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Assessment of ground water quality and associated health risk through heavy metals contamination due to municipal solid waste landfilling practices in Mardan, Pakistan

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Abstract

This study was conducted in urban region of district Mardan. Pakistan is among the world arid regions and it is facing scarcity of fresh water sources. Groundwater is the major source of drinking water in the country and therefore, its quality protection and sustainability matters a lot. In the study area locals were using municipal solid waste (MSW) as fill material for leveling their residential lands from the last 10 to 20 years. Total 20 groundwater samples were collected from the area. 13 samples were collected from MSW landfill sites and 07 samples from safe zones. These samples were assessed for selected heavy metals (HMs) of Chromium (Cr), Lead (Pb), Cadmium (Cd), Copper (Cu), Zinc (Zn), Iron (Fe), Nickel (Ni) and Manganese (Mn). Among HMs Cr, Cd and Ni were found above the NEQS-Pak and WHO permissible limits. For health risk assessment (HRA), an average daily dose (ADD) and health quotient (HQ) were calculated. Highest ADD was found for Zn (3.74×10^{-3}) while highest average HQ was recorded for Ni (2.20×10^{-2}). Combined HI of all the HMs was found ≤1 which indicated potential health risk to the locals. Except Pb-Cd (r =0.537) and Zn-Mn (r=0.681) no specific correlation was observed among HMs. This behavior can be attributed to the input of HMs from anthropogenic sources rather than their natural sources. The study concluded that using MSW as fill material has deteriorating impacts on groundwater quality and ultimately pose threats to human health. Therefore, this practice should be stopped.

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Introduction

Among all the natural resources water is the most important and vital source as without it existence of life is not possible on earth. It supports all forms of life. Though there are some organisms which can survive without oxygen but no organism can live without water (Jalal and Kumar, 2013). All sources of water are essential for life but quality of water matters a lot when it intends to drinking water (Raju *et al.*, 2009). Water is an important factor for life sustainability on earth and without it existence of life is not possible. Hence, assessment and monitoring of water quality is important and variability of drinking water quality is directly concerned with human life (Muhammad *et al*, 2010). Significance of safe drinking water can be understood from the press release of United Nations (UN) Secretary General in 2002 on world water day which states "an estimated 1.1 billion people lack access to safe drinking water, 2.5 billion people have no access to proper sanitation, and more than 05 million people die each year from water-related diseases - 10 times the number killed in wars, on average, each year. All too often, water is treated as an infinite free good. Yet even where supplies are sufficient or plentiful, they are increasingly at risk from pollution and rising demand. By 2025, two thirds of the world's population is likely to live in countries with moderate or severe water shortages". Now when we are in few years distance from 2025 the words of UN Secretary General seems true. In 2016, natural disasters affected a population of 411 million wherein 94% were drought affected. Currently more than 40% of the world population is facing water crises (Guppy and Anderson, 2017). These crises may cause displacement of large scale population. The water supply and demand gap is projected to grow50% till 2030 (Guppy and Anderson, 2017). These conditions are very alarming, especially for underdeveloped and developing nations like Pakistan. This situation needs global attention to cope with effectively.

Besides, due to growing population and increased industrialization the demand for fresh water has been increased recently throughout the world, especially in arid and semi-arid regions of developing world (Guppy and Anderson, 2017). Pakistan is among the world's arid regions. Most of its population is dependent on ground water to fulfill their requirements for drinking and other domestic purposes (UNDP, 2016). On the other hand, due to prevailing climatic conditions water scarcity is an alarming issue in the country. According to a research study conducted in 2006 Pakistan has been listed among the countries having renewable water resources below the calculated threshold of 1500m^3/capita/year by the year 2030 (Rijsberman, 2006). Finding of the study proved correct as the availability of water has been dropped from 2172 m^3/capita in 1990 to 1306m^3/capita in 2015 (UNDP, 2016). In these circumstances protection of water sources is of utmost importance for Pakistan.

This already depleted ground water source is facing another serious concern of quality in Pakistan over the last few decades. Dependence on groundwater and indiscriminate usage has led to overexploitation of this resource. Moreover, threats to water contamination have been increased with the increase in population and industrialization (Khan *et al*, 2018). The increased population has resulted in poor management of municipal and industrial effluents as well as solid wastes management problems. Poor management of these wastes contaminates the very basic and already depleted natural water resources. Large amount of toxins along with other chemicals are mixed with water bodies when these waste come in contact with it (Jain *et al.*, 2005; Leoni *et al.*, 2005). Predominantly in urban areas solid waste dumps pose great threats to the ground water source. Lack of awareness among the general public regarding proper management of municipal solid waste (MSW) further worsen the situation. In some areas of the country MSW is used as fill material for leveling lands without adopting any mitigation measures. In such environment ground water is at high risk of contamination. This contamination level may be easily monitored and assessed through the presence of heavy metals (HMs) in such waters (Lee and Jones-Lee, 1993; Christensen *et al.*, 2001;

Tengrui *et al*., 2007; Ogundiran and Afolabi, 2008). Absorption of heavy metals in human body can create various complications and diseases in humans. For example Cadmium (Cd), a heavy metal can be fatal to human cell by displacing essential minerals like vitamins E and C from their active sites and prevent them to perform its normal function in the human body (Sukumar, 2006). Heavy metals like nickel (Ni), Fe and manganese (Mn) are required by enzymes for their activities. However, if entered in higher quantity to the human body through any of the pathways is proven toxic for human health (Ab-razak *et al*., 2015). In comparison ingestion of heavy metals is more health concern than other pathways (Cornelis and Nordberg, 2007). However, in Pakistan, the study on impacts of solid waste mismanagement on ground water quality is in infancy stage. Very limited research work has been conducted on this topic.

Usman *et al*, 2017 analyzed the ground the water quality near open dumps of MSW and found high level of contamination including heavy metals as compared to the water samples collected far away from the site. The research study concluded that MSW dumps poses threats to nearby ground water sources. Similar results were obtained by Muhammad and Zhonghua, 2014. Butt and Ghaffar, 2012 conducted a research study on the impacts of MSW dumps on ground water quality in Lahore city of Pakistan and found that the ill practices of waste management carried out at landfill site and the absence of leachate collection system has a great impact on the ground water quality of local aquifers. The analytical results showed most of the selected physicochemical parameters above the WHO permissible limits. Aiman *et al*, 2016 analyzed the ground water samples for presence of selected HMs near a landfill site in Lahore city and the results revealed that concentration of Pb, Fe, Cd, Mn and Cu surpassed the permissible limits of World Health Organization (WHO) and US Environmental Protection Agency (USEPA). Health risk factor of the HMs was found >1. The results confirmed that the MSW dumps pose greater threats to the exposed human population.

In view of all the above facts this research study was designed in the urban region of district Mardan, Pakistan because in the study area the locals were in practice of using MSW for leveling their residential lands. There was no check on this practice. The locals have drilled bore directly above such sites for extracting ground waters for their domestic use. Therefore, the selected study area was best suited for assessment of the MSW landfilling impacts on ground water quality. Basic aim of the study was;

i. To assess the impacts of aforementioned practice of the locals on ground water quality, the basic need of life, and associated health risk.
ii. To clear concept of the concerned government institution and general public regarding the ill impacts of using MSW for leveling residential lands.
iii. To enhance the level of awareness among the general public regarding the protection of precious ground water resources.

Material and methods

Study area

This study was conducted in urban region of district Mardan, Khyber Pakhtunkhwa (KP) province. The districtis situated between latitude 34°05´ to 34°32´ North and longitude 71°48´to 72°25´ East. Main focus was on the small residential settings comprising of Tariq colony, Makka colony and Gulibagh areas. This area was falling within jurisdiction of TMA Mardan. The locals were in practice of using municipal waste for leveling their residential lands.

No leachate collection system or other safety measures were adopted while leveling their lands using MSW. Total population of sampling area was ranged from 4000 to 5000 inhabitants. Locals were extracting water through hand and pressure pumps. Representative water samples were collected from the area.

Samples collection and preservation

As shown in table 1. total 20 drinking water samples were collected from the area. 13 (S1-13) representative water samples were collected from the region where MSW was used for leveling residential lands and 07 (S14-20) reference samples were collected from the surrounding safe zones for comparative study. All the samples were collected in 1.5 litter (L) poly propylene bottles. The bottles were rinsed with 1:1 concentrated HNO_3 and triple distilled water of Milli Q prior to utilization. For analysis of HMs the samples were acidified with 10% nitric acid. The samples were stored in a container having ice packs to keep the temperature in standard range (<08°C) and then stored at 4°C in the laboratory for further analysis. Locations and sources.

Table 1. Particulars of sampling points.

S. No	Sample ID	Location	Source
1	S-1	Gulibagh	Pressure pump
2	S-2	Gulibagh	Pressure pump
3	S-3	Gulibagh	Hand pump
4	S-4	Gulibagh	Pressure pump
5	S-5	Gulibagh	Hand pump
6	S-6	Gulibagh	Pressure pump
7	S-7	Gulibagh	Pressure pump
8	S-8	Gulibagh	Pressure pump
9	S-9	Makka colony	Pressure pump
10	S-10	Makka colony	Pressure pump
11	S-11	Makka colony	Pressure pump
12	S-12	Tariq colony	Pressure pump
13	S-13	Tariq colony	Pressure pump
14	S-14	Hajiabad	Pressure pump
15	S-15	Hajiabad	Pressure pump
16	S-16	Hajiabad	Pressure pump
17	S-17	Munshiabad	Hand pump
18	S-18	New bagh colony	Hand pump
19	S-19	Shamsi road	Pressure pump
20	S-20	Officer colony	Pressure pump

Analysis of heavy metals

The samples were analyzed for the presence and quantification of selected HMs of Cr, Pb, Cd, Cu, Zn, Fe, Ni and Mn. The analysis was carried out through flame atomic absorption spectrophotometer (Perkin Elmer AAS 700) under standard operating settings as per American public health association (APHA), 1998 guidelines at the centralized resource laboratory (CRL), University of Peshawar, Khyber Pakhtunkhwa. Double distilled water was used throughout the analysis. The delay and integration time of AAS was 05 seconds. The HMs results were compared with national environmental quality standards (NEQS) of Pakistan Environmental Protection Agency. Where the standard was not fixed for any particular HM in NEQS then the result was compared with WHO standard.

Health risk assessment

According to the US national academy of sciences (US-NRC) the risk assessment defines the possibility of an incidence occurrence and estimation of the magnitude of adverse impacts on human health upon exposure to the incidence (Lee *et al*, 2005). HRA can be applied to all possible pathways of human exposure; however, in the current study focus was only on the ingestion of HMs through drinking water.

For HRA average daily dose (ADD) of HMs through drinking water was calculated using US EPA, 1992 equation as;

$$ADD = C \times IR \times ED \times EF / BW \times AT$$

Where, *C* represents concentration of metals (mg/L), *IR* represents ingestion rate (2L/day), *ED* is the exposure duration to the metal (72 years), *EF* represents the exposure frequency in days per year (365 days), *BW* stands for average body weight in kilo grams (72 kg), and *AT* shows the average time period in days, equal to the life expectancy (72 years × 365 = 26,280 days) (Gul *et al.* 2015).

The ADD values of HMs was divided by reference dose (RfD) to calculate the health quotients (HQ) for selected HMs using US EPA,1992 equation as following;.

$$HQ = ADD/RfD$$

Reference dose values for HMs were 5×10^{4}, 1.5, 2×10^{-2}, 3×10^{-1},3.6×10^{-2},3.7×10^{-2}, 1.4×10^{-1}, and $3\times 10^{-1}$for Cd, Cr, Ni, Zn, Pb, Cu, Mn and Fe respectively(Lee *et al*, 2005). HQ values of all the HMs were combined to find out the overall toxic effect which is termed as the hazard index (HI).

$$HI = \sum HQi = 1 \ldots \ldots n$$

Where HI is the hazard index showing overall risk to the individual exposed to all the toxicants in the media.

Statistical analysis

Statistical software SSPS 20 was used for statistical analyses.

Results and discussion

Concentration of Heavy Metals

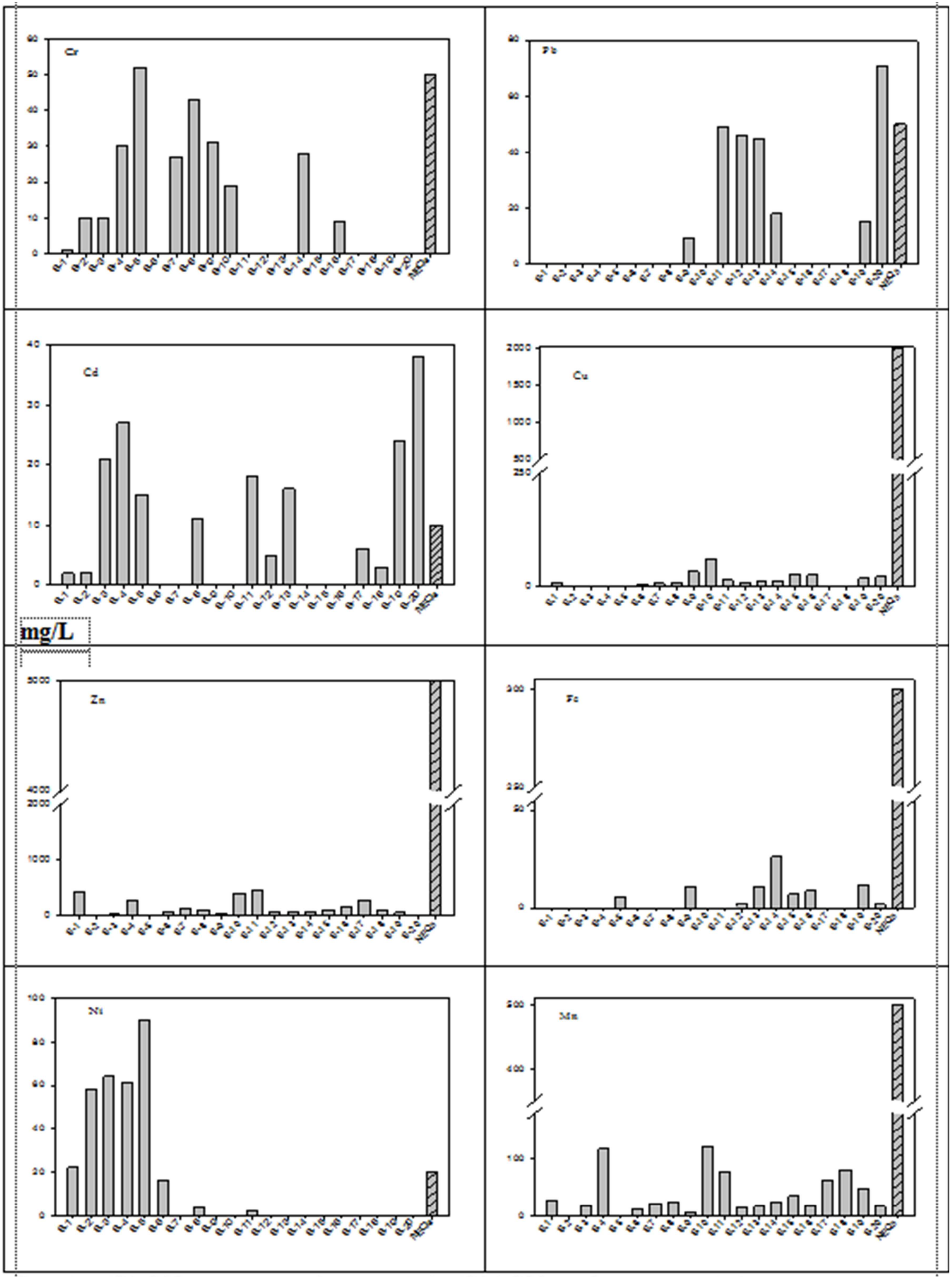
Table 2. and Fig. 1. show results of HMs concentrations in the samples. The concentration in representative sample followed the increasing order as Zn> Mn> Ni> Cu > Cr >Pb> Cd> Fe, with average concentration of 0.448mg/L, 0.122mg/L, 0.090mg/L, 0.061mg/L, 0.052mg/L, 0.049mg/L, 0.027 and 0.011mg/L, respectively. In the reference samples average concentration of Zn (0.103mg/L) was found the highest followed by Mn (0.040mg/L), Cu (0.0157mg/L), Pb (0.0148mg/L), Cd (0.010mg/L), Fe (0.008mg/L), and Cr (0.0052mg/L). Ni was not detected in any of the reference samples. The results of representative samples were compared with reference water samples (table 2). Concentrations of Cr and Ni were found above the maximum permissible limits of WHO and NEQS-Pak (0.050mg/L and 0.020mg/L respectively) only in representative samples. Concentrations of Cr and Ni were also found higher than those reported by Gul *et al*., 2015. Pb was found above the permissible limit (0.050mg/L) only in reference samples. This could be the possible reason of the presence of sulfide minerals in the study area (Gul *et al*, 2015). Cd was found above the NEQS-Pak permissible limits (0.010mg/L) in both reference and representative samples of the study area. Concentrations of Cu, Zn, Fe and Mn were found within the NEQS-Pak permissible limits of 2.00mg/L, 5.00mg/L, 0.300mg/L, and 0.500mg/L respectively in both the reference and representative samples. Almost all the HMs, except Pb, Cd and Fe, were found in relatively higher concentration in the representative samples (Fig. I). This indicates the contamination of ground water sources due to solid waste landfilling practices in the area. HMs are described as undesirable metals in drinking water and therefore, regular monitoring of the ground water in the study area is proposed.

Health risk assessment

Chronic (non-carcinogenic) risk data is given in table 3. which is calculated for representative samples (S1-S13) only. For Cr the highest ADD value was recorded as 1.44×10^{-3}mg/Kg-day with average value of 3.61×10^{-4}mg/Kg-day. In case of Pb1.97×10^{-3}mg/Kg-day was recorded as the highest value with an average of 3.51×10^{-4}mg/Kg-day. Cd had maximum ADD value of 1.05×10^{-3}mg/Kg-day having average of 2.61×10^{-4}mg/Kg-day. Cu highest value was recorded as 1.69×10^{-3}mg/Kg-day having average value of 3.85×10^{-4}mg/Kg-day. For Zn highest ADD value was found as 1.24×10^{-2}mg/Kg-day while average value for the metal was 3.74×10^{-4}mg/Kg-day. For Fe 7.22×10^{-4}mg/Kg-day was the highest recorded value having average of 1.19×10^{-4} mg/Kg-day. ADD for Ni was calculated and the highest value was recorded as 2.50×10^{-3}mg/Kg-day with an average value of 4.40×10^{-4}mg/Kg-day. For Mn highest ADD value was found as 3.39×10^{-3}mg/Kg-day having average value of 1.02×10^{-3}mg/Kg-day. Highest ADD value for HMs were found in order of Zn > Mn> Ni> Cu> Cr> Pb> Cd>Fe. The ADD values of the HMs were found proportional to their concentrations in the samples.

Highest average HQ value of 2.20×10^{-2} was found for Ni with highest range of 1.25×10^{-1} followed by Zn1.25×10^{-2}having highest range of 4.15×10^{-2}, Cu with average value of 1.04×10^{-2}having highest range of 4.58×10^{-2}, Pb average of 9.76×10^{-3} and highest range of5.48×10^{-2}, Mn 7.29×10^{-3} having highest range of 2.42×10^{-2}, Fe 3.98×10^{-4} having highest range of 2.41×10^{-3}, Cr 2.41×10^{-4} with highest range of $9.63\times 10^{-}$

[4] and Cd 5.22×10[-8] having highest range of 2.11×10[-7]. The HQ value of all the HMs is summarized in table 3.. Contamination of drinking water with HMs is considered as potential health risk for humans if HQ>1 (Muhammad *et al*, 2010). In the present study the HQ values for all the HM were found<1, hence the ground water of the area doesn't pose any risk to health of the people exposed to it.



Samples (S1-S13 = representative samples), (S14-S20= reference samples)

Fig. 1. Concentrations of HMs in the samples.

Table 2. Concentrations of the HMs in samples (mg/L).

HMs	Representative samples (n = 13)		Reference samples (n = 7)		Pak-NEQS/WHO Limits
	Range	Average	Range	Average	
Cr	0-0.052	0.017±0.017	0-0.028	0.0052±0.010	0.050
Pb	0-0.049	0.011±0.020	0-0.071	0.0148±0.026	0.050
Cd	0-0.027	0.009±0.009	0-0.038	0.010±0.015	0.010
Cu	0-0.061	0.0128±0.017	0-0.028	0.0157±0.011	2.000
Zn	0-0.448	0.151±0.170	0.011-0.273	0.103±0.086	5.000
Fe	0-0.011	0.00230±0.0042	0-0.026	0.008±0.009	-.-/0.300
Ni	0-0.090	0.0243±0.032	0.000	0.00±0.00	0.020
Mn	0-0.122	0.035±0.042	0.016-0.080	0.040±0.024	0.500

Table 3. HRA through drinking water consumption in the representative samples.

HMs	ADD (mg/kg-day)		HQ	
	Range	Average	Range	Average
Cr	0-1.44×10^{-3}	3.61×10^{-4}	0-9.63×10^{-4}	2.41×10^{-4}
Pb	0-1.97×10^{-3}	3.51×10^{-4}	0-5.48×10^{-2}	9.76×10^{-3}
Cd	0-1.05×10-3	2.61×10^{-4}	0-2.11×10^{-7}	5.22×10^{-8}
Cu	0-1.69×10^{-3}	3.85×10^{-4}	0-4.58×10^{-2}	1.04×10^{-2}
Zn	0-1.24×10^{-2}	3.74×10^{-3}	0-4.15×10^{-2}	1.25×10^{-2}
Fe	0-7.22×10^{-4}	1.19×10^{-4}	0-2.41×10^{-3}	3.98×10^{-4}
Ni	0-2.50×10^{-3}	4.40×10^{-4}	0-1.25×10^{-1}	2.20×10^{-2}
Mn	0-3.39×10^{-3}	1.02×10^{-3}	0-2.42×10^{-2}	7.29×10^{-3}

As the laboratory results showed the presence of more than one HMs in ground water of the study area therefore, hazard index (HI) value was calculated for each of them to check the health risk posed by each metal individually and their combined health effect. HI calculation results are given in table 4. Maximum HI value was recorded for Ni (4.40×10^{-1})followed by Zn (1.82×10^{-1}), Cu (1.25×10^{-1}), Pb (1.15×10^{-1}), Mn (9.01×10^{-2}), Cr (4.13×10^{-3}), Fe (2.78×10^{-3}) and Cd (6.5×10^{-7}). If the HI value is <1 for the toxicants then chronic risk to human health due to exposure through the selected pathway is considered negligible. However, the overall HI value for all the toxicants was found as 9.60× 10^{-1} which is close to the risk factor limit as HI$\leq$ 01. This shows that potential risk exists for local population if they are exposed to all the selected toxicants (HMs) through the selected pathway for longer period.

Table 4. Hazard indices for the heavy metals.

HMs	HI
Cr	4.13×10^{-3}
Pb	1.15×10^{-1}
Cd	6.5×10^{-7}
Cu	1.25×10^{-1}
Zn	1.82×10^{-1}
Fe	2.78×10^{-3}
Ni	4.40×10^{-1}
Mn	9.01×10^{-2}
HI	9.60×10^{-1}

Correlation among HMs

Table 5. summarizes the correlation among the HMs. The correlation matrix showed positive correlation among Cr-Fe (r = 0.162), Cr-Ni (r = 0.431), Pb-Cu (r = 0.109), Pb-Fe (r = 0.139), Cd-Ni (r = 0.265), Cd-Mn (r = 0.132), Cu-Zn (r = 0.282), Cu-Fe (r = 0.218) and Cu-Mn (r = 0.283). However, the only significant positive correlation was observed among Pb-Cd (r = 0.537) at the 0.05 level (2-tailed) and Zn-Mn (r = 0.681) at the 0.01 level (2-tailed). Negative correlation was found between the pairs of Cr-Pb (r = -0.366), Cr-Cd (r = -0.042), Cr-Cu (r = -0.021), Cr-Zn (r = -0.168), Cr-Mn (r = -0.083), Pb-Zn (r = -0.066), Pb-Ni (r = -0.336), Pb-Mn (r = -0.155), Cd-Cu (r = -0.184), Cd-Zn (r = -0.11), Cd-Fe (r = -0.116), Cu-Ni (r = -0.475), Zn-Fe (r = -0.344), Zn-Ni (r = -0.189), Fe-Ni (r = -0.228), Fe-Mn (r = -0.273), and Ni-Mn (r = -0.123). This correlation behavior of the HMs is non-natural and indicates the contamination of HMs from anthropogenic sources. As the samples were collected from the MSW land fill sits therefore it could be assumed from the correlation analysis that the ground water sources were being affected by the MSW landfilling practices. Though the impacts are not severe yet however, the contamination of ground water sources due to MSW landfilling practices cannot be ruled out. Continuation of these practices by locals in the study area may pose greater health risk in near future.

Table 5. Correlation matrices among HMs in the drinking water samples.

	Cr	Pb	Cd	Cu	Zn	Fe	Ni	Mn
Cr	1	-0.366	-0.042	-0.021	-0.168	0.162	0.431	-0.083
Pb		1	0.537*	0.109	-0.066	0.139	-0.336	-0.115
Cd			1	-0.184	-0.11	-0.116	0.265	0.132
Cu				1	0.282	0.218	-0.475*	0.283
Zn					1	-0.344	-0.189	0.681**
Fe						1	-0.228	-0.273
Ni							1	-0.123
Mn								1

*. Correlation is significant at the 0.05 level (2-tailed).

**. Correlation is significant at the 0.01 level (2-tailed).

Conclusion

This study concludes that the relatively high concentrations of HMs in representative water samples can be attributed to the leaching of HMs from MSW landfill sites. Correlation analysis of HMs also supports the non-natural occurrence of the metals in ground water sources closer to the landfilling sites. Except Pb-Cd (r = 0.537) and Zn-Mn (r = 0.681) no specific correlation was found among the HMs. This behavior of HMs indicates inputs from anthropogenic sources rather than their natural grouping. Overall HI value of all the toxicants in the representative samples was found ≤1 which revealed potential health risk. It is crystal clear from this research study that using MSW as fill material is a potential threat to ground water sources and ultimately to human health.

Recommendations

To ensure safety of precious plus already scarce ground water sources in the country and related human health, the practice of using MSW as fill material should be discouraged. Awareness level should be enhanced among the local population in this regard. Being an issue of public welfare, regular monitoring and assessment of ground water sources is recommended for the study area. Environmental Protection Agencies, being mandated for protection of natural resources, shall search out and implement mitigation measures to prevent the ground waters from further deterioration due to such practices.

Conflict of interest

The authors declare that there are no conflicts of interest.

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