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A Comparative Study of Total and Bioavailable Cadmium and Zinc Concentrations and Distributions among Different Land Use Types within Baghdad City

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Abstract

Total and bioavailable levels of Cd and Zn in topsoil (0–20) cm taken from four different land use types (residential, commercial, industrial and mixed) of the urban area of Bagdad, Iraq, were analyzed by the flame atomic absorption spectrophotometer. This was with a view to appraising the influence of a variety of anthropogenic activities on heavy metal contamination of the urban soil. Results showed that the range of Cd and Zn were (0.00-6.35) mg/kg and (5.20-219.95) mg/kg respectively. As compared with the calculated world average of unpolluted soils, cadmium displays higher concentrations while the zinc concentration was within this common world range. The level of pollution was assessed using geoaccumulation index (Igeo), for all land use types Igeo decreasing order, followed the order of (Cd> Zn). The relative bioavailability for Cd, and Zn has been observed as 2.46-5 % and 2.80-9.78 % respectively. It can be concluded that, although total concentrations of the examined heavy metals were generally high, but at the same time the bioavailable ones were relatively low. This can be an indicator that with the recent environmental factors (alkaline to sub-alkaline soil pH), the toxicity of heavy metals to humans was in its minimum level.

Key Words: Heavy metals, Bioavailability, Geoaccumulation index, soil pollution

توزيع محتوى العناصر الثقيلة الكلية والجاهزة حيويا في مختلف انواع استخدامات الاراضي في مدينة بغداد، العراق

الخلاصة

تم تحديد المستويات الكلية والجاهزة حيويا من الكادميوم، الكروم، النحاس، النيكل، الرصاص والزنك في التربة السطحية (٠-٢٠ سم) التي تم جمعها من أربعة أنواع مختلفة لاستخدامات الأراضي (السكنية والتجارية والصناعية والمختلطة) من مدينة بغداد، العراق، باستخدام جهاز طيف الامتصاص الذري، بهدف تقييم تأثير الأنشطة البشرية المختلفة على تلوث التربة بالعناصر الثقيلة. النتائج تشير إلى أن محتوى التربة من الكادميوم يتراوح بين ٠.٠٠-٦.٣٥ ملغم/كغم، الكروم ٠.٠٠-٢٢٣.٧٥ ملغم/كغم،

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النحاس ٧.٨٥-١١٥.٠٠ ملغم/كغم، النيكل ٢٢.٩٠-٢٣٦.٢٢ ملغم/كغم، والرصاص ٠.٠٠-٢٣٠.٥٠ ملغم/كغم، والزنك ٥.٢٠-٢١٩.٩٥ ملغم/كغم. وباستثناء عنصر الزنك في جميع أنواع استخدامات الأراضي وعنصر النحاس في المناطق السكنية والصناعية، فقد سجلت جميع العناصر الثقيلة تركيز أعلى من المتوسط العالمي للتربة غير الملوثة. وباستخدام مؤشر التراكم الارضي تم تقييم مستويات التلوث بالعناصر الثقيلة في منطقة الدراسة، حيث كانت نتائج الترتيب التنازلي لقيم مؤشر التراكم الارضي كما يأتي:

الكادميوم>الرصاص>الكروم>النيكل>الزنك>النحاس،

الكادميوم>الرصاص>النيكل>الكروم>الزنك>النحاس،

الكادميوم>الرصاص>النيكل>الزنك>الكروم>النحاس

والكادميوم>الرصاص>الكروم>النحاس>النيكل>الزنك للمناطق السكنية والتجارية والصناعية والمختلطة على التوالي. النمط المكاني لتحليل المخاطر يشير إلى أن الكادميوم هو من الملوثات الأكثر أهمية المساهمة في المخاطر على صحة الإنسان. وقد لوحظ ان الجاهزية الحيوية النسبية للكادميوم، الكروم، النحاس، النيكل، الرصاص والزنك تتراوح بين ٢.٧١-٥.٩١٪، ٠.٥٦-٢.٣٪، ٣.٧٦-١١.٧٥٪، ١.٠-١.٨٧٪، ٢.٣٦-٩.٠٥٪، ٣.٩-١٠،٦٧٪ على التوالي. ويمكن أن نستنتج إلى أنه على الرغم من أن التراكيز الكلية للعناصر الثقيلة كانت مرتفعة بشكل عام، ولكن في الوقت نفسه لوحظ ان التراكيز الجاهزة حيويا منخفضة نسبيا. وهذا يمكن أن يكون مؤشرا مع العوامل البيئية (الدالة الحامضية التربة)، على ان سمية المعادن الثقيلة للإنسان هي في مستواها الأدنى.

الكلمات المرشدة: المعادن الثقيلة، التوافر الحيوي، مؤشر التراكم الارضي، تلوث التربة

INTRODUCTION

In recent years, with the development of the global economy, both type and content of heavy metals in the soil caused by human activities have regularly increased, resulting in the deterioration of the environment [1-3]. Heavy metals are extremely hazardous to the environment and organisms. Urban land use and structure have an influence on soil contamination. The urban area processes an extensive range of different land uses, such as traffic, industry, business, residential uses, gardens and public green spaces, differing in their patterns of human activity and their possible impacts on soil quality [4]. Land use and cover may serve as an indicator of disturbance, site history, management, and the urban environment; these factors result in a mosaic of soil patches [5]. According to spatial analysis, it was observed that the areas with higher metal concentrations were generally located in industrial and residential areas, roadsides and crowded commercial districts [6].

The major sources of heavy metal contamination in urban soils include vehicle emissions, industrial wastes, household garbage, building and weathered particles of sidewalk and precipitation in the atmosphere, etc. [7]. Oil refineries can be a major source of heavy metal pollution due to poor management, faulty equipment, and lack of environmental controls, and may have a significant impact on the surrounding environment [8]. Heavy metals in the environment can be divided into two classes: bioavailable (soluble, nonsorbed, and mobile) and nonbioavailble (precipitated, complexed, sorbed, and nonmobile). Many researches on heavy metal bioavailability has been conducted in soil systems as understanding the fate of heavy metals in soil and sediments is critical in examining the heavy metal effects on biota, metal leaching to groundwater, and metal move to the food chain [9]. Bioavailability is the extent to which a contaminant in a source is free for uptake. Misra, defined metal bioavailability as the fraction of heavy metal in the soil that is accessible to the food chain and to the plants [10]. Metal's bioavailability in soil is largely influenced by metal nature and soil properties. It is well known that the availability of a number of trace elements becomes greatest at low pH and the reasonable result is that plants absorb more quantities of toxic elements [11]. Organic matter in soil is another important factor affecting heavy metal availability. It was reported that heavy metal adsorption onto soil constituents declined when organic matter content in soils decreased [12]. Organic matter influences the availability of heavy metals in soils through its binding effect on soil components [13].

Materials and Methods

Study area

Baghdad city (33°14'-33°25'N, 44°31'-44°17'E), is situated in the Mesopotamian alluvial plain. The city was divided by the Tigris River into the right section (Karkh) and left section (Risafa) with a flow direction from north to south. It is described by arid to semiarid climate with cold winter and dry, hot summer, where the average annual rainfall is about 151.8 mm [14]. According to the ministry of planning, Baghdad covers an area of 4555 km^2, with a population density of 7,180,889 capita in 2009 [15].

In this study, four different land use types were selected within Baghdad city viz. residential (Al-Utayfiya), commercial (Palestine Street), industrial (Al-Duraa Refinery) and mixed area (Al-Karrada). Figure (1) shows the study area (Baghdad city) and the locations of the selected land use types.

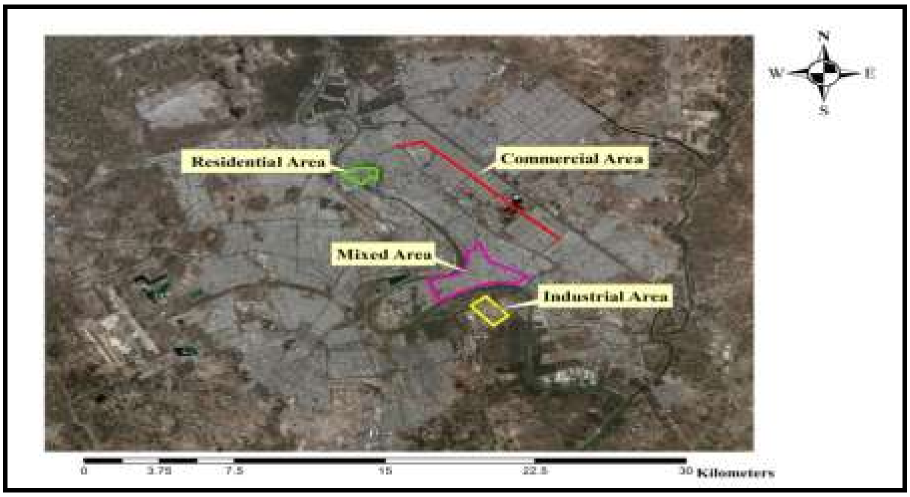

Figure (1). Study area and the selected land use type

Soil sampling and analyses

The selected locations of the samples were as follows: a total of 80 soil samples were collected within Baghdad city, including twenty samples (each sample representing a composite sample of at least 3 subsamples) from open areas and roadsides of each of the previous mentioned land use type areas. The sampling points were randomly distributed to cover the entire study sites. Soil sample was prepared by collecting about 1 kg of the surface soil (0-20 cm) by hand digging with a stainless steel spatula in labeled sacks and transported directly to the laboratory. All the samples were taken during the period from November 2013 to January 2014, and the sampling sites were documented using Global Positioning System (GPS) device type GARMIN. Soil samples were air-dried, and passed through a 2 mm sieve, after they had been dis-aggregated with a porcelain pestle and mortar. Then these soil samples were stored in clean self-sealing plastic bags for further analysis.

Electrical conductivity (EC) and pH were measured in a soil water suspension (soil: water, 1:2.5 by volume) by a calibrated conductivity meter and pH meter respectively (HANNA/ HI 9811-5). Determination of heavy metals included finding both total and bioavailable concentrations in soil samples. Wet-digested with a combination of HCl and HNO_3 was used to analyse the total concentration of heavy metals [16], while for the determination of the bioavailable portions, ten grams of air dried soil in 250 ml conical flask was extracted using 20 ml of diethylen-triamine pentacetic acid (DTPA) buffered at pH 7.3. The contents were shaken for 2 hours. The mixture was filtered through filter paper (Whatman No. 42). The filtrate was diluted with distilled water for 100 ml volume and stored in clean plastic bottle for metal determinations [17]. Heavy metal analysis was made by Atomic Absorption Spectrophotometry (AAS 6300, Shimadzu, Japan).

Results and discussion

Physical characteristics

Table (1) shows the main physical parameters determined for soil samples. Results indicate that the urban soil pH was ranging from 7.20 to 8.80 with mean and median values were between 7.92 and 7.90 respectively, it was near the neutral to sub-alkaline condition. Higher pH values were recognized to a high percentage of carbonate contents, which caused the neutralization of soil acidity, these findings are matched with what Al-Zubaidy [18] pointed out in his study on Iraqi soil acidity, with the limits of pH (7-8) because they contain calcium carbonate (lime: $CaCO_3$) and calcium sulfate ($CaSO_4.2H_2O$) in large quantities. Soil in the urban areas with an alkaline reaction has been reported to be quite common [19,20]. Closeness of the pH values observed for the soils may recommend an indication that the pH effect on the availability of the metals is in minimal and so do not *affect* site characterization.

The EC values varied from 70.00 to 4550.00 µS/cm with mean and median values of 1330.00 and 1205.00 respectively. This high EC is familiar in Iraqi soils which contain high content of calcium, magnesium and sodium salts [18], and these salts have high solubility in soil solution [21]. Variation in EC values shows that soluble salt concentration varies considerably. The higher values observed in the industrial area (Al-Duraa Refinery), where the lowest values were in Al-Utayfiya residential area.

Table (1). Statistical summary of pH and EC (µS/cm) of urban soils

	Land Use Type	Minimum	Maximum	Mean	Median	Std.
pH	Residential	7.40	8.80	8.10	8.10	±0.29
	Commercial	7.60	8.80	7.95	7.80	±0.32
	Industrial	7.20	8.00	7.62	7.60	±0.23
	Mixed	7.30	8.60	8.04	8.10	±0.31
	Baghdad City	7.20	8.80	7.92	7.90	±0.34
EC	Residential	70.00	2430.00	1021.00	1190.00	±574.03
	Commercial	80.00	2990.00	1248.00	1180.00	±713.13
	Industrial	450.00	4550.00	1894.00	1630.00	±1147.56
	Mixed	130.00	2570.00	1157.00	1125.00	±678.70
	Baghdad City	70.00	4550.00	1330.00	1205.00	±862.00

Total heavy metal content

The measured heavy metals show higher concentrations than the computed world mean of unpolluted soils [22,23]. The descriptive statistics of the heavy metal total data sets relative to surface soil samples are given in Table (2) followed by a brief discussion about each one of them.

For Cd (only samples within the mixed area), Zn, total concentrations were not normally distributed, showing a skewed distribution. Therefore, for these samples, medians instead of means were used in the calculations of the total concentrations since they would describe such distributions more precisely (see Table (2), values in bold are the used ones to present the discussions and results).

Table (2). Statistical summary of heavy metals concentrations (mg/kg)

	Land use	Range of	Mean	Median	Std.	Skewness
Cd	Residential	0.00-6.20	**2.99**	3.40	±2.20	-0.16
	Commercial	0.00-6.35	**3.42**	3.30	±2.16	-0.43
	Industrial	1.25-5.98	**4.00**	3.78	±1.55	-0.22
	Mixed	1.20-5.80	4.18	**4.48**	±1.24	-0.94
Zn	Residential	18.95-115.20	60.26	**58.00**	±24.16	0.63
	Commercial	5.20-155.95	58.80	**51.25**	±37.95	1.203
	Industrial	37.40-219.95	108.05	**106.90**	±39.81	0.83
	Mixed	30.80-169.65	79.64	**77.53**	±36.64	0.74

*In bold: values used to present the discussions and results.

Cadmium distribution

The Cd concentration in the study area varies from 0.00 mg/kg to 6.35 mg/kg, with mean values of 2.99 mg/kg, 3.42 mg/kg, 4.00 mg/kg, in the residential, commercial and industrial areas respectively, and a median value of 4.48 mg/kg in the mixed area. The observed values exceed the computed worldwide mean of unpolluted soil 0.53 mg/kg [22].

Figures (2) and (3) depict the spatial distribution and a comparison of Cd levels in soil among various land use types respectively. It can be noted that the effect of land use type for Cd was very clear following the decreasing order of mixed> industrial> commercial> residential. Cd is generally present in the environment at low levels. However, human activity can increase Cd levels accordingly to the urban industrial activity [24]. This study indicates that 75%, 80%, 100% and 100% of all soil samples in the residential, commercial, industrial and mixed areas respectively containing cadmium concentrations greater than the standard limit in soil.

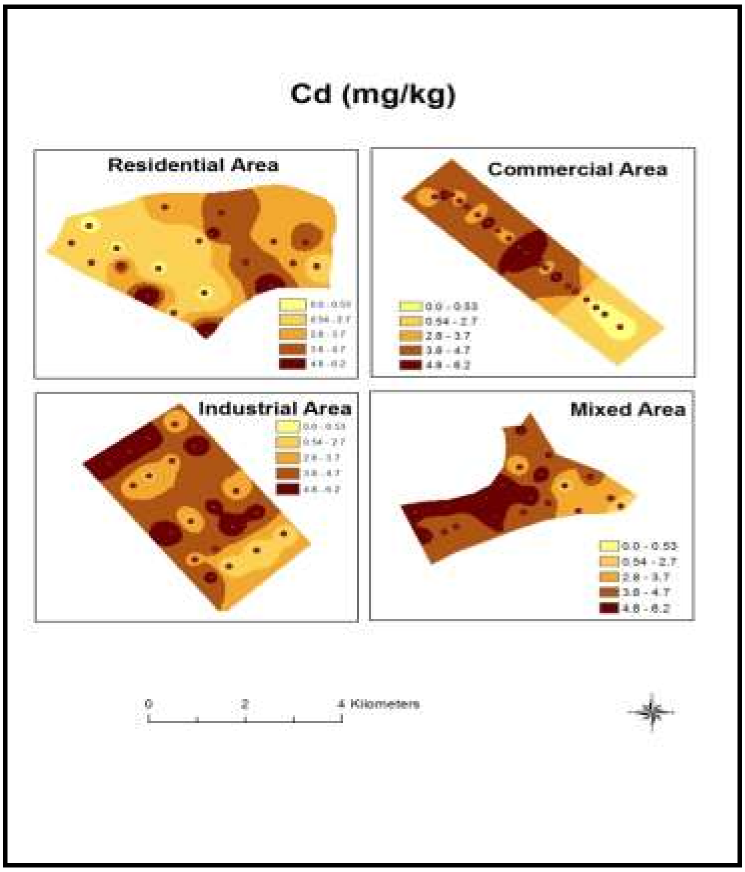

Figure (2). Spatial distribution of cadmium in the study area

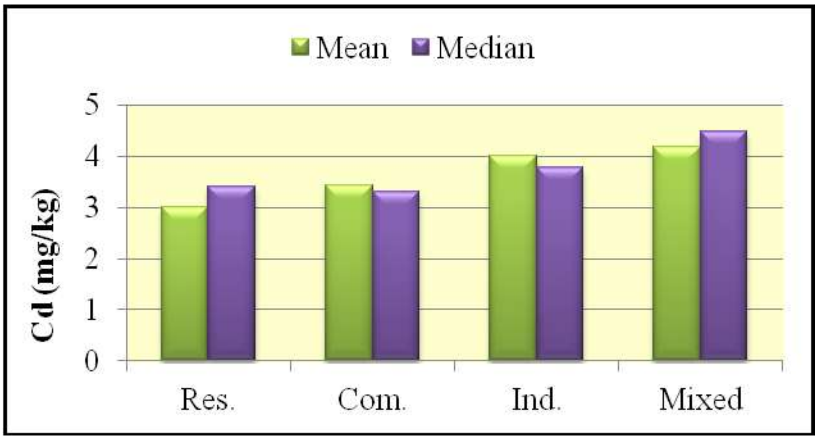

Figure (3). Comparison of Cd levels (mg/kg) among various land use types

Maximum concentrations of Cd in the mixed and commercial areas may be related to the wear and tear of tires and the higher traffic density on the busy roads of these two areas as compared to the residential area. Also, cadmium can travel for long distances from the source of emission by atmospheric transport [25]. It can be found in nickel-cadmium batteries, diesel fuel, Tires, cigarettes, PVC plastics, and paint pigments, insecticides, fungicides. Moreover, furniture, cars, trucks, agricultural tools, industrial tools and various kinds of fasteners including bolts, nuts, wrenches and nails have cadmium covering [24].

For the industrial area, maximum concentrations were near to the refining units, flare, tanks of oil products and cross roads. The results of this district were much higher than those of [21] who reported a mean value of (0.035 mg/kg) for cadmium near to the flare. [26], stated that refining of ores is the largest source of industrial atmospheric cadmium emissions, followed by waste incineration.

Zinc distribution

The Zn concentration in the study area varies from 5.20 mg/kg to 219.95 mg/kg, with median values of 58.00 mg/kg, 51.25 mg/kg, 106.90 mg/kg and 77.53 mg/kg in the residential, commercial, industrial and mixed areas respectively. The values of Zn concentration were reported within the regular global range for total Zn concentrations in soil (10-300 mg/kg) [23], and so for the common mean for unpolluted soil, which is reported as 100 mg/kg [22], except that for the industrial area which was little higher 106.90 mg/kg.

Figures (4) and (5) show the spatial distribution and a comparison of Zn levels the in soil among various land use types respectively. Results showed that 10%, 15%, 70% and 30% of all soil samples in the residential, commercial, industrial and mixed areas respectively contain zinc concentration greater than the standard limit in soil.

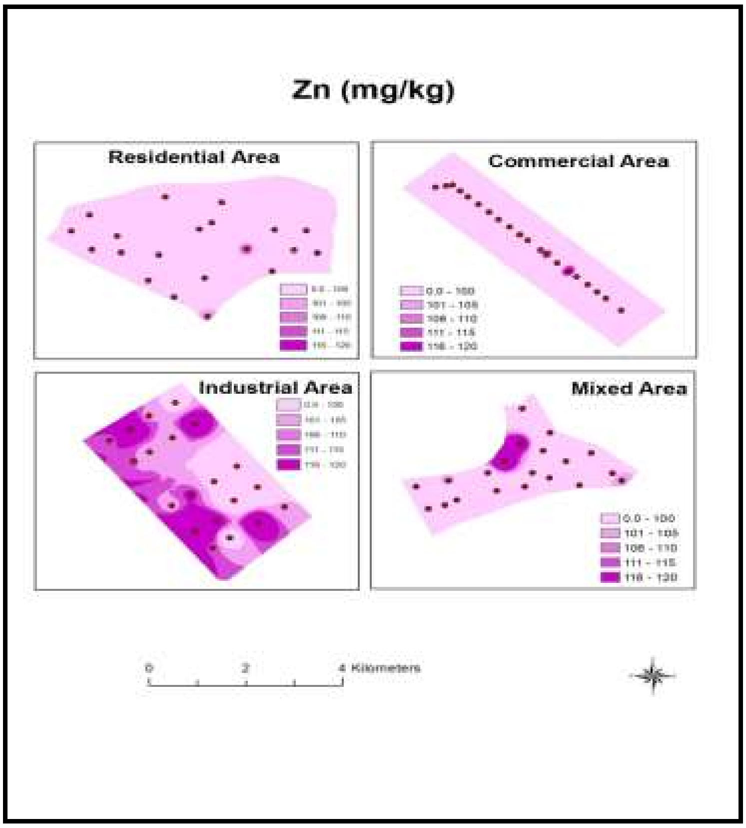

Figure (4). Spatial distribution of zinc in the study area

For the industrial area maximum concentrations of zinc were near to the refining units, industrial water treatment plant, heavy oil boards and old tanks of oil products, while for the mixed area they were near to the fuel station and artesian automobile workshops. These results are committing with [27] who reported that the accumulation of heavy metals such as Zn in the topsoil of industrial area is mainly affected by anthropogenic activities. The anthropogenic sources of Zn are associated with industries, waste combustion and may be resulting from mechanical abrasion of vehicles, as they are used in the manufacture of brass alloy itself and come from brake linings, oil leak sumps and cylinder head gaskets [28].

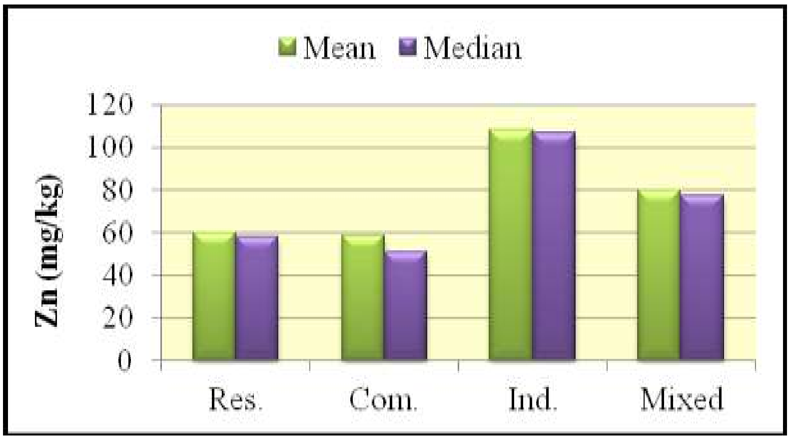

Figure (5). Comparison of Zn levels (mg/kg) among various land use types

Assessment of urban soil pollution

To assess the extent and severity of heavy metal contamination within each land use type of this study, the geoaccumulation index (I*geo*) was applied. It is used to assess metal contamination in urban soil by comparing current and preindustrial concentrations. It is also engaged in pollution estimation of the heavy metals in urban road dust. Geoaccumulation index is computed using the following equation [29,30]:

$$I_{geo} = log_2 (C_n/1.5B_n) \quad (1)$$

Where C_n is the measured concentration of the examined metal in the soil and B_n is the geochemical background concentration of the same metal. The constant 1.5 is introduced to minimize the effect of possible variations in the background values, which may be attributed to anthropogenic influences [31].

The following categorization is given for geoaccumulation index: <0 = practically unpolluted, 0-1 = unpolluted to moderately polluted, 1-2 = moderately polluted, 2-3 = moderately to strongly polluted, 3-4 = strongly polluted, 4-5 = strongly to extremely polluted, and >5 = extremely polluted [32]. Reference values (Earth crust averages) of Cd and Zn are (0.2 and 70) mg/kg [33].

Table (3) gives I_{geo} values of Cd and Zn in the urban soils, results are also described in Figure (6). The pollution levels of these metals in the environment expressed in terms of geoaccumulation index indicated $3>I_{geo}>4$ for Cd which may interpret that the soil samples in the study area were strongly polluted with this element. The obtained I_{geo} for Zn in the industrial area revealed that most of the soil samples examined fell into the class of unpolluted to moderately polluted, while negative I_{geo} values of Zn in the residential, commercial and mixed areas, is an indicator of the lack of contamination in these areas with these metals.

Table (3). I_{geo} values with classes of soil quality

	Residential	Commercial	Industrial	Mixed
Cd	3.32	3.51	3.74	3.90
Zn	-0.86	-1.03	0.03	-0.44

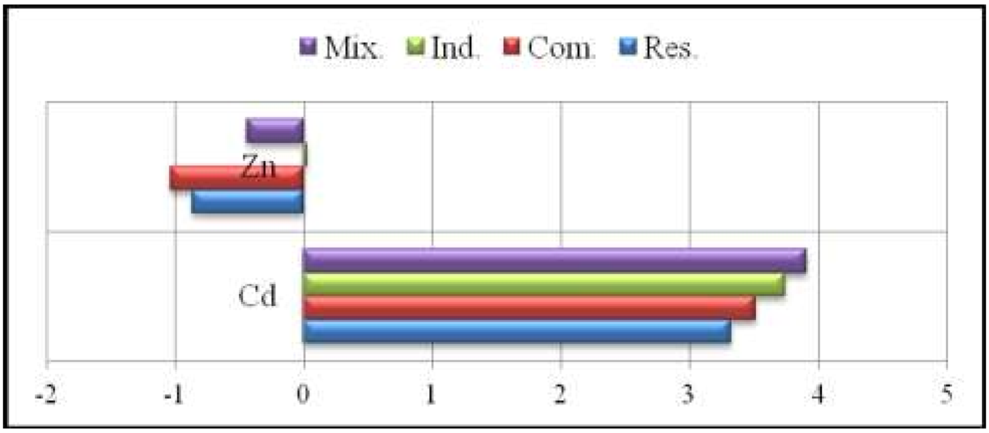

Figure (6). Geoaccumulation indices in different land use types

Metals bioavailability

Heavy metal total concentrations in the soil can be a helpful parameter representing pollution intensity. So, it is documented that the particular behavior of heavy metals is determined by their available forms rather than by total concentration. Table (4) shows the statistics of bioavailable concentrations of the examined heavy metals in this study. For extracting the heavy metals in this study, 20 ml of DTPA extracting reagent was used which covers water soluble, exchangeable and some oxide fractions among many heavy metals in the soil. The relative bioavailability (RA) of metals (%) was calculated as follows:

$$(RA)(\%) = \frac{Conc.of\ metal\ in\ DTPA\ extractant}{Conc.of\ metal\ in\ HNO3+HCl\ extractant} * 100 \qquad (2)$$

The results plotted in Figure (7) reveal that Zn is relatively more available than Cd. The relative bioavailability for Cd and Zn has been observed as 2.46 -5 % and 2.80-9.78 % respectively. Approximate results have been reported by [34], who observed Cd: 10.06 % and Zn: 0.66%, with a mean value of pH: 7.8, which is also very close to the pH of this study (7.92). The difference in the relative bioavailability and potential mobility between these two metals may be explained considering the fact that Zn is an essential micronutrient in many plants, and as a sequence, is likely to be more soluble and mobile in soil-plant ecosystem than others [35].

Table (4). Statistical summary of metals bioavailability in different land use types

	Land Use	Mean	Median	Std.	Skewness	*RA%*
Cd	Residential	0.09	0.08	±0.07	0.19	3.01
	Commercial	0.12	0.12	±0.07	-0.41	3.51
	Industrial	0.20	0.19	±0.09	-0.12	5
	Mixed	0.11	0.11	±0.03	0.05	2.46
Zn	Residential	2.56	2.17	±1.83	0.65	3.74
	Commercial	2.10	1.75	±1.31	0.44	4.10
	Industrial	10.79	10.46	±6.84	0.54	9.78
	Mixed	4.42	4.28	±2.61	0.19	2.80

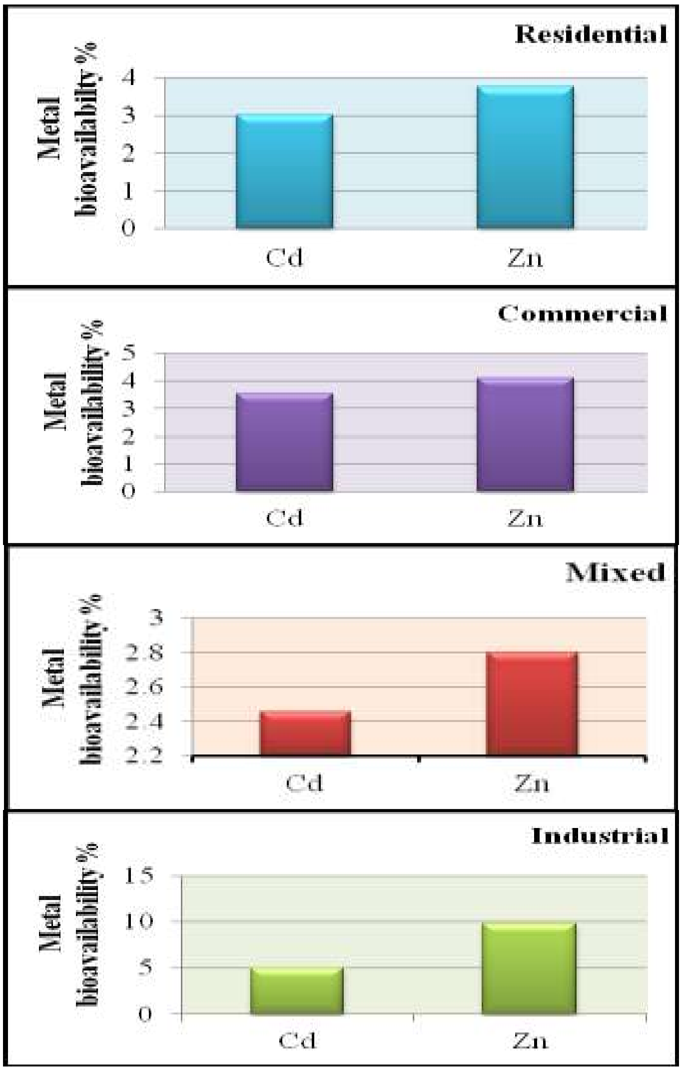

Figure (7). Comparison of percentage urban soil metal bioavailability among various land use types

The soil is a dynamic system, subject to short-term fluctuations for example, variations in moisture status, pH and redox conditions and also undergoes slow alteration in response to changes in management and environmental factors. These changes in soil properties may affect the form and bioavailability of heavy metals. In general, heavy metal cations are most mobile under acid conditions.

It can be concluded that, although total concentrations of the examined heavy metals were generally high, but at the same time the bioavailable ones were relatively low. This can be an indicator that with the recent environmental factors (alkaline to sub-alkaline soil pH), the toxicity of heavy metals to humans is in its minimum level.

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