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Concentration of heavy metals in leachate, soil, and plants in Tehran's landfill: Investigation of the effect of landfill age on the intensity of pollution

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ABSTRACT

Heavy metals are one of the important pollutants in landfill leachate. Plants and soil near the landfill may be contaminated by leachate. In this study, by evaluating the heavy metals in the leachate of two landfills and the soil and plants near them, the amount of pollution caused by the leachate in the environment around the landfills in Tehran was investigated. This study was conducted in three stations, soil and plant samples were prepared according to a specific protocol. Also, three indexes including PI, PI_{Nemerow}, and BF were used to interpret the results. The results showed that the concentration of total metals in the old landfill leachate and new landfill leachate was only 12% different and was 24.13 mg/L on average. In the new landfill leachate, iron had the highest concentration among metals, which was 22.94 and 17.01 mg/L in two samples. In the old landfill leachate, the concentration of manganese was 15.71 mg/L, which was the highest among the studied metals. The concentration of heavy metals in the soil of the old landfill was 24.6% lower than the concentration of metals in the soil of the new landfill. In all samples, the highest metal concentration in the soil was related to manganese, which was 33.65-34.14 mg/L. Cadmium had the lowest concentration in soil compared to other metals. The concentration of total metals in the studied plants was 29-60 ppm. The PInemerow for studied stations was 0.1711, 0.1708, and 0.1463. The highest PI in the case of lead was observed at the second station equal to 0.54. The highest BF in case of Atriplex Undulata was more than 6 and related to cadmium, while the highest BF in case of Atriplex Cinearea was more than 3.5 related to cadmium. This study showed that the soil and plants of the landfill were contaminated with heavy metals under the influence of leachate, and the ability of plants to uptake and accumulate metals can be used to manage soil pollution near the landfill.

1. Introduction

Municipal solid waste generally refers to materials such as plastic, glass, food, metals, paper, and the like, which are produced as solid waste by commercial, office, household, industrial, and the like [1–3]. The expansion of the use of technology in the last hundred

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years and the variety of consumption caused by it have caused significant changes in the amount and composition of municipal solid waste [4]. The production of millions of tons of solid waste per day in the world is a constant challenge in human life. In developing countries, the problems of municipal solid waste management are more due to population growth, urbanization, and little awareness and participation of citizens [5]. This phenomenon is a very serious challenge, especially in megacities. In big cities, there are different paths in solid waste management, which mainly lead to two destinations, including recycling or disposal [6].

Landfilling is the oldest method of solid waste disposal which can be a threat to the environment and health [7]. Due to its easy operation and cost-effective, landfill is the most widely used method of municipal solid waste disposal in the world [8,9]. This method is widely used in developing countries, but it can be a threat to the environment [10]. Biological, physical, and chemical transformations occur in buried solid waste. In fact, the landfill site is known as a reactor [11]. Due to these reactions, gas and leachate are produced in the landfill sites. Although methods such as insulation of the landfill and daily covering are used to reduce the environmental consequences of the landfilling, the characteristics of the leachate cause the effects of the landfill on the environment to be very serious and diverse [12]. Leachate is a liquid from the solid waste landfill, which is produced under the influence of the amount of moisture in the landfill site and due to chemical and physical reactions. Leachate is known as an important source of environmental pollution, including groundwater, due to the presence of organic and inorganic substances, including heavy metals [13]. Landfill leachate contains many pollutants, including toxic substances and heavy metals, and usually has high COD [14]. Soil and water pollution in the areas near the landfill site and the areas affected by the landfill leachate is a serious concern in the management of municipal solid waste [15]. Pollution in the soil and water of the areas under the influence of landfill leachate affected by the technology. In developing countries, due to the use of old methods and non-use of pollution control technology, the amount of pollution may be higher.

The level of pollution in the soil and water sources near the landfill site is influenced by the factors that affect the amount of leachate and also the type and quantity of pollutants. The most important parameters affecting the amount of leachate are the quantity and composition of the buried solid waste, the hydrological conditions, and the height of the landfill cell. Also, in addition to the composition of the solid waste, parameters such as the age of the landfill, degradation stage, and the applied technology, are effective in the quality of the landfill leachate [16]. Heavy metals are one of the most important contaminants in landfill leachate [17]. Regarding the effect of solid waste separation on the concentration of leachate heavy metals, in developing countries, due to low waste separation before landfill, leachate heavy metals may be higher [18]. Therefore, there is a possibility of soil and water pollution in areas affected by landfill leachate. In this study, the soil contamination status of the affected areas in old and new landfill site in Tehran by heavy metals in the surrounding environment, which can be an ecological and health risk. Sampling of wild plants in the study area was done in order to evaluate the accumulation of metals in plants as an adverse consequence of landfill leachate and the possibility of using them in the phytoremediation process.

2. Method

2.1. Landfill sites description

This study was conducted in two landfills, including an old landfill and a new landfill in Tehran. These landfills were located in the south of Tehran in the Kahrizak region, which is known as Aradkoh landfill. At the time of conducting this study, 5200 tons of municipal solid waste was sent to this landfill every day. A part of the solid waste in Aradkoh was placed in various processes to energy and compost, and about 2500 tons of the solid waste was landfilled [19]. According to previous reports, it is estimated that 250 cubic meters of leachate are produced daily in the Aradkoh landfill [19].

2.2. Sampling and laboratory analysis of samples

Samplings of leachate, soil, and plants were done in three stations. The first station on the new landfill was at the leachate return point, the second station was at the downstream of the old landfill, and the third station was at the downstream of the new landfill.

2.3. Leachate

Leachate samples were regularly collected from the stations once a month [20]. Two liters of leachate per sampling and for six months. HDPE bottles were used for sampling and before sampling, the bottles were rinsed with leachate [21]. In each sampling, 0.5 L of leachate was collected and transported to the laboratory at 8–15 °C [22]. To keep the characteristics of the leachate constant, the samples were stored in a refrigerator at 4 °C in a dark environment [20,22]. All the samples were acidified using nitric acid for reducing the pH to less than 2 to prevent the precipitation of metals [21].

2.4. Soil

Soil sampling was done in all three stations at a depth of 10–50 cm [21]. Sampling was done manually, each sample weighed approximately 2 kg and the samples were placed in polyethylene bags and sent to the laboratory. The soil samples were dried in the laboratory at room temperature and sieved to obtain a particle size smaller than 2 mm [21]. For heavy metal analysis, sieved samples were kept in HDPE containers. In the investigation of landfills, two wild plants, including Atriplex Undulata and Atriplex Cinearea

(1)

were identified as the most abundant plants and their sampling was done. Plant samples were collected from the same locations as soil samples.

2.5. Plant

Plant samples were collected with appropriate permissions and that all plant experiments were conducted in accordance to relevant regulations and guidelines. Washing with distilled water was used to clean plants from soil particles [23]. The plant samples were dried at 60 °C for 48 h before grinding [23]. 2 g of the samples were placed in a 50 mL porcelain dish. Then, 10 mL HNO₃ (68% w/w) was poured into the dish and heated for oxidation on a hot plate for 30–45 min. 2 mL HClO₃ (37% w/w) was added to the sample after cooling and reheating to obtain a clear, semi-dry solution. The sample was then cooled and passed through grade 42 Whatman® cellulose filter paper. Then, it was transferred to a 50-mL volume balloon to reach the desired volume, and distilled water was added [24,25]. Analysis of heavy metal concentrations in the plant was performed from three parts: root, stem, and leaf. Cd, Pb, Ni, Fe, Zn, and Cu were selected to assay the contamination. Heavy metal concentration measured by Atomic Absorption Spectrometer (Perkin-Elmer® AAnalyst 200). The concentrations of the measured metals were compared with the permissible limits of valid international standards. Then the severity of pollution due to the presence of leachate on the surrounding environment is measured by BF, PI, and Nemerow Pollution Index (NPI) [26].

2.6. Indexes

In this study, three indexes were used to interpret the data. Bioavailability factor (BF) is obtained from the ratio of metal concentration in plant components to soil to investigate the metal transfer rate from soil to plant environment [27,28]. This factor has been used in researches to investigate the relationship between the aerial part of the plant and the soil [29,30]. Some sources refer to the upper part of the soil [31] leaf to soil [32,33], stem to soil [34], and roots to soil [35]. This study has only focused on investigating the amount transferred from the soil environment to the plant. The following equation shows how to calculate this factor:

$$BF = C_{Plant}/C_{soil}$$

where C_{plant} is the total metal concentration in the plant and C_{soil} is the total metal concentration in the soil.

The pollution index (PI) is used to determine which heavy metal poses the greatest threat to the soil environment. This index is also needed to calculate some complex indices, for example, the PI_{Nemerow} [36,37]. The following equation was employed to PI index [38, 39]:

$$PI = C_{Sample}/B_n$$
⁽²⁾

where C_{Sample} is the sample concentration and Bn is the amount of metal concentration in the soil geochemical background. The extracted values for the Bn parameter based on the soil type of the area are shown in Table 1 [40].

According to this factor, the soil is classified in terms of contamination into five classes. In the healthiest condition, and when (PI < 1), the soil is clean and uncontaminated. Also, in the worst case, if (PI > 5), the intensity of soil contamination will be extreme. This classification is shown in Table 2.

The Nemerow Pollution Index (PI_{Nemerow}) allows assessing the overall degree of soil pollution and includes the contents of all analyzed heavy metals [26]. It is calculated for both the O and A horizons, based on the following formula.

PI _{Nemerow} =	$\left(\frac{\left(\frac{1}{n}\sum_{i=1}^{n}PI\right)^{2}+PI_{max}^{2}}{2}\right)$	(3)
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where PI is the calculated value for the Single Pollution Index, PI_{max} is the maximum value of the Single Pollution Indexes of all heavy metals, and n is the number of heavy metals. According to PI_{Nemerow}, five classes of soil quality are presented in Table 3.

3. Results and discussion

The results of investigating the concentration of heavy metals in leachate are shown in Table 4. As the results show, although the amount of studied metals in the stations was the same, but the proportion of the studied metals was different. The total studied metals in each of the stations were 25.18, 22.13, and 25.1 mg/L, respectively. In station 1 and station 3, iron had the highest concentration

Table 1

Moon	concentration	of me	tal in	coil	bedrock	nor	nnm i	B	١
wiean	concentration	or me	tai iii	SOIL	Demock	per	ppm v	Dn	J

Metal	Fe	Си	Ni	Mn	Cd	Pb
B _n	47,200	45	68	850	0.3	20

Table 2	
Classification of soil contamination based on PI.	

The value of PI	Soil contamination
PI < 1	_
$1 < \mathrm{PI} < 2$	Low
2 < PI < 3	medium
3 < PI < 5	strong
PI > 5	Very strong

Table 3
Soil pollution classification by the Nemerow Pollution Index (PI _{Nemerow}).

Class	T	п	ш	n/	N/
Class	1	11	111	10	v
PI _{Nemerow}	≤ 0.7	0.7–1	1–2	2–3	≥ 3
Soil quality	uncontaminated	Danger range	Low contamination	Moderate contamination	Severe contamination

among metals, which was 17.01 and 22.94 mg/L, respectively. But in station 2, the concentration of manganese was 15.71 mg/L, which was the highest among metals. The ratio of studied heavy metals in different stations is shown in Fig. 1. In all stations, cadmium had the lowest concentration among the studied metals. Also, the results showed that cadmium, nickel, and copper had the lowest concentration variation in different stations, while the concentration variation of iron and manganese was significant.

The average concentration of heavy metals in landfill leachate in Iran can be compared with the results of this study. Torkashvand et al. 2016 reported the concentrations of copper, cadmium, lead, iron, and nickel in the studied landfill leachate in Iran as 1, 0.45, 0.85, 14, and 1.1 mg/L, respectively [41]. Also, Pasalari et al. 2019 reported that the concentration of manganese in the Iran's landfills leachate was between 3.2 and 8.1 mg/L [19]. Therefore, the concentration of copper in the leachate of station 2 was similar to the average concentration in landfills in the country, while the concentration of this metal in stations 1 and 3 was much lower than the average concentration in the studied leachates in Iran. Also, the concentration of cadmium in all the studied stations was lower than the reported average in the country. Iron and manganese are two common metals found in landfill leachate in Iran, which is caused by the composition of Iran's municipal solid waste, so iron and manganese formed the highest concentration of metals detected in this study [19,41]. The concentration of lead in stations 2 and 3 was slightly lower than the average of other studies, while the concentration of lead in station 1 was higher than the average of other studies. The nickel concentration is station 3 was similar to the average reported in other studies, while it was 3 and 2 times higher in stations 1 and 2, respectively.

The concentration of heavy metals in the leachate is one of the important pollutant that is considered in the selection of the treatment process. In general, the quality characteristics of landfill leachate depend on two important factors, which are the composition of the buried waste and the age of the landfill [19]. Based on landfill age classification, landfills less than 5 years old are young, landfills between 5 and 10 years old are middle-aged, and landfills older than 10 years are old [19,42]. Therefore, one of the reasons for the difference in the concentration of heavy metals in the leachate of stations 1 and 3 with the concentration of metals in the leachate of station 2 can be the difference in the age of the two landfills. Even if the composition of the buried wastes is similar, the concentration of heavy metals in old landfills can be different compared to young landfills due to the change in pH. Also, the reduction of biodegradability in old landfills is an indirect factor in changing the concentration of heavy metals in leachate compared to the young landfills. Therefore, one of the reasons for the lower concentration of the total metals studied in Station 2 could be the age of this landfill. On the other hand, one of the reasons for the difference in leachate metal concentration in this study compared to other studies in Iran could be the different composition of buried waste. Even the difference in the composition of waste buried over time in a landfill can be effective in the concentration of heavy metals in the leachate. The acidic condition of the leachate causes the leaching of heavy metals to be slow, so the leachate is one of the sources of heavy metal pollution [43]. Considering the leakage of heavy metals from some solid waste compounds such as used batteries, electrical waste and cigarette butts, the change in the composition of these components is effective in the concentration of heavy metals. For example, Torkashvand et al. 2021, studied the effect of the proportion of cigarette butts in the waste mass on the quality of leachate. They reported an increase in the concentration of heavy metals in the leachate proportional to the increase of cigarette butts in the buried waste [44].

Heavy metals are one of the important pollutants that leak into the environment from various sources such as wastewater and solid waste. The presence of heavy metals such as iron and manganese in leachate will cause contamination of soil and underground water resources. There is a possibility that irrigation of agricultural crops with water under the influence of leachate will cause the entry of metals such as manganese into the food chain [45]. These pollutants can cause various diseases in humans by entering the food chain.

 Table 4

 Concentration of studied heavy metals in different leachate (mg/L).

	Pb	Cd	Mn	Ni	Cu	Fe	Total
Station 1	2.17	0.32	1.95	3.36	0.37	17.01	25.18
Station 2	0.76	0.16	15.71	2.70	1.02	1.78	22.13
Station 3	0.65	0.1	0.29	1.04	0.08	22.94	25.1



Fig. 1. Ratio of metals in studied leachates.

Several studies have investigated soil contamination with heavy metals, and their results have shown that many soils near industries, landfill sites are contaminated [46,47]. The toxicity of leachate on human cells shows the importance of this source of pollution, which releases pollutants into the soil and water sources [48]. Landfill leachate is a source of heavy metal pollution in soil and water sources [43]. The results of the analysis of soil samples are shown in Table 5. These results showed that the amount of heavy metals in the soil of the old landfill is 24.6% lower than that of the new landfill. In all stations, manganese had the highest concentration compared to other metals, and cadmium was the lowest metal among the studied metals. However, as Fig. 2 shows, the proportion of metals measured in the soil of different stations is not the same.

The high ratio of manganese in the studied stations (63.26%–87.25%) is due to the presence of solid residues that are the origin of this metal. Wastes such as blade, bottle caps, galvanizing goods, insecticides, pigments, and paint are known as the source of manganese in landfill leachate, which can cause soil pollution [43]. Considering the low rate of source separation of solid waste in Iran [6], the presence of manganese-origin waste in the landfill has increased the concentration of this metal in leachate. The ability to absorb metals by soil has caused the concentration of metals such as lead and copper in the studied soils to be about 1 mg/kg. Although the presence of these metals in electrical waste is emphasized, used batteries, pipes, and paint are also known sources of lead and copper emissions [49,50]. The potential for metal absorption by the soil, the distance of water sources from the landfill, and topography are factors that affect the concentration of heavy metals in the soil and even the water sources around the landfill [25]. Therefore, the transfer of various pollutants, including heavy metals, to water resources near the landfill site is a serious concern in landfill leachate management, which has been analyzed in many developing countries [51]. In general, the concentration of heavy metals in the depends on factors such as the difference in the concentration of metal accumulation in the soil in a long time [52]. Increasing the concentration of metals in the leachate [49], the absorption rate of metals by soil particles in the soil. However, it has been reported that the amount of heavy metals in the soil affected by leachate is lower during the rainy season [52].

The order of metal concentrations in soil samples is as follows: Cd < Cu < Pb < Ni < Fe < Mn. In this study, PI and PINemerow indices were used to determine the extent of contamination (Table 6). PI and PINemerow indices of soil samples are classified in non-contaminated classes. Fortunately, the landfill of hazardous wastes has been minimized due to proper efficiency separation operations in the Aradkooh Complex. The result can be seen in the soils near the landfill with low concentrations of heavy metals such as lead, nickel, and cadmium. Proper management of hazardous and industrial wastes has eliminated the flow of household waste from this type of waste. Therefore, proper disposal and management of industrial waste is another reason for the low concentration of heavy metals. The results of another study also showed that there is heavy metal contamination in the soil around active landfills and abandoned landfills, but it is more in the soil around active landfills, so there is a health and ecological risk in the soil around landfills [53]. The impact of leachate heavy metals on soil pollution can be identified by comparing their concentration with the geochemical background. The results of such investigations in the soil around the landfill site show an increase in the concentration of heavy metals in the soil, so that in one sample in Morocco, these conditions caused moderate to high contamination level in the soil, which was mostly caused by metals such as lead and cadmium [54].

Measurement of heavy metals in plants affected by leachate is important from two aspects. First, the accumulation of pollutants, including heavy metals, in plants can be a sign of their entry into the food chain and lead to subsequent adverse consequences. The origin of pollutants including heavy metals in plants can be soil, water, and air. So, the presence of heavy metals in plants can indirectly be a reason for the intensity of pollution in soil or water [52]. Second, the ability to remove heavy metals by plants can be considered as a solution for pollution management in landfills. Phytoremediation technique is a well-known method to reduce pollutants in the

 Table 5

 Concentration of studied heavy metals in different soil samples (mg/Kg).

	Pb	Cd	Mn	Ni	Cu	Fe	Total
Station 1	1.02	0.07	33.92	2.83	0.86	14.92	53.62
Station 2	1.09	0.07	34.14	1.59	1.15	1.09	39.13
Station 3	0.71	0.06	33.65	1.58	0.96	14.94	51.90



Fig. 2. Ratio of heavy metals in the soil of different stations.

Table 6 The calculated PI and $\ensuremath{\text{PI}_{nemerow}}$ for different stations.

	PInemerow	PI							
		Pb	Cd	Mn	Ni	Cu	Fe		
Station 1	0.1711	0.0510	0.2333	0.0399	0.0416	0.0191	0.0003		
Station 2	0.1708	0.0545	0.2333	0.0402	0.0234	0.0256	0.0000		
Station 3	0.1463	0.0355	0.2000	0.0396	0.0232	0.0213	0.0003		

environment, but the ability of different plants to absorb different pollutants is not the same [55,56]. The results of the analysis of heavy metals in the studied plants are shown in Table 7.

The results showed that the amount of metals in the plants of Station 3 was much lower than the metals in the plants of other stations. Also, the proportion of metals in the plants of different stations was not the same. In station 1 and station 2, manganese was the most abundant metal, while in station 3, iron was the most abundant metal in plants. In station 1, manganese constituted 50% of the total concentration of studied metals in plants, and in station 2, this ratio was 60.6%. However, iron accounted for 57.7% of the total metals in station 3 plants. These results showed that according to the amount of studied metals in the soil of the stations, the ability to uptake iron and manganese by the studied plants was significant. However, the highest accumulation of metal in the plant compared to its concentration in the soil was seen in the case of cadmium. This ability has caused plants to be used to manage pollution, including heavy metals. In previous studies, the reduction of manganese, copper, and zinc from landfill leachate by plants has been reported as 33, 28, and 36%, respectively, but the amount of accumulation of metals in soil and water should not be so high as to affect plant growth [58]. The proper growth of plants in this study and the accumulation of metals in them showed that the tolerance threshold of these plants was higher than the concentration of metals in the soil affected by leachate and they can be used in phytoremediation method. In addition, proven methods can also be used to increase the accumulation of metals in plants. For example, the use of EDTA will increase the accumulation of lead in plants and increase the efficiency of phytoremediation [59].

The order of metal concentrations in plants is the same as their concentration in soil. As shown in Figs. 3 and 4, both plant samples had better performance in cadmium uptake than other elements and did not have significant uptake of lead (ratio less than 1). According to the BF index results in different components of the plant, the ratio of metal concentrations in the samples are the root to soil> leaf to soil> stem to the soil, respectively. This indicates the tendency of both plant species to store metals in the roots, which is consistent with the studies of Jay et al. (2009) [60]. Copper and manganese were used as micronutrients for various metabolic components of the plant. Therefore, they are mainly transported from the roots to the upper part of the plant, and their concentration is higher in the aerial organs [61].

The amount of lead is considered the highest concentration of metal transferred from stem to leaf, but in the transfer from root to stem or leaf to root ratio, Ni is the most transferred element. Pb is commonly known as a metal with less movement, and in this study, the results of all samples are consistent with it [62]. Therefore, considering the effect of leachate on increasing the concentration of pollutants including heavy metals in the soil, water and plants surrounding the landfill site, it is imperative to control the environmental and health consequences of the leachate. The use of liners to prevent leakage of leachate into the surrounding environment and the continuous monitoring of pollution in the surrounding soil and water have been proposed as suggestions for landfill leachate risk

Table 7 Concentration of studied heavy metals in different plant samples (ppm).

	Pb	Cd	Mn	Ni	Cu	Fe	Total
Station 1	0.41	0.17	30.01	1.12	1.24	27.41	60.36
Station 2	0.33	0.27	33.11	1.64	1.05	18.22	54.62
Station 3	0.34	0.19	8.85	2.08	1.16	17.25	29.87



Fig. 3. Ratio of metal concentrations in Atriplex Undulata species to soil (BF factor).





management [63].

4. Conclusion

The effect of leachate on the concentration of heavy metals in soil and plants affected by it was investigated. The concentration of heavy metals in the leachate of the old landfill was 22.1 mg/L, while the concentration of heavy metals in the leachate of the new landfill was 25.1 mg/L. The proportion of heavy metals in the old landfill leachate was not the same compared to the new landfill leachate. In the leachate of the new landfill, iron was the most metal, which constituted 92.3% of the total metals, while in the leachate of the old landfill, manganese was the most metal, which constituted 70.9% of the total metals. The concentration of total metals in the soil of the old landfill was 39 mg/kg, while the concentration of total metals in the soil of the new landfill was 51 mg/kg. Iron and manganese had the highest concentrations and lead and cadmium had the lowest concentrations among the metals accumulated in the studied plants. The concentration of metals in the leachate was influenced by the composition of buried waste. The age of the landfill was also effective in the concentration and proportion of metals in the leachate. The soil was contaminated with heavy metals due to leachate, but the investigated indexes showed that the level of contamination is not serious. The accumulation of heavy metals in the plants grown near the landfill was significant, which could be one of the reasons for the reduction of pollution in the studied soil. These

plants can be used in phytoremediation as a pollution reduction technique.

Declarations

Author contribution statement

Seyyed Mahdi Hosseini Beinabaj: Conceived and designed the experiments; Performed the experiments.

Ali Hosseinzadeh: Conceived and designed the experiments; Contributed reagents, materials, analysis tools or data; Wrote the paper; Analyzed and interpreted the data.

Hossein Heidarian: Contributed reagents, materials, analysis tools or data.

Hamed Mohammad Aleii: Contributed reagents, materials, analysis tools, data.

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Data availability statement

Data will be made available on request.

Additional information

No additional information is available for this paper.

Declaration of competing interest

The author(s) declared no potential conflicts of interest.

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