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State of knowledge and future research needs on microplastics in groundwater

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ABSTRACT

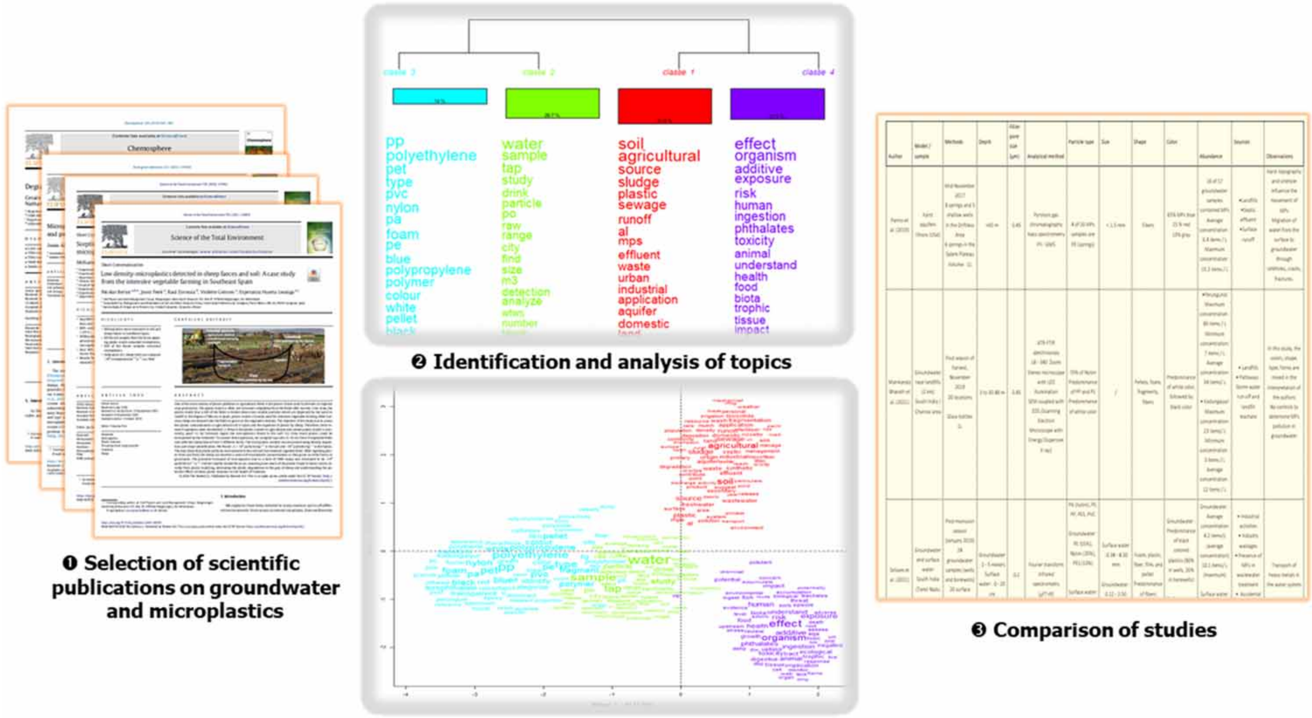
Microplastics (MPs) are widespread in aquatic and soil environments. This study targets the issue of MPs' transfer from soil to groundwater. Scientific papers were collected and analyzed using a text-mining approach that classifies text segments. This allowed the identification of four research topics and the organization of the results into a summarizing table. Those four topics are sources of groundwater MPs, main types of MPs (physico-chemical properties, polymer units, shapes, and size), human exposure (mainly drinking water), and potential environmental and human effects. Compared to the research of MP on aquatic or soil compartments, scientific data on MP in groundwater are less substantial. Current results show a divergence due to differences in context (alluvial aquifer, fractured rock aquifer, karst aquifer, etc), collecting, sampling, and analytical methods. This divergence requires further research with standardized analytic protocols and reference materials. The associated research gaps were identified by using the same approach. The following five topics emerged: (1) the transfer of MPs from soil to underground, (2) the contribution of groundwater to drinking water microplastic pollution, (3) the interaction with other contaminants, (4) the human and environmental effects, and (5) the protective and remediation solutions.

Key words: environment, groundwater, health, microplastics, text mining

HIGHLIGHTS

- A text-mining tool was used to distinguish sources, types, exposure pathways, and effects of microplastics (MPs).
- There is a lack of common criteria to compare the behavior of MP in groundwater and their potential environmental and health impacts.
- There is a need for standardized sampling, extracting, and analytical methods on microplastics in groundwater and biological matrices.

GRAPHICAL ABSTRACT



ABBREVIATIONS

BPA	bisphenol A
EDCs	endocrine disrupting chemicals
HDPE	high-density polyethylene
LDPE	low-density polyethylene
MP/MPs	microplastics
NP/NPs	nanoplastics
NOAA	National Oceanic and Atmospheric Administration
PA	polyamide
PBT	polybutylene terephthalate
PE	polyethylene
PES	polyester
PET	polyethylene terephthalate
PS	polystyrene
PU	polyurethane
PVC	polyvinyl chloride
PVF	polyvinyl fluoride
PCB	polychlorinated biphenyl
PAH	polycyclic aromatic hydrocarbon
ECUs	elementary context units

1. INTRODUCTION

Plastics have been widely used, usually as single-use, for their physical–chemical properties, and their low cost. Their production reached about 370 million tons in 2019 (Plastics Europe 2020). It is estimated to reach 33 billion tons in 2050 from which 10% would be transported into the ocean (Rochman *et al.* 2013). Plastics are usually classified into two types: primary plastics from industrial products and secondary plastics from the fragmentation of macroplastics (UV degradation, abrasion, biodegradation, etc) (Syberg *et al.* 2015).

Usually, the term microplastics (MPs) translate to small plastic fragments. Specifically, MPs are considered to be particles smaller than 5 mm in size, while nanoplastics (NPs) are generally defined by a diameter of less than 100 nm (EFSA Panel on Contaminants in the Food 2016). However, there is an absence of consensus regarding the definition as introduced by Hartmann *et al.* (2019). National Oceanic and Atmospheric Administration (NOAA) proposes a broad definition of MP as polymers with a size lower than 5 mm not distinguishing them from NP (Arthur *et al.* 2009). The absence of consensus on MP size makes it difficult not only to define standardized protocols for collecting, sampling, and analyzing MP but also to compare the results of (eco)toxicological impacts of MP between different studies. Further to the size consensus, there are also different morphologies of plastics: fibers, spheres, fragments, and films. As for the size, the morphology of MP can influence the media properties (de Souza Machado *et al.* 2019). They can also determine their ability to interact with living organisms and to accumulate within them (Prata *et al.* 2021). These effects are further emphasized by the fact that MPs have low degradation and a wide dispersion through all environments (Zhang *et al.* 2021). Further to the effects of MP components, due to their high hydrophobicity, charge, and reactive surface, MPs can be also vectors of other contaminants such as polychlorinated biphenyls (PCBs), heavy metals, polycyclic aromatic hydrocarbons (PAHs), acting as 'Trojan Horses'.

The three main plastics produced and most often identified in environments are polyethylene (PE), polystyrene (PS), and polypropylene (PP) (Brachner *et al.* 2020). Besides, polyethylene terephthalate (PET), a type of polyamide, is often found in textiles and in washing machine wastewater, and usually ends up in the waterways (Yang *et al.* 2019). These types are also found at high altitudes, at the poles, in oceans, soils, groundwater, drinking water, and organisms (Abbasi *et al.* 2019). The effects of these MPs on ecosystems differ between MPs (Zhou *et al.* 2020). For instance, PE and polyvinyl chloride (PVC) increased the metabolic activity of microorganisms, whereas PS and PET did the opposite (Fei *et al.* 2020).

MPs in soils are less studied compared to an aquatic environment like oceans and freshwater (Qin *et al.* 2020; Zhang *et al.* 2020; Zhou *et al.* 2021a). However, the major sources of MP, like sewage sludges, agricultural mulching, and tire abrasion on highways, are located in continental areas. Indeed, MP occurrence in soils is estimated to be 4–23 times higher than in the ocean (Horton *et al.* 2017). When soils are completely saturated by rainfalls, additional water goes down the groundwater to replenish it. When soils contain pollutants, they can partly be transferred to groundwater. Groundwater quality thus depends on soil quality (Arias-Estévez *et al.* 2008; Keesstra *et al.* 2012) but also on soil leaching ability (Djodjic *et al.* 2004). Groundwater is an integral part of the hydrological cycle and an important source of drinking water (Song *et al.* 2020). Some regions, such as northwestern Germany, are supplied with drinking water only from groundwater (Mintenig *et al.* 2019). Groundwater can be then a source of MP for human exposure through drinking water. Only a few studies investigated exclusively the presence, abundance, and transfer pathways of MP in groundwater. Regarding the study of MP in freshwater, there is a lack of research regarding human exposure from groundwater MP compared to surface waters (Re 2019).

The aim of this review is to establish a state of knowledge on MPs in groundwater, based on two approaches: (1) the elaboration of a bibliographic database applying the PRISMA methodology developed by Moher *et al.* (2009) (58 scientific papers have been selected, prior to 24 March 2021); (2) the data classification on the state of the art based on lexicometric analysis on the scientific papers. Lexicometric analysis is a text-mining technique belonging to statistical analysis, here used to identify, in scientific papers, different topics related to MPs in groundwater. The used tool was IRaMuTeQ version 0.7 alpha 2 (Interface de R pour les Analyses Multidimensionnelles de Textes et de Questionnaires) (Ratinaud 2014). This tool organizes the word content of the scientific papers into different classes or utterance use contexts based on Reinert's hierarchical descending classification (Reinert 1983). The results are the distance χ^2 between clusters, the frequency, and the contingency coefficient. Besides, in order to visualize the classes, a Correspondence Factorial Analysis (CFA) is performed, based on a contingency table and the distance between the words. The analysis makes it possible to represent the oppositions and links between words and classes (Teil 1975). Moreover, for each cluster, the similarity analysis (ADS) developed by Ratinaud & Marchand (2012) was used to highlight lexical communities. Similarity analysis is a relevant analysis to determine the lexical relationships between each term and to reveal how the different communities are related to the main topics of the relevant class. For more information concerning the software, refer to Supplementary Material.

The four classes obtained are related to (1) sources of groundwater contamination from MPs, (2) the human exposure pathways of MPs, (3) the types of MPs, and (4) the potential effects of MPs. In addition, another statistical analysis was conducted to highlight the research gaps and emerging concerns related to MPs in groundwater.

2. RESULTS

2.1. Elaboration of the database

The literature search based on keywords resulted in 4,950 articles. After removing the papers that were not peer-reviewed, the symposia or seminars or publications solely dealing with MP in the aquatic environment, and the papers lacking references on groundwater, the database contained 58 remaining articles that were included in our meta-analysis. We proceeded to a lexicometric analysis which is a text-mining technique comprising statistical analysis. It consists in studying the words used in 40-word segments or elementary context units (ECUs) in terms of frequency and associations. The objective is to organize the word content of large texts into different classes or utterance use contexts. In these articles, the ECUs dealing with current knowledge and the ones dealing with research gaps were extracted and constituted two different databases. For more information, the methodology is included in the annex. For more information, the methodology is included in Supplementary Material.

2.2. Classification results on the current knowledge

The 58 articles contained 5,472 ECUs, including 3,757 forms and 39,150 total occurrences dealing with current knowledge on groundwater MP.

Reinert's classification resulted in four different classes as represented in Figure 1. In this figure, the first words in each column are the active forms with the highest χ^2 in the class that represents them. In other words, the higher the χ^2, the more discriminating the form and the stronger the link between the form and the class.

Class 1 (in red) contains the most information (31.8%), followed by Class 4 in purple (27.5%), Class 2 in green (26.7%), and Class 3 in light blue (14%). The first class is the main class which reveals the meaning of the body of the text. It is worth noting that it is not necessarily the most voluminous in Iramuteq analyses. The fact of obtaining these different classes contributes to the understanding and interpretation of the terms mainly approached by the researchers. As a result, each part corresponds to a different lexical term and refers to specific publications and authors (Figures 1 and 2).

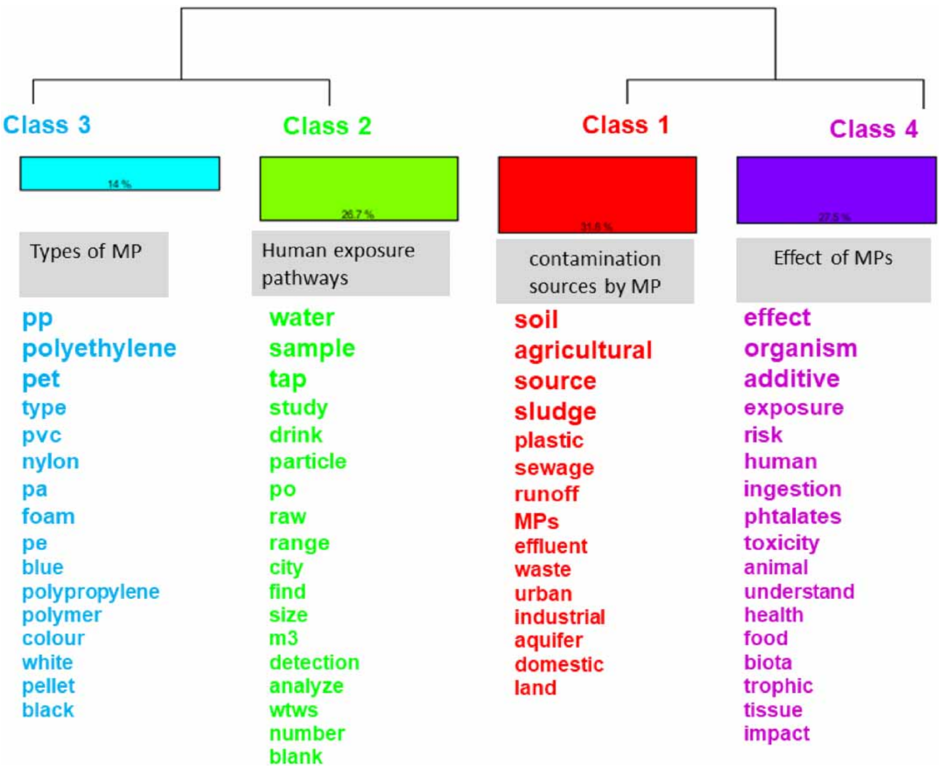


Figure 1 | Dendrogram related to the ECU on groundwater MP (only significative words are shown for each class: $p < 0.001$). Words with the higher χ^2 association to the cluster are with a larger character size. Please refer to the online version of this paper to see this figure in color: http://dx.doi.org/10.2166/wh.2022.048.

Class 1 is marked by the active forms 'soil', 'agricultural', 'source', 'sludge', 'plastic' which refer to the contamination sources by MP. It appears that agricultural, industrial sources, and water runoff are the few studies sources of MP pollution in groundwater.

Class 4 is scored by the active forms 'effect', 'organism', 'additive', 'exposure', 'risk'. This class is related to the effects of MP on soil functions, microbiota, plants, and human health.

Class 2 is branded by the active forms 'water', 'sample', 'tap', 'drink'. This class is related to the human exposure pathways of MP deriving from groundwater.

Class 3 is tagged by the words 'PP' for polypropylene, 'polyethylene' (PE), 'PET' for polyethylene terephthalate, 'type', 'PVC' for polyvinyl chloride, 'nylon'. This lexical field is related to the types of MP which have been detected in groundwater and tap water.

Besides, the Factorial Correspondence Analysis allows us to visualize the four classes, with the corresponding forms. It completes the hierarchical descending classification by representing the class organization along the first two-dimensional axes (Figure 2).

These figures show for each class, the main discriminants ECUs. The figures indicate that axis 1 (+) is discriminated by Class 4 on the effects (also discriminating axis 2 (−)) and by Class 1 on the sources (also discriminating axis 2(+)), whereas axis 1(−) is discriminated by Class 3 on the MP types. Class 2 on the exposure is less discriminant than the other since it is more concentrated around 0.

In other words, the active words are gathered in distinct and discriminating themes. Except for Class 2 and Class 3 dealing with the types of plastics and exposure of the human population to MPs. The question is often raised as to what type of MPs are analyzed and detected in the groundwater and whether there is a risk of exposure to these MPs through drinking water.

Besides, each topic of active words is associated with several publications and authors. The χ^2 statistical test is applied to categorize the publications and authors according to their significance for a specific lexical field.

Thereby, for each class, the main characteristic ECUs could be visualized to better understand the content of each class and identify the most relevant publications to gather the main outcomes in a synthetic table (Table 1).

2.2.1. Sources of MP (Class 1)

Few studies have analyzed the contamination sources of MPs in groundwater, usually because soil is supposed to filter and trap pollutants, thus, limiting the MP in groundwater. The groundwater is then supposed to not be a microplastic reservoir.

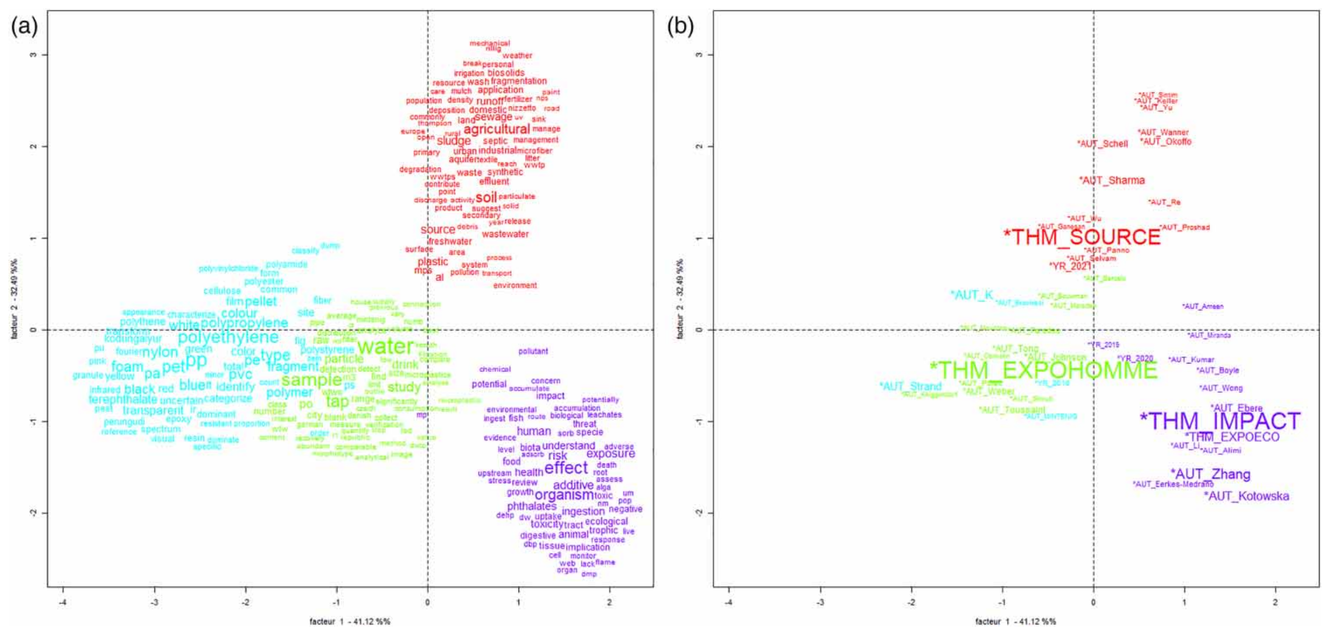


Figure 2 | (a) Distribution of active words along the two first axes of the correspondence analysis (Class 1 in red, Class 2 in green, Class 3 in light blue and Class 4 in purple (only the forms with a p-value lower than 0.05 are represented; the two axes represent 73.61% of the total variance). (b) Clustering of active words (a) into search themes (for instance: THM_SOURCE) associated with the most relevant authors (for example: AUT_Zhang). Please refer to the online version of this paper to see this figure in color: http://dx.doi.org/10.2166/wh.2022.048.

Table 1 | Groundwater MP sources, abundances, and characterization methods

Author	Model/sample	Methods	Depth	Filter pore size (μ m)	Analytical method	Particle type	Size	Shape	Color	Abundance	Sources	Observations
Panno *et al.* (2019)	Karst aquifers Illinois (USA)	Mid-November 2017 8 springs and 3 shallow wells in the Driftless Area 6 springs in the Salem Plateau Volume: 1 L	<65 m	0.45	Pyrolysis gas chromatography mass spectrometry PY – GMS	4 of 20 MP samples are PE (springs)	<1.5 mm	Fibers	65% MP blue 15% red 13% gray	16 of 17 groundwater samples contained MP Average concentration: 6.4 items/L Maximum concentration: 15.2 items/L	• Landfills • Septic effluent • Surface runoff	Karst topography and sinkhole influence the movement of MP Migration of water from the surface to groundwater through sinkholes, cracks, fractures.
Manikanda Bharath *et al.* (2021)	Groundwater near landfills (2 km) South India/ Channai area	Post season of harvest, November 2019 20 locations Glass bottles 1 L	3–30.48 m	0.45	ATR-FTIR spectroscopy LB – 340 Zoom Stereo microscope with LED illumination SEM coupled with EDS (Scanning Electron Microscope with Energy Dispersive X-ray)	70% of Nylon Predominance of PP and PS Predominance of white color	/	Pellets, foam, fragments, fibers	Predominance of white color, followed by black color	• Perungundi Maximum concentration: 80 items/ L Minimum concentration: 7 items/L Average concentration: 34 items/L • Kodungaiyur Maximum concentration: 23 items/ L Minimum concentration: 3 items/L Average concentration: 12 items/L	• Landfills • Pathways: Storm water runoff and landfill leachate	In this study, the colors, shape, type, forms are mixed in the interpretation of the authors No controls to determine MP pollution in groundwater
Selvam *et al.* (2021)	Groundwater and surface water South India (Tamil Nadu state)	Post-monsoon season (January 2019) 24 groundwater samples (wells and borewells) 20 surface water samples (Punnakayal estuary) Volume: 20 L	Groundwater: 2–5 m Surface water: 0–20 cm	0.2	Fourier transform infrared spectrometry (μFTIR)	PA (nylon), PE, PP, PES, PVC Groundwater: PE (55%), Nylon (35%), PES (10%) Surface water: PA (38%), PE (30%), PP (22%), PVC (5%), cellulose (5%)	Surface water: 0.34–4.30 mm Groundwater: 0.12–2.50 mm (34% < 1 mm)	Foam, plastic, fiber, film, and pellet. Predominance of fibers	Groundwater: Predominance of black colored plastics (80% in wells, 20% in borewells) Surface water: Predominance of blue color (45%)	Groundwater: Average concentration 4.2 items/L (average concentration) 10.1 items/L (maximum) Surface water: 7.8 items/L (average concentration) 19.9 items/L (maximum)	• Industrial activities • Industry wastages • Presence of MP in wastewater treatment • Accidental discharge of industrial raw water • Sewage sludge	Transport of heavy metals in the water system
Poleć *et al.* (2018)	Surface water Groundwater Tap water with plastics input Poland	Volume: 5 L Polish river: 2 samples (Rudawa river, Vistula river) Groundwater: 2 samples. One with HDPP (High-Density Polypropylene) One with glass bottles	/	0.4	SEM–EDS DXR Raman	Rudawa river: no MP Vistula river: cosmetic pellet	No mentioned Microbeads are considered	Fragments Groundwater: irregularly shaped particles	Samples glass bottles from groundwater: blue and green particles	No quantified	/	HDPP bottles used to study the MP migration and verification of analytical methods The non-detection of MP in surface waters can be explained by low analysis volumes, river dynamics, topography, depth.
Mintenig *et al.* (2019)	Drinking water Germany Water at the outlet	August 2014 Sampling: 4 locations	30 m	0.2	Hyperion 3000 FTIR microscope	PVC, PE, PA, PEST, Epoxy resin	50–150 μm	Fragments	Black Transparent	14/ 24 samplings = No MP 5 samplings = 1 items/ m^3	• Distribution systems: • HDPE, PVC pipes	The presence of plastics would be more due to the water supply

(*Continued.*)

Table 1 | Continued

Author	Model/sample	Methods	Depth	Filter pore size (µ m)	Analytical method	Particle type	Size	Shape	Color	Abundance	Sources	Observations
	of groundwater wells Water at the DWTP Water at the water meter Tap water	40 m^3 of raw water and drinking water			Tensor 27 FTIR spectrometer					4 samplings = 1–3 items/m^3 1 sampling = 7 items/m^3 Average: 0–7 /m^3	● Storage tank with epoxy ● Rings of polypropylene (PP) in storage tanks	related to the distribution systems in PVC or PE Low contamination Contamination observed on controls, due to a sampling manipulation
Shruti *et al.* (2020b)	Drinking water fountains using groundwater Mexico City	July–August 2019 42 stations 3 samples for each station Volume: 1 L	/	0.22	SEM–EDS	PTT and epoxy resin	0.1–5 mm <0.5 mm (50%) 0.5–1 mm (25%) 3–5 mm (3%)	Fibers and fragments (mostly fibers)	Transparent fibers (69%) Blue fibers (24%) Red fibers (7%)	Average concentration: 18 ± 7 items/L	● Effluents discharges	Contamination of <1 mm MP in all stations
Strand *et al.* (2018)	Tap water/ Groundwater Danmark	17 samples	/	0.2	Fourier transform infrared spectrometry (µFTIR)	PET, PP, PS, ABS, PU	20–100 µm	Fibers (82%) Fragments (14%) Films (4%)	Pink, blue, black	Average concentration: 0.3 items/L	/	/
Weber *et al.* (2021)	Drinking water in Germany House connections and transfer station	Volume: 0.25–1.3 m^3 samples from a station using groundwater as a source of water	/	1.2	Raman microspectroscopy	PET, PP, PS, PE	MP < 10 µm no detected	Fibers and fragments	Phthlocyanine pigment was detected is one of five taps	<1 items/L Concentrations was below the average blank value (0.65 ± 0.54 items/blank)	/	Low concentration No microplastics were detected at consumption taps
Pivokonsky *et al.* (2018)	Raw and treated drinking water Czech Republic	Volume 1 L	/	5 and 0.2	FTIR Raman spectroscopy	PET, PP, PE	1–10 µm Raw water: 1–5 µm (40–60%) 5–10 µm (30%–40%) Treated water: 1–5 µm (25%–60%) 5–10 µm (30–50 µm)	Fragments Fibers	/	Raw water: 1,473 ± 34 to 3,605 ± 497 items/L Treated water: 338 ± 76 to 628 ± 28 items/L	/	The treatment plants are less efficient at removing the smallest MP
Tong *et al.* (2020)	Drinking water/Tap water China	38 samples of tap water in different cities HPPE bottles	/	0.2	Raman spectroscopy	14 types of plastics polymers detected: Majority of PE and PP	Size: 3 µm–445 µm with a mean size of 66 µm Predominance of 50 µm particles	Predominance of fragments, followed by fibers and spheres	/	0–1,247 particles/L of 38 samples Average concentration: 440 items/L	Hypotheses: drinking water distribution systems	Abundance of particles might be overestimated due to the extrapolation
Kirstein *et al.* (2021)	Drinking water/Tap water Skane Sweden	Sample volume: 1 m^3	/	5 and 0.7	µFTIR Py-GCMS	PA, PES, acrylic Blank: PE (67%), PA (24%), PET (5%), acrylic (3%), PP (2%)	<150 µm <20 µm (32%)	Fragments (81%) Fibers (19%)	/	Average concentration across the distribution system: 0.174 items/L Average concentration in one station: 0.809 ± 0.688 MP/L (highest number) Blank: Average contamination of 46 ± 63 MP/blank	/	Low MP proportion in drinking water processed in a high-performance drinking water treatment plant

However, MP migration from soil to groundwater could occur through leaching from a landfill or plastic mulching, surface runoff, wastewater effluents, septic effluent, and sewage sludge (de Souza Machado *et al.* 2018, 2019; Mintenig *et al.* 2019; Panno *et al.* 2019; Lau *et al.* 2020; Shruti *et al.* 2020b; Manikanda Bharath *et al.* 2021; Selvam *et al.* 2021). Other sources are car tire debris, abrasion of clothes and textiles in washing machines, cosmetic and care products, substance coating and atmospheric deposition, and fragmentation of plastic litter (Figures 1 and 2 and Table 1). As shown by Ng *et al.* (2018), the most contaminated areas are related to agricultural soils. However, as indicated in Table 1, these results should be interpreted with caution since most studies on soil and groundwater pollution concern agricultural areas. Soil can receive sewage sludge amendments enriched with plastics during land application processes. More than 99% of MP in wastewater is retained in sewage sludge, with a majority of plastics smaller than 300 μm (Schell *et al.* 2020). Boyle & Örmeci (2020) estimated that agricultural soils receive 63,000–430,000 tons of MP from sewage sludge and agricultural composts. In addition, flooding with lake water (0.82–4.42 plastic items m^3) or river water (0–13,751 items km^2) can provide major input pathways for plastic into the soil. Additional sources comprise littering along roads and trails, illegal waste dumping, road runoff as well as atmospheric input (Bläsing & Amelung 2018). One of the only studies to have found evidence of MP in karst groundwater is the American study of Panno *et al.* (2019). Karst aquifers are formed by the erosion of carbonate rocks by surface and groundwater flows, leading to the formation of a groundwater aquifer. These authors reported on 16 of 17 groundwater samples with an average concentration of 6.4 particles/L, with a maximum concentration of 15.2 particles/L. They suggested a positive correlation between the concentration of MP in karst groundwater and the wastewater components triclosan, phosphate, and chloride. This relationship indicates that wastewater is one of the sources of MP pollution in groundwater. Another study shows the impact of landfills on groundwater quality (Manikanda Bharath *et al.* 2021). They found evidence of MP in groundwater near landfill sites (lower than 2 km). Poor management of landfills and wastes is a major source of MP pollution in aquifers. They reported a concentration of MP between 2 and 80 particles/L in 20 groundwater samples. However, no controls were conducted. This makes the confirmation of groundwater contamination difficult. Indeed, contamination can occur due to handling failures, and lack of precaution when analyzing samples, such as the use of nylon clothing (Li *et al.* 2019).

2.2.2. Types of MP (Class 3)

In the few studies that have analyzed the composition of MP in groundwater, the most common types of plastics found are PE, PET, and PP (Panno *et al.* 2019; Manikanda Bharath *et al.* 2021; Selvam *et al.* 2021). These types of plastics have also been identified in drinking water samples from groundwater (Strand *et al.* 2018; Mintenig *et al.* 2019; Weber *et al.* 2021), in drinking water (Tong *et al.* 2020), in raw and treated waters (Pivokonsky *et al.* 2018). These three compounds are mainly found in food packaging, water bottles, and textiles (Martínez Silva & Nanny 2020). PP and PE are often used in the cosmetics industry and in the production of microbeads (Jiang 2018). PS, polyamide (PA), ABS, polyurethane (PU), PVC, PES, and epoxy resin were also identified in groundwater and/or drinking water samples (Strand *et al.* 2018; Mintenig *et al.* 2019; Panno *et al.* 2019; Shruti *et al.* 2020b; Zhou *et al.* 2020; Manikanda Bharath *et al.* 2021; Selvam *et al.* 2021; Weber *et al.* 2021).

Most forms in soils, groundwater systems, and drinking waters are fibers and fragments (Pivokonsky *et al.* 2018; Strand *et al.* 2018; Mintenig *et al.* 2019; Panno *et al.* 2019; Shruti *et al.* 2020b; Tong *et al.* 2020; Kirstein *et al.* 2021; Selvam *et al.* 2021; Weber *et al.* 2021). The fragments are often derived from the degradation of various plastics (Pivokonsky *et al.* 2018). Fibers are the most common particles found in sediments, living organisms, and atmospheric fallout (Dris *et al.* 2016). Besides, washing machine effluents are the main source of plastic fibers in the environment due to the degradation of textile particles (Pirc *et al.* 2016). Washing machines can generate 1,900 plastic fibers in a single cycle (Browne *et al.* 2011) and enrich sewage sludge with plastic fibers. Indeed, the presence of fibers in soils and waters is evidence of the amendment of sewage sludge, fertilizers, and other industrial processes (Zubris & Richards 2005; Li *et al.* 2019; Liu *et al.* 2019; Brahney *et al.* 2020; Zhou *et al.* 2021b). Panno *et al.* (2019) reported that all samples from karst aquifers analyzed were contaminated with microfibers and suggested contamination by septic system effluents.

Regarding the reported size of MP in groundwater (Table 1), Panno *et al.* (2019) found MP with a size lower than 1.5 mm in karst aquifers with an average concentration of 6.4 particles/L. Selvam *et al.* (2021) identified a wide range of MP size in groundwater (0.12–2.50 mm), with a predominance of MP lower than 1 mm (34% of the total number of MP). In this study, the mean concentration reported is 4.2 particles/L. In drinking waters using groundwater as a source, the size ranges are generally smaller, with a predominance of MP lower than 500 μm. Several fractions have been identified in tap water: greater than 10 μm (Weber *et al.* 2021), 20–100 μm (Strand *et al.* 2018), 50–150 μm (Mintenig *et al.* 2019). However, Shruti *et al.* (2020b) identified MP ranging in size from 0.1 to 5 mm in public fountains, but with a majority of MP lower than 0.5 mm (50% of the total number of MP). Actually, in treated waters, MP does not exceed a certain size range, particles with a size larger than 50 μm seem to be removed by the treatment plants, with relatively lower concentrations of MP in treated waters than in raw waters (338 ± 76 to 628 ± 28 particles/L and $1,473 \pm 34$ to $3,605 \pm 497$ particles/L, respectively) (Pivokonsky *et al.* 2018). In the same study, a size range of 1–10 μm was also identified with a majority of 1–5 μm MP for treated and raw waters (25–60% and 40–60% of total MP, respectively). Smaller sizes were also identified in two other studies: 3–445 μm (Tong *et al.* 2020), lower than 150 μm with a majority of MP lower than 20 μm (Kirstein *et al.* 2021). These studies show that the smallest ranges are often not removed and are detected in drinking water, posing a real risk to human health. However, other studies show negligible concentrations of MP with concentrations of 0.7 items m^{-3}, 0.174 items/L, lower than 1 item/L, 18 ± 7 items/L, respectively (Mintenig *et al.* 2019; Shruti *et al.* 2020b; Kirstein *et al.* 2021; Weber *et al.* 2021). This large difference in concentration can be due to the lack of standardization of analytical protocols (Zhang *et al.* 2019) with different choices of pore sizes during filtration, volume to be collected, separation and analysis methods (μFTIR, Raman, SEM–EDS), quantification, identification and comparison of the characteristics and abundance of plastics (Perez *et al.* 2022). This makes the inter-comparison of MP concentrations in the different media very difficult.

2.2.3. Environmental exposure pathways of MP (Class 2)

Knowledge of the transfer pathways of MP from soil to groundwater is very limited. Vertical transport, infiltration, percolation, and leaching play a role in the distribution and transport of MP in groundwater (Huerta Lwanga *et al.* 2016; Ganesan *et al.* 2019; O'Connor *et al.* 2019; Panno *et al.* 2019; Shruti *et al.* 2020a; Gao *et al.* 2021; Ren *et al.* 2021b; Selvam *et al.* 2021). This distribution depends on the physico-chemical characteristics of soils, groundwater, and MP.

Several factors have been reported regarding the distribution of plastics in soils and groundwater (Ganesan *et al.* 2019; Panno *et al.* 2019; Wang *et al.* 2019; Hou *et al.* 2020; Luo *et al.* 2020; Shruti *et al.* 2020b; Wu *et al.* 2020; Selvam *et al.* 2021). Ionic strength, freeze–thaw cycle, temperature, pH, microbial activity, and soil texture influence MP transport in the different soil layers (Li *et al.* 2018; Bradney *et al.* 2019; Luo *et al.* 2020; Mammo *et al.* 2020; Menéndez-Pedriza & Jaumot 2020; Yu *et al.* 2020; Rai *et al.* 2021; Yin *et al.* 2021). For example, an increase in soil pH tends to extend particle transport (Ren *et al.* 2021b). The transport of MP can take place through soil pores and cracks, if the size of the MP is smaller than the size of the soil pores. The smaller the MP size, the larger the surface area, and the more transferable the MP will be in the soil. O'Connor *et al.* (2019) reported that smaller PE particles (21 μm) were more mobile than larger PE particles (181, 349, and 535 μm). Thus, the size of MP has a significant effect on vertical transport, in conjunction with other transport-enhancing parameters such as wet–dry cycles (O'Connor *et al.* 2019; Gao *et al.* 2021; Ren *et al.* 2021b). O'Connor *et al.* (2019) reported that as a result of an increase in the number of wet–dry cycles, PE–MP (21 μm) penetrated the deep layers of the sandy soil more easily. Finally, not all soils can act as a barrier that prevents pollutants from leaching into groundwater. Zhou *et al.* (2021b) analyzed 29 soil samples along the Yangtze River (China), taking into account a high mountain site with low anthropogenic activity and a site near an urban area. They reported a predominance of PA, characteristic of domestic wastewater, with more microfragments in the lower layers (10–15 cm) than in the upper soil layers (0–5 cm). Their study shows not only the contamination of areas characterized by low human presence but also the ability of MP to transfer into subsoils. They reported that MP with a size lower than 200 μm constitutes most of the contamination (63% of 10–100 μm MP identified in the samples) with the majority being microfragments and microfibers.

Density, solubility, hydrophobicity, size, type, and shape of plastics are parameters to be taken into account in the behavior of plastics. In particular, density, solubility, hydrophobicity are important parameters concerning the distribution of MP in the water column. The migration of plastic pollution is influenced by the preferred flow paths, diffusion, dispersion, adsorption, and chemical and biological transformation of plastics. Other groundwater factors to be considered when studying the transfer of MP within groundwater are the water table level, effective porosity, hydraulic conductivity and hydraulic gradient. Panno *et al.* (2019) reported that hydrogeological characteristics, topography of karst environments influence the mobility of MP. Some karst landscapes are characterized by the presence of sinkholes, which are excavations generated by erosion of a karst environment. Through sinkholes, water migrates from the ground surface to the groundwater, which favors the migration of MP. Fractures and crevices contribute to the mobility of plastics, amplified by the characteristics of the plastics, as previously stated. Larger plastic particles would be more likely to be retained in crevices or fractures while smaller particles will reach groundwater more quickly.

Furthermore, the ageing of MP which depends on soil, climate, and MP characteristics, is also a relevant variable to consider in MP migration from soil top groundwater since they can enhance changes in surface topography, charge, polarity, the chemical structure of plastic polymers (Ren *et al.* 2021b). Ageing can then favor the MP binding with other pollutants and the release of MP additives in the environment (Ren *et al.* 2021b). Wang *et al.* (2021) studied the behavior of aged PE in contact with air, soil, and water. Functional groups, –OH, –CO, –CH appeared after PE exposure to UV light. Functional groups are binding sites and therefore active sites for the absorption of organic pollutants and heavy metals. In addition, the UV-aged PEs showed a strong capacity to remobilize phthalates, known to be endocrine disruptors (Habert *et al.* 2009). In the study, phthalates could influence the adsorption of copper and tetracycline (antibiotics) on the surface of the PE. There was also the formation of a biofilm on the surface of PE cultivated in soil or water, which may promote the adsorption of heavy metals and antibiotics. The study by Selvam *et al.* (2021) confirms the previous results regarding the ability of MP to act as transport vectors to enhance mobilization and potential exposure of metals to other organisms. They reported different adsorption depending on the type of plastic, metal, and the analytical medium. Five types of plastics were analyzed: PP, PE, PA, PVC, and cellulose. PVC, PP, and PE showed the highest adsorption of heavy metals (adsorption rate is calculated as the concentration of metals on the surface of MP (μg/g) versus the concentration of metals in water (μg/mL). In surface water samples collected from the vicinity of a sewage treatment plant in India, PP showed high adsorption of metals, particularly cadmium, followed by manganese and arsenic. For PE, only the adsorption of arsenic and cadmium was reported. In surface water samples from an estuary, the adsorption of manganese, zinc and arsenic by PP was more important. For PVC, the highest adsorption rates were for manganese, copper, and lead for both test sites. Thus, as MP are vectors of pollutants in surface waters, it is relevant to assume that they will also be vectors in groundwater and thus will play the role of 'Trojan horse' in drinking water and organisms.

In addition, macrofauna can be vectors for the transfer of MP into groundwater, especially bioturbation by *Lumbricus terrestris* which contributes to the vertical transport of MP in the lower terrestrial layers (Huerta Lwanga *et al.* 2016, 2017; Rillig *et al.* 2017). Rillig *et al.* (2017) reported the ability of earthworms to transport spherical PE–MP of different sizes (first fraction: 710–850 μm, second fraction: 1,180–1,400 μm, third fraction: 1,700–2,000 μm, fourth fraction: 2,360–2,800 μm) in the deepest levels of potted soil. The number of plastic particles transported through the layers depends on the size of the MP. The smallest fraction (710–850 μm) was most often identified in the lower layers (10 cm), while the other fraction sizes were most often observed in the middle layers (3.5–7 cm). Furthermore, earthworm activity has an influence on the transport of MP, as in the absence of earthworms, MP was retained on the surface. The study by Huerta Lwanga *et al.* (2017) confirms that the smallest particles are the most mobilizable and bioavailable to earthworms. In this study, earthworms were cultured in a mesocosm in the presence of low-density polyethylene (LDPE) (with a size lower than 50 μm and between 63 and 150 μm). The concentration of LDPE–MP lower than 50 μm increased by 65% in the burrows, compared to a concentration of 40% at the soil surface. Earthworms bury the MP and are found in the walls of the burrows. These tunnels are the preferred pathways for transporting MP to groundwater, new organisms, and plants. Transfer to plants can occur via root–groundwater interactions or via root–soil contact. However, the transport mechanisms of MP in the various terrestrial, aquatic, biotic, and atmospheric media remain poorly known and little studied. The ability of MP to be transferred through the entire food chain poses significant health and ecological risks.

2.2.4. Effects of MP from groundwater (Class 4)

The studies related to the effects of MP on groundwater fauna and flora, dealing more with hazards than risks, usually extrapolate the results obtained in the laboratory, on soil or aquatic organisms, and/or conclude there is no effect of groundwater MP pollution on organisms. However, this conclusion can be misleading due to a lack of studies on exposure assessment and a lack of knowledge on groundwater biodiversity. Nonetheless, groundwater is home to biodiversity called stygofauna, crustaceans such as copepods, isopods, amphipods, decapods, fungi, worms, snails, and amphibians (Hérivaux *et al.* 2013). However, no study has been published on the action of MP on the biodiversity of groundwater. Nevertheless, stygofauna would contribute to good water quality by degrading pollutants (Hérivaux *et al.* 2013) but what about the degradation of plastics? What are the effects of MP on groundwater organisms? *Daphnia magna* is often used as a model in the toxicological study of MP in surface waters and could provide some answers to the effects of MP in groundwater. Exposure of *D. magna* to MP has been reported to result in decreased growth rate, decreased reproduction, and increased mortality (pristine microspheres 1–5 μm, dose: 0.1 mg/L (Martins & Guilhermino 2018), inhibition of mobility (irregularly shaped fragments of PE 10–75 μm, dose: 0.0001–10 g/L (Frydkjær *et al.* 2017). An increase in mortality was also reported in the study by Jemec *et al.*

(2016) (ingestion of PET microfibers 62–1,400 μm, dose: 12.5–100 mg/L) and Aljaibachi & Callaghan (2018) (PS 1–2 μm, dose greater than 0.01 mg/L). In addition, groundwater contamination poses a contamination risk to plants that use their root systems to draw water (Ebere *et al.* 2019; Yang *et al.* 2021). The knowledge of such contamination and the mechanisms by which this contamination is possible needs to be better understood.

Currently, there is no study on the impacts of groundwater MP on environmental attributes. Most studies concentrate on the impacts of MPs on soil properties such as permeability, bulk density, texture, evapotranspiration, wetting rate, water retention rate, infiltration rate, bacterial community diversity, and temperature (Steinmetz *et al.* 2016; de Souza Machado *et al.* 2018, 2019; Wan *et al.* 2019; Qi *et al.* 2020; Prata *et al.* 2021). Other studies focus on the consequences of soil MP on plants and microorganisms with or without the addition of other compounds (antibiotics, heavy metals, pesticides) (Rillig 2012; Dong *et al.* 2020; Jiang & Li 2020; Ren *et al.* 2021a).

Regarding the impacts of groundwater MP on human health, Hwang *et al.* (2020) estimated the human exposure to 3 μm PS MP particles to 4 μg per year by consuming drinking water. Furthermore, in France groundwater sources represents about 66.4% of water intended for human consumption (Dequesne *et al.* 2021), whereas in the United States, half of the drinking water comes from groundwater (USGS 2017). Thereby, humans are supposed to be exposed to groundwater MP. Indeed, according to Cox *et al.* (2019), Americans would be exposed to 90,000 particles through ingestion of bottled water (based on the daily recommendation of 2 L). However, parts of MP coming from the bottles and from groundwater were not specified.

2.3. Future implications and perspectives

The research gaps were identified by collecting the gaps sections of the 58 articles dealing with MPs in groundwater. It resulted in 228 ECUs representing 1,592 forms and 8,065 total occurrences. As for the analysis of main topics, the Reinert classification was processed, leading to five classes of topics presented in Figure 3.

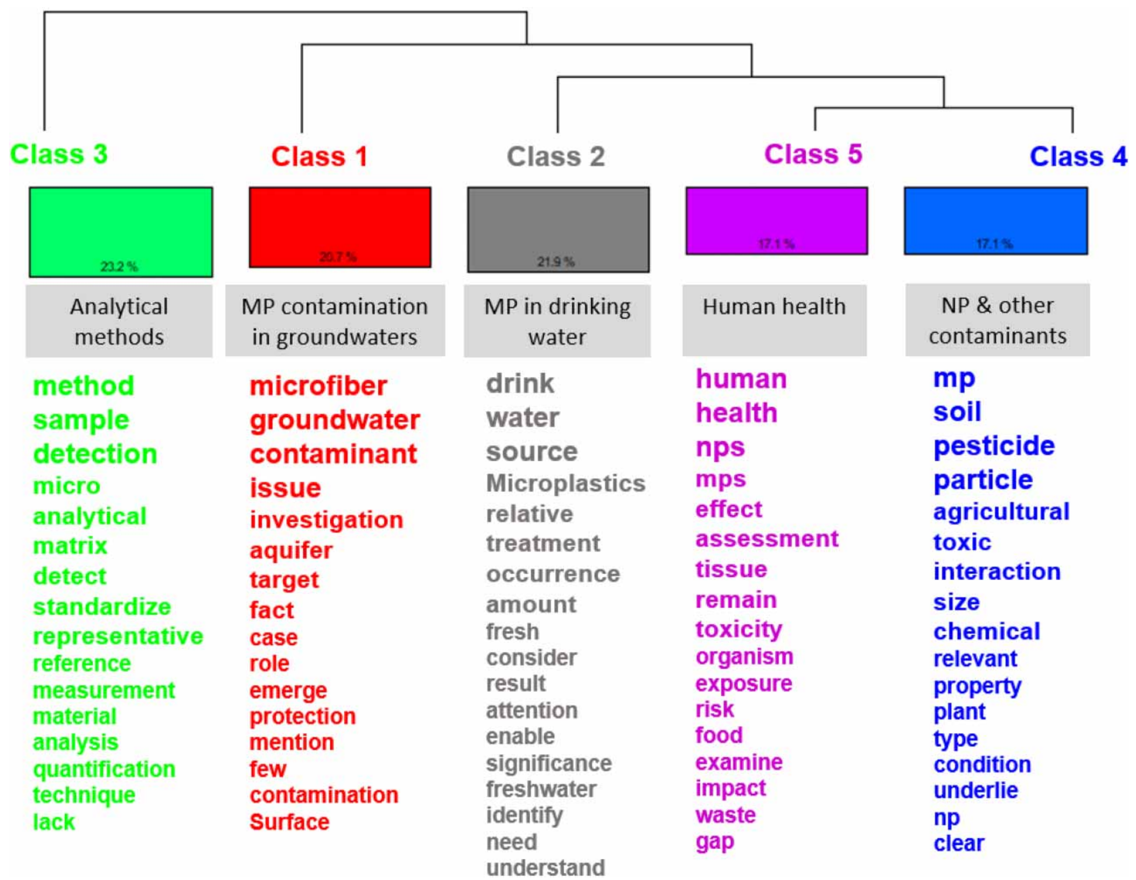


Figure 3 | Dendrogram of the ECU related to research gaps (only significative words are shown for each class: $p < 0.001$). Words with the higher χ^2 association to the cluster are with a larger character size.

Class 3 represents the lack of analytical standards. Class 1 is about the lack of knowledge on groundwater MP contamination and protection solutions. Class 2 concerns the lack of knowledge on the contribution of groundwater to drinking water MP pollution. Class 5 is about the need for research on human exposure and health impacts. Class 4 deals with MP and NP interaction with other contaminants in agricultural contexts.

Regarding the lack of analytical standards and protocols (Class 3), different protocols and methodologies affect the quality of the results (Toussaint *et al.* 2019). Standardization of protocols would avoid contamination in the laboratory and provide representative samples of plastic pollution in the environment. As for all monitored substances in polluted sites, the harmonization depends on the number of samples, a relevant variable which affects the result representativeness. Indeed, too few samples may lead to a wrong interpretation and not reflect the current contamination and risks. It also requires a representative volume of water to be sampled, as a small volume reduces the chance of finding particles in the sample, leading to a bias in the analysis. Koelmans *et al.* (2019) recommend taking a volume of 500 L of groundwater to obtain interpretable results. However, to achieve a standardization of the analytical method, it is required to have a consensus related to the definition of MPs, especially the size of plastics. Further to the lack of protocols, the low accessibility makes difficult groundwater MP sampling. Up to date, the question of which piezometer to use is not answered. Alternatives to PVC like stainless steel piezometers are then encouraged but can be much more expensive. The sampling protocols should also ensure the lack of MP contamination from clothing, airborne, distilled water, and plastic bottles. Research should also focus on MP traceability during transport by water so that to confirm groundwater contribution to environmental and human exposure. Protocols are then necessary not only for sampling, detection, and analyzing but also for studying the behavior and the effects of MP on environmental and biological matrices.

Regarding the lack of knowledge on groundwater MP and protection measures (Class 1), the research about MP has been widely discussed in marine environment, followed by surface waters, while groundwater knowledge is very scarce. Future research should focus on the transport mechanisms of MP according to MP types and groundwater and soil conditions to identify which factors influence most of the vertical transport. Since many cities use groundwater as a source of drinking water, more research should be dedicated to the contribution of groundwater as a source of human exposure to MP. Other research should focus on substitutions for harmful materials such as biodegradable plastics. However, some biodegradable plastics like PLA have been shown to be more harmful than common plastics (Qi *et al.* 2018, 2020). Biodegradable plastics can be degraded more easily and can then penetrate in all media more quickly. This raises the question of biodegradability and safe-by-design criteria. These measures to protect groundwater from MP contamination should be then developed in collaboration with plastic producers, landowners and managers, researchers on materials, socio-economists, and environmental risk experts.

Regarding the need to understand the occurrence and the sources of MP in drinking water from groundwater (Class 2), as emphasized by Miranda *et al.* (2020), studies should be focused on the overall water treatment and the release of MP in drinking water. The main questions to answer are: How effective are water treatment techniques in removing MP? Which treatment steps contribute the most to the MP removal? Because of health risks, the presence of MP in drinking water deserves more attention. However, data on water contamination from MP in tap water are insufficient and are unreliable due to a lack of standardized analytical protocols.

Regarding the knowledge of human and environmental exposure to MP and potential effects (Class 5), no published study has directly examined the effects of MPs on human. The risk to human health associated with MPs is further studied for additives, with potential effects on the endocrine and reproduction systems. Human health risks are poorly understood because it is difficult to extrapolate results from mammalian models to humans due to the lack of standardization of protocols and because humans are not 'giant rodents'. The effects observed in rodents may be different from the effects that will be observed in humans, and it is possible that there will be no effect. However, one must be very careful with studies that claim no effects, as the models may be 'false negatives', i.e. show no toxicological effects but prove dangerous for humans. From mammalian models, we cannot guarantee the same effects on humans. Moreover, the effects of plastics depend on the exposure dose, the exposure time of the species, the individual, and the sensitivity window. Besides *in vitro* models can determine the toxicity of the substance at the cellular level, but they do not take into consideration all the biological processes that will influence the transport, degradation, and toxicity of MPs. For example, the digestion process still needs to be studied.

In addition, MPs can highly interact with other contaminants (heavy metals, antibiotics, pesticides, etc) to form highlighting pollutant mixtures by playing the role of 'Trojan horse'. This concept, the mechanism and the impacts that can arise from it are poorly studied and requires further research. Thereby, the research should be oriented to the elaboration of standardized protocols for being able to compare the effects of MP on different biological matrices and to identify the main transfer pathways and mechanisms that results in toxicity.

Research on the mobility of MP in soils and their ability to migrate into groundwater (Class 4) should concentrate on the soil buffering. Wanner (2021) have shown that plastics can interact with other contaminants, such as pesticides. Although Castan *et al.* (2021) found that MP would hardly co-transfer organic contaminants in lower soil layers, the role of bioturbation should be more studied. Biota such as plants, micro- and macro-fauna, could also act as factors of MP stabilization or remediation. That is why, it is crucial to study MP (including its type, shape, composition) in the overall ecosystem. But, again, this can be done only by elaborating first standardized analytical protocols and reference materials.

In terms of policy knowledge, few policy guidelines and frameworks are available. In Europe, the directive 98/83/CE related to quality of water destined for human consumption, aims to ensure that information on water quality is more transparent and to increase vigilance regarding the presence of pollutants including MPs. Besides, the European Commission developed a policy to reduce the emission of MPs and the environmental impact of certain plastic products, through the directive 2019/904, the REACH regulation and the Green Deal and Circular economy action plan. In France, the government aims to reduce our dependency on plastics and control plastic pollution. There are decrees on waste related to reduction of single-use plastics, and in 2025, washing machines should be equipped with filters to retain plastic microfibers. However, there are no regulations on the release of MPs from wastewater treatment plants and the input of MPs through sewage sludge. Could the size, flow, and quantity of MPs in the water and sludge be regulated? There is an important need to establish a link between the directives and the gaps regarding MPs in soil, groundwater, and drinking water.

3. CONCLUSIONS

MPs are widespread in the aquatic and soil environment. The question of the transfer of MP from soil to groundwater and their potential transfer to drinking water was raised in this study. After a pre-selection of scientific papers, the texts were segmented with Iramuteq to identify research topics. Text mining is an interesting approach for the bibliographic research and can be used in other research fields while the literature is consequent. This software permits to organize the results and visualize the research priorities related to MPs in groundwater. Research must be encouraged related to knowledge gaps:

- Compared to research on aquatic or soil compartments, scientific data on groundwater are more limited. The existing results show divergence due to differences in context (alluvial aquifer, fractured rock aquifer, karst aquifer, etc) collecting, sampling, and analytical methods. This divergence, particularly regarding the presence of MP in the drinking water, requires further research with standardized analytic protocols and reference materials.
- MPs represent a risk to terrestrial ecosystems. The physico-chemical characteristics of the soil, of groundwater and MPs should be more considered to demonstrate main parameters influencing the vertical transport of MP and the co-transport contamination through 'Trojan horse' process. The transfer models developed in the laboratory should be more developed to better understand the transport mechanisms on soil, groundwater, and organism. The impact of co-transport contamination by MPs and persistent organic compounds (POPs) on organism should be more assessed.
- The question of plastic regulation and decision criteria for developing safe-by-design alternative materials should be raised and answered through a collaboration between plastic producers, landowners and managers, researchers on materials, socio-economists, and environmental and health risk experts.

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.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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