VERTICAL DISTRIBUTION OF MICROPLASTICS & HEAVY METALS IN LANDFILL SOIL AND GROUNDWATER

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ABSTRACT

Microplastic(MP), an emerging environmental contaminant, along with heavy metals are raising concerns due to their potential negative impacts on ecosystems and human health. As the ultimate repository for municipal solid waste, landfills have grown to be a substantial sink and hence, a further point source of these harmful pollutants for the nearby environment. In Bangladesh, despite having a high possibility of MP contamination, the evidence on water and soil environment is less explored. To better understand their destiny and possible co-transportation, this study aims to portray the vertical distribution of MPs and heavy metals in landfill soil and groundwater at the Anandabazar dumping site in Halishahar, Chattogram which is close to the new residential and commercial developments. Soil samples were collected from different depths ranging from 3m to 20m in three boreholes. Groundwater samples were collected from shallow tube wells located within a 500m radius of the dumping site. Additionally, agricultural soil and product samples were collected from two lands in the vicinity. All the soil and groundwater samples were preserved, prepared, and analyzed maintaining standard protocol followed by FT-IR Spectroscopy for MPs and Atomic Absorption Spectrometry for selected heavy metals. Though quantification of MPs could not be executed, dominant MPs identified were Aliphatic Nitriles, Vinyl Polymers, and Silicon-Oxy Polymers with their substantial vertical stratification. Heavy metal concentrations were shown in increasing quantities in soil, and agricultural products than water in order of magnitude. The highest concentration of 272.8 mg/kg of Fe was found in borehole soils, while the lowest of 0.8mg/kg of Cr. The groundwater samples do not exhibit heavy metal concentrations of concern. The findings show that although there exists no structured pattern in the vertical distribution of MPs in different soil layers, the dominant group was mostly found in silt layers whereas clay layers contained Silicon-Oxy polymers. Also, the horizontal distribution of MPs in the case of groundwater and agricultural soils showed no significant variation concerning distance from the landfill site. The study is continued and hope to discuss further outcomes in the future. However, the present investigation revealed the potential of MPs and heavy metal-induced pollution that demand immediate attention in designing city-wise sanitary landfill sites, that are now acting as dumping sites and are prone to induce environmental hazards with time.

Keywords: *AAS, FT-IR spectroscopy, Heavy metals, Landfill, Micro-plastic,*

1. INTRODUCTION

Microplastics (MPs) are tiny plastic particles (less than 5mm in size) that are present throughout the environment, including terrestrial ecosystems, aquatic systems, and even the atmosphere. Their pervasiveness in the environment has sparked worries about their lingering effects on aquatic and terrestrial ecosystems, as well as human health. There are MPs in the world's oceans, rivers, lakes, sediment, air, and even the most remote and pristine regions. The staggering statistics indicate that billions or even trillions of MP particles are scattered throughout the environment. MPs have also been found in terrestrial ecosystems, such as urban systems and agricultural soils. Compared to aquatic habitats, there has been less research on MPs in soil, although newer studies indicate that they are common. In Bangladesh, an investigation of microplastic contamination in urban and rural soils discovered considerable microplastic contamination in both settings (Hossain et al., 2020). MPs were found in soil from agricultural areas (Pérez-Reverón et al., 2022), demonstrating their prevalence in soil ecosystems. And this presence of MPs in the soil ecosystem greatly puts the soil ecosystem in jeopardy.

Landfills are one of the potential sources of MP which endangers its surrounding environmental components. Landfills experience chemical and physical changes brought on by the disintegration of solid waste refuse with the soil matrix. Rainwater's biological, chemical, and physical processes within solid waste refuse are aided by liquid percolating through the matrix of the debris consequently forming leachate. Leachate is a chemical mixture made up of dissolved organic matter, xenobiotic organic compounds, various anions and cations, and heavy metals (Kamaruddin et al., 2017). The primary threat associated with municipal landfills is the prospect of pollution posed by leachate mobilization into surface and groundwater via the subsoil. But it is also evident in some studies that soil acts as a filter medium for leachate which retains MPs in its complex matrix and reduces groundwater contamination (Viaroli et al., 2022).

Landfills also being a potential source of metal contamination, produces harmful metals during the decomposition of garbage in landfills, contaminating nearby soil, groundwater, and surface water. These heavy metals come from a number of natural and man-made sources. The leachate collects these toxic elements and takes it into the soil acting as a carrier. Eventually the leachate can permeate deep enough to reach the ground water and contaminate it. Several studies done globally revealed that landfill leachate is a dominant source of heavy metals in groundwater (Qi et al., 2018) (Alemayehu et al., 2019). An investigation on the distribution of heavy metals in a dumping site in Sylhet, Bangladesh found that the amounts of heavy metals varied significantly across the dump site, with the concentrations found increasing towards the dumping area, specially, the top layer of soil (Alam et al., 2020) (Essien et al., 2022). In another study, the distribution of heavy metals in groundwater close to an active landfill in Dhaka City revealed the quantity of heavy metals in groundwater to be higher in the vicinity of the dump site and decreased with increasing distance (Azim et al., 1970). There are also several studies on agricultural soil and products that detected heavy metal that exceeded the allowable limit set by the international guideline (Ahmad & Goni, 2010).

Leachate, surface runoff, and wind dispersion are three ways that MPs might leave landfills and enter nearby ecosystems (Horton et al., 2017). This emphasizes the need for efficient waste management techniques that can reduce their discharge into the environment of MPs and heavy metal pollution. Proper landfill management, such as efficient leachate treatment, lining beneath the landfill and regular monitoring, can limit the release of pollutants into the environment and prevent the contamination of groundwater. But in developing countries like Bangladesh, most of the dumping sites are non-engineered landfills. The most common method of waste disposal in developing nations, including Bangladesh (Azim et al., 1970) (Parvin & Tareq, 2021), and even in parts of developed nations (Dotaniya et al., 2017), is the dumping of non-segregated solid waste into landfill sites. The surrounding environmental components are significantly impacted by the inappropriate management of landfills and production of hazardous leachate (Dotaniya et al., 2017) (Megremi et al., 2019).

This study aims to assess pollution around landfills by analysing MPs and heavy metal concentrations in soil and groundwater at various depths. The effectiveness of pollutant filtration is influenced by soil type. The main focus is on identifying MPs, establishing heavy metal concentrations, and examining their potential to enter into the human food chain through groundwater and agricultural products.

2. METHODOLOGY

2.1.1 Study Area

The landfill area is located at Anandabazar, Halishahar in the northern part (22.312839°N, 91.772701°E) of Chattogram, a designated site for waste disposal by Chattogram City Corporation. It serves as one of the repositories for solid waste generated within the city. It is small in size and has an area of ten acres. The dumping yards have no provision of leachate collection. The Anandabazar landfill site receives about half of Chittagong's daily generated waste, currently reaching a peak height of 53', well above the safe height of 13'.

2.2 Sample Collection

2.2.1 Borehole Soil

The soil samples were collected from three boreholes dug near the landfill in the post-monsoon season. The boreholes B-1, B-2 and B-3 in the site are shown in Figure 1. This particular site is located within 500 meters of the targeted landfill. The soil stratification of the boreholes is shown in Table 1. The surface soils are generally fine-grained soils with low plasticity.

Table 1: Soil Stratification

Borehole Number	Vertical Distance From Ground Surface (meter)	Soil Classification
B-1	0 m – 3 m	Clayey Soil; Soft
	3 m - 19 m	Silty Soil; Loose
	19 m – 22 m	Silty Sand; Medium Dense
B-2	0 m – 5.5 m	Clayey Soil; Soft
	5.5 m - 20 m	Silty Soil; Loose
B-3	0 m – 6 m	Clayey Soil; Soft
	6 m – 18 m	Silty Soil; Medium Dense
	18 m – 21 m	Clayey Soil; Soft

2.2.2 Groundwater

All the samples were collected from ground water sources. Figure 1 shows the distribution of the sampling points across the sampling area. The water samples were collected in glass containers from shallow tube wells. Approximately 500 ml of ground water samples were collected from each source, designated as TW-1, TW-2, TW-3, TW-4, TW-5 and carried to the laboratory. Specific site details of the mentioned sources are given in Table 2 below.

Table 2: Details of groundwater sources at site

Sample Designation	Distance from Landfill (m)	Location
TW-1	0	22.313086N, 91.771688E
TW-2	112.65	22.313811N, 91.771122E
TW-3	160.93	22.313923N, 91.771688E
TW-4	257.5	22.315552N, 91.771466E
TW-5	450.65	22.316610N, 91.770833E

2.2.3 Agricultural Soil & Products

The agricultural lands are adjacent to the landfill. The local residents are directly involved with farming in these lands and the products are consumed across the city. The agricultural soil and product (Tomatoes) samples were collected from two fields using shovel. A total of six soil sample and two sets of Tomatoes (three soil samples and one set of tomatoes from each field) were collected from both the lands. Afterwards, those were carried in alum foils and taken to the laboratory. The locations of the samples at site are white marked in Figure 1.

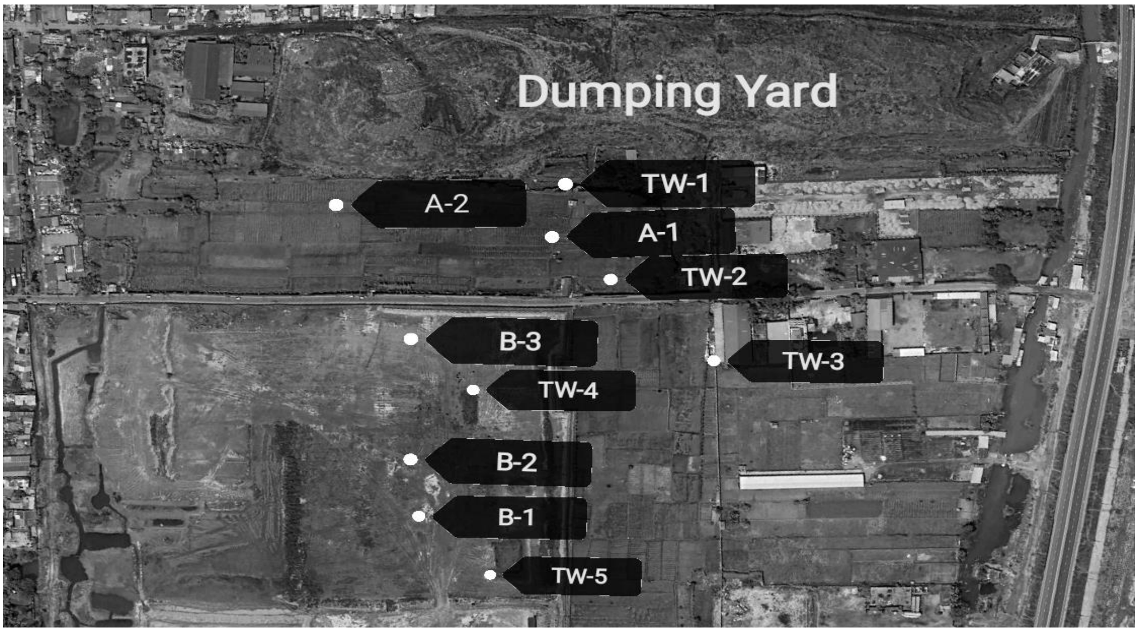

Figure 1: Designated Sampling points at site

2.3 Sample Preparation

2.3.1 Soil Samples

The general sample preparation steps for MPs in soils include drying, grinding, and sieving. Air-drying (25–35 ℃) was carried out for all the soil samples of boreholes and agricultural lands. Then 200g of samples were sieved through no. 200 sieve (4.75 mm) followed by fine grinding into powder. Composite samples were prepared by vigorous mixing of soil samples from two consecutive depths until homogeneity was achieved. Table 3 and Table 4 represent the details about composite sampling of borehole and agricultural soils respectively. Finally, 10mg from each sample were wrapped with alum foil and stored in clean, tiny Polyethylene pouches for further analysis.

In case of heavy metals, individual samples were used for borehole soils and composite samples for agricultural soils. The soil samples from both sources were dried in room temperature, ground to homogenized powder and sieved through no. 200 sieve (4.75mm). There are numerous and varied methods of sediment bound metal extraction procedures reported in the literature. To keep the metal extraction procedure consistent and logistically efficient, this study undertook the digestion process incorporating Aqua regia, a mixture of HNO_3 & HCl in 3:1 ratio, with the soil samples and heated at 60ºC till white flames were visible. The samples were then cooled, filtered through Whatman filter paper no. 42 and stored in plastic bottles.

Table 3: Composite sample details of borehole soils

Borehole	Depth of individual Sample (m)	Depth Span of Composite Sample (m)	Sample Designation
B1	5	5 to 15	BS-11
	15		
	30	30 to 45	BS-12
	45		
	55	55 to 70	BS-13
	70		
B2	3	3 to 7.5	BS-21
	7.5		
	10.5	10.5 to 13.5	BS-22
	13.5		
	16.5	16.5 to 19.5	BS-23
	19.5		
B3	5	5 to 15	BS-31
	15		
	25	25 to 40	BS-32
	40		
	55	55 to 65	BS-33
	65		

Table 4: Composite Sample details of agricultural Soil

Agricultural Field	Collected Soil Sample	Designation of Composite Soil Sample
A1	1	AS-1
	2	
	3	
A2	4	AS-2
	5	
	6	

2.3.2 Groundwater Samples

Though quantification of MPs demands filtration process prior to microscopic analysis, this study is limited within qualitative identification only. We collected two replicas of each sample and stored in 50 mL glass jars rinsed with distilled water and subsequently refrigerated at 4ºC for FT-IR analysis.

Same procedure is applicable for heavy metals detection in water samples. As visible suspended particles were present, the samples were analysed by Atomic Absorption Spectrometry (AAS) following a prior filtration process by Whatman filter paper no. 42.

2.3.3 Agricultural Products

The agricultural products (Tomatoes) were washed with tap water to eliminate soil and dust particles. The samples were chopped into small pieces and oven dried at 70ºC for 96 hours (Murthy HC, 2019). Then the dried samples were ground into fine powder and stored in clean plastic pouches for chemical analysis. The digestion procedure is same as soil samples.

All the materials and vessels were rinsed for three times with distilled water before their use and they were covered with aluminium foil after each step. Cotton lab coats and nitrile gloves were worn during the whole process. Cross contamination was avoided as much as possible.

2.4 Analytical Methods

A widely employed approach for detecting and categorizing MPs in soil and water samples is Fourier Transform Infrared (FTIR) analysis. Target MPs in a sample and their constituent polymer types can be identified by comparing referenced spectrum libraries. FT-IR spectra were processed by KnowItAll@Wiley's software. The spectra were enhanced by using baseline correction and normalization approaches. The sample spectra were compared to reference spectra in the software library to identify plastic materials, such as polymers like polyethylene (PE), polypropylene (PP), polystyrene, or PS, and polyethylene terephthalate (PET) etc. The distinctive peaks and absorption bands that signify the presence of different types of MPs in the sample spectra were identified.

All the digested soil samples from boreholes and agricultural fields and tomato samples were made up to 50ml volumetric flask using deionized water. The filtered water samples were directly analysed. Calibration standards were prepared through serial dilution of standard stock solution of multi-elements having concentrations of 1,000 mg/L. Blank solutions (solvent only) were aspirated into the AAS instrument and instrument was made to zero. Then the prepared sample solutions were placed into the AAS instrument and absorbance were measured. Using the calibration curve, the concentrations of the elements in the sample were measured based on individual absorbance. Any sample that gave a reading beyond the calibrated ranges was diluted until a satisfactory reading was obtained. Deionized water was used throughout for cleaning apparatus, preparing standard solutions and other related purposes of laboratory work in order to avoid cross contamination.

3. RESULTS & DISCUSSIONS

3.1 Microplastics

3.1.1 MP's In Groundwater

The data generated from FT-IR spectroscopy is represented in a graphical manner and a sample graph is shown below in Figure 2 for TW-2 water sample.

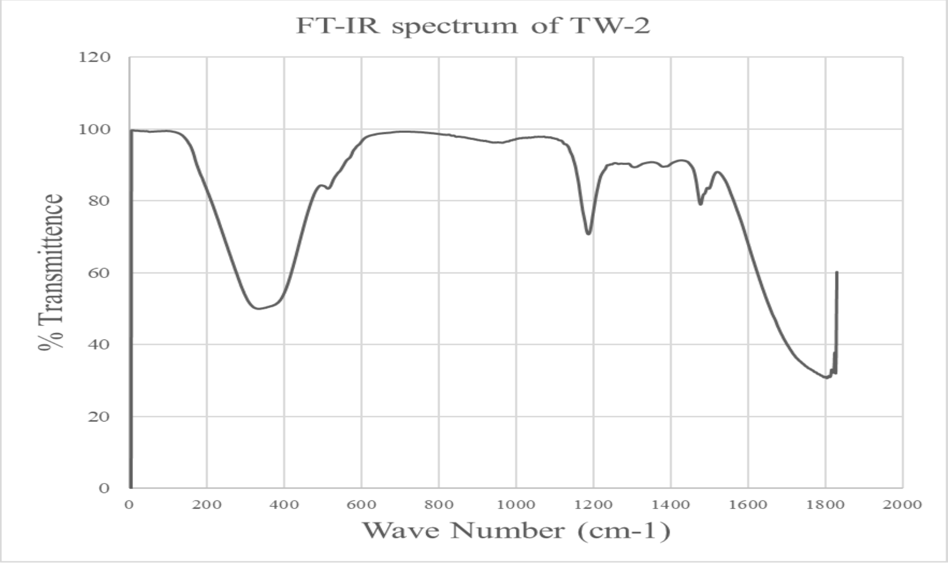

Figure 2: FT-IR spectrum for TW-2 water sample

All the FT-IR data were analyzed by KnowItAll@Wiley’s software and spectrum library. An overall representation of MPs found in all 5 groundwater samples is given in the Table 5 below.

Table 5 Identification of MPs in Groundwater Samples

Sample Designation	**TW-1**	**TW-2**	**TW-3**	**TW-4**	**TW-5**
Distance from Landfill (m)	0	112.65	160.93	257.5	450.65
MPs Identified	LDPE	LDPE	LDPE	LDPE	LDPE
		HDPE	HDPE		HDPE
		Polyamides	Polyamides	HDPE	Polyamides
	HDPE	Polyureas	Polyureas		Polyureas
		Fluoro-Polymer	Fluoro-Polymer	Polyamides	Fluoro- Polymer
		Resins	Resins	Polyureas	Resins

Table-6 shows the ubiquity of MPs in groundwater samples. These plastics may be originated from varieties of sources from the dumped wastes some of which have been mentioned in Table 6. A careful insight in the results leads to the understanding that the water samples are contaminated with almost similar type of MPs irrespective of their distance from landfill site. It can be presumed that the MPs found in the water samples may be more susceptible to fragmentation compared to other ones which makes it more vulnerable in terms of intrusion in human body or plants. Also, it shows that the landfill leachate can percolate deep into the soil layers and consequently reach groundwater systems. Another important issue that needs to be addressed is that groundwater should not be withdrawn from a certain radius of the dumping point. If necessary, it can be lined at the bottom depending on the type of soil and wastes dumped. More detailed investigations and quantification of MPs are required to confirm the raised problems and proposed suggestions.

3.1.2 MPs in Borehole Soils

Table 7 below depicts the current picture of MPs pollution in landfill soils. Varieties of plastics are likely to be present in the soils from the boreholes B-1, B-2 and B-3. Though analysis was confined to particular range of depths only, MPs were still found to be ubiquitous throughout the boreholes up to 20m depth. Individual analysis revealed that the topmost layers of boreholes B-1 and B-3 (1.5m to 4.5m) contains Silicon-Oxy compounds probably originating from rubber and resin type polymers whereas it is LDPEs and HDPEs in case of B-2. The second examined layers in B-1 and B-2 are abundant with Aliphatic Nitrile polymers, Vinyl polymers, Silicon-Oxy polymers in contrary of which the similar layer of B-3 contains Aliphatic Epoxides and Phosphazenes which were unique among all the soil and water samples. The next tested layers of all three boreholes show different results. In B-1, a wide range of layer (16.5m to 21.5m) carries halogen bound compounds probably indicating halogen polymers like PVCs, PVDCs, PBDEs, BPS and so on. B-2 was again found to be carrying LDPE and HDPEs whereas B-3 had bounty of Aliphatic Nitrile polymers, Vinyl polymers, Silicon - Oxy polymers.

Table 6: Identification of MPs in borehole soil samples

Borehole No	**Depth (m)**	**Functional Groups**	**Probable Sources**
B-3	15-20	Aliphatic Nitriles	Pharmaceuticals, Agrochemicals, Acrylic Fibres
		Vinyl Polymers	PVC, PVDC from Pipes fittings, Medical Wastes
		Silicones	Resin, Rubbers, Gels
	7.5-12	Aliphatic Epoxides	Polyurethane foams, , Electrical Insulators
		Polyphosphazenes	Automobile & Biomedical Products
	1.5-7.5	Not Found	Not Found
B-2	16.5-19.5	Aliphatic Hydrocarbons	LDPE, HDPE
	10.5-13.5	Aliphatic Nitriles	Pharmaceuticals, Agrochemicals, Acrylic Fibers
		Vinyl Polymers	PVC, PVDC from Pipes fittings, Medical Wastes
		Silicones	Resin, Rubbers, Gels
		Alkyl Phosphonates	Chealatings agents, Surface modifiers
	3-7.5	Aliphatic Hydrocarbons	LDPE, HDPE
B-1	16.5-21.5	Halogen based Polymer	PBDE, BPS (Polyester), Brominated Epoxy Resins.
		Phosphazenes	Automotive & Biomedical products
	9-13.5	Aliphatic Nitriles	Pharmaceuticals, Agrochemicals, Acrylic Fibers
		Vinyle Polymers	PVC, PVDC from Pipes fittings, Medical Wastes
		Silicones	Resin, Rubbers, Gels
	1.5-4.5	Silicones	Rubber, Resin, Gels

The figure below shows the vertical stratification of MPs with the details of soil layers. A thorough analysis of the stratification shows that there is neither any common trend of MPs distribution pattern in the soil layers nor it varies with soil type. Still a generalization can be made by considering hypothetical horizontal layers and that is the topmost layer of each borehole has clayey soils and contains Silicon-Oxy polymers. The examined mid layers of B-1 and B-2 mostly contain Aliphatic Nitrile polymers, Vinyl polymers, Silicon -Oxy polymers where the soil type is silt, though it slightly varied in B-3 may be due to other geological or environmental factors. Further analysis is required to find the types of MPs and their relationship with soil type by testing soil samples from more boreholes and depths and MPs quantification considering all other factors.

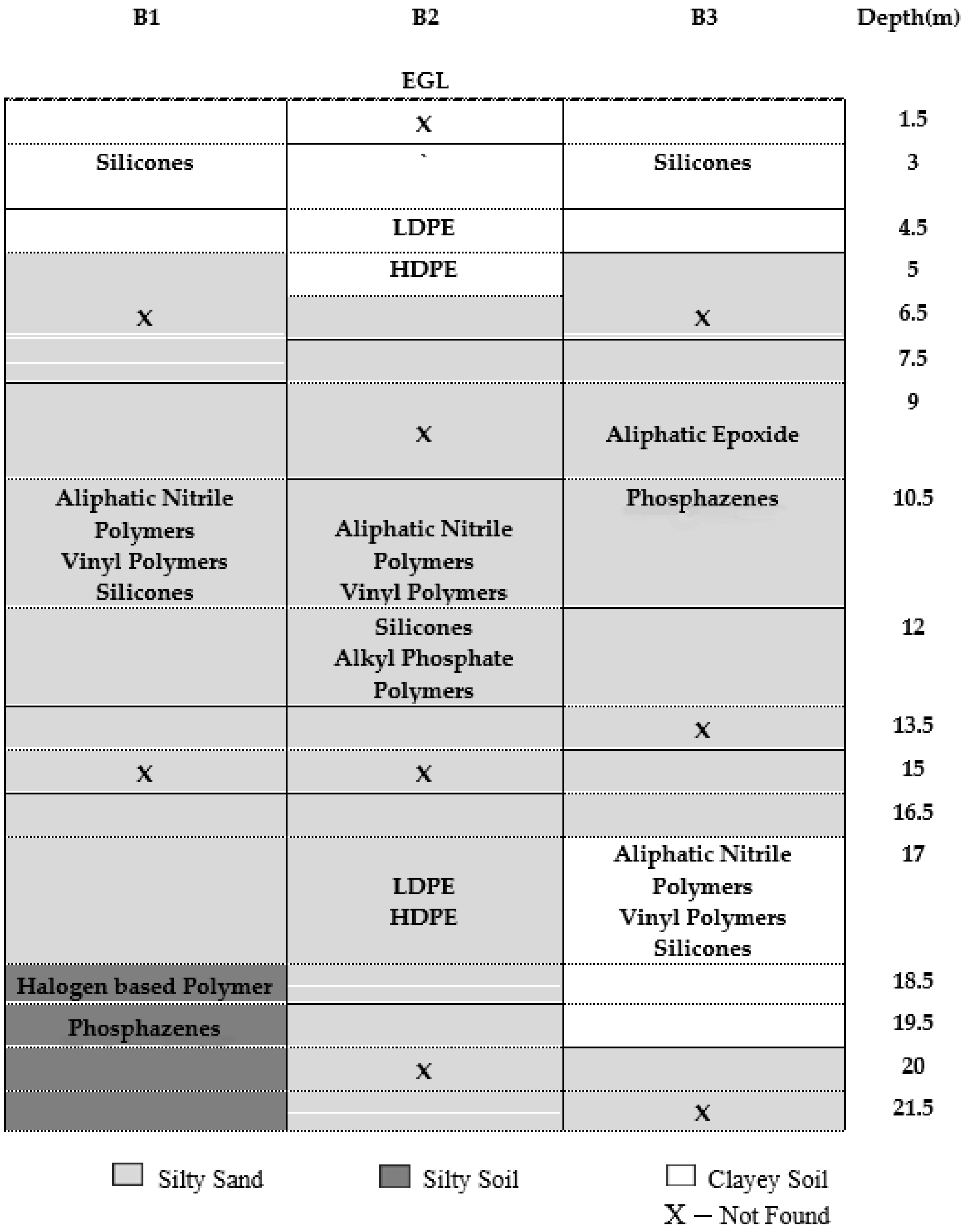

Figure 3: Vertical stratification of MPs in different soil layers

3.1.3 MPs in Agricultural Soils

Both the agricultural fields A-1 and A-2 irrespective of distance from landfill were found to have Silicon-Oxy polymers. The reasonable assumption of it is that the samples were collected from terrestrial soils only which are mostly clayey which resembles with the MPs distribution pattern in soil samples from boreholes previously discussed.

Table 7: Identified MPs in Agricultural Soils

Designated Samples	**Functional Groups**	**Probable Sources**
AS -1	Silicones	Rubber, Resin, Gels
AS -2	Silicones	Rubber, Resin, Gels

3.2 Heavy Metals

3.2.1 Heavy Metals in Groundwater

Though the groundwater samples were found to have varieties of MPs, the concentration of tested heavy metals were relatively low. The concentrations of Chromium (Cr) were 0.025, 0,018, 0.018, 0.010 and 0 mg/L, Iron (Fe) of 0.2, 0.14, 0.12, 0.15, 0.16 mg/L and Copper (Cu) concentrations were 0.09, 0.05, 0.04, 0.14, 0.15 mg/L. Nitrate (NO_3) contents were found to be 7.4, 4.5, 1.8, 0.5 mg/L in the water samples TW-1, TW-2, TW-3, TW-4, TW-5 respectively. Cr and NO_3 concentrations demonstrate a decreasing manner with distances from the landfill. The probable cause of less accumulation of heavy metals in groundwater samples might be that metals are more likely to be attracted by sediments rather than water. That's why the heavy metal contamination level is much lower than the contamination level in soils.

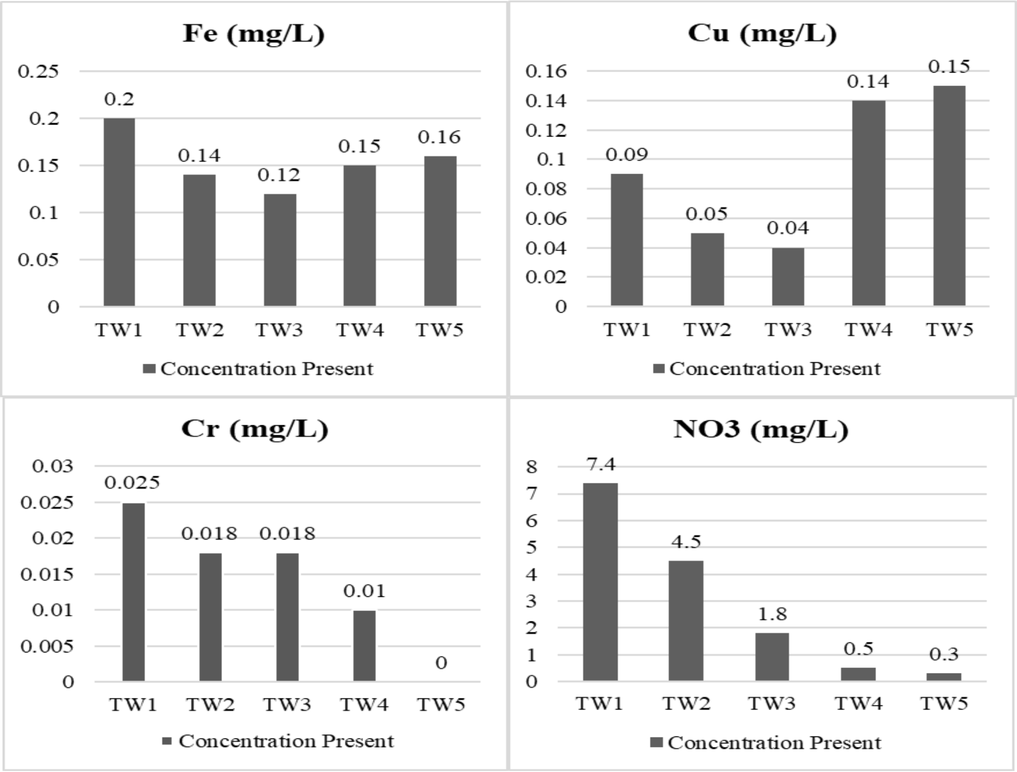

Figure 4: Heavy Metal Concentrations in Groundwater Samples

3.2.2 Heavy Metals in Borehole Soils

The borehole soil samples were found to be substantially contaminated with heavy metals. The heavy metals contamination levels at corresponding depths in the three boreholes are shown by bar charts in Figure 5. An in-depth vigilance in the obtained results shows opposite accumulation tendencies of Cu and Fe at different depths. Cu were higher in concentration where Fe found to be the least and vice-versa. Another important observation was that Cu was found to be in higher concentration in clay layers whereas the amount of Fe was higher in silt soils. The most probable reason might be that Fe is more likely to be sand bound metal than Cu. The same trend as Cu is noticed for Cr. The overall concentration of Fe seemed to be uniform in all the boreholes. Cr concentration decreased with depth.

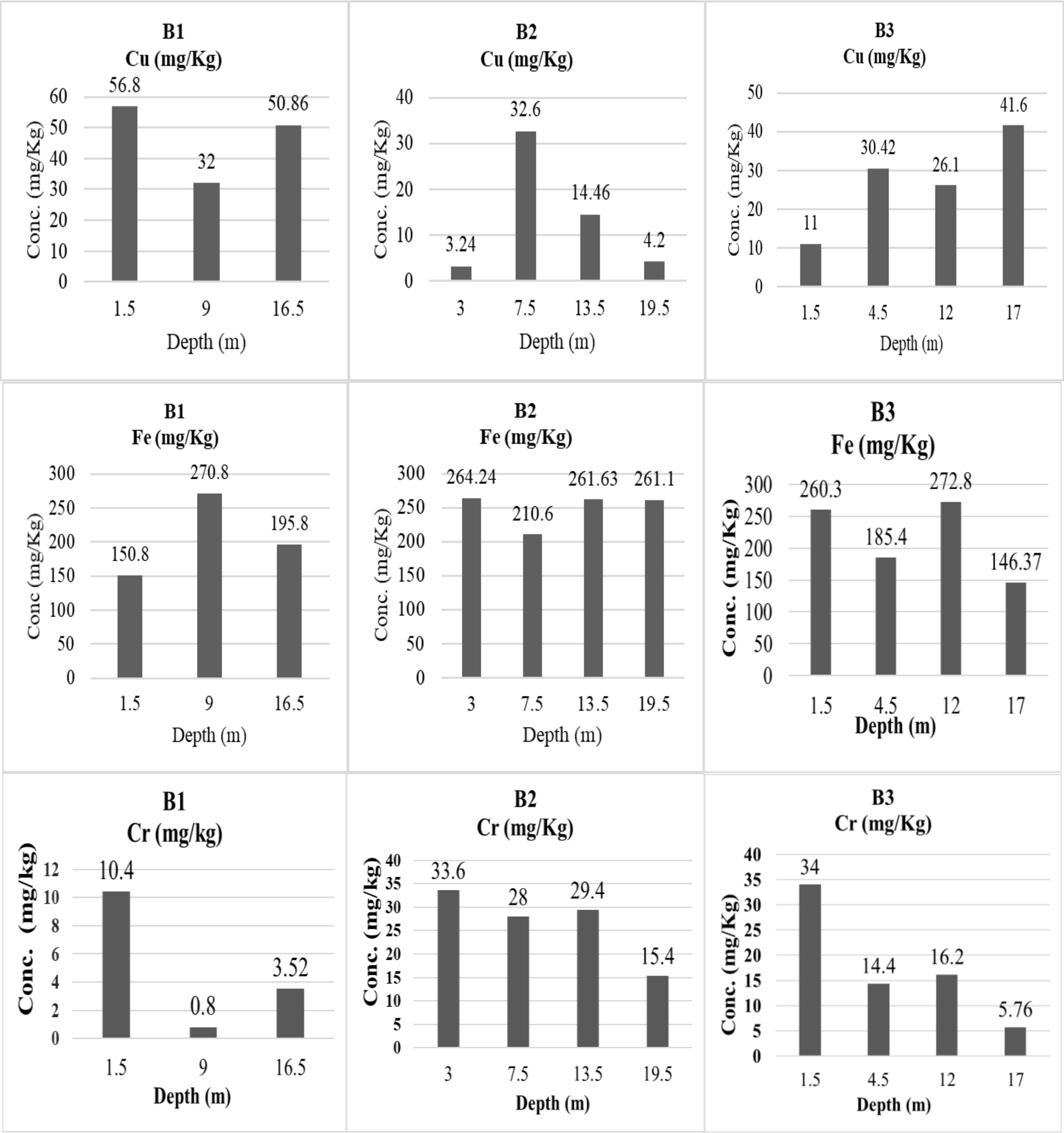

Figure 5: Concentration of Cu, Fe & Cr in Borehole Soil

3.2.3 Heavy Metals in Agricultural Soils & Products

The agricultural soils were found to be conspicuously contaminated having higher concentrations of heavy metals. The two sample locations have almost similar amount of Cu, Fe and Cr as it is seen in the bar charts in Figure 6. As the samples were collected from terrestrial soils closer to the dumping site than the boreholes, the metal concentrations were significantly greater.

In case of tomatoes, the concentrations are almost close to permissible values indicating higher contamination level and further potential of health risks. The concentration of Cu, Fe and Cr were found to be 105.2 mg/Kg, 320 mg/Kg and 0.47 mg/Kg respectively. The recommended values by FAO and WHO for these three metals are 73.3 mg/kg, 425.5 mg/Kg and 1.3 mg/Kg. Concentration of Cu has already exceeded the standard values. The generated leachate from landfill flows through the nearby soils as run-off and the existing plants absorb the heavy metal containing water for their growth. It can be considered as a signal to imminent danger of serious health hazards if cultivation is continued beside landfill.

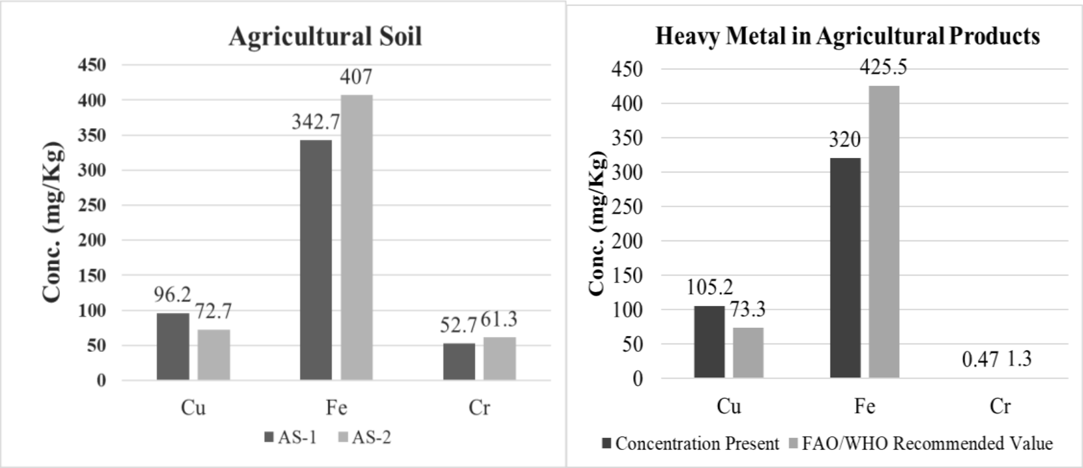

Figure 6: Concentration of heavy metals in agricultural soil & products

4. CONCLUSIONS

The study comprehensively assessed traits, analytical approaches, and implications of Microplastics (MPs) and heavy metal contamination in landfill soils and groundwater in Bangladesh. MPs were found both in all the soils and groundwater samples at different depths. A more extensive study is needed, including quantification of MPs considering factors like soil properties, structures, sample types, aging processes, and polymer composition to understand their variability in the context of soil type. The ubiquity of MPs and heavy metals in nearby landfill soils and groundwater suggest that landfill leachate carries these toxins and contaminate groundwater. Vertical distribution of MPs in soil lacks any specific pattern, but assumptions made as silt and clay layers may have affinity towards specific polymer kinds. Horizontal distribution of MPs in groundwater shows a consistent pattern regardless of distance from the landfill.

Heavy metals were found in both borehole soils and groundwater with higher contamination levels in soils. Iron (Fe) was uniformly concentrated in soils with highest and lowest concentrations of 272.8 mg/L and 146.37 mg/L respectively, while other metals showed variations. In groundwater, Cr and Nitrate concentrations showed a descending order with average values of 0.0142 mg/L and 2.9 mg/L, while Fe and Cu showed the opposite with averages 0.154 mg/L and 0.094 mg/L. Heavy metals were present in alarming concentrations in agricultural soil and products, with 105.2 mg/kg of Cu in tomatoes exceeding FAO-recommended thresholds of 73.3 mg/kg, Fe and Cr close to permissible limits with average concentrations of 320 mg/kg and 0.47 mg/kg.

MPs persist in the water table, urging regulatory agencies to establish safe distances from landfills for drinking water purposes. Heavy metals in borehole soils also showed significant affinity towards specific soil type suggesting that landfill locations should be considered according to waste types. Agriculture near landfills may need restrictions due to heavy metal accumulation and consumption of such products for a long time may threaten public health, but exploring heavy metal treatment in soils through plant uptake, such as tomatoes, requires extensive experimental analysis.

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