Bioaccessibility and health risk assessment of heavy metals pollution in street dust from urban areas in Diwaniya, Iraq

A Dissertation Submitted

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In

Biology (Microbiology)

By

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Abstract

This study sought to define the metals bioaccessibility from street dusts in Diwaniyah streets for the first time. The samples were collected in order to research the levels, sources, and pollution indices of Cd, Ni, Zn and Cu. The geoaccumulation Index (Igeo) found that the contamination level for Cd was moderately polluted to strongly polluted status in Al-Forat, Naseej and Um-Alkail streets. Hence, it is significant to focus on the observation of cadmium in the local environment. The relative bioaccessibility of cadmium was high in Alorat Street (90.2%). and EF has employed to know possible sources of heavy metals studied in the street dusts, Cd and Pb are the same sources and originated from tire wearing, brake wearing and heavily traffic, while Ni and Cu were originated from metal processing industries. The ecology risk of heavy metals results were carried out for the calculation of risk dust in environment. These results can help us to get a better sight of potential health risks to Diwaniyah residents.

Pollution indices in Diwaniyah streets were used to measure population exposure suggested that both of metals studied in Diwaniyah streets dusts were harmful effects of human environment.

CHAPTER ONE

1. Introduction

Diwaniyah city is an urbanized area that located southern Iraq where the factories increasing recently as a result to economic prosperity and population growing. Street dust particles can be contain heavy metal and organic pollution, which can possibly re-suspended in the air due to movement of vehicles, the sources of heavy metal in street dust is due to motor vehicle emissions, the wear and tear of tires, and industrial emissions among others, which all related to the dust particles and could enter the body by ingestion or inhalation (Gu 2014; Li et al. 2015; Yang et al. 2016).

Almost half of the world's population lives in urban areas, and their health issues and livelihoods are a major concern (Shi et al. 2008). Hence, assessment of heavy metals pollution in the streets of Diwaniyah city is important which can causes serious problems to human health of local residents. Heavy metals is non-biodegradable, toxicity,persistence and tend to accumulate in human tissue(Huang et al., 2013; Liu et al., 2016; Varol, 2011).

Using bioaccessibility through SBET method is accurate and more actual to measure the possibility health risks due to heavy metals can dissolve in gastrointestinal fluid and simulated the biochemical conditions in the human gastrointestinal tract (Ruby et al., 1999; Bruce et al., 2007; Schroder et al., 2004).

Many of previous studies have focused on heavy metals contamination in urban streets from different countries (Chon et al., 1995; De Miguel et al. 1997;

Charlesworth et al., 2003), a limited studies have explored the heavy metals pollution in the urban streets of Iraq. However, there is a lack of data on the bioaccessibility of heavy metals in Diwaniyah streets dust based on in vitro digestion model. This highlights the importance of implement more bioaccessibility researches of heavy metals in streets dust in order to improve and control risk assessments.

1.2. Objectives of Study:

The main objectives of this study were:

1- To assess the total concentrations and the bioaccessibility of heavy metals in streets dust of Diwaniyah city

2- To evaluate heavy metals contamination degrees in streets dust using pollution indices

CHAPTER TWO

2. Literature review

2.1. Heavy metals

Recently, heavy metals are increasing in different urban area which are related to chemical hazards and human health. Metal is potential toxicity occur naturally or anthropogenic (Duffus, 2002). In the same time heavy metals could be useful for nutrition of plants, animals and human being such as Fe, Mg, Na, and V, and in other hand some metals like Hg, Pb, and Cd have positive nutrition effect and may tend to accumulate in the tissue (Spiegel, 2002).

2.2.1. Copper

A copper considerate one of the common heavy element with atomic number 29 and with a density of 8.96 g\cm^3 It is ductilemetal with a very high electrical and thermal conductivity. However, copper is one of the few metallic from the copper.Later , through thousands of years , copper was the first metal does not react with water , but it does slowly react with atmospheric oxygen to form a layer of black brown copper oxide and which also recently related with a dust particles within air

Sources of copper (Cu)

The most total amount of copper on earth is vast with around 1014 ton in the top kilometers of earth,s crust , that is around 5 milliom year,s worth at the current of extraction. Also , the human activities taked avast space through the industrial processes which used most copper by minning or extracted as a copper sulfides from a large open pit mines. In addition, chile was the top of copper producer, copper could also be relcoverd through the origin [insitu] leach process.

The available quality is barely sufficient to allow all countries to reach developed level of copper usage. Copper is used in roofing , currency and for photograghic technology. Copper plating and copper sheating were widely used to protect the under-water hulls of ships Plants , animals and some microorganisms used copper as an essential trace element, The natural ratio of copper within human body at level of about 1.4-2.1 mg per kg of body mass Absoebed of copper begin in the gut , then transported to the liver bound to albumin after processing in the liver.

In the second phase, copper is distributed to other tissues that involved the protein ceruloplasmin, which carrying most of copper in blood. This protein also carries the copper that is extracted in milk. Copper in the body mass through enterophepatic circulation, and the body is able remove some excess copper as a necessary by excreating it via bile that carries som copper out of liver which is not then reabsorbed by the intestine.

The health effects of increase copper consumption

Peradventure increasing of copper consumption it will cause sever illnesses which involve kideney failure and death can occure with as alittle ratio as 1 gram of copper sulfate. Eithr , the symptoms which copper causes are vomiting, nausea , bloody diarrhea , stomach pain , fever , low blood pressure , anemia and finally heart problems which all result from copper toxicity .

2.2.2. Nickel (Ni)

Nickel is a chemical element with the atomic number 28 and with a density of 8.908g\cm^3 . it is white silvery lustrous element with a slight golden tinge Nickel is a hard ductile because it is transition metal. The pure nickel is used to maximize the reactive surface area. Even though, pure native nickel is found in earth's crust only in tiny amounts, in the altramafic crust but it present related with iron almost Nickel – iron meteorites that were not exposedto oxygen when outside earths atmosphere. The uses of nickel are summarized by a natural meteoric nickel ironalloy has been traced as for back as 3500 BCE. It was first isolated and classified as the chemical element in 1751, who initially mistook the ore for a copper mineral. An economically important sources of nickel include is the iron ore limonite, which often contains 1-2% nickel The other mineral ores of the nickel include pentlandite and the mixture of NI- rich natural silicates known as garnierite.

Nickel is one of many elements that is include iron, cobalt and gadolinium which characterize by the silicon burning process and later set free in largr quantities. This metal are mostly used about 68% of world production stainless steel. A percentage 10% is used for nickel-based and copper-based a illony steels , 3% in foundries , 9% in plating and 4% in other applications including fast growing , battery sector. Nickel is a necessary nutrient element for some microorganisms and plants that have enzymes which contain as an active sites. The bulk of nickel is mined from two types of ore deposites , the first is a laterite, where the principal or mineral mixtures are a nickel ferouslimonate.

2.2.3. Cadmium (Cd)

A heavy chemical element with atomic number of 48 and with a density of 8.65 g\cm^3 It is soft bluish-white metal , it demonstrates oxidation states +2 in most of its compounds and like mercury , it has a low melting point than the transition metals.

Cadmium not considered transition metals. Cadmium in earth's crust is between 0.1 and 0.5 parts per million (ppm). Cadmium occurs as a minor component in most Zinc ores and is abyproduct of zinc production. Cadmium was used for a long time as a corrosion- resistant plating on steel , and cadmium compounds are used as red – orange and yellow pigments , to colour glass and to stabilize plastic. Cadmium use id generally decreasing because it is toxic and it has replaced with one of its new few uses is for solar panels. Although cadmium has no known biological function in higher organisms, and it dependent carbonic an hydrase has been found in marine diatoms. Animal kidney is the first organ which reach the limit of safe of human consumption

It is not worthy that the highly exceeded maximum limit levels of Cd were identified I animal kidneys from both inside and outside the local contaminated area .

2.2.4. Lead (Pb)

Is a heavy element with atomic number about 82. It is denser than more common materials, lead is malleable and soft and often has a relatively low melting point. It has gray colour when exposed to air, lead has the highest atomic number of any stable element and three of isotopes each include amajor decay chain of heavier elements Lead is easily extracted from its ores, a principal ore of lead, often bears silver. Production of lead was about ten million tonnes, over half of which was from recycling. Lead's high density , low melting point , ductility and relative inertness to oxidation make it useful. these properties combined with its relative abundance low coast , resulted in its extensive use in construction , pumping , batteries , and shot weights , silders , wight paints , gasoline and radiation shielding. lead's toxicity was recognized , and its use has since been phased out of many applications , the steps of lead produced are still use by many countries to applicate it. The toxin of a lead accumulate in soft tissues and bones , it acts as aneuro toxin damaging the nervous system and interfering with the function of biological enzymes , causing neurological disorders , such as brain damage and behavioral problems.

2.3. Bioaccessibility of Heavy metals

The bioavailability of heavy metals can be determined by different ways, including in vivo experiment and in vitro digestion method, bioaccessibility can be defined as the quantity or fraction which is released from the food matrix in the gastric-intestinal tract and turn out for absorption in human body (Heaney 2001). The bioaccessibility can be actual in human health and accurate experiment results in humans or experimental animals can support knowledge on the bioavailability of ingested heavy metals contaminations in the media (Oomen et al., 2002). In other hand, in vivo method is expensive, more efforts needed, waste time, and ethical considerations (Versantvoort et al., 2004). Meanwhile, in vitro digestion method is easy, fast, cheap, and can provide mimic situations for animals and humans researches (Hur et al., 2011; Juhasz et al., 2007). In addition, in vitro digestion method could reduce experimental animals used. Determining the bioavailability of heavy metal concentration can facilitate more realistic exposure and health risk assessments (Guney et al., 2012). The Simple Bioavailability Extraction Test (SBET) method

(British Geological Survey, UK) was used in this study due to give accurate results and is easy with experimental in the lab.

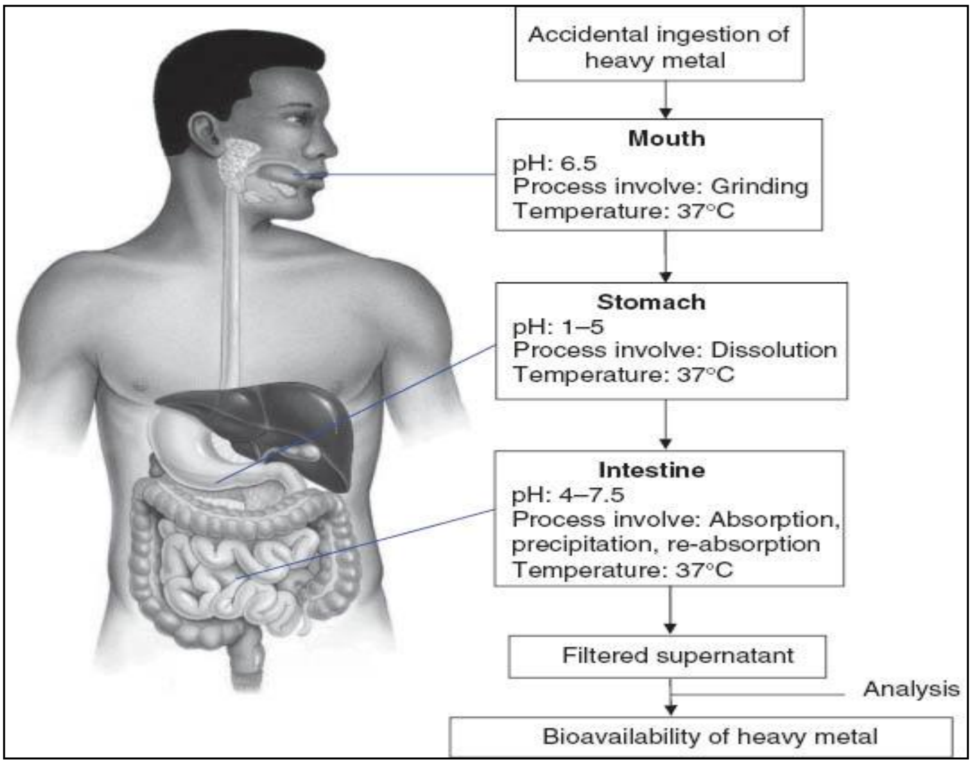


Figure 1 In vitro digestion model process in the human body (Source; Yuswir et al., 2013.

CHAPTER THREE

3. Materials and methods

3.1. Study area

Diwaniyah city is the important and the largest city in Iraq, the population of about 1.2 million. The urbanization and industrialization in the city has made vehicular growth and increasing numbers of vehicles in the roads. Moreover, Iraq rebuilding efforts have been devote to the recuperation and repair of sorely damaged urban infrastructure can contribute dusts in the streets. In Diwaniyah city has a power plant and around 3 industrials that include oil refinery, chemical, steel mill, cement plant, textile, plastic manufacturing. The climate characterized in Diwaniyah is arid to semiarid and dry hot in summer (50 ˚C) and cold in winter (4 ˚C); the annual rainfall mean is approximately 112 mm. The samples collected from different urban cities including: Al- Forat (S1), Um-Al-Khail (S2), Naseej (S3), Orouba (S4), Eskary (S5),

3.2. Street dust sampling

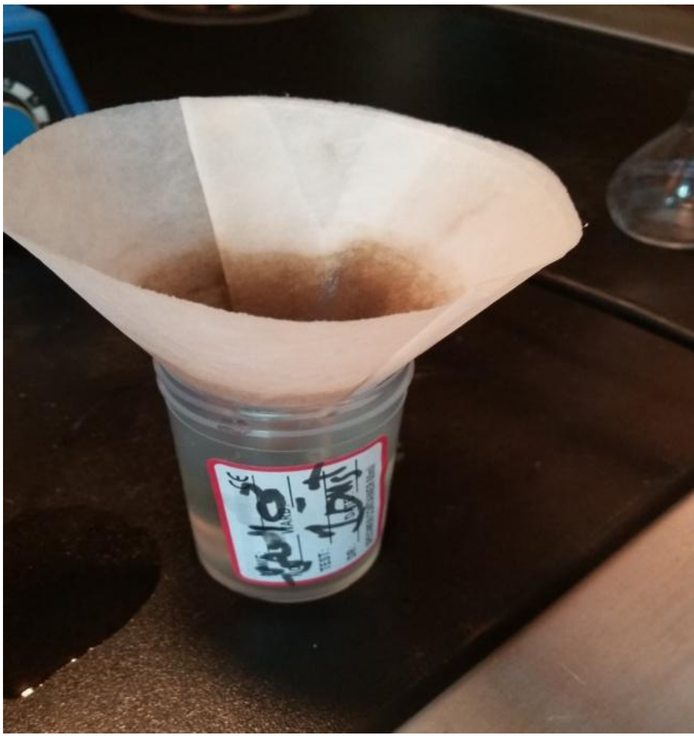
The street dust was collected according to Trujillo-González et al. (2016). The streets dust samples (n=5) were collected on February, 2019, the sites, designation, description, and coordinates of sampling collected are shown in Table 1. The frame of 1 m, brushes and a plastic hand-shovel were used to collect the dust samples from Diwaniyah streets. The polyethylene bags were used to store dust samples collected, labeled and carefully transported to the laboratory.

Table 1 The coordinates and description for each sampling station

Station	Sampling site	Coordinates	Description
1	Al-Forat	32° 1'26.23"N 44°54'16.50"E	Power electric plant
2	Um-Al-Khail	32° 0'10.49"N 44°54'10.79"E	Commercial
3	Nassej	31°57'57.76"N 44°57'16.68"E	Rubber factory, Textile factory
4	Orouba	31°59'29.96"N 44°54'53.67"E	Commercial
5	Eskary	31°59'55.03"N 44°20'49.72"E	Commercial

3.3. Samples digestion and analyses

The dust samples (0.5 g) were digested in 10 ml solution of a mixture HNO_3 (AnalaR grade, R&M 65%) and $HClO_4$ (AnalaR grade, R&M 70%) in the ratio of 4:1(v/v), into heater at low temperature (40˚C) for 1 h and then at 140˚C for 3 h (Ismail, 1993). The digested samples were then diluted to 40 ml with double-distilled water (DDW) and filtered through Whatman No.1 filter paper into pre-cleaned 40 ml volumetric flasks. The samples were measured for trace metal concentration using Atomic Absorption Spectrophotometer. The data are presented on dry weight basis (mg/kg dry weight). In order to avoid contamination, all glass wares were soaked in acid wash (10 % HNO3) for at least 24 h and later rinsed with double distilled water and air dried before use. To ensure precision and accuracy of the analytical method, quality control calibration curves were generated by analyzing multiple-level calibration standards.



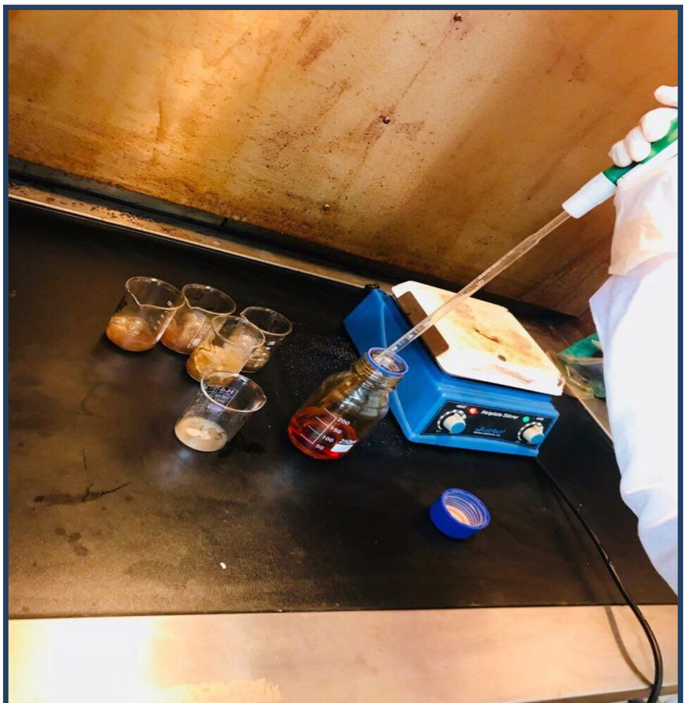


Figure 2: Procedures analysis in the lab

4.1. Geo-accumulation index (Igeo)

The geo-accumulation index (Igeo) was used to assess the metals pollution degree in the dust (Santos Bermejo et al., 2003). The global average shale (Turekian and Wedepohl, 1961) as background for metals references in geo-accumulation index (Igeo) values which were using the follow equation,

$$Igeo = \log_2 \left(\frac{Cn}{1.5Bn}\right)$$

$$EF\ metal = \frac{(Cx / Fe)\ sample}{(Cx / Fe)\ shale}$$

4.2.Contamination factors (CF)

The contamination factor (CF) measured as:

CF= C sample/ C background

where, Cm Sample is the concentration of a given metal in river sediment, and Cm Background is value of average shale. (CF < 1) low degree, (1 ≤ CF < 3) moderate degree, (3 ≤ CF < 6) considerable degree, and (CF ≥ 6) very high degree. Therefore,

Potential ecological risk (E_r^i)

The possible risk to the ecology is also calculated to assess the dangers of heavy metals in the dust and. Potential ecological risk factor (E_r^i) was propsed by Hakanson (1980)

CHAPTER FOUR

4. Results and Discussion

4.1. Total and bioaccessibility of metals levels in the street dusts of Diwaniya

The mean concentrations of total and bioacccessibility of Cd, Cu, Pb and Fe from all the collection sites are presented in Fig 2. Pb was the most abundant metals in the streets dust followed by Ni, Cu, Cd and Fe, It has been noted that streets dust was significantly contributed to heavy metal concentration levels in streets of urban areas (Karanasiou et al., 2009; Athanasopoulou et al., 2010). In general, Pb concentrations were found to be higher in streets of Al-Forat street (111.6 mg/kg) and the lowest level for Cd was in streets of Eskary street (0.03 mg/kg). However, the results were lower than those reported in Tehran streets (Iran), Delhi streets (India) and Baoji streets (China) (Kamani et al., 2017; Lu et al., 2009; Sezgin et al., 2004). The measured of Cu concentrations was highest in Naseej (65.8 mg/kg) due to oil lead that emission from power electric plant. In addition, the highest concentration for Cd (1.33 mg/kg) was also recorded in Al-Forat Street. The results of bioaccessible of Pb concentrations (Fig 3) were highest in Al-Forat street (96.1 mg/kg) and ranging from highest in (96.1 mg/kg) to lowest in Eskary Street (10.43 mg/kg), increasing of Bio-Pb in study area is due to high anthropogenic activities such as oil leaded and petroleum which are concentrated in these locations. Cd were also higher in Al-forat streets than in other locations. In contrast, the minimum concentration of Cd (0.0003 mg/kg) was in Orouba Street. Moreover, Bio-Cu was high in the Naseej streets (25.8 mg/kg) due to these sites being closer to commercial activity or chemical industries which are more affected by extensive human activities. The concentrations of total and bioaccessibility of Cu were high in Naseej and Al-Forat streets (42.04; 16.3 mg/kg) respectively that because metal-processing and power plant generation in the Al-Forat ans Naseej Streets as well as brake abrasion from frequent brake use can be associated with increasing Cu level in the street (Kamani et al., 2017; Duong and Lee 2011; Roubicek et al. 2008; Thorpe and Harrison 2008).

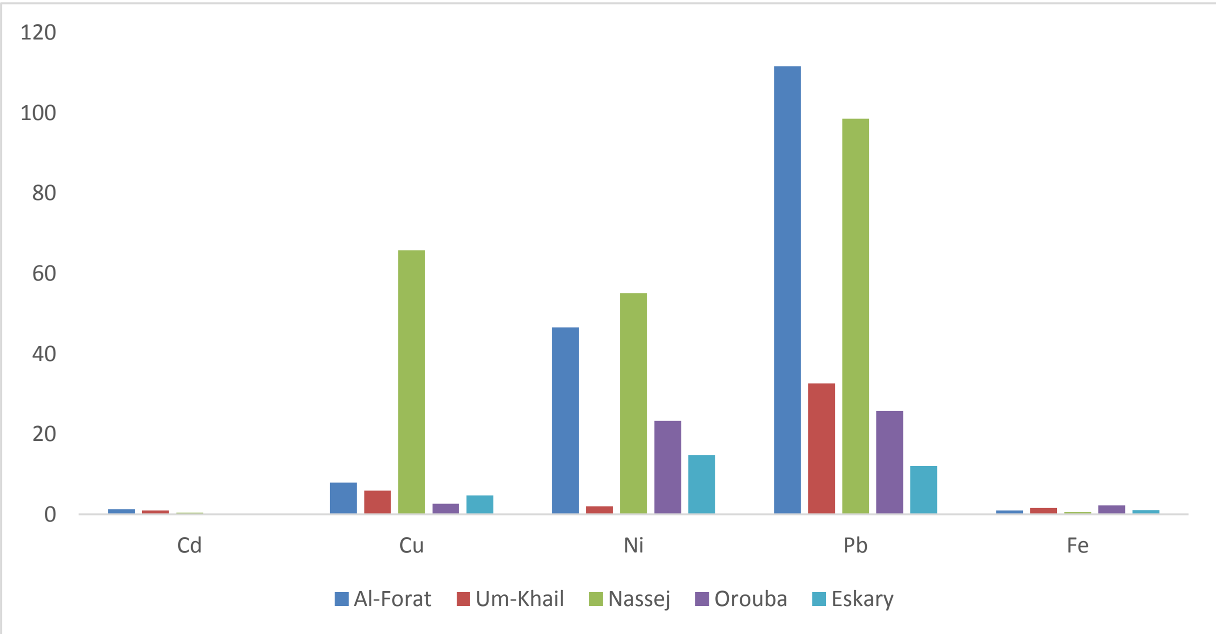


Figure. 3: The **c**oncentrations of five heavy metals in dust (mg/kg), Fe (%) of Diwaniyah City.

The relative bioaccessibility of heavy metals defined as the percentage of bioaccessible of metals with respect to its the total metals, the results showed metals contamination absorbed through the ingestion of dusts as shown in Fig. 3. The R-Bio-Cd in streets dust of Diwaniyah city ranges as of 5-76 % and the high

R-Bio-Cd among these sampling sites showed at Al-Forat Street. For R-Bio-Cu ranged from 18% to 52% and the high R-Bio-Cu was recorded in the Um-Al-Khail Street, this could be related to the socioeconomic actions that take place in the commercial sector, where a large amount of tall buildings and a high population density in Diwaniyah city (Trujillo-Gonzalez et al., 2016). There are not reported had investigated the metals bioaccessibility in the street dusts from Iraq in order to compare with previous studies. Therefore, the results of this current study were compared with different universal studies. Relative bioaccessibilty of Pb and Cu which are showed higher than the Cu bioaccessibility (>90%) was previously recorded from road dust and roadside soils along a peri-urban transect, Italy (Padoan et al., 2017).

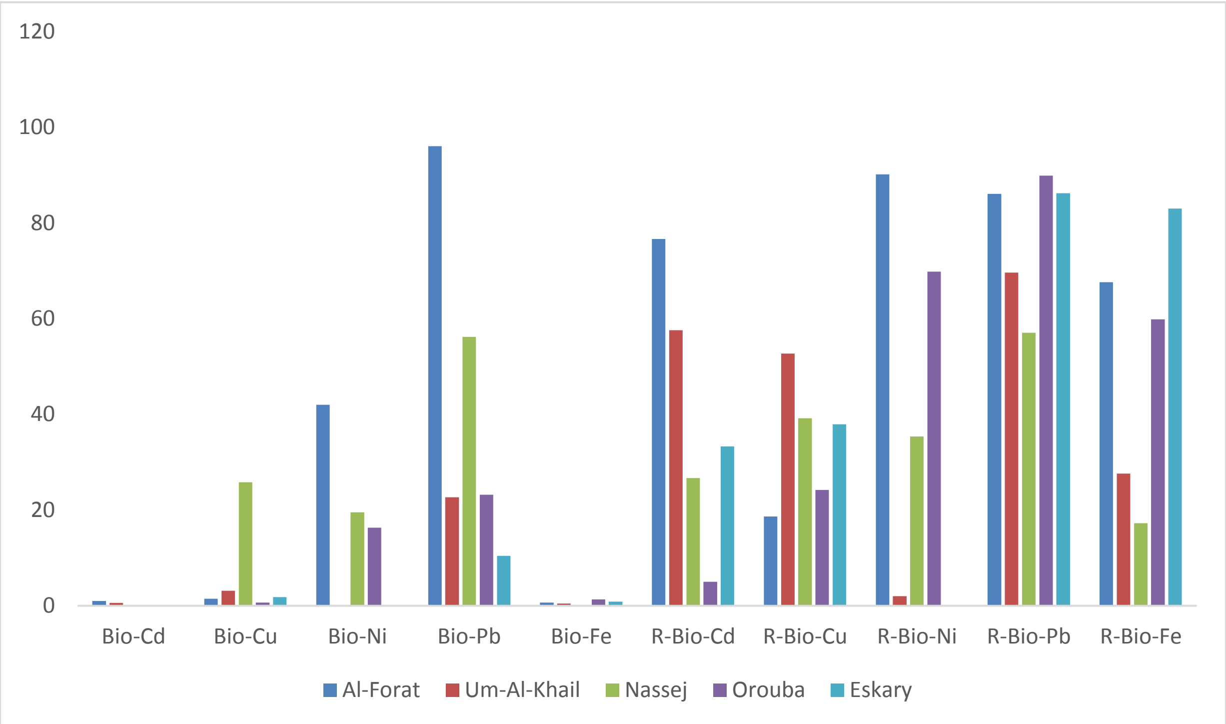


Figure. 4: The concentrations of Bio-Metals and R-Bio-Metals in dust (mg/kg), Fe (%) of Diwaniyah City.

4.2. Streets dust pollution indices

In order to know level of metals contaminations in street dusts, we used the Geoaccumulation Index (Igeo) proposed by Müller (1979) and which also utilized to assess degree of metals pollution from urban street dusts (Lu et al., 2009, Wei and Yang, 2010; Wei et al., 2015). The Igeo is calculated by using the following formula (Müller,1979):

$$\text{Igeo} = \log2\left(\frac{\text{Cn}}{1.5\text{Bn}}\right)$$

Where Cn is the calculated concentration of the studied element n in the dust and Bn is geochemical background value of the element n of average shale. The seven different classes of geo-accumulation index along with associated dusts street pollution are given in the Table 3.

It has showed that Cd concentrations in Um-Al-Khail and Al-Forat streets were much higher than average shale and based on Igeo found moderately polluted to strongly polluted in the same streets suggesting that the anthropogenic sources have a severe effect on the quality of these functional areas such as commercial and vehicle exhausted emission (Kamani et al., 2017).

Table 2: The Igeo and classes of metals in Diwaniyah streets dust

Sites	Cd	Class	Cu	Class	Ni	Class	Pb	Class
Al-Forat	1.56	2	-5.66	0	-6.26	0	-4.49	0

Um-Al-Khail	1.13	2	-6.09	0	-6.68	0	-4.92	0
Nassej	0.0	1	-7.82	0	-7.82	0	-6.05	0
Orouba	-6.22	0	-15.0	0	-14.0	0	-12.2	0
Eskary	-3.90	0	-11.3	0	-11.7	0	-9.96	0
Average[a] shale	0.30	0	45	0	68	0	20	0

Igeo value	Igeo class	Dust quality
<0	0	Unpolluted
0-1	1	Unpolluted to moderately polluted
1-2	2	Moderately polluted
2-3	3	Moderately polluted to strongly Polluted
3-4	4	Strongly polluted
4-5	5	Strongly polluted to extremely polluted
>5	6	Extremely polluted

[a]Turekian and Wedepohl (1961)

The contamination factor (CF) of Cd in the dust of Al-Forat and Um-Al-Khail streets were very high > 2. The total mean of contamination factors in the dust of sampling locations were decreased in the order of Cd > Pb> Cu > Ni. In this research,

Table 3: Contamination factor (CF) of heavy metals contamination in sampling locations

Stations	CF				Dust quality			
	Cd	Cu	Ni	Pb	Cd	Cu	Ni	Pb
1	4.43	0.17	0.68	5.58	high	low	low	high
2	3.30	0.13	0.02	1.63	high	low	low	moderate
3	1.5	1.46	0.81	4.92	moderate	moderate	low	high
4	0.02	0.05	0.34	1.29	low	low	low	moderate
5	0.1	0.10	0.21	0.60	low	low	low	low

The enrichment factor values (EF) for Cd was 20.5, 9.31 and 11.8 in Al-Forat, Um-Al-Khail, and Nassej streets respectively, were found to be severe enrichment ($5 \le EF \le 10$). While, EF value for Cd in other streets of Diwaniyah city was no enrichment. In addition, The EF value for Cu was also high in Nassej street (11.8), and also EF values for Pb were high in Al-Forat, and Nassej streets indicating severe enrichment, EF values less than 1.5 suggested that metals from natural sources, whereas, EF values higher higher than 1.5 evidenced that metals from anthropogenic emissions . Thus, in this study, the values of EF for metals in present study in all streets of Diwaniyah city were higher than 1.5.

Table 4: Enrichment factor (EF) of heavy metals contamination in sampling locations

Stations	EF				Dust quality			
	Cd	Cu	Ni	Pb	Cd	Cu	Ni	Pb
1	20.5	0.83	3.18	25.9	Severe	Low	Moderate	Severe
2	9.31	0.37	0.08	4.9	Severe	Low	Low	Moderate
3	11.8	11.8	6.42	39.0	Severe	Severe	Severe	Severe
4	0.04	0.12	0.71	2.67	Low	Low	Low	Minor
5	0.43	0.46	0.94	2.62	Low	Low	Low	Minor

Table 5 showed the potential ecological risk factor of the Diwaniyah streets . The Cd mainly contributed to the potential ecological risk factor of the Al-Forat street which is considerable risk in that station and Um-Al-Khail street can be categorized moderate potential ecological risk factor. Moreover, Ni, Cu and Pb can be categorized no risk were recorded in all stations.

Table 5: Enrichment factor (Er) of heavy metals contamination in sampling locations

Stations	Er				Dust quality			
	Cd	Cu	Ni	Pb	Cd	Cu	Ni	Pb
1	133	0.09	3.42	27.9	Considerable	Low risk	Low risk	Low risk
2	99	0.04	0.14	8.15	moderate	Low risk	Low risk	Low risk
3	45	1.31	4.04	24.6	Low risk	Low risk	Low risk	Low risk

4	0.6	0.01	1.7	6.45	Low risk	Low risk	Low risk	Low risk
5	3	0.05	1.08	3.02	Low risk	Low risk	Low risk	Low risk

Conclusion

The street dusts of Diwaniyah city has elevated four elements (Cd, Ni, Zn and Cu) concentrations.

The findings of this study indicated that Al-Forat, Um-Alkail and Naseej are highly contaminated by Cd in the street dusts. In vitro digestion model was used to measure bioaccessible fraction of metals in the street dusts that R-Bio-Cd was quite low in the Orouba Street and higher for R-Bio-Ni in Al-Fora Street, the different bioaccessibility values in Diwaniyah streets are associated with the differences anthropogenic activities sources. The contamination factor was very high in Al-Forat street and Naseej street and EF has employed to know possible sources of heavy metals studied in the street dusts, Cd and Pb are the same sources and originated from tire wearing, brake wearing and heavily traffic, while Ni and Cu were originated from metal processing industries. The ecology risk of heavy metals results were carried out for the calculation of risk dust in environment. These results can help us to get a better sight of potential health risks to Diwaniyah residents.

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