Sokoto Journal of Geographical Studies (SJGS)

Volume 3, Issue 1, December, 2025 Edition

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Published by the Department of Geography, Sokoto State University, Sokoto P.M.B. 2134, Along Birnin Kebbi Road, Sokoto State-Nigeria

Department of Geography, Sokoto State University, Sokoto P.M.B. 2134, Along Birnin Kebbi Road, Sokoto State-Nigeria

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E-ISSN: 3115-5812, PRINT ISSN: 3034-551X

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Acknowledgements

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TABLE OF CONTENTS

SOKOTO JOURNAL OF GEOGRAPHICAL STUDIES (SJGS) *E-ISSN: 3115-5812, PRINT ISSN: 3034-551X*

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Volume 3, Issue 1, December 2025 Edition

MICROPLASTICS POLLUTION IN THE GROUNDWATER OF THREE LAND USE TYPES, SOUTHEASTERN HUNGARY

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Abstract

https://doi.org/10.65760/sjgs.v3.i1.21

Groundwater is one of the sources of freshwater for drinking and agricultural activities, yet it is continuously threatened by emerging contaminants and pollutants such as microplastics (MiP). Contrary to the surface water bodies, there is a paucity of knowledge about the rate and distribution of the horizontal movement of microplastics in the groundwater of different land use areas. This study examined 18 groundwater samples collected from three land use types greenhouse, grassland, and landfill across four locations in southeastern Hungary (Szeged, Szarvas, Szentes, and Hódmezővásárhely). The research aimed to quantify microplastic contamination levels, analyze their morphological characteristics, and compare pollution levels among the land uses. After filtering 1 litre and digesting organic matter with hydrogen peroxide, microplastics were identified and counted under a microscope. The mean microplastic concentration across all samples was 7.6 ± 3.4 pieces/L, with the landfill area recording the highest level (27.50 ± 11.29 pieces/L), followed by the greenhouse (2.88 ± 0.65 pieces/L) and grassland (0.20 ± 0.20 pieces/L) areas. Statistical analysis revealed a significant difference in microplastic abundance among the three land use types, indicating that landfill areas are the primary contributors to groundwater contamination. The dominant microplastic forms were green and red fibers and fragments measuring between 1.0 and 2.1 mm. No evidence of horizontal movement of microplastics between the areas was found. The study concludes that groundwater from all land use types contains microplastics, recommending treatment before consumption or irrigation and emphasizing the need for proper plastic waste management and further research on groundwater contamination dynamics.

Keywords: *Groundwater, Land Use, Microplastics, Landfill, Greenhouse, Grassland, Hungary.*

Introduction

Plastic use in all sectors has led to large amounts of generated waste and microplastic contamination. Global plastic production has declined slightly, from 368 tons in 2019 to 367 tons in 2020, with a similar trend observed for Europe: plastic production declined from 57.9 to 55 tons (Plastic Europe, 2021).

Plastic contaminants range from large-sized particles (i.e., 1-5 mm), medium-sized microplastics (1 µm to 1 mm) to nanoplastics (≤1 µm). Plastic contaminants in the environment come from primary sources, such as sewage sludge, organic and inorganic fertilizers, irrigation water, and atmospheric and wind deposition (Allen *et al*., 2019; Bigalke *et al*., 2022; Sussana, 2018). They also come from secondary sources due to the disintegration of larger plastic materials, such as greenhouse films found in mulch (Babagyayou *et al*., 2020; Huang, 2020; Kumar *et al*., 2021; Mo *et al*., 2021; Schothorst *et al*., 2021). Microplastic waste can be transported vertically through cracks in soil. Other transfer methods include the application of irrigation water, soil microorganisms, and leaching (Bigalke *et al*., 2022; O'Connor *et al*., 2019; Rezaei *et al*., 2019; Zhang *et al*., 2018). Plastic contaminants cause imbalances in ecosystems and affect soils, plants, water bodies, aquatic organisms, insects, animals, and human health (Li *et al*., 2021; Mora *et al*., 2021; Rondoni *et al*., 2021; Serrano-Ruiz *et al*., 2021; Zhang *et al*., 2021).

Groundwater is one of the sources of global freshwater, supplying a substantial amount of the world's drinking water in rural and urban communities (UNESCO, 2018). The groundwater percolates and is stored in the groundwater aquifers is filtered through the rock layers and soil, which improves its quality compared to other surface water (Foster & Chilton, 2003). It is more reliable during the period of seasonal fluctuations and drought making it very important in regions with shortages of surface water such as parts of Central and Eastern Europe (Kløve *et al*., 2011). Beyond domestic use, groundwater supports industry, agriculture, and ecosystem services, yet its value as a drinking water source is most critical for public health and sustainable development. However, increasing pressures from land-use changes, over-abstraction, and contamination including emerging pollutants such as microplastics pose significant risks to its long-term safety and availability (Lapworth *et al*., 2017). Protecting groundwater quality is therefore essential to secure safe drinking water for present and future generations.

The primary sector industry, greenhouse farming, consumes large quantities of plastics. Thus, it generates plastic waste (macro and micro) in large quantities. Globally, greenhouse farming covers 220,000 ha of land and consumes 250,000–350,000 tons/year of plastic film (Dilara & Briassoulis, 2000). In Hungary, greenhouse farming covers more than 6,500 ha (Scarascia-Mugnozza *et al*., 2011). Plastic contamination in agricultural soils is mainly in the form of plastic films, but it also includes other contaminants, and plastic wastes have been found on soil profiles, and groundwater (Saadu & Farsang, 2022).

Similarly, the forest environment is not free from contamination by plastic materials. The ubiquitous nature of plastic has led to the contamination of forest soil. Choi *et al*. (2020) found 160 pieces/kg of MiP in less anthropogenically affected forest soils. MiP recorded in virgin land may be due to atmospheric fallout, wind deposition, and precipitation runoff (Choi *et al*., 2020; Rezaei *et al*., 2019; Schell *et al*., 2022). Landfills are the world's plastic reservoirs. They store thousands of tons of different types of waste. According to Nizeto *et al*. (2016), landfills serve as a means for waste disposal; they can store 20%–40% of plastic waste produced globally. Likewise, Afrin *et al.'s* (2020) study indicated that landfills are significant sources of MiP entering the environment.

MiP coming from different sources within the ecosystem penetrates the soil and enters groundwater aquifers through the leaching of contaminated groundwater (Zhang *et al*., 2022). The movement of soil organisms, such as earthworms and rodents, aids in the distribution of MiP in the environment (Maaß *et al*., 2017). Notably, MiP enters groundwater due to soil texture, infiltration, and repeated wet–dry cycles (Oßmann, 2021; O'Connor *et al*., 2019; Schell et al., 2022).

Moreover, there is a lack of knowledge about the abundance of MiP in groundwater associated with different land uses, although this knowledge is imperative because it reveals the pollution level of groundwater. Recent studies on microplastic pollution in the freshwater ecosystem are more concerned with surface water than groundwater. Similarly, few studies have compared levels of microplastic contamination. Recently, the World Health Organization (2021) lamented the lack of studies on MiP in drinking water. They emphasized that although the scant data do not currently indicate a threat to human health, there is a need to collect more data to draw proper conclusions. Hence, the present case study aimed to i) quantify the level of microplastic distribution and contamination in groundwater associated with different land uses, ii) examine the morphological structures of microplastic contaminants, and ii) compare the level of contamination associated with different land uses.

Materials and methods

Study Site

This research was conducted on groundwater associated with three land uses in the Békés (Szarvas) and Csongrád (Szeged, Szentes, and Hodomosóhely) counties of southeastern Hungary: greenhouse, landfill, and grassland. The greenhouse farmlands are located in Szeged, Szarvas, and Szentes. The landfill and grassland areas are located in Hodomesovaherly, which has been in a landfill since 1990. Currently, some part of the area is used for residential purposes while the other part is abandoned. The climatic condition of the study area is warm and dry (mean annual temperature: 10.5°C; mean annual precipitation: 520 mm), with an average annual radiation of 2,080–2,090 h/annum. The area, 84 m above sea level, is underlain by perched groundwater at a depth of 100 cm.

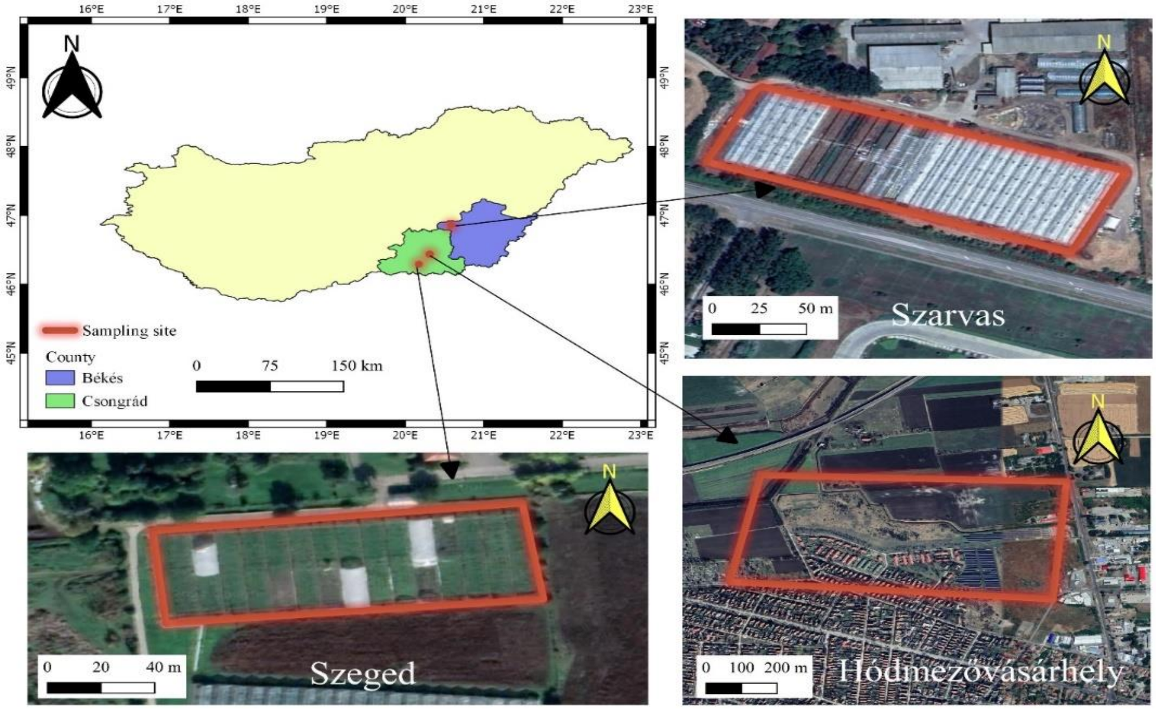

Figure 1: Location of the Study Area

The sample area was a plain with loess bedrock, and the natural soil type is Phaeozem, according to the World Reference Base for Soil Resources (Szolnoki *et al*., 2013). Before being abandoned 5–15 years ago, the greenhouse areas were used for tomato, pepper, and cucumber production. The groundwater sampling occurred at different times. For example, in the greenhouse farmlands of Szeged, sampling occurred in March 2021, in Szarvas, it occurred in February 2022, and in Szentes, it occurred in August 2022. Three groundwater samples were collected each from Szeged, Szarvas, and Szentes, respectively, and eight were collected from Hodomesovaherly. A control sample was collected from Szentes. A total of 18 shallow groundwater samples were collected.

Depth to Groundwater

The physical characteristics of the groundwater sampling locations are summarized in Table 1. The groundwater sampling depth ranged from 70 to 560 cm below ground level, and perched aquifers were present at all the sampling locations, 40–160 cm below ground level. The waiting period for perching water was 30 minutes in all the sampling sites. The samples were stored in a fridge at 4°C before analysis.

Microplastic Sample Preparation

The methods developed by Su *et al*. (2020) and Panno et al. (2019) were used to extract MiP from the samples. Briefly, 1 Litre of groundwater sample was filtered through a Whatman filter (0.45 µm) using a vacuum pump. The filters were washed in Petri dishes with 10 ml of 30% H_2O_2. The solution was left for 24 h at room temperature to digest any organic matter. The samples were filtered for a second time using a Whatman filter (0.45 µm) and a vacuum pump. The filter papers were later air-dried for microscopic identification and quantification.

Identification, Classification, and Quantification of Microplastics

The extracted MiP were observed using a digital microscope (Inspex II; software version 1.06; firmware version F001-001-011; ring light version 1.03; Ireland) at 50 × magnification. Suspected microplastic particles were confirmed using the heat and needle method. These experiments were conducted at the analytical laboratory of the Department of Geoinformatics, Physical and Environmental Geography, University of Szeged. Pieces of different macroplastics and 5% of suspected MiP were later confirmed by Raman analysis performed at the Department of Mineralogy, Geochemistry, and Petrology, University of Szeged. The compositions of the plastic materials were accurately determined by comparison with the Raman library.

Table 1. Physical Characteristics of the Groundwater Sampling Locations

S/N	Location	Land Use	Coordinates		Actual Depth (cm)	Perched Depth (cm)
			Latitude	Longitude		
1	Szeged	Agriculture (Greenhouse)	46.172864292	20.10212364	160	100
2	Szeged	Agriculture (Greenhouse)	46.172861988	20.10207858	120	100
3	Szeged	Agriculture (Greenhouse)	46.17285093	20.10198282	120	100
4	Szarvas	Agriculture (Greenhouse)	46.852400	20.583519	450	350
5	Szarvas	Agriculture (Greenhouse)	46.852381	20.583589	400	350
6	Szarvas	Agriculture (Greenhouse)	46.852364	20.583682	500	350
7	Szentes	Agriculture (Greenhouse)	46.3615	20.1732	550	460
8	Szentes	Agriculture (Greenhouse)	46.3615	20.1726	560	500
9	Szentes	Agriculture (Greenhouse)	46.3614	20.1751	530	355
10	Hódmezővásárhely	Landfill	46.437236	20.317545	350	150
11	Hódmezővásárhely	Landfill	46.434204	20.316234	320	180
12	Hódmezővásárhely	Landfill	46.436829	20.313057	340	190
13	Hódmezővásárhely	Landfill	46.436610	20.310665	320	240
14	Hódmezővásárhely	Grassland	46.436196	20.307521	500	270
15	Hódmezővásárhely	Grassland	46.438144	20.306687	330	200
16	Hódmezővásárhely	Grassland	46.439024	20.308571	400	250
17	Hódmezővásárhely	Grassland	46.437774	20.312524	280	170
18	Hódmezővásárhely	Grassland	46.435331	20.312563	230	150

Statistical Analysis and Quality Control

This analysis used both descriptive and inferential statistics. Descriptive statistical analysis was performed using Microsoft Excel, whereas inferential analysis was conducted using SPSS (Version 22). ANOVA was employed to determine the difference in the abundance of MiP among the three different land uses. Contamination prevention techniques, such as rinsing the apparatus with distilled water three times, were adopted throughout the laboratory processes, during which researchers always wore cotton lab coats and gloves. The bare minimum plastic material was used during the sampling and laboratory analysis. A blank sample was created using distilled water to determine the procedural and environmental contamination levels. To prevent atmospheric contamination, aluminum foil was used throughout, from sampling to the final analysis stages, to cover the samples.

Results

Abundance of MiP

A total of 137 microplastic pieces were found in the study areas. The average microplastic contamination was 7.6 ± 3.4 pieces/L (mean ± standard error). The number of MiP varied across the sampling points. Figure 2 shows that most plastic contaminants were recorded in the landfill (27.50 ± 11.29 pieces/L). The maximum number recorded in any one landfill location was 48 pieces/L in sampling 8, followed by 46 pieces/L in drill 9. The minimum number recorded was 6 in drill 5. The second-highest rate of microplastic contamination was recorded for greenhouse land use (2.88 ± 0.65 pieces/L). Of the nine greenhouse farmland drills, only one recorded zero pieces/L. The highest number, nine pieces, was recorded for both Szeged and Szarvas farmland, while eight pieces were recorded in Szentes greenhouse farmland. One-way ANOVA revealed that there was significant difference in the abundance of MiP in groundwater among the three land use areas: F (2, 15) = 9.87, p = .00. Post hoc testing revealed significant differences for landfill land use (M = 27.50, SD = 22.59), which had a higher abundance of MiP than the greenhouse (M = 2.88, SD = 1.96) and grassland land use areas (M = 0.20, SD = 0.44).

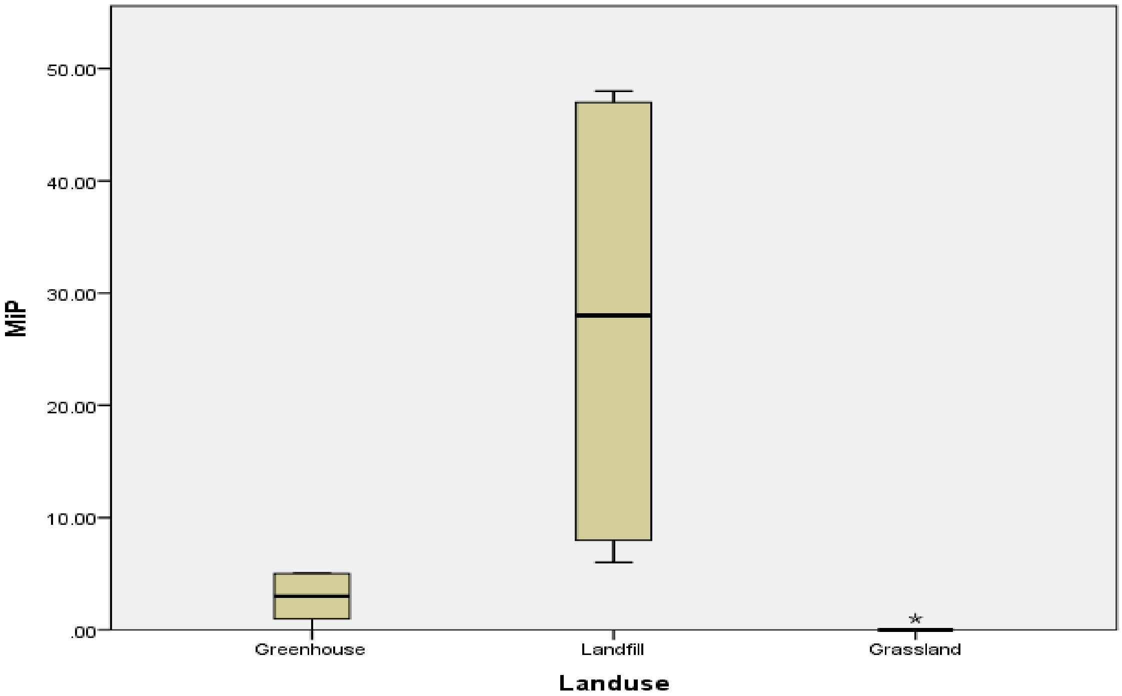

Figure 2: The Abundance of Microplastics in the Groundwater of the Three Land Uses

Morphological Structure of MiP

Figure 3 presents the morphological structure (size, structure, and color) of the plastic contaminants. The size of plastic contaminants was classified into five classes. In the greenhouse farmlands, only two groups were recorded. The 1.2- 2.1 mm size was more common (55%) than the 0.1 -1.0 mm (45%). On the contrary, five classes of microplastic sizes were found in the landfill, 2.2-3.2 mm were the most common structure (32%), followed by 1.2- 2.1 mm (23%), then 0.1 -1.0 mm (20%), and 3.3- 4.3 mm (15%). The least size recorded was 4.3-5 mm (10%). However, only the 0.1 -1.0 mm category (100%) was recorded in grassland farmlands.

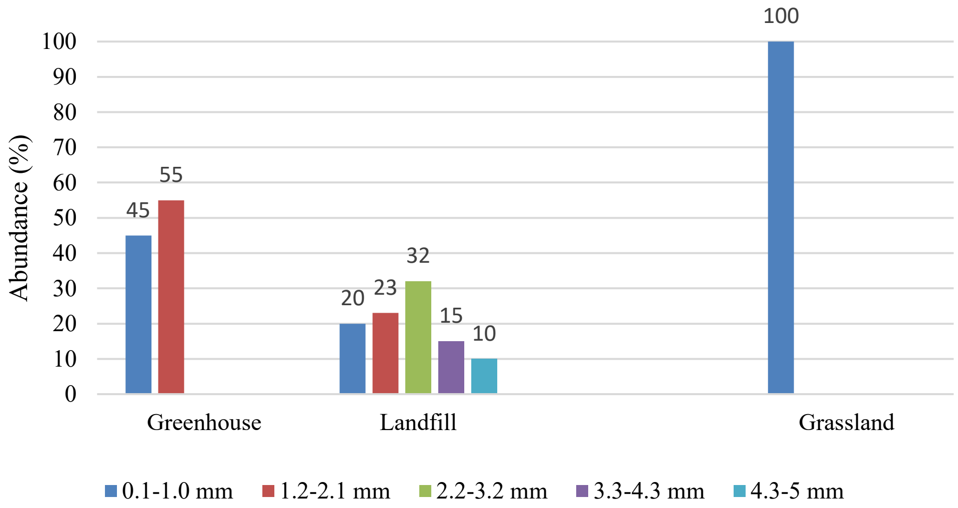

Figure 3: The Sizes of MiP in the Grassland, Greenhouse, and Landfill

Furthermore, the research shows that the microplastic contaminants' structure includes fiber, fragments, and foam (Figure 4). Only two structures were recorded in the groundwater of greenhouse farmlands; microplastic fibers are the most common, which is four times higher than fragments. This research also shows that the landfill microplastic shape includes films, fragments, and fibers. The most contaminants observed were plastic fiber, accounting for 85% while fragments and foam account for 7% each. Only the fiber structure was recorded in the grassland.

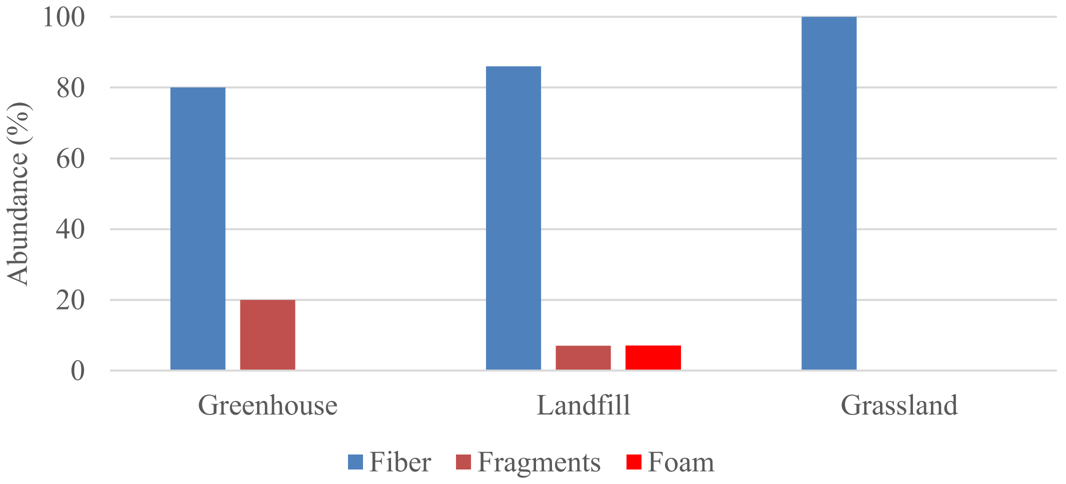

Figure 4: Morphological Structure of Microplastics in the Groundwater

A total of seven colors were recorded throughout the area: Blue, red, grey, white, black, green, and transparent (Figure 5). In the greenhouse land use, blue is the most common color recorded (31%), followed by red and white (25% each), and black was the least recorded (19%). However, more colors were recorded in the landfill land use. Of which, green was the most abundant (60%), followed by yellow (13%). The least abundant color was red (2%). Conversely, only a single color (red) was recorded in the grassland land use.

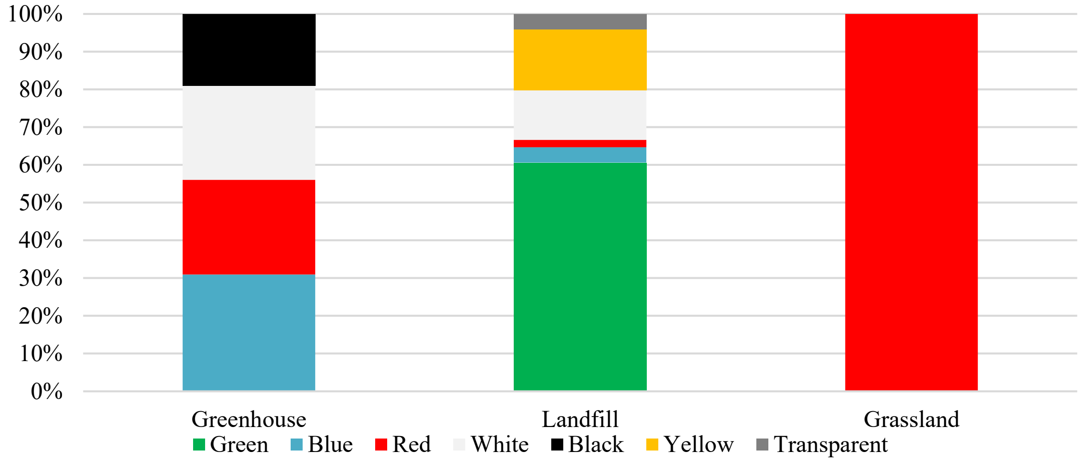

Figure 5: The Colors of MiP in the Grassland, Greenhouse, and Landfill Landuses

Horizontal Movement of MiP

An independent t-test was used to compare the means of landfill and grassland land uses. The results showed that the landfill with four observations (M = 27.50, SE = 11.29) is significantly different with grassland with five observations (M = .20, SD = .20), t (7) = 2.75, p = .02. The individual sample comparisons showed that the landfill samples (sample numbers 10, 11, 12, and 13) contained a substantially higher MiP compared to the grassland samples (sample numbers 14,15,16, 18, and 17).

Discussion

Abundance of MiP

The result of this research shows that the overall average microplastic contamination was 7.6 ± 3.4 pieces/L in the entire area. One-way ANOVA revealed that there are significant differences in the abundance of MiP in the groundwater. Higher MiP contaminants were recorded in the landfills, followed by the greenhouse, and then the grassland. The result of the presence of an abundance of microplastics in landfill land use agrees with the findings of Afrin *et al* (2020), which was conducted in the urban landfill landuse of Bangladesh. The result found that it contained a high abundance of microplastic contaminants. However, the high concentration of MiP in the landfill area was related to the contribution of landfill to the MiP storage as well as the homogenous nature of the landfill materials (Afrin *et al*, 2020). Another reason for the abundance might be because of the long depth of landfill samples in the study area which is 320 cm – 350 cm (Table 1). Lastly, the reason might be attributed to the duration of landfills in the area, which has been in existence since the 1990s. This study shows that the landfill is the main source and pathway of transporting MiP contaminants to the groundwater.

On the other hand, the result on the presence of MiP in greenhouse land use confirmed the finding of previous finding that reported the presence of MiP in the greenhouse, vegetable production and mulch farmlands. (Saadu and Farsang, 2022; Bigalke *et al*., 2022; Wang *et al*., 2022). The reason of high accumulation of MiP in the greenhouse farming system is that it is characterized by generating huge plastic contaminants in different shapes and sizes (Wang et al, 2022). Also, the plastic contaminants enter the greenhouse environment as a result of direct applications of water and fertilizer, and the fragmentation of larger plastic materials, such as films and fibers for greenhouse structure (Saadu & Farsang, 2022). The low abundance of MiP in the greenhouse land use is compared to landfill because the landfill is one of the major sinks of plastic waste contaminants, as it can store 21-42% of plastic waste globally (Afrin *et al*., 2020).

The result of the low incidence of MiP contaminants in the grassland agrees with the findings of Haixin *et al*. (2022), which was conducted in the Qinghai-Tibetan Plateau, China. The result found that grassland contained less microplastic contaminants compared to orchard and farmland land uses. Similarly, Ding *et al*. (2021) reported that grassland contained less microplastic than sand land but had higher microplastic content compared to woodland. The reason for abundance may have been the result of the low impact of anthropogenic activities in grassland areas. Another important pathway of MiP to grassland is surface water runoff from the nearby roads to the grassland areas as this encourages high inflow of MiP contaminants into the grassland land use (Choi *et al*., 2021).

However, MiP in the greenhouse and grassland enrironment penetrates to the groundwater because of infiltration of irrigation water, natural rainfall, leaching, soil texture and presence of cracks in the soil (Bigalke *et al*., 2022; Hiaxin *et al*., 2022). However, no significant amount of MiP was recorded in the control samples of both areas.

Characteristics of MiP Debris

The result of morphological structure in the greenhouse landuse shows that 1.2 – 2.1 mm category was higher (55%) compared to 0.1-1.0 mm (45%). This data shows that the plastic materials use for greenhouse farming fragment and form smaller pieces. The main reason for this fragmentation is because of the easily ageing of plastic materials in the greenhouse environment due to the effect of climatic variables such as solar radiation, wind, precipitation, and atmospheric contaminats (Andrady, 2003). The rate of ageing is also increased by the stress caused by agrochemicals (Vox et al, 2008). However, only fiber and fragment were recorded in the greenhouse land use in terms of structure. Fiber accounted for 80% while the fragments accounted for 20%. This result confirmed with many findings that recorded plastic fiber in the surface and groundwater environment (Panno *et al.*, 2019, Shen *et al.*, 2020; Bikalge *et al.*, 2022). Microplastic fibers were frequent observed because the fibrous materials have the highest mobility in the vertical direction of soils (Zhang *et al*., 2022). Moreover, a large number of microplastics in the greenhouse groundwater appeared in blue then white and red. This might be as result of fragmentation of greenhouse larger plastic materials.

On the other hand, more size categories were found in the landfill land use. The most abundant size was 2.2 -3.2 mm (32%) followed by 1.2 -2.1 mm (23%). The least size recorded was 4.3 -5 mm (10%) followed by 3.3 - 4.3 mm (15%). Smaller size MiPs have high chances of migration into the groundwater ecosystem. However, of the three structures found in the MiP of landfill, fragments accounted for over 85% of the total structures followed by foam and fiber. Unlike other land uses, different color of MiP were recorded in the landfill land use, the dominants followed by white and yellow. The reason for high disparity in size, shape and colour of MiP in the landfill landuse is because of heterogeneity nature of landfill materials (Afrin *et al*., 2020).

Contrarily, only one size category was recorded in the grassland land 0.1 – 1.0 mm. Similarly, only a red MiP fiber were recorded. The reason for high disparity compared to the land uses may be due to the very few contaminants found in the greenhouse farmland as reported by previous findings (Ding *et al*., 2021; Hiaxin *et al*., 2022). Also, the presence of MiP in grassland groundwater might have been the result of the ubiquitous nature of plastics. In support of this result, Su *et al*. (2022) and Panno *et al*. (2020) found MiP in the groundwater of uncultivated lands in the United States and South Korea, respectively. Similarly, MiP have been found at depth in forest, grasslands, and facility soils of South Korea and China respectively (Choi *et al*., 2020; Haixin *et al*., 2022). MiP have been found to penetrate soils via cracks in the soil, infiltration, and other contamination sources (Mora *et al*., 2021; O'Connor *et al*., 2019; Panno *et al*., 2019; Su *et al*., 2021).

Horizontal Movement of MiP

The horizontal movement of MiP was assessed by comparing the results for the landfill and grassland land use areas. The two areas were sampled at the same time to minimize errors due to temporal differences. The two land uses are located close to each other of about 450 meters, but a river and waterways separate them. An independent t-test was used to compare the means of these land uses. The results showed that the landfill with four observations (M = 27.50, SE = 11.29) is significantly different with grassland with five observations (M = .20, SD = .20), t(7) = 2.75, p = .02. Additionally, the morphological structures of the contaminants were studied to determine the relationship between the two land uses. The individual sample comparisons showed that the landfill samples (sample numbers 10, 11, 12, and 13) contained a substantially higher amount of MiP compared to the grassland samples (sample numbers 14,15,16, 18, and 17).

The result of the landfill and grasslands were compared because the two land uses are located close to each other of about 450 meters, but they are separated by a river and waterways. There appeared to be no apparent connection or relationship between landfill land use and grassland land use. The MiPS found in the landfill were quite different from the one found in the grassland land use because large fragments and variety of colors were found in the landfill land use. However, in contrast, only minimal fiber was found in the grassland groundwater and control samples. Therefore, the data shows that there is no horizontal movement of MiP from landfills to grassland.

Conclusion

This study is among the first to quantify, characterize, and compare microplastic contamination in groundwater in greenhouse farmlands, landfill land use and grasslands areas. MiP were found in all three land use areas. The concentration was greatest in the landfill area, followed by the greenhouse and grassland areas. The plastic contaminants were mainly microplastic fibers, fragments, and foam of different sizes and colour. The results of this study indicate that there is no evidence for the horizontal movement of MiP from high contaminated regions to low contaminated regions. Given that microplastic particles were found in the groundwater of the areas (mostly a landfill region). Hence, groundwater from such areas must be treated before human consumption and irrigation use to reduce the microplastic load in humans and agricultural soils. Additionally, farmers and stakeholders must dispose of plastics from greenhouse farming areas. Finally, this research provides insights that could lead to further research on microplastic contamination in groundwater.

Acknowledgments

We thank the owners of the Flóratom Kft. (Szeged) greenhouse farmlands at Szarvas and Szentes for granting us access to their respective farmlands. We also acknowledge the efforts of Gabor, Fabian, and Szilveszter, who helped with the fieldwork. Lastly, we thank Dr. Krisztián Fintor of the Department of Mineralogy Geochemistry and Petrology, University of Szeged, for his assistance with the Raman spectroscopy analysis.

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