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Primary Treatment of Landfill Leachate Effects on Heavy Metal and Soil Chemical Properties in Kwashe Industrial Area in Duhok Province, Kurdistan Region of Iraq

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ABSTRACT

Leachate is a major problem created from municipal solid waste (MSW) landfills and is considered the worst source of surface water and groundwater pollution. Kwashe industrial area as one of the most fast developing and polluted industrial area in Kurdistan region with many factories has a large solid waste separation factory that receives approximately 900 tons of municipal solid waste, out of which about 50% is organic kitchen waste. Samples of leachate were randomly collected from different sites downward drain of landfill and treated by aeration and filtration processes over a month. The results revealed that the soil was affected highly by soluble salts like Na and K from landfill; consequently, the electrical conductivity was 6.37(ds/m), 6 folds greater than the acceptable ranges. The primary treatment of landfill leachate by aeration and filtration reduced all heavy metals to acceptable ranges except Fe and Al, which remained relatively high over allowable ranges because the behavior and reaction of heavy metals in leachates is complicated as many acids are formed during the acidogensis of organic waste biodegradation that increases the solubility of these cationic heavy metals. The extractable forms of calcium, magnesium, sodium, potassium, sodium, and lithium are within acceptable ranges, which indicated not being enough to cause a threat in deteriorating soil and environmental chemistry. The results also showed that there was a close correlation between landfill leachate content of heavy metals and soil content affected by this leachate of Zn, Fe, Cu, Al, and Co at ($P \leq 0.01$), but the soil downward landfill leachate fortunately did not reach the hazardous limits of their content in heavy metals and also did not exceed the standard ranges in soil.

GRAPHICAL ABSTRACT

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Introduction

A landfill is an engineering place where rubbish and municipal waste are disposed and buried, considered the oldest form of waste treatment. In early history, it was referred to as a maiden. Landfills refer to controlled waste disposal which cause minimum environmental injuries. Now landfills are the most common methods of organized waste disposal in many places around the world [1].

Kwashe industrial area in Summel district, Duhok city, Kurdistan region-Iraq was constructed in 2012, including many factories such as more than 50 petroleum refineries, dying, tanning, cement, recycling of steel in addition to large municipal waste separating factory that produce a huge amount of wastes that are collected in open dumping landfill producing high amount of leachate as more than half of this waste are wet organic kitchen food waste in a natural valley near waste separating factory [2].

The worst kind of waste dumping is open dumping like Kwashe open dumping landfill where loads of waste are dumped in an uncontrolled condition in an open area with no soil burial, no leachate collection system, no drainage pipes, bad access to the tipping area, mostly in a raining season, accessible to scavenging, and uncontrolled open burning. Open dumping creates a threat for both surrounding environment and public health as well [3].

Leachate is defined as an aqueous dark brown to black heavy solution when waste breaks down in the landfill, and drains from a landfill and contains mainly dissolved and suspended materials. Leachate is a major problem created from municipal solid waste (MSW) landfills and considered the worst source of surface water and groundwater pollution. Leachate results mainly from excess water in wet organic wastes when it is decomposed and also from precipitation in the rainy season which enters the landfill. This heavy liquid is highly toxic and contains many hazardous pollutants such as toxic heavy metals, industrial solvents, ammonia, toxic organic compounds, pathogens, and household cleaners, micro organic pollutants as pharmaceutical and pesticide residues, and electronic wastes which contain highly toxic lead, mercury and cadmium. These hazardous pollutants in leachate can pollute the soil, ground water and waterways [4].

Amongst the non-biodegradable pollutants in leachate are recalcitrant heavy metals. The behavior and reaction of heavy metals in leachates are complicated as many acids are formed during the acidogensis of organic waste biodegradation that increase the solubility of cationic heavy metals. So the removal of heavy metals from leachate is difficult and shrouded in a lot of mysteries. Biological treatment by inducing aeration of landfill leachate is mainly used for the removal organic pollutant, but considerable amount of heavy metal is also removed simultaneously with biological treatment by bio-sorption mechanisms. Bio-sorption of heavy metals are done by immobilization to new biomass by microbial communities and other chemical mechanisms as chelating and ion exchange as well as physical adsorption. The removal range of heavy metal relies on many factors such as their concentrations in leachate, their speciation, and existence of other alkaline earth metals [5].

As two-third of landfill waste is biodegradable, organic materials comes mainly from households and restaurants. When these materials are degraded by microbial communities in landfill sites, they produce methane gas, which is a very potent greenhouse gas and self-igniting gas, causing continuous fires in open landfills. Methane is 20 times more effective than CO2 in trapping heat in the atmosphere, and considered the second greenhouse gas next carbon dioxide. The most health implications noticed in individuals near landfill areas are birth defects, low birth weight, and cancers in numerous studies. So the aim of this study is to evaluate the primary treatment of leachate by aeration and filtration in heavy metal content and physic-chemical properties of treated leachate in the Kwashe industrial area in Duhok City, Kurdistan Region of Iraq.

Material and methods

Study Area

As shown in Figure 1, the study area was located in the Kwashe industrial area as one of the most fast developing and polluted industrial area in Kurdistan region, which includes many factories as oil refineries, tanneries, dying, steel, and cement. Kwashe is an industrial area which far 20km west of Duhok's city. This area lies between latitude 36.9906°N longitudes 42.7894°E [2].

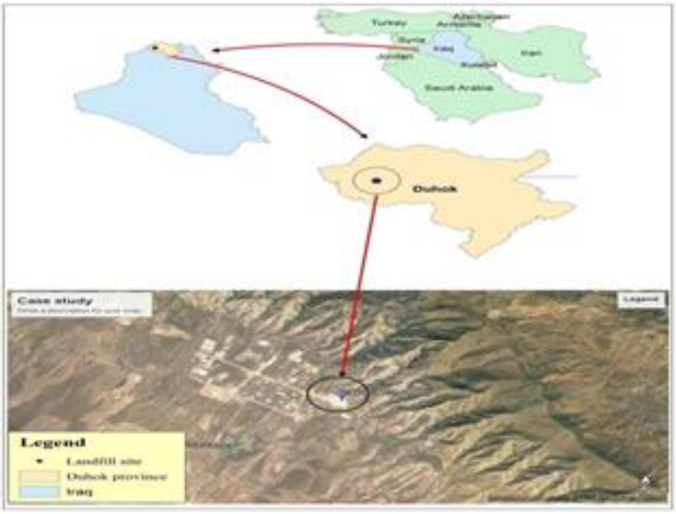

Figure1: The location of Kwashe industrial area and landfill in Kurdistan Region of Iraq

Kwashe industrial area has also a large solid waste separation factory that receives approximately 900 tons of municipal solid waste and about 50% organic kitchen waste. After waste separation, about 40% are thrown into an open landfill in a nearby valley without any separation and treatment because the load of municipal garbage is more than the capacity of the waste separation factory, a huge amount of these wastes is thrown directly into the open landfill as shown in figure 2. Figure 2 also shows release of self- igniting gasses, attraction of wild bird to organic food waste and many drains of leachate under the heap of landfill waste [6].

Figure 2: Different views of Kwashe open landfill, showing direct thrown of garbage, release of self- igniting gasses, attraction of wild bird to organic food waste and many drains of leachate under the heap of landfill waste

Samples Collection and Treatment of Leachate

Samples of leachate were randomly collected from different sites downward drain of landfill and taken in plastic bottles. Some of them were sent directly for analysis in the laboratory and others for leachate treatment by aeration and filtration processes over a month. 50L leachate was poured in 70L plastic jars. Small water propulsion motor was inserted and generated in leachate jars to start aeration processes. This process continued for weeks until the dark reddish black returned to slight brown in color indicating that there had been significant decomposition of organic waste. Then a few samples were then sent off to be investigated for analysis.

The filtration process was carried out by using big bottles of water 20L, by putting sequential layers of sand, coals and silt in 5 cm thickness respectively and replicated till the bottle was filled to about three quarters. Tiny holes were placed at the bottom of the bottle to allow filtered leachate to drain. Aerated leachate was then poured into the filter and filtered leachate that cleansed was received in clean vessels. Samples of filtered leachate were analyzed for studied parameters.

Determination of Heavy Metals

Samples of landfill leachate and soil affected by this leachate were dried in an oven at 75 °C for 72 h. 0.5 gram of each sample was digested in the presence of aqua regia (a mixture of nitric acid HNO3 + HClO4 in the ratio 3:1) for heavy metals, lead (Pb), cadmium (Cd), chromium (Cr), copper (Cu), cobalt (Co), manganese (Mn), Aluminum (Al), Iron (Fe), Nickel (Ni), and zinc (Zn). Then, the samples for heavy metals determination in the filtrate samples was carried out by using atomic absorption spectrophotometer (AA-7000) graphitefurnaceatomizer (GFA-7000) Shimadzu at the Environment directorate in Duhok city, according to standard analytic methods [2,7].

Soil Chemical Properties

The electrical conductivity EC of soil saturation extracted after equilibrium for 24 hours was measured by the EC-meter as described by another study [21]. The pH of the saturation extract was measured by the pH-meter. Soluble Ca+2 and Mg+2 were determined by complex metric titration with versenate (EDTA) while extractable Ca+2 and Mg+2 were determined at pH (8.2) by complexometric titration with EDTA according to a past inquiry [8]. The extractable and soluble Na+, Li+, Ba2+ and K+ were measured by flame photometer [9]. Chloride Cl- was measured by titration with AgNO3 and K2Cr2O4 as an indicator, as described by an earlier study [8], sulfur was determined by ammonium acetate method [10], and phosphorous was determined according Olsen method [11].

Statistical Analysis

The collected data were fed into SPSS software [12] using one-way ANOVA method for analysis according to the following model:

$У_{ij}= \mu+T_i+\delta_{ij}+e_{ij}$

Where: $У_{ij}k$= observation of studied heavy metals.

μ= overall mean

T_i= the effect of treatment.

e_{ij}= experimental error.

Also, the difference between the means was tested using Duncan's multiple range test [13].

Result and Disscusion

As indicated in Table 1, the most chemical properties of soil affected by landfill leachate are the soluble salts like soluble sodium and potassium over the acceptable ranges. Consequently, these soluble salts will increase the electrical conductivity of the leachate about 6 folds greater than the acceptable ranges in the soil. These huge amounts of soluble salts are mainly due high consumption of salts in food cooking in Duhok city and the food waste contributes over half of dumped waste in Kwashe open landfill. Table 1 also shows considerable increases in the other alkaline soluble calcium and magnesium to the maximum permissible limits in the soil, while the alkaline extractable barium reaches over acceptable ranges.

The extractable forms of calcium, magnesium, sodium, potassium, and lithium are within acceptable ranges indicating not being enough to cause a threated in deteriorating soil and environmental chemistry. However, these values are also greater than the average content of soil not affected by landfill leachate in this area [14,15].

Table 2 revealed that the soil downward landfill leachate fortunately did not reach the hazardous limits of their content in heavy metals and did not exceed the standard ranges in soil. Especially the two main toxic, non-essential health damaging, lead and cadmium were below detection limits. This low content of heavy metals in leachate is due to the main food, organic origin of landfill wastes that originally contain low amounts of heavy metals. It also indicates that the landfill is less affected by hazardous industrial wastes, like electronic and car batteries separated in the first stage in Kwashe waste separated factory.

Table 1: Some chemical properties of soil affected by landfill leachate

No.	Parameters	Concentration mg/kg	Acceptable ranges
1	pH	8.4	5.5-10
2	Sodium adsorption ratio, %	4.7	0-13
3	Electrical Conductivity (ds/m)	6.37	0.6-1.2
4	Chloride (ppm)	505.5	8-1800
5	Sulfur (ppm)	466.8	40-500
6	Phosphorus (ppm)	487.6	35-5300
7	Soluble calcium (ppm)	108.4	1-150
8	Soluble magnesium (ppm)	49.1	1-50
9	Soluble sodium (ppm)	238	0. 5-120
10	Soluble potassium (ppm)	158	1-10
11	Soluble barium (ppm)	77.6	-
12	Soluble lithium (ppm)	0.1	-
13	Extractable calcium (ppm)	20970	700-500000
14	Extractable magnesium (ppm)	2365	400-9000
15	Extractable sodium (ppm)	2750	150-25000
16	Extractable potassium (ppm)	3840	80-37000
17	Extractable barium (ppm)	3600	100-3000
18	Extractable lithium (ppm)	40	3-350

Table 2: The heavy metals content of soil affected by landfill leachate

No.	Heavy metals sample	Concentration mg/kg	Acceptable ranges
1	Zn	149.9	1-900
2	Pb	BDL	2-300
3	Cd	BDL	0.01-2
4	Mn	1348.6	20-10000
5	Cr	63.1	5-1500
6	Fe	17325.3	2000-550000
7	Cu	82.68	2-250
8	Ni	286.3	2-750
9	Al	15466.1	10000-300000
10	Co	0.944	0.05-65

In Table 2, the statistical analysis of treated landfill leachate by aeration and filtration indicates a significant influence of primary treatment in reducing all heavy metal content comparing to the original content in the non-treated leachate. However, the original content in most cases did not reach the risky levels and reduced further by aeration ad filtration. The rational explanation of these positive effects is that the aeration decomposes most degradable organic matter in the leachate to reduce the amount of suspended solid which contain heavy metals by precipitation and settle to the bottom of aeration vessels to be removed with sludge formed after aeration. Also, filtration absorbs a considerable amount of this heavy metal in the surface of bio film created in sand and gravel used as filters and to prevent them to transfer with filtered effluents and to allow maximum purification of leachate in this stage [16].

Heavy metals are broadly categorized to three groups according to their physiological role in living tissues of plants and human being, to essential micronutrients that have a vital positive functions in metabolism pathways as they act mainly as enzyme cofactor, like Zn, Mn, Cu, and Fe and have minimum health effects compared with the other two groups as living cell utilize widely in there functions. The other categories of heavy metals are called probably essential as they have positive effects on certain plant species and absent in others, as Ni, Co, Vn, and Cr, which their negative effects are greater than the first categories as its role in the metabolism is still unowned and many environmental issues have been recorded with this group. The third group is called toxic, non-essential heavy metals which do not have any positive metabolic actions in living cells, as Cd, Pb, As, and Hg and have the tendency of bioaccumulation in living cells to cause serious health effects at minimum cancer [17,18]. The ranges of heavy metals concentrations in landfill leachates in various countries around the world are for Cd 0.2–20 mg L-1, Cr 5–600 mg L-1, Mn 0.01–70 mg L-1, Fe 0.3–220 mg L-1 and close to the results stated in this study [19,20].

Table 2 also revealed that there were little amounts of the studied essential micronutrients like Zn, Mn, and Cu, except Fe normal that elevated over ranges but reduced with final filtration treatment to be near acceptable ranges. Also, probably essential micronutrients in origin that do not treated leachate are few and further reduced by primary treatment. They did not exceed almost global standards of heavy metals in leachate, while the two very toxic heavy metals Cd and Pb studied in this research fortunately, were below detection limits BDL. Aluminum is called an innocent element and its environmental effect is very low; however, its content is very high in leachate especially with primary treatment and more about 6 thresholds of global standards. This high level of Fe and Al is due to huge amounts of their impurities in municipal trash consumed by Duhok city citizenships.

Table 3: The average heavy metals (mg/l) ± standard error in untreated and treated leachate by aeration and filtration samples

Heavy metals	Mean concentration mg/l in untreated control ±S.E	Mean Concentration mg/l in treated by Aeration ±S.E	Mean concentration mg/l in treated by filtration ±S.E	Significance (p)	Maximum limit mg/l
Zn	0.092±0.001a	0.078± 0.008b	0.0048±0. 007c	0.0001	2.3
Pb	BDL	BDL	BDL	-	0.1
Cd	BDL	BDL	BDL	-	0. 01
Mn	0.987±0.0508a	0.759±0.042b	0.536±0.013c	.000	5
Cr	0. 054±0.001a	0. 051±0.005a	0. 042±0.013c	.001	1
Fe	8.994±0.637a	8.176±1.802a	6.2780±1.690c	.000	5
Cu	0.707±0.087a	0.6187±0.837a	0.45033±1.943c	.000	0.5
Ni	0.1968±0.0136a	0.1647±0.027b	0.1292±0.019c	000	1
Al	10.986±0.584a	9.849 ±0.786b	7.809±0.961c	000	2
Co	0.00073±0.00002a	0.00068±0.00005b	0.00050±0.00006c	.003	1

Means that do not share letter in a column are significantly different at probability level ($P \leq 0.05$), BDL= Below Detection Limit.

Zinc (Zn)

It is an essential micronutrient, both for metabolisms of plants and human beings, especially in the cell division, embryonic formation and genetic expression of individuality. Findings have revealed that the zinc concentration are close to homeostatic level and don not emit any hazardous effects both for the environment and human wellbeing.

Manganese (Mn)

As, Zn is an essential micronutrient for almost all species, found in food staff as grains, herbs and seafood as its essential for diatoms proliferation, which is considered the base of the food chain in aquatic ecosystems. The high amount of Mn can damage the central nervous system and cause Parkinson disease, also cause serious respiratory disease. However, its concentration also lay with homeostatic level and does not emit any hazardous effects both for the environment and human wellbeing. The leachate content of Mn in Kwashe landfill leachate is minimum and safe for the environment.

Chromium (Cr)

The most abundant sources of Cr+4 and Cr+6 are tanneries of leather, textile, electro painting, and chemical and also steel manufacturing. While Cr+3 is vital for better human cardiovascular functioning and metabolism as well as fighting diabetes, Cr+6 is carcinogenic, mutagenic, immune system destructor, liver and kidney damage, and also causes many well documented human and animal diseases. The human body can consume 1 g of Cr+3 in day without negative effects. Cr content in leachate under study area is also permissible.

Iron (Fe)

Iron is an essential element for both plant growth and human nutrition. Fe plays a gracious role in the metabolism of all living organisms, especially plants, as it acts as a cofactor with many enzymes as in ferredoxin with nitrogenase enzyme that convert inert atmospheric nitrogen to ammonia to be utilized by plants. Iron deficiency in human blood also can cause serious health defeats as anemia and neurodegenerative diseases. Although there are no health-based guideline values proposed for iron in human, ranges of daily intake of 10 to 50 mg/day is sufficient for better iron bio availability. The leachate in Kwashe landfill even with aeration and filtration contain relatively higher than slandered and need further advanced treatment to reduce its level in allowable levels [17].

Copper (Cu)

Copper is another essential micronutrient for plant majority and a woman's pregnancy good brain health and its deficiency lead to blood, bone and skin diseases as well as the two main economical crops of wheat and rice will not yield in soil deficient in copper. And, many worldwide standards do not consider copper as a carcinogenic agent for humans. Copper in Kwashe landfill leachate contains a little bit of copper, but with aeration and filtration treatments it fell to acceptable ranges [18].

Nickel (Ni)

Nickel is essential for primitive microorganisms as bacteria and archaea, while probably essential for higher organisms as well, including plants. Higher concentration of nickel causes many kinds of cancer and other diseases for human being and retard the biosynthesis of chlorophyll in plants. However, the leachate in this study is low and adsorbed in organic matter so it reduced considerably by filtration with coal filter in this study [2].

Aluminum (Al)

The Aluminum and most heavy metals behavior and effects on human being healthcare are not fully understood yet. Al and Co are among the heavy metal that are believed to have some beneficial physiological role in living organisms as plants but yet classified among harmful metals. Al solubility increases in acidic soil and causes chemical tress in plants resulting in shallow roots and lowing yield, also excess Al in human blood causes disruption of both kidney dialysis and neurological system. The Al in Kwashe leachate is above permissible ranges and even with treatment by aeration and filtration, it remains significantly higher than the allowable ranges. So, further advanced treatment is needed to reduce their levels. However, the alkaline nature of the sounding soil will precipitate Al and prevent it to enter in the first trophic levels, which are grazing herbs across leachate drains.

Cobalt (Co)

The Co has little toxic characteristics, but excessive amounts of Co irate respiratory system, inhibit the formation of vitamin B12, aggravate goiter and cause cardiomyopathy inhuman. Co has a beneficial role in many living cells as it is a part of B12 vitamins and its deficiency in human blood causes anemia and has adverse effects on the nervous system. The leachate in this study contain negligible amounts of Co that is a thousand times lower than allowable ranges, so it does not emit any risks in the environment and human health [18].

Table 4 demonstrates that there a close correlation between landfill leachate content of heavy metals and soil content affected by this leachate of Zn, Fe, Cu, Al,and Co at ($P \leq 0.01$) as the leachate content of these metals easily leaches with effluents, especially Fe and Al compared with other metals. While no correlation is observed between Mn and Ni content in leachate and the soil affected by this leachate as the origin content in leachate are low, there is a negative correlation between Cr content in leachate and soil.

Table 4: The correlation coefficient between landfill leachate content of heavy metals and soil content affected by this leachate ppm

	Content of Heavy metals ppm of soil affected by leachate	Correlation coefficient
Leachate content of heavy metals ppm	Zn	0.91**
	Pb	-
	Cd	-
	Mn	0.19
	Cr	-0.36*
	Fe	0.81**
	Cu	0.79**
	Ni	0.09
	Al	0.99**
	Co	0.99**

Where*(P≤0.05)**(P≤0.01)

Conclusion

The most chemical properties of soil affected by landfill leachate are the soluble salts like soluble sodium and potassium over the acceptable ranges. Consequently, these soluble salts will increase the electrical conductivity of the leachate about 6 folds greater than the acceptable ranges in the soil. The two main toxic, non-essential health damaging, lead and cadmium were below detection limits in leachate. The treatment of landfill leachate by aeration and filtration indicated a significant influence of primary treatment in reducing all heavy metal content comparing to the original content in the non-treated leachate. There were little amounts of almost essential micronutrients in Kwashe landfill leachate like Zn, Mn, and Cu, except Fe normal that were elevated over ranges but reduced with final filtration treatment to be near acceptable ranges. Aluminum is called an innocent element as its environmental effects is very low; however, its content is very high in leachate and even with primary treatment not match and more about 6 thresholds of global standards. And even with treatment by aeration and filtration, Al remains significantly higher than the allowable ranges, so further advanced treatment is needed to reduce their levels. However, the alkaline nature of the sounding soil will precipitate Al and prevent it from entering in the first trophic levels, which are grazing herbs across leachate drains. There is a close correlation between landfill leachate content of heavy metals and soil content affected by this leachate of Zn, Fe, Cu, Al, and Co at ($P \leq 0.01$) as the leachate content of this metal easily leaches with effluents, especially Fe and Al compared with other metals. And in most cases the filtration process is more efficient than aeration to reduce heavy metals in landfill leachate.

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Authors' contributions

All authors contributed toward data analysis, drafting and revising the paper and agreed to be responsible for all the aspects of this work.

Conflict of Interest

The authors declare no conflicts of interest.

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