and 51.8% respectively. In industrial areas, 78% of groundwater samples exhibited extremely high pollution, with the remaining 22% showing low pollution. Residential areas had 80% of groundwater samples indicating extremely high pollution, with high and medium pollution levels each accounting for only 10% (Fig. 4a). Most sampling points were clustered in Asia, where all samples from China and most samples from India had extremely high levels of groundwater MP contamination (Fig. 4a).

Different types of polymers exhibit varying degrees of toxicity to the environment. In the OG, the proportion of level I, II, and III was 27.6%, 65.5%, and 6.9%, respectively. In the CG, the proportions of groundwater in landfills classified into *HI* risk categories I, II, III, and IV were 4.3%, 73.9%, 8.7%, and 13.0%, respectively. Nearly half of the samples from nature reserves were classified as hazard level I. In agricultural areas, the groundwater samples fell into categories I, II, III, and IV at rates of 36.5%, 33.8%, 23.4%, and 6.2%, respectively, compared to 25.6%, 37.2%, 34.9%, and 2.3% in industrial areas. In residences, 87.5%

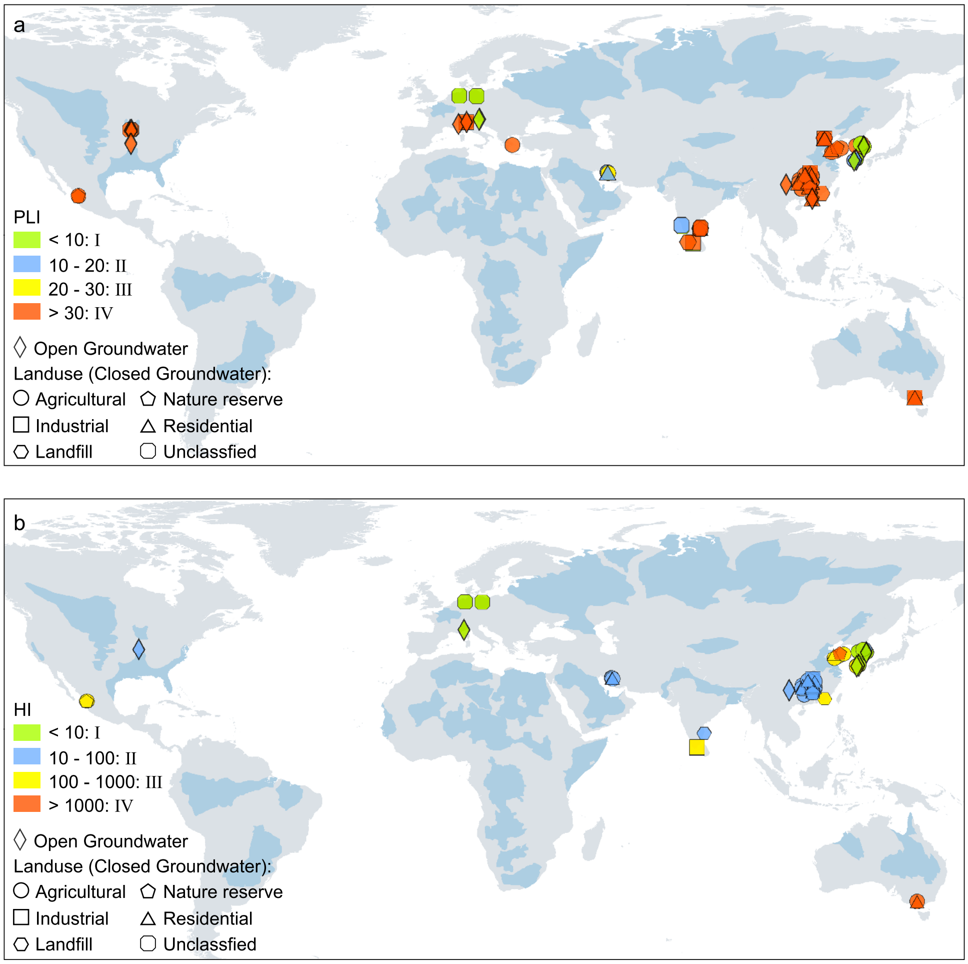

Fig. 4. Risk of MP pollution in groundwater. Pollution load index (*PLI*) (a), Polymer hazard index (*HI*) (b).

of samples were classified as hazard level II, with the remainder falling into categories III and IV (Fig. 4b). Additionally, 74.4% of samples provided both abundance and polymer types of groundwater MPs. Areas with high *PLI* values in groundwater were common, but the associated *HI* were not elevated, particularly in samples from Asia (Fig. 4b). The levels of MP pollution do not correlate with polymer risk, highlighting the importance of considering both the abundance and polymer types when studying MPs.

3.3. Local scale results: sources and movement of groundwater MPs from various land-use types

Analysis of MPs on a local scale provides further insights into the spatial distribution of groundwater MPs under the influence of surface contamination sources. In both OG and CG, MPs exhibit a proclivity for aggregate downstream of the sampling area due to the water flow direction (Fig. 5). For the OG, a particular area situated in karst caves in Guizhou (China) was selected. Except for an upstream groundwater sample that was likely recharged by a lake (and therefore influenced by atmospheric deposition and consequently exhibiting heightened MP abundance), the remaining sampling points were situated within an underground karst river. The point furthest downstream within the sampling area exhibits the highest abundance of MPs (Fig. 5a).

Within the CG, the abundance of MPs exhibited variations spanning one to three orders of magnitude among different land-use types. Regardless, the distribution of MPs remained consistently aligned with the prevailing flow direction. Notably, two landfills situated in India emerge as the most contaminated areas, featuring wastewater treatment plants as additional pollution sources alongside the impact of the landfills. Wastewater treatment plants contribute to the detection of MPs in the adjacent sampling points, therefore sampling points nearest to this place were not considered. Upon mitigating the influence of this contamination, our analysis revealed that, in the context of the southeastward groundwater flow trend in these areas, downstream locations from the landfill exhibit higher MP abundance compared to upstream locations. Moreover, the southeast direction was identified as being more prone to MP accumulation compared to other directions (Fig. 5b and c). In contrast, agricultural, residential areas, and nature reserves manifested a narrower range of overall MP abundance when there were no other pollution sources present. Even when separated by distances exceeding ten kilometers, MPs demonstrate a proclivity to aggregate downstream in concordance with the groundwater flow field (Fig. 5d-f).

The flow of groundwater has led to variations in the morphological characteristics of MPs between the upstream and downstream sections of the sampling area (Fig. 5g-j). In the downstream, there was a higher richness of MP colors compared to the upstream (Fig. 5g). In terms of shapes, from the upstream to the downstream, there was a decrease in the percentage of fragments from 50.78% to 30.49% ($p < 0.05$), accompanied by an increase in the percentage of pellets from 1.85% to 8.47%, and fibers remained basically unchanged (Fig. 5h). Regarding polymer types, there was a decline in the presence of PP and PE in the downstream compared to the upstream of the sampling area, while PA in the downstream was on the rise, but none of the trends were significant (Fig. 5i). Additionally, the particle size of MPs showed little variation ($p > 0.05$), with sizes smaller than 0.1 mm and between 0.1-5 mm accounting for approximately 60% and 40% respectively, regardless of the

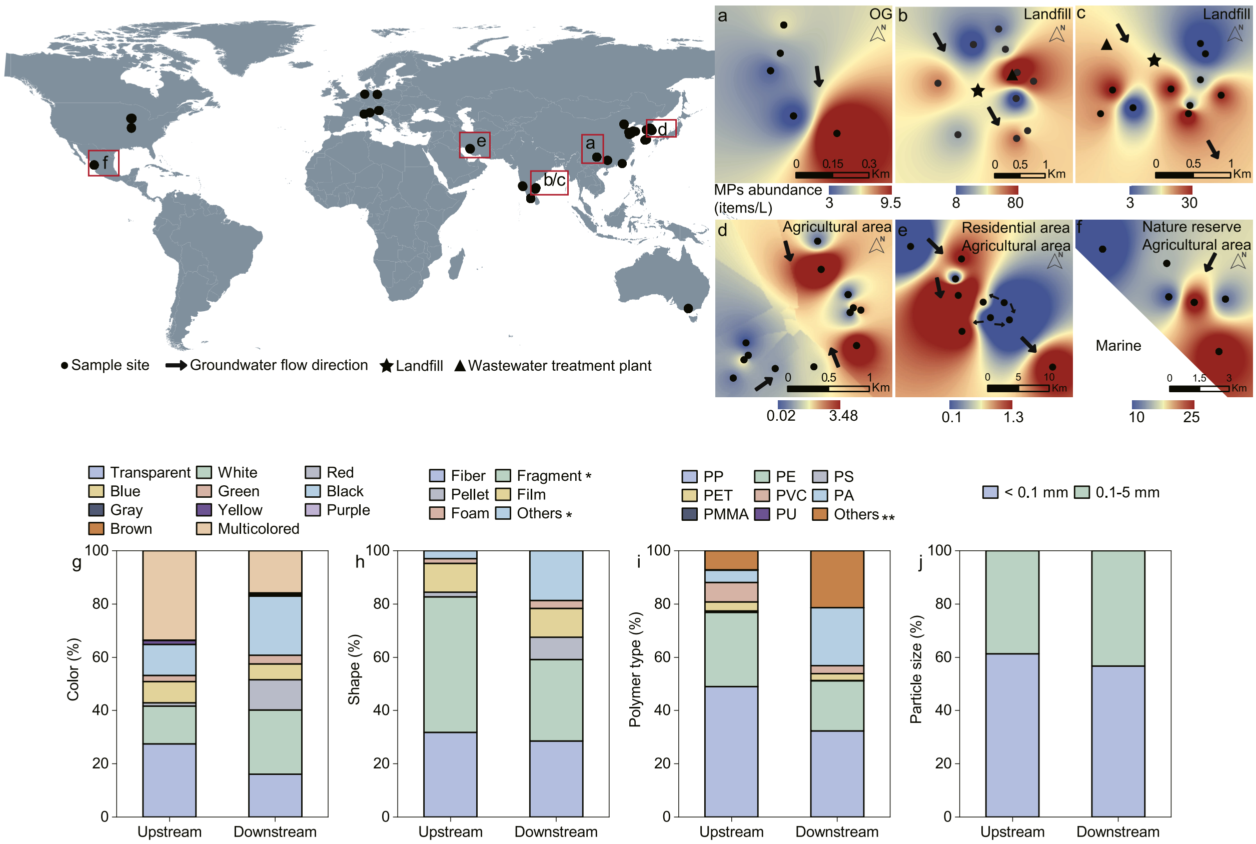

Fig. 5. Changes in MP abundance with groundwater flow direction (a-f) and differences in MPs color (g), shape (h), polymer type (i), and particle size (j) between upstream and downstream of six study areas. * and ** indicates $p < 0.05$ and $p < 0.01$ respectively.

location upstream or downstream (Fig. 5j).

4. Discussion

4.1. Overall occurrences

Studies, although limited, have shown that groundwater contamination with MPs was higher than anticipated. OG, which are natural groundwater discharge points, are more susceptible to evapoconcentration of pollutants (particularly under drought conditions; van Vliet et al., 2023) and the accumulation of surface-borne contaminants in dissolved and particulate forms (Goeppert et al., 2020). However, probably due to the small sample size of the OG, this difference is not significant in abundance. In terms of morphological characteristics, the openness of OG yielded a more diverse array of MP colors, albeit with a lower abundance of polymer types compared to CG, potentially a result of more sampling points and land-use types encompassed within the CG. The marginally higher particle size of MPs in the shallower OG compared to the deeper CG can be attributed to reduced filtration through aquifer materials as well as the accumulation of MPs transported as aerosols or runoff into groundwater-fed springs. Soil and rock pores form continuous channels that range in size from nanometers to centimeters, which regulate the size of MPs that can pass through them and restrict the entry of larger-sized MPs into groundwater (Viaroli et al., 2022).

Elucidating the pathways through which MPs disperse into CG and OG is crucial for tracing the occurrence of contamination and deciphering the global fate of MPs. We hypothesize that the primary reasons for MPs occurrence in OG are surface runoff and atmospheric deposition, consistent with aquatic bodies like rivers and lakes. In addition, human activities such as waste disposal and the exchange of water with underground aquifers are also pollution pathways of MPs in OG. However, CG has little or no exposure to the atmosphere and most of the MPs in it are subsamples of surface MPs filtered through the soil. Human activities (e.g., agricultural cultivation, and waste dumping) allow MPs to accumulate in the soil and reach groundwater through surface water infiltration and soil faunal transport (Guo et al., 2020). At the same time, direct downward injection of reclaimed water may also lead to MP contamination in aquifers (Hartmann et al., 2021) (Fig. 6). However, MP contamination in CG from these factors is reduced compared to OG due to the compartmentalizing influence of soil. These transport pathways can be combined with the global transport of MPs at large scales to guide the delineation of the global MP cycle.

4.2. Pollution sources

The influence of the different surrounding land-use types on the abundance of groundwater MPs is direct and strong, particularly noticeable between landfills, nature reserves, and agricultural areas (Fig. 3b). The high abundance of MPs in landfill groundwater can be attributed to landfills acting as sources and sinks for MPs, with approximately 5,000 metric tonnes of plastic waste accumulated here or in the natural environment as of 2015 (Geyer et al., 2017). In nature reserves, the high level of MP pollution may be attributed to most sampling sites being coastal, facilitating the accumulation of MPs from both terrestrial and marine origins (Ouyang et al., 2022). In contrast, agricultural areas show a greater range in MP abundance but generally have lower median levels (nature reserve median = 17.5 items/L, landfill median = 13.4 items/L, agricultural median = 1.0 items/L), likely due to plastic pollution being localized. Although the use of mulches, wastewater irrigation, and biosolid fertilizers leads to residual MPs in soil (Weithmann et al., 2018; Khalid et al., 2023), there are regions where these initiatives are lacking or prohibited, such as South Korea, where biosolid fertilizers are prohibited, resulting in extremely

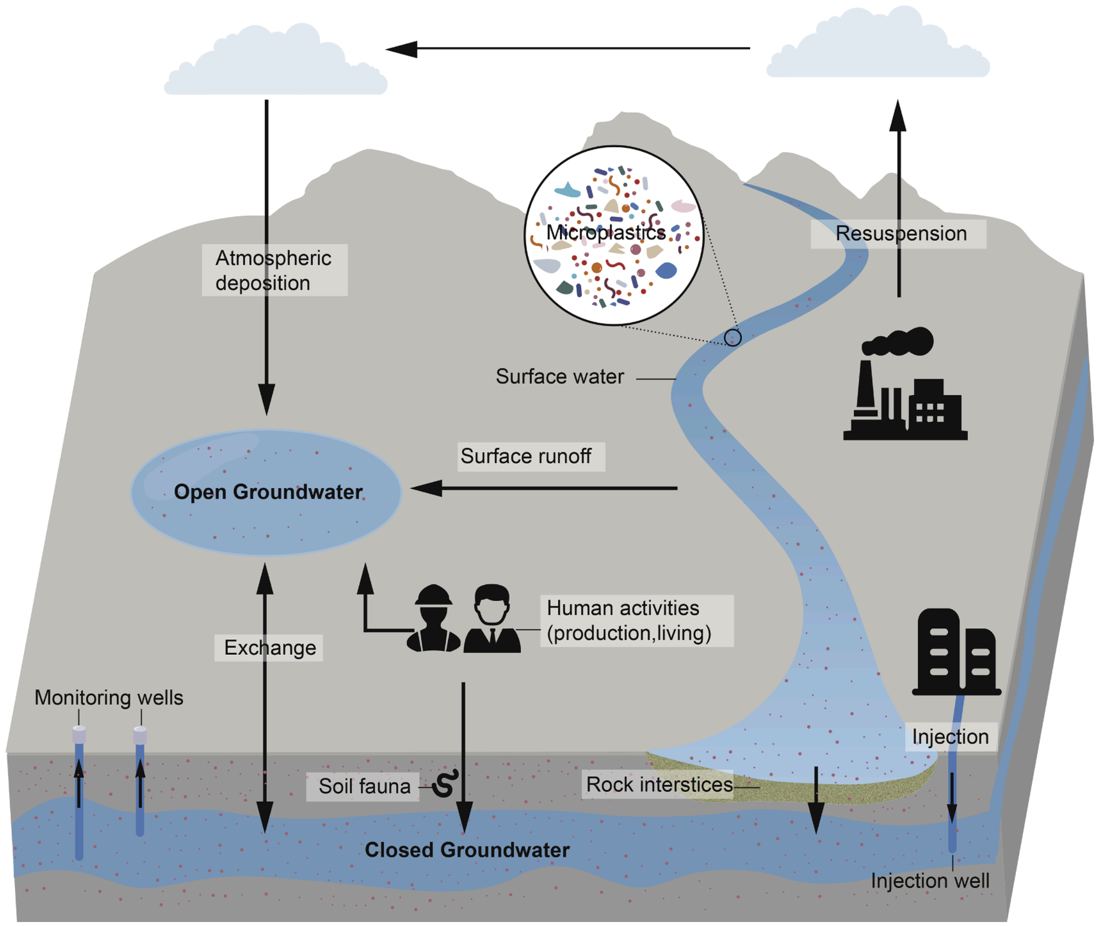

Fig. 6. Main pathways of MPs into open groundwater (OG) and closed groundwater (CG).

low MP abundance in agricultural areas (Jeong et al., 2023).

Variations in land-use types also manifest in MP morphological characteristics. For instance, transparent MPs were significantly more prevalent in the groundwater of agricultural areas than in landfills ($p < 0.001$), likely due to the predominant use of transparent mulch in agriculture. Concurrently, fragments, along with PP and PE, were the primary shapes and polymer types after the degradation of mulches (Khalid et al., 2023), their high proportion in agricultural soils also leads to an increased abundance of these specific forms of MPs in groundwater, significantly exceeding that found in landfills ($p < 0.001$). The MP particle sizes in groundwater from agricultural areas, landfills, nature reserves, and residential areas were predominantly less than 0.1 mm, except for industrial areas (Fig. 3e). In industrial areas, where plastic-related industrial production is a major contributor to regional MP load (Long et al., 2019), we hypothesize that a higher proportion of primary MPs contributes to larger detected particle sizes here compared to others.

MPs in groundwater present a high level of ecological risk across various land-use types, with overall contamination levels exceeding those of reservoir waters, which are also sources of drinking water (Guo et al., 2021). Furthermore, the geographic clustering of pollution is notable, especially in China. The policies on plastic manufacture and usage, as well as human habits in the region, can influence the background pollution levels of environmental MPs (Knoblauch and Mederake, 2021; Munhoz et al., 2023). In terms of polymer hazard, the environmental toxicity of PVC, PMMA, and PAT is much higher than that of other types (Lithner et al., 2011). Groundwater in residential areas containing these polymers warrants long-term attention. The ecological risks associated with nanoplastics are higher than those of MPs (Jeong et al., 2016), and the size distribution of MP particles has been shown to follow a power law with a negative exponent (Koelmans et al., 2022). Therefore, the actual ecological risk of MPs in groundwater may be higher than assessed in this study.

4.3. *MP flow*

The potential alterations in MP abundance and morphological characteristics induced by groundwater flow represent a significant concern, particularly following the elucidation of the regional contamination severity. The hydrophobic properties of MPs reduce concerns about their mobility, however, when some compounds aggregate with MPs, hydrophobicity decreases and mobility increases (Jiang et al., 2022). Currently, the transport distance of MPs in groundwater is still unknown. This is due to low availability of groundwater MP data, which is likely limited by the complexity (requirement for bores to be available) and time-consuming nature of groundwater sampling, and the high costs of MP analysis. Similar to surface water, alterations in groundwater levels and variations in flow rates likely alter the rate and extent of MP transport (Drummond et al., 2022). However, the lack of comprehensive hydrogeological information in most sampling areas makes practical validation of this difficult.

The flow of groundwater does not give each MP an equal opportunity to move, as factors such as shape, density, and size contribute to variations in travel distance. Studies have shown that different shapes of MPs have high to low deposition rates in rivers as fragments, fibers, and pellets (Hoellein et al., 2019). Pellets, characterized by smoother surfaces, experience less resistance in water flow and tend to accumulate downstream. In contrast, fragments face greater resistance due to their irregular shapes and larger surface areas. Thus, similar to rivers, fragments are more likely to be deposited instead of moving with the current, leading to a decrease downstream. The sedimentation patterns of fibers differ from those of fragments and pellets, including horizontal, inclined, and vertical settlement (Dai et al., 2024). Their elongated and slender characteristics allow them to twist into various shapes within the groundwater, positioning their transport distances between those of fragments and pellets (Hoellein et al., 2019). In terms of polymer type, although less dense MPs are thought to be transported over longer distances (Hoellein et al., 2019), our findings reveal a decrease in the proportion of PP and PE downstream, which have lower densities than water, whereas PA, which has a relatively higher density, increased as a proportion, although these differences were not statistically significant. This result may be influenced by the porous characteristics of groundwater, where shape impacts on travel distance might overshadow the differences in polymer types. For instance, at sampling points predominantly in agricultural areas, PP and PE, prevalent in mulches, may be present in groundwater as fragments, while PA is more likely to be in the form of fibers. Moreover, inconsistent land use between upstream and downstream areas can affect the MP characteristics at sampling points. MPs of different colors should theoretically have an equal probability of being transported by groundwater flow. A richer variety of colors was detected downstream of the sampling area than upstream, which may be attributed to the fact that the contamination in the downstream area is a cumulative result of the transport of groundwater from the upstream area as well as infiltration from the downstream surface sources. The size variation of MPs between upstream and downstream was not significant, potentially due to aquifer pores, such as fractured rock, which might be relatively homogeneous in nearby areas, thereby promoting a uniform distribution of similar-sized MPs within the water body.

4.4. Challenges and perspectives

MP contamination of groundwater is a pressing concern as groundwater systems are likely to become important sinks of MPs. Our study identifies MPs in 29 cities' aquifers across the world where groundwater MP data is available and summarizes the current status of groundwater MP contamination and potential risk areas. However, we acknowledge that due to the high variability of groundwater MP abundance, the limitation in sample size for this study, resulting from low numbers of available groundwater MP data in the literature could lead to biases in our results. To assess the extent to which this may be the case, we used an incremental sampling method on MP abundance data to determine the median change rate. We identify that the median change rate in MP abundance fluctuates greatly with increasing sample size, with a marked improvement in the stable of change rate once the sample size reaches approximately 180, stabilizing at ±30% (Fig. S4). The monitoring of MPs in groundwater is often unevenly distributed, predominantly in regions with significant human footprints but scarce groundwater resources (Fig. 2a). In contrast, field monitoring of MPs in groundwater-rich regions is limited. To better estimate the abundance and distribution of MPs in groundwater, it is necessary to expand field monitoring, particularly in groundwater-rich regions where human activities, such as landfills and agriculture, are intensive (Rochman, 2018). Additionally, we recommend enhancing data comprehensiveness (e.g., complete sampling site information and MP morphological data) and promoting open data sharing (Musen, 2022) to strengthen the exchange and integration of groundwater MP detection results worldwide.

A significant challenge encountered during data collection was the lack of standardized protocols for collecting MPs from environmental samples. The abundance of MPs is influenced by particle size (Koelmans et al., 2022). The variability of sampling and identification instruments affects the range of MP particle sizes detected and the precision of the results (Prata et al., 2019). A review of the methods used to identify MPs in groundwater includes Fourier-transform infrared spectroscopy (FTIR), Raman spectroscopy, near-infrared spectroscopy, optical stereo microscopy, electron microscopy, and gas chromatography-mass spectrometry. Many studies have utilized combinations of microscopy with FTIR (62.1%) or microscopy with Raman spectroscopy (17.25%) to enhance the accuracy and efficiency of identification. This combined analytical approach should be advocated. Meanwhile, it is noted that FTIR is limited to a minimum particle size of 20 μm, whereas Raman spectroscopy has a higher resolution and can detect MPs as small as 1 μm (Tirkey and Upadhyay, 2021). Therefore, when conducting combined analyses, we recommend opting for Raman spectroscopy. Additionally, to minimize environmental and procedural contamination impacts on results, experiments should be conducted while wearing cotton clothing, avoiding the use of plastic utensils, and establishing both positive and negative controls.

Regular monitoring and standardized detection protocols are essential for accurately assessing MP pollution in groundwater. It is also necessary to implement measures to reduce MP contamination. Given that groundwater MP pollution is exogenous, controlling surface sources of pollution is particularly crucial. For example, enhancing the recycling and reuse of plastic products (Kwon, 2023), reducing the accumulation of plastics in landfills, and improving landfill treatment facilities are vital steps. In agricultural areas, the use of biodegradable films instead of plastic mulches can decrease the accumulation of plastics in the soil (Serrano-Ruiz et al., 2021).

5. Conclusions

We utilise a CAS approach to systematically understand the distribution differences of MPs in groundwater at multi-scales. A total of 386 groundwater MP samples from 29 cities were integrated. At the full data scale, the median abundance of MPs in OG and CG was 4.4 and 2.5 items/L, with no significant difference in abundance, but significant differences in colors, shapes, polymer types, and sizes. The CG exhibited a high coefficient of variation in abundance, possibly due to differences in land-use types at sampling points. Sampling points in nature reserves and landfills exhibited extremely high MP pollution loads, with 3-6 times higher MP abundance than industrial and residential areas. Agricultural areas had an extremely high coefficient of variation in abundance, but the median was only 1.0 items/L, which was significantly different from nature reserves and landfills. Different anthropogenic activities also led to regional uniqueness in the morphological characteristics of MPs. Through the comparison of upstream and downstream MPs, we found that the direction of groundwater flow propels the movement of MPs, inducing their downstream aggregation, but each particle does not travel equidistantly. The contamination risk of groundwater MPs needs to be assessed from multiple perspectives, including abundance and polymer type. The results indicate that groundwater MP pollution is mostly regional, and its severity should not be underestimated. In light of these results, we urgently call for an expansion of research on groundwater MPs, especially in groundwater-rich regions with an intensive human footprint, and the adoption of standardized detection protocols for better analysis of global groundwater MP pollution in the future.

CRediT authorship contribution statement

Yu-Qin He: Writing – original draft, Visualization, Methodology, Investigation, Data curation. **Liza K. McDonough:** Writing – review & editing, Conceptualization. **Syeda Maria Zainab:** Writing – review & editing. **Zhao-Feng Guo:** Writing – review & editing, Funding acquisition. **Cai Chen:** Visualization, Methodology. **Yao-Yang Xu:** Writing – review & editing, Supervision, Resources, Project administration, Funding acquisition, Data curation, Conceptualization.

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.

Data availability

I have shared my data at the Attach File step

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.watres.2024.121808.

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