Assessment of Heavy Metal Concentrations in Roadside Soils from Mafraq, Jordan

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Abstract: *In the present study, the concentrations of nine heavy metals (Cr, Cd, Cu, Mn, Pb, Co, Fe, Ni, and Zn) in roadside soils were measured by flame atomic absorption spectroscopy (FAAS). Roadside soils were collected during the summer semester from Mafraq, Jordan (N = 97). Four sampling sites were selected for this study, including commercial area, industrial area, residential area, and Irbid-Mafraq highway. The average metal concentrations (±SD) in the collected soils were 16840 (± 9479), 40 (± 40), 99 (± 44), 478 (± 155), 60 (± 62), 2 (± 1), 73 (± 23), 14 (± 5), and 45 (± 13) mg/kg for Fe, Cu, Zn, Mn, Pb, Cd, Cr, Co, and Ni, respectively. The results of this study revealed that the highest Mn, Cu, Co, Ni, Zn, Fe, and Pb concentrations were found in the industrial area, and the highest Cd concentration was found at the Irbid-Mafraq highway. The values of the enrichment factors for Zn, Cr, Cd, and Ni in the collected roadside soils were found to be higher than 10, confirming the presence of anthropogenic pollution in the studied areas. The results of this study were also compared with other studies performed in different countries in the world.*

Keywords: *roadside soil; heavy metals; spatial distribution; Mafraq, Jordan*

■ INTRODUCTION

The pollution of the environment by different classes of toxic pollutants became dramatic due to rapid development in industry, agriculture, and urbanization that has taken place in different countries of the world in recent years [1-3]. Soil is one of the environmental components that is needed for plant growth, clean water, healthy food, and other human and development activities [4-5]. Soil is an important natural resource that is used for transporting useful minerals and organic materials found on the earth's surface to plants by absorption through their roots, stems, and leaves [3,6]. During the last years, soil become heavily polluted by a large number of toxic pollutants like pesticides (herbicides, insecticides, and fungicides), polyaromatic hydrocarbons, and heavy metals due to rapid industrialization and urbanization [3-5,7-10]. Heavy metals are chemical substances that are classified as hazardous pollutants due to their toxicity to humans, animals, and plants at a certain concentration. Commonly, they are classified into two types: The first type is an important group of heavy metals (e.g. Fe, Cu, Mo, V, Zn, Mn, or Mg) that are important for the health of human beings, animals, and plants [1,3,11]. However, these metals become toxic when their level exceeds the permissible limits. The second type is metals like Co, As, Cr, Cd, Hg, Pb, Sn, or Ni [3,12-13]. These metals are hazardous and toxic even at low concentrations and have no beneficial biological requirement in humans, animals, and plants [12,14].

During the last years, soils have been heavily polluted with toxic heavy metals due to natural sources such as abrasion of basaltic rocks, forest fires, sea-salt spray, wind-borne soil particles, and volcanic eruptions. The pollution was also caused by different anthropogenic sources like mining and smelting, automobile emissions, fertilizers, electroplating industry, oil spills, disposal of municipal waste, disposal of industrial wastes, thermal power plants, tire and brake wear, fossil fuel combustion, waste burning, road paint degradation, atmospheric deposition, sewage irrigation, and corrosion of batteries [1–3,5,7,9-10,15]. Soil pollution due to heavy metals has become a worldwide health and environmental issue that has attracted special attention in the last years, which is documented in a large number of review articles [2-4,16-21]. This is because soil acts as a medium for the deposition of heavy metals and other toxic pollutants due to irrigation with contaminated water, and then it transports them to plants by absorption through the roots and foliar surfaces [3-4,6]. This means that heavy metals in soil will accumulate in higher concentrations in plants. Once these toxic metals accumulate in food crops, they can pose serious adverse effects to human health via dermal contact with contaminated soil, ingesting contaminated grains and vegetables, or drinking contaminated groundwater [16]. These metals pose a significant threat to human beings, animals, and plants by polluting natural resources such as soil, atmosphere, and water [3-4]. Unlike many other organic pollutants, heavy metals are thermostable, have a long persistence time, non-biodegradable, and thus readily accumulate in different environment components to toxic levels [2,7,11,22]. In urbanized areas, toxic and essential heavy metals accumulate in the soil for different reasons, such as atmospheric deposition, leaching of dust, traffic activities, and slag [1,5,23]. It is reported that several factors strongly affect the bioaccumulation of heavy metals in soil like physical and chemical properties of soil (e.g., pH and electrical conductivity), type of soil, type and concentration of metal, mobility and bioavailability of metal, metal speciation, type and content of organic matter in soil [7,9-10].

The significant accumulation of heavy metals in the human body results in several health problems. For example, the toxic symptoms caused by Pb include damage to neurons, insomnia, reduced fertility, damage to kidneys and renal system, respiratory disorders, anorexia, hyperactivity, cardiovascular impairments, high blood pressure, and mental disorders [2,20-21,24]. The toxic symptoms caused by Cd include hypertension, bone disease, coughing, headache, lung and prostate cancer, kidney diseases, bone diseases, reproductive deficiencies, and hypochromic anemia [2,20,24-25]. Chronic exposure to Ni causes chest pain, dizziness, dry cough, lung and nasal cancer, low blood pressure, nausea, skin sensitivity, asthma, heart attack, cardiovascular diseases, and kidney diseases [2,24,26]. High concentrations of As in the human body lead to human diseases such as brain damage, respiratory disorders, and skin cancer [2,20].

In Jordan, the contamination of the different environmental components (air, water, and soil) with heavy metals has increased dramatically because of the recent development in many fields, such as industry, agriculture, and population. Therefore, several environmental studies were carried out to study heavy metal pollution in many samples like agricultural soils, roadside soils, house dust, street dust, plants, water, and air [5,8,11-12,14,27-32]. For example, Eid Alsbou and Al-Khashman [31] determined the levels of selected metals (Cd, Cu, Fe, Pb and Zn) in street dust and roadside soil samples collected from Petra, Jordan. The authors observed that the highest concentration of the investigated metals found in the eastern parts of the roads was due to the prevailing western winds in the study area. The authors concluded that traffic emissions are the main source of these metals in the studied area [31]. In a similar study, Al-Momani [32] measured the concentrations of six heavy metals (Pb, Cd, Zn, Cu, Cr, and Ni) in soils collected from four different sampling locations (busy and main roads, industrial, tunnels and residential roads) in the Greater Amman area, Jordan by inductively coupled plasma atomic emission spectroscopy. The results showed that the concentrations of the investigated metals in the industrial zone were higher than in other regions, whereas maximum Pb concentrations were observed in tunnels and congested roads. The author concluded that burning fossil fuels, vehicle emissions, and wear brake lining materials are the main sources of emissions of these metals [32].

During the last few years, soil contamination with heavy metals has been studied extensively in Jordan, but there is not enough information available on the heavy metal pollution of roadside soils in Mafraq, Jordan. Therefore, this is the first study to investigate the content of the studied heavy metals in roadside soils collected from Mafraq, Jordan. This city was selected for this study because it has been suffering from serious environmental problems due to rapid and uncontrolled growth in many fields like industry, agriculture, and population due to the high number of refugee camps in the city. This, in turn, increases the load on different environmental elements (soil, water, and atmosphere) in the city. The main objectives of this study were: (1) to estimate the concentrations of nine heavy metals (Pb, Fe, Co, Cd, Ni, Cr, Cu, Mn, and Zn) in roadside soils collected from four sampling sites including commercial area, industrial area, residential area, and Irbid-Mafraq highway, (2) to define the possible sources of the studied metals in the studied areas, and (3) to compare the results of this study with similar studies performed in Jordan and other countries in the world.

■ EXPERIMENTAL SECTION

Materials

All chemicals, solvents, and reagents used in the present study were of analytical grade. Standard solutions of 1000 mg/L of each studied metal (Mn, Cd, Cr, Fe, Cu, Pb, Co, Ni and Zn) were from Merck KGaA (Darmstadt, Germany), 70% (v/v) HNO_3 from Carlo Erba Reagents (France), 35% (w/w) H_2O_2 and 40% (v/v) HF were from Scharlau Chemie (Barcelona, Spain). Deionized water was used to prepare working standards and sample solutions.

Instrumentation

In the present study, a flame atomic absorption spectrophotometer (FAAS) (Type: Varian Spectr AA-55B, Australia) equipped with a deuterium background correction was used for measuring the concentrations of nine heavy metals in the tested roadside soils. Analysis using FAAS was carried out at the most sensitive and analytical spectral lines of the metals (Zn 213.9 nm, Cu 324.8 nm, Fe 386.0 nm, Cr 357.9 nm, Cd 228.8 nm, Co 240.7 nm, Mn 279.5 nm, Pb 217.0 nm, and Ni 232.0 nm).

Procedure

Study area

Mafraq city (26551 km^2 total area, 142401 population) is about 60 km north of Amman (the capital of Jordan), at the crossroads of Syria to the north and Iraq to the east [11]. Mafraq has a dry and hot climate and most of the rainfall is in the winter semester. In recent years, Mafraq has been highly affected by environmental pollution due to rapid and uncontrolled growth in many fields, such as commercial, industrial, agricultural, and residential areas. In addition, the extensive migration of refugees from neighboring courtiers to Mafraq increases the load on soil, atmosphere, and groundwater in the city. During the last few years, different industrial activities have been developed in Mafraq such as agricultural, detergents, pesticides, concrete, food, herbicides, and fertilizer factories. In the present study, roadside soils (N = 97) were collected from four sampling locations, as shown in Fig. 1. The commercial area, which is a crowded urban area, represents the center of Mafraq city (Downtown area). This sampling site has the two main Mafraq transit bus stations (Western and Eastern bus stations), gas stations, high traffic volume, and a large number of shopping centers. In addition, this sampling site is considered a crossroad for all cars coming from Jarash city and all the villages in the west of Mafraq. The second sampling site is the main industrial area in Mafraq city, which comprises many mechanical and electrical garage repairs and car paint workshops. The residential area represents the third sampling site in Mafraq city. This area is composed of Hussein and officer's neighborhood, with heavy traffic and high population density. The fourth sampling site is the Irbid-Mafraq highway (the first 1–2 km of the highway). This sampling site is a crossroad of cars coming from north Badia, East Badia, and Irbid cities, which means that thousands of cars go through this road daily.

Samples collection and pretreatment

In this study, roadside surface soils (the upper 0–10 cm) were collected during the summer semester (June–October 2019) from four sampling sites, including a commercial area (49 samples), industrial area (10 samples), residential area (18 samples), and Irbid-Mafraq highway (20 samples). Roadside soils were collected in dried and labeled polyethylene bags using a clean and new dustpan and brush. In order to remove stones, glasses, wires, and large parts of the plant from the collected soils, they passed through a 2 mm mesh

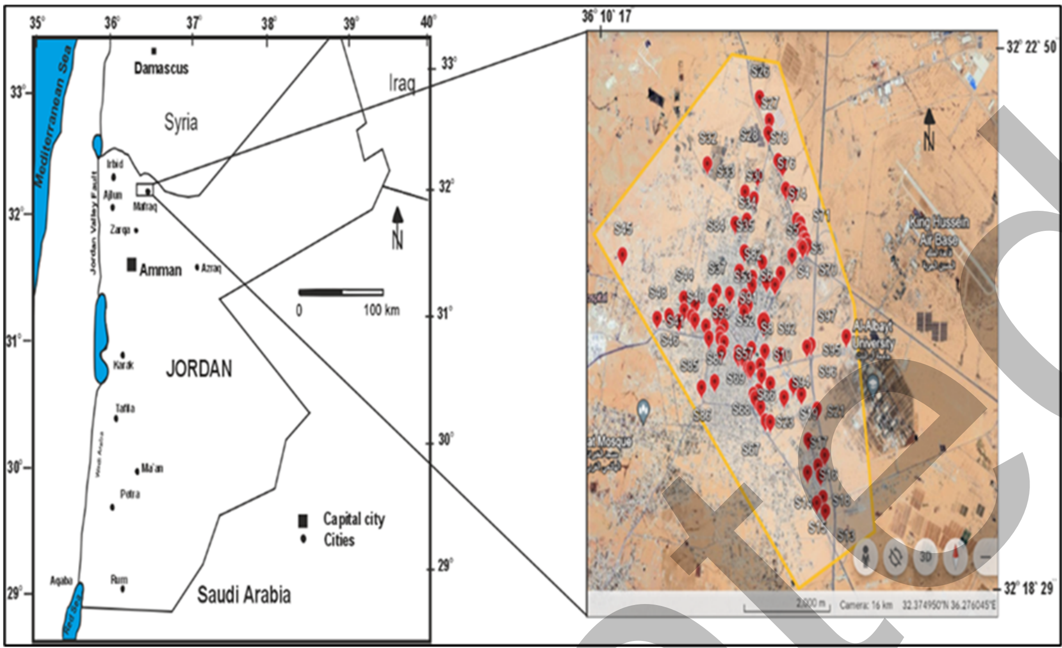


Fig 1. The sampling sites in Mafraq

plastic sieve. The sieved soils were then dried using electrical oven (ca. 85 °C). Finally, the soil samples were homogenized using a mortar and pestle and stored in clean plastic bottles.

Sample preparation

In the present study, the collected roadside soils were prepared as described by Al-Serhan et al. [11]. Briefly, a weight of 0.50 g of dry soil sample was placed into a microwave Teflon tube. To each Teflon tube, 8 mL HNO_3, 2 mL HF, and 2 mL H_2O_2 were then added. The solution was left in the Teflon tube for about 10 min. The microwave digestion program was worked at a power of 850 W and the Teflon tube was heated to about 200 °C for 10 min. After cooling, the solution was filtered into a 50 mL volumetric flask and the volume was completed to the mark with deionized water.

Calibration

A series of working standard solutions for Fe, Cu, Zn, Mn, Pb, Cd, Cr, Co and Ni were freshly prepared by diluting an appropriate portion of the stock solution containing 1000 mg/L using 0.5% (v/v) HNO_3. From a linear calibration curve for each metal, the values of correlation coefficient (R^2), intercept with standard error ($a \pm S_a$), and slope with standard error ($m \pm S_b$) were calculated using the linear least squares method. These values were then used to calculate the lowest limit of detection LOD (mg/kg of solid sample) and lowest limit of quantitation LOQ (mg/kg of solid sample) according to the following equations (Eq. (1) and (2)) [5,11]:

$$LOD = \frac{3.3 \times S_a}{m} \quad (1)$$

$$LOQ = \frac{10 \times S_a}{m} \quad (2)$$

where S_a is the standard error of the y-intercept of the regression line, and m is the slope of the calibration curve. The values of LOD and LOQ for all the studied metals are listed in Table 1.

Quality control and assurance

It is reported that the analysis of blank is an essential step used to correct any contamination problem during a validation of the analytical method. In order to obtain the correct concentrations of the studied metals in the collected roadside soils, the results obtained in this study were blank-corrected [11,28]. With the aim to confirm the validity and accuracy of the FAAS analysis method used in this study, three standard reference materials (SRM) for these metals, including (i) SRM-1646a (Estuarine sediment), (ii) SRM-1633b (trace elements in coal fly ash), and (iii) SRM-2702 (inorganics

Table 1. The LOD, LOQ, R^2, and percent recovery of each studied metal

Metal	LOD (mg/kg)	LOQ (mg/kg)	R^2	Recovery (%)		
				SRM-1646a	SRM-1633b	SRM-2702
Zn	1.396	4.655	0.988	97	95	91
Cu	0.083	0.276	0.998	94	96	92
Fe	1.262	4.206	0.984	96	93	90
Cr	0.246	0.821	0.996	96	94	93
Cd	0.046	0.155	0.994	98	96	92
Co	0.062	0.208	0.996	94	96	92
Mn	0.047	0.157	0.992	96	93	95
Pb	0.759	2.530	0.981	95	92	90
Ni	0.199	0.665	0.996	93	89	92

Table 2. Statistical summary of Pb, Zn, Fe, Cd, Cr, Cu, Co, Ni, and Mn concentrations (mg/kg) in roadside soils in all sampling sites

Metal	Average	SD	Median	C.V (%)	Range
Cd	2.4	1.3	2.5	0.50	2.0–3.0
Co	14.0	5.2	13.0	0.40	13.2–16.7
Cr	72.9	22.8	75.0	0.30	64.9–83.1
Cu	40.0	40.0	35.0	1.00	32.8–87.5
Fe	16840.0	9479.0	20257.0	0.56	13880.0–24653.0
Mn	478.0	155.0	491.0	0.32	431.8–576.5
Ni	44.6	13.0	44.0	0.30	40.3–50.2
Pb	59.8	62.4	48.0	1.00	41.1–90.7
Zn	99.4	44.2	89.0	0.44	83.7–105.4

in marine sediment) were prepared and analyzed along with roadside soils. Results presented in Table 1 demonstrate that there is a good agreement (> 90%) between the measured values of the studied metals and their certified values, which confirms the validity and accuracy of the FAAS analysis method used in this study. In addition, duplicate samples were used during the analysis to confirm the precision of the results.

■ RESULTS AND DISCUSSION

LOD and LOQ

Results listed in Table 1 confirm that the calibration curves for all studied metals were linear, with $R^2 \geq 0.981$. The lowest detection limits of the studied metals ranged between 0.046 mg/kg for Cd and 1.396 mg/kg for Zn, while the lowest quantitation limits ranged between 0.155 and 4.655 mg/kg for the same heavy metals (Table 1). The relatively low values of the detection limits demonstrate the sensitivity of the FAAS analysis method used in this study.

Metal Concentrations in Roadside Soils

In the present study, roadside soils were collected from four sampling sites including commercial area, industrial area, residential area, and the Irbid-Mafraq highway, Mafraq, Jordan (N = 97). These soils were evaluated using FAAS to determine the concentrations of Fe, Cu, Zn, Mn, Pb, Cd, Cr, Co, and Ni. The average concentrations (mean ± S.D) of each metal are presented in Table 2 (for detailed information about the concentration of the studied metals in all sampling sites, refer to Table S1). All metal concentrations were determined on a dry-weight basis. Metal concentrations (± S.D) in the analyzed soils were found to be 16840 (± 9479), 40 (± 40), 99 (± 44), 478 (± 155), 60 (± 62), 2.4 (± 1.3), 73 (± 23), 14 (± 5), and 45 (± 13) mg/kg for Fe, Cu, Zn, Mn, Pb, Cd, Cr, Co, and Ni, respectively (Table 2). Moreover, the average metal concentrations were found to be decreased in the following order: Fe > Mn > Zn > Cr > Pb > Ni > Cu > Co > Cd.

Tables S2-S5 show the average concentration, standard deviation, median, coefficient of variation, minimum concentration, maximum concentration, and number of analyzed samples for the studied metals in all sampling sites. The relatively high values of standard deviation for the studied metals can be attributed to the variations in the nature of the sampling sites.

Spatial Distribution of Heavy Metals

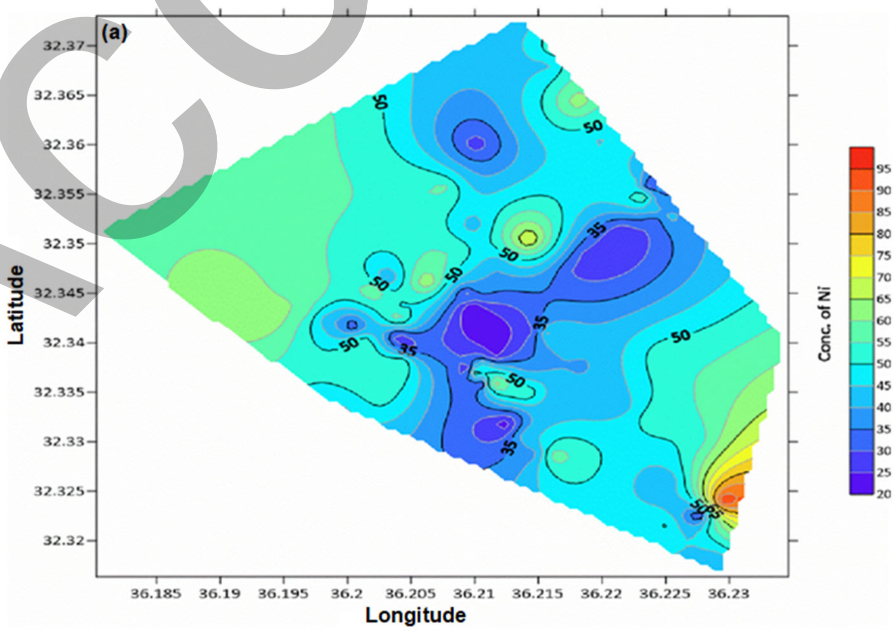
For a detailed discussion of the distribution of the examined metals in the studied areas (commercial area, industrial area, residential, and Irbid-Mafraq highway), all statistical data for Pb, Ni, Zn, Cr, Fe, Cd, Cu, Co, and Mn were evaluated and presented in Tables 2 and S2-S5.

Nickel

The average Ni concentrations in the analyzed soils ranged from 40.3 to 50.2 mg/kg with a mean of 44.6 mg/kg, as shown in Table 2 (for details, refer to Tables S2-S5). The maximum Ni concentrations permitted in soil are 35 mg/kg according to the Dutch and Nigeria standards [33-34] and 50 mg/kg according to the Food and Agricultural Organization/World Health Organization (FAO/WHO) [35]. This means that the Ni concentrations obtained in this study were found to be within the permitted limits set by FAO/WHO, but higher than those limits set by Dutch and Nigeria standards. Data show that the highest Ni concentration was found in the industrial area with a mean of 50.2 mg/kg, where Co, Cu, Fe, Mn, Zn, and Pb were also at maximum (Table S3, Fig. 2). The spatial distribution of Ni in the analyzed soils in the studied areas is shown in Fig. 2(a). The red color indicates the highest Ni concentrations, as shown by the legend on the right side of Fig. 2(a). The average Ni concentrations in the analyzed soils are in the following order: industrial area > Irbid-Mafraq highway > commercial area > residential area (Fig. 2(b)). The higher Ni concentrations obtained in this study might be due to engine oil, engine wear, brake dust, tire abrasion, lubricant corrosion of cars, Ni-Cd batteries, heavy oil combustion, and the incineration of waste and sewage [5,16,23,36]. The average Ni concentration in the analyzed soils was higher than those found in Pakistan [37] and China [38], but lower than those found in Turkey [7], Jordan, Al-Zarqa city [8], and Egypt [9], as shown in Table 3.

Zinc

Results achieved in this study show that the average Zn concentrations in the analyzed soils ranged from 83.7 to 105.4 mg/kg with a mean of 99.4 mg/kg (Table 2) (for details, refer to Tables S2-S5). The maximum Zn concentrations permitted in soil established by the European Union (EU) and FAO/WHO is 300 mg/kg [35,39], while Dutch and Nigeria standards set this level



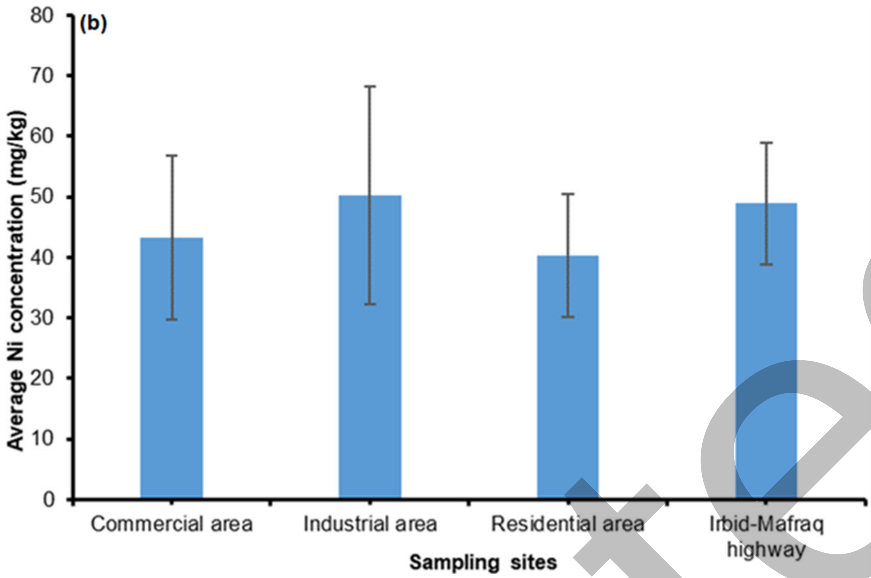


Fig 2. (a) Spatial distribution and (b) average concentration of Ni in the studied areas

Table 3. Average concentrations of Pb, Zn, Cd, Cr, Cu, Co, Ni, and Mn (mg/kg) in roadside soils collected from Mafraq city compared with other studies in different countries

Country	Cd	Co	Cr	Cu	Mn	Ni	Pb	Zn	Reference
Jordan (Petra city)	1.00	----	----	19.00	----	----	177.00	129.00	[31]
Jordan (Al-Zarqa city)	6.60	----	88.2	21.70	492.2	113.1	58.90	122.00	[8]
China	0.12	----	----	27.00	----	32.0	44.00	71.00	[38]
Pakistan	0.84	6.0	---	13.00	174.0	8.8	36.50	56.70	[37]
Turkey	3.60	15.0	127.0	52.67	817.0	62.4	10.75	66.25	[7]
India	----	----	34.4	19.00	32.5	----	34.40	5.70	[43]
Egypt	1.19	----	182.0	92.00	----	111.0	54.00	----	[9]
England	1.40	----	----	87.30	----	----	233.00	174.60	[44]
Jordan (Mafraq city)	2.40	14.0	72.9	40.00	478.0	44.6	59.80	99.40	This study

at 140 mg/kg [33-34]. These findings indicate that the average Zn concentrations obtained in this study were found to be within the permissible limits for this metal. The highest Zn concentration was found in the commercial and industrial areas with a mean of 105 mg/kg (Tables S2 and S3, Fig. 3). In addition, it is observed that the commercial and industrial areas were heavily polluted by Zn, as highlighted by the red color in the map (Fig.3(a)). The average Zn concentrations in the analyzed soils are in the following order: commercial area, industrial area > Irbid-Mafraq highway > residential area (Fig.3(b)). The higher Zn concentrations in the analyzed soils might be attributed to traffic emissions, abrasion of brake linings, paints, waste combustion, lubricating oils, fertilizers, and pesticides [23,40-42]. The average Zn concentration determined in this study was higher than those found in India [43], Pakistan [37], China [38], and Turkey [7], but lower than those found in England [44], Jordan, Petra city [31], and Jordan, Al-Zarqa city [8] as described in Table 3.

Lead

Lead is a non-essential and persistent metal. It is extremely toxic, even at low concentrations. The results presented in Table 2 show that the average Pb concentrations in the analyzed soils ranged from 41.1 to 90.7 mg/kg with a mean of 59.8 mg/kg (for details, refer to Tables S2-S5). The maximum Pb concentrations permitted in soil are 85 mg/kg according to the Dutch and Nigeria standards [33-34] and 100 mg/kg according to EU and FAO/WHO standards [35,39]. This implies

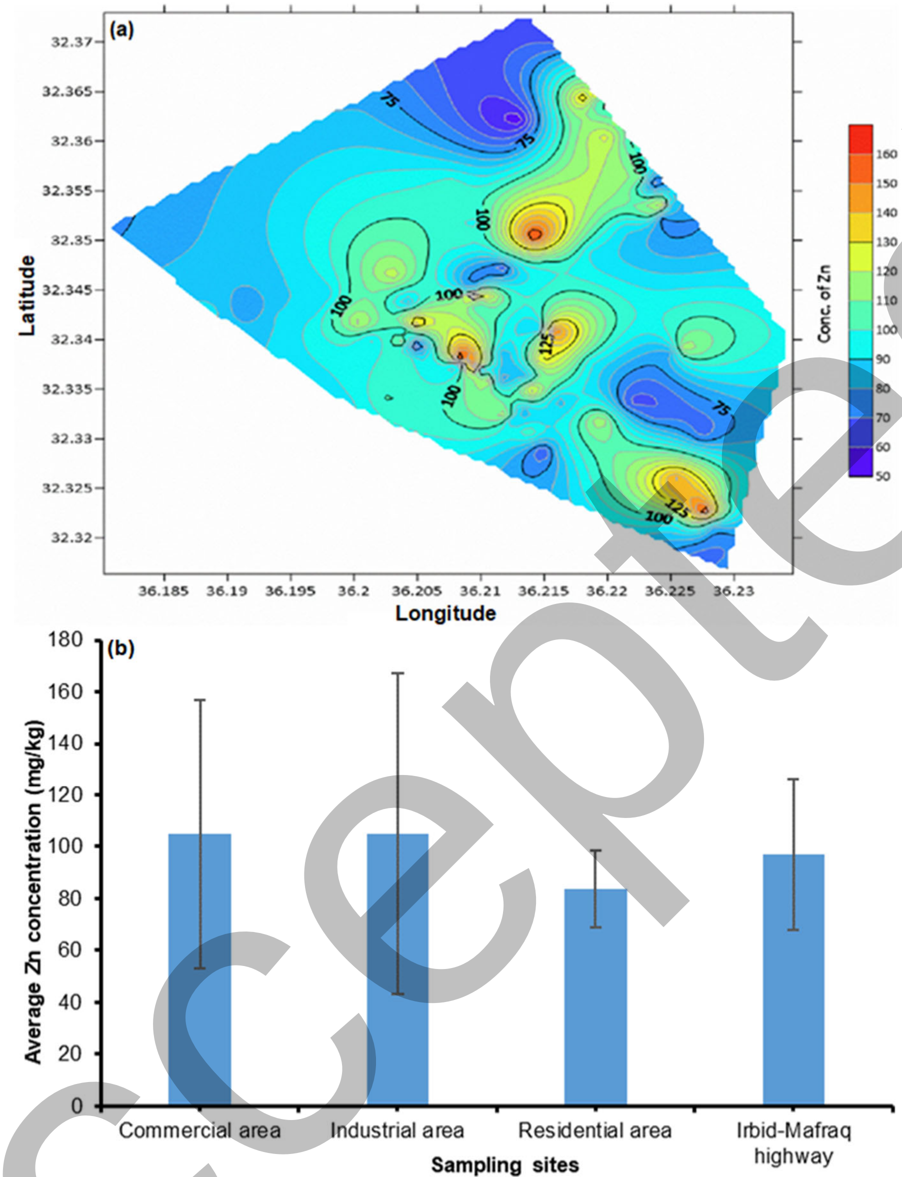


Fig 3. (a) Spatial distribution and (b) average concentration of Zn in the studied areas

that the average Pb concentrations in most of the analyzed soils were found to be within the permissible limits for this metal. The highest Pb concentration was found in the industrial area with a mean of 90.7 mg/kg (Table S3, Fig. 4). This is because Pb mostly originates from the combustion of leaded fuel, spills from batteries, engine wear, and brakes abrasions [5,42]. The major sources of Pb in urban soil reported in the literature include cable covers, pigments, plumbing, plating, gasoline additives, wear and corrosion of vehicle parts, and the paint on roads [2,5,15-16,45-46]. The highest Pb concentration is indicated by the red color, as shown by the legend on the right side of Fig.4(a). As can be seen from Fig. 4, the industrial area exhibited maximum concentration levels, followed by the Irbid-Mafraq highway. In addition, it is observed that all the studied areas exhibited similar spatial distribution patterns (Fig. 4(a)). The average Pb concentrations in the analyzed soils are in the following order: industrial area > Irbid-Mafraq highway > commercial area > residential area (Fig.4(b)). The average Pb concentration obtained in this study was higher than those found in India [43], Pakistan [37], China [38],

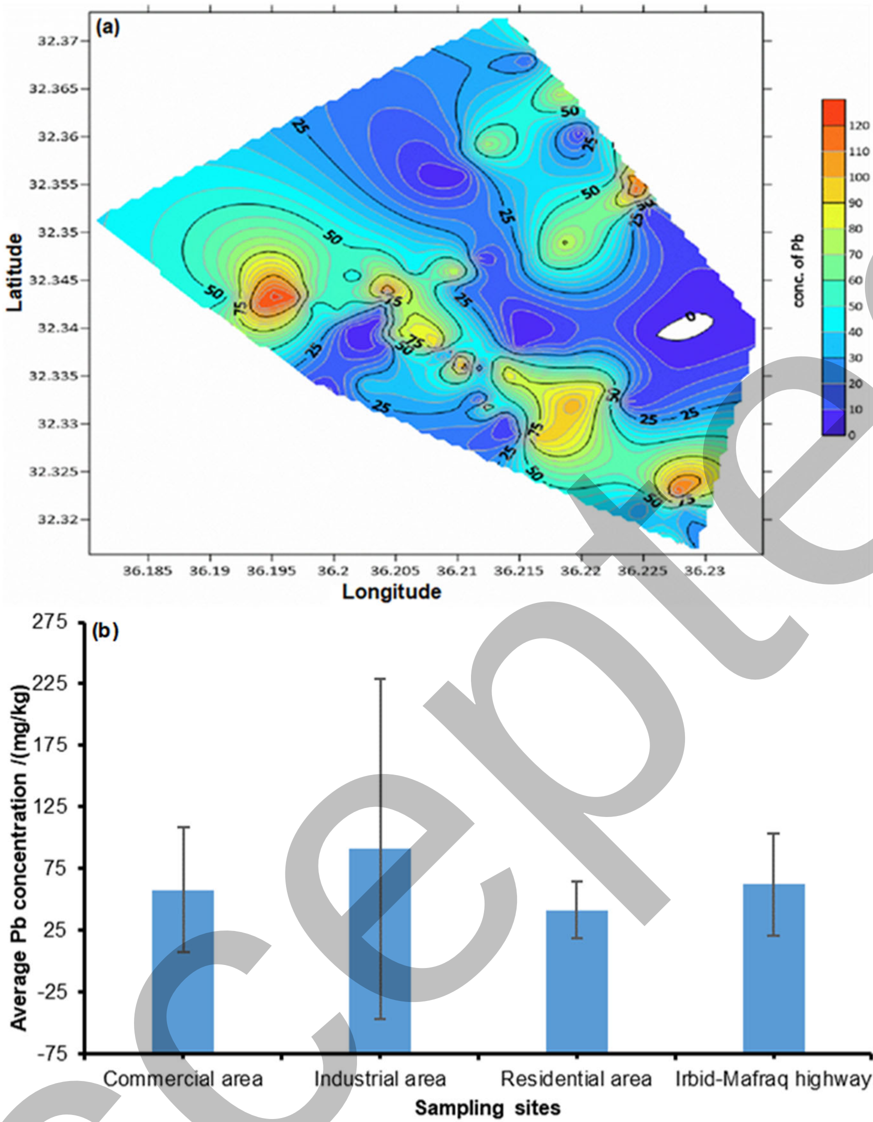


Fig 4. (a) Spatial distribution and (b) average concentration of Pb in the studied areas

Jordan, Al-Zarqa city [8], Turkey [7], and Egypt [9], but lower than those found in England [44], and Jordan, Petra city [31] as described in Table 3.

Cadmium

The average Cd concentrations in the analyzed soils ranged from 2.0 to 3.0 mg/kg with a mean of 2.4 mg/kg, as presented in Table 2 (for details, refer to Tables S2-S5). The maximum Cd concentration permitted in the soil is 3.0 mg/kg according to FAO/WHO [35], 1.0 mg/kg according to EU [39], and 0.8 mg/kg according to the Dutch and Nigeria standards [33-34]. This means that the average Cd concentrations in the analyzed soils were found to be within the permissible limit set by FAO/WHO, but higher than those limits set by Dutch, EU, and Nigeria standards. Results show that the highest Cd concentration was found at the Irbid-Mafraq highway with a mean of 3.0 mg/kg, which was also polluted with Cr, Ni, Co, and Mn (Table S5, Fig. 5). The high Cd levels obtained in this study might be due to old car tires, engine oil leakage, traffic emissions, and abrasion of brake linings [5,11,23,47]. The highest Cd concentration is observed on the Irbid-Mafraq highway, followed by the residential area, as highlighted by the red color in the map (Fig. 5(a)). The average Cd concentrations in the analyzed soils are in the following order: Irbid-Mafraq highway > residential area > industrial area > commercial area (Fig. 5(b)). It is reported that the major sources of Cd in urban soil are incineration of municipal wastes containing plastics, Ni-Cd batteries, using Cd as pigments and stabilizers for polyvinyl chloride (PVC), spills from batteries, combustion of leaded fuel, old car tires, engine oil, abrasion of brake linings, road surface wear, waste disposal from different industrial activities, and vehicle and road component degradation [10,23,25,42,48]. The average Cd concentration determined in this study was higher than those found in England [44], Pakistan [37], China [38], Jordan, Petra city [31], and Egypt [9], but lower than those found in Jordan, Al-Zarqa city [8], and Turkey [7] as shown in Table 3.

Iron

Iron is the fourth most abundant metal found in the soil. The data presented in Table 2 indicate that the average Fe concentrations in the analyzed soils ranged from 13,880 to 24,653 mg/kg with a mean of 16,840 mg/kg (for details, refer to Tables S2-S5). Results show that the highest Fe concentration was found in the

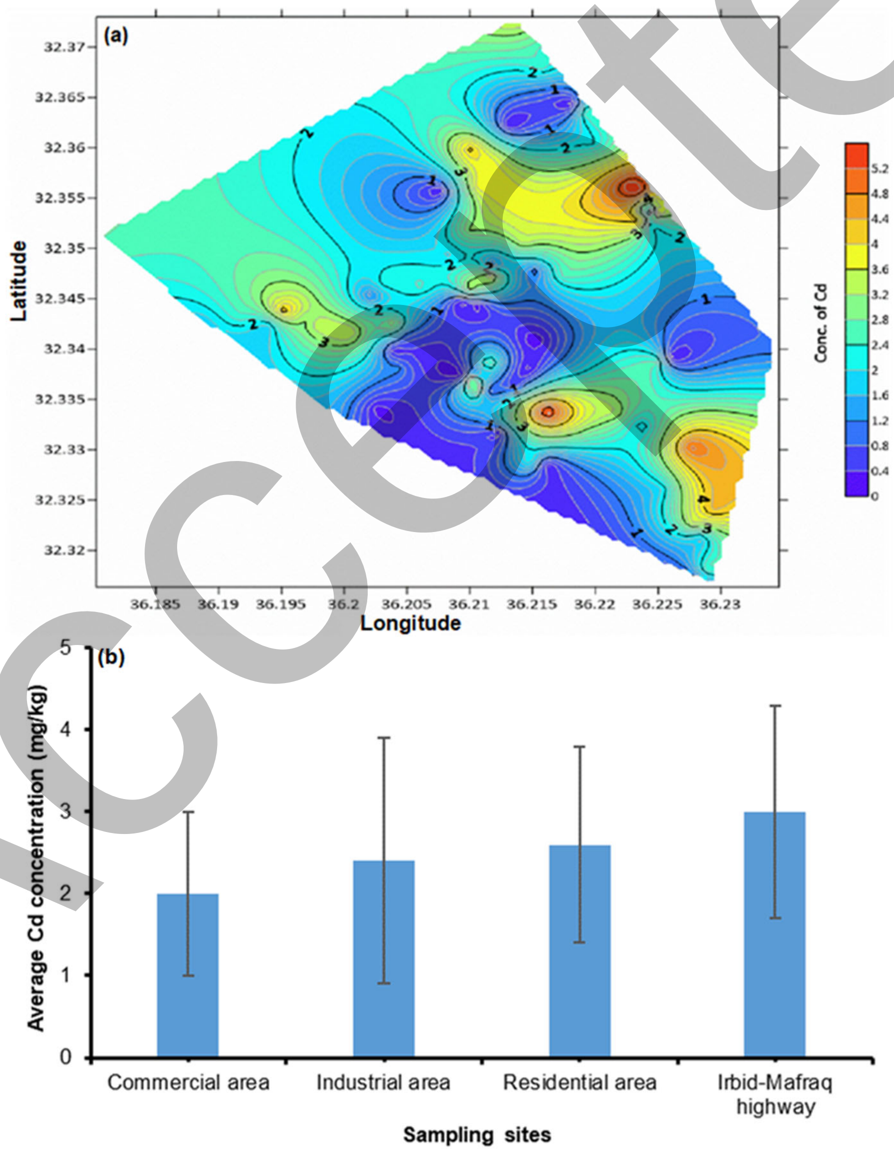


Fig 5. (a) Spatial distribution and (b) average concentration of Cd in the studied areas

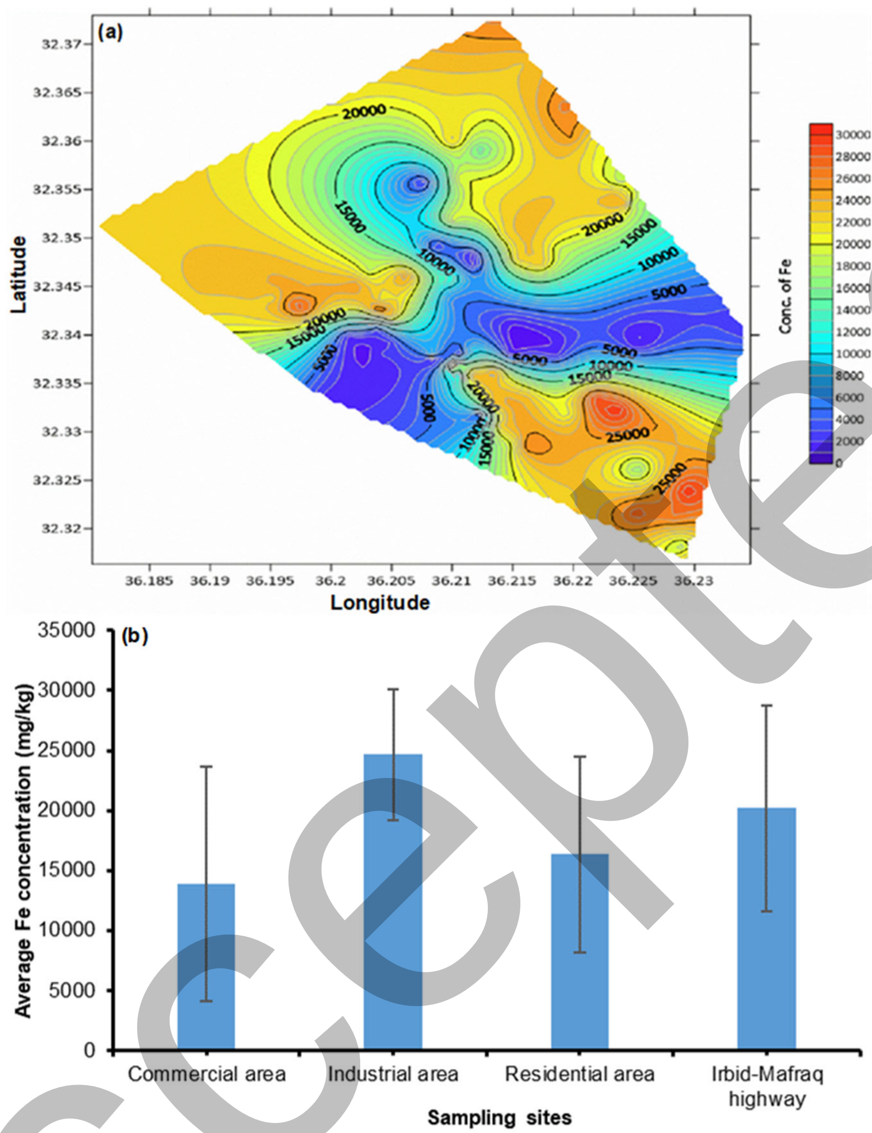


Fig 6. (a) Spatial distribution and (b) average concentration of Fe in the studied areas

industrial area with a mean of 24,654 mg/kg, which was also polluted with Cu, Ni, Co, Pb, Zn, and Mn (Table S3, Fig. 6). The highest Fe concentration is observed in the industrial area followed by Irbid-Mafraq highway as highlighted by the red color in the map (Fig. 6(a)). The average Fe concentrations in the analyzed soils are in the following order: industrial area > Irbid-Mafraq highway > residential area > commercial area (Fig. 6(b)). The major sources of Fe in urban soils reported in the literature include brake linings, vehicle wear, storage of discarded vehicles, and the presence of steel factories [5,11,49].

Copper

For the concentration of Cu in the analyzed roadside soils, the results of this study reveal that the average Cu concentrations ranged from 32.8 to 87.5 mg/kg with a mean of 40 mg/kg (Table 2) (for details, refer to Tables S2–S5). The permissible Cu concentration recommended by Dutch and Nigeria standards is 36 mg/kg [33-34], while FAO/WHO and EU standards set this value at 100 mg/kg [35,39]. These findings confirm that the average Cu concentrations in the analyzed soils were within the permissible limits set by FAO/WHO and EU standards but higher than those set by Dutch and Nigeria standards. Results show that the highest Cu concentration was found in the industrial area with a mean of 87.5 mg/kg, where Ni, Co, Pb, Fe, and Mn were also at maximum (Table S3, Fig. 7). This is because Cu is mainly originated from tires abrasion, engine wear, power wires, and corrosion of vehicle parts [23,29,41]. The spatial distribution of Cu in the roadside soils is shown in Fig. 7(a). It is observed that the residential area exhibited maximum Cu concentrations followed by the Irbid-Mafraq highway, as highlighted by the red color in the map (Fig. 7(a)). In addition, it is shown that other studied areas (residential and commercial areas) exhibited similar spatial distribution patterns. The average Cu concentrations in the analyzed soils are in the following order: industrial area > Irbid-Mafraq highway > residential area > commercial area (Fig. 7(b)). The high Cu concentrations detected in the analyzed soils might be due to brake pads, vehicles, engine wear, paints, tire wear, car lubricant wear, and brake dust [5,11,41]. In addition, power wires are the major sources of Cu in residential areas [11]. The average Cu concentration determined in this study was higher than those found in Pakistan [37], China [38],

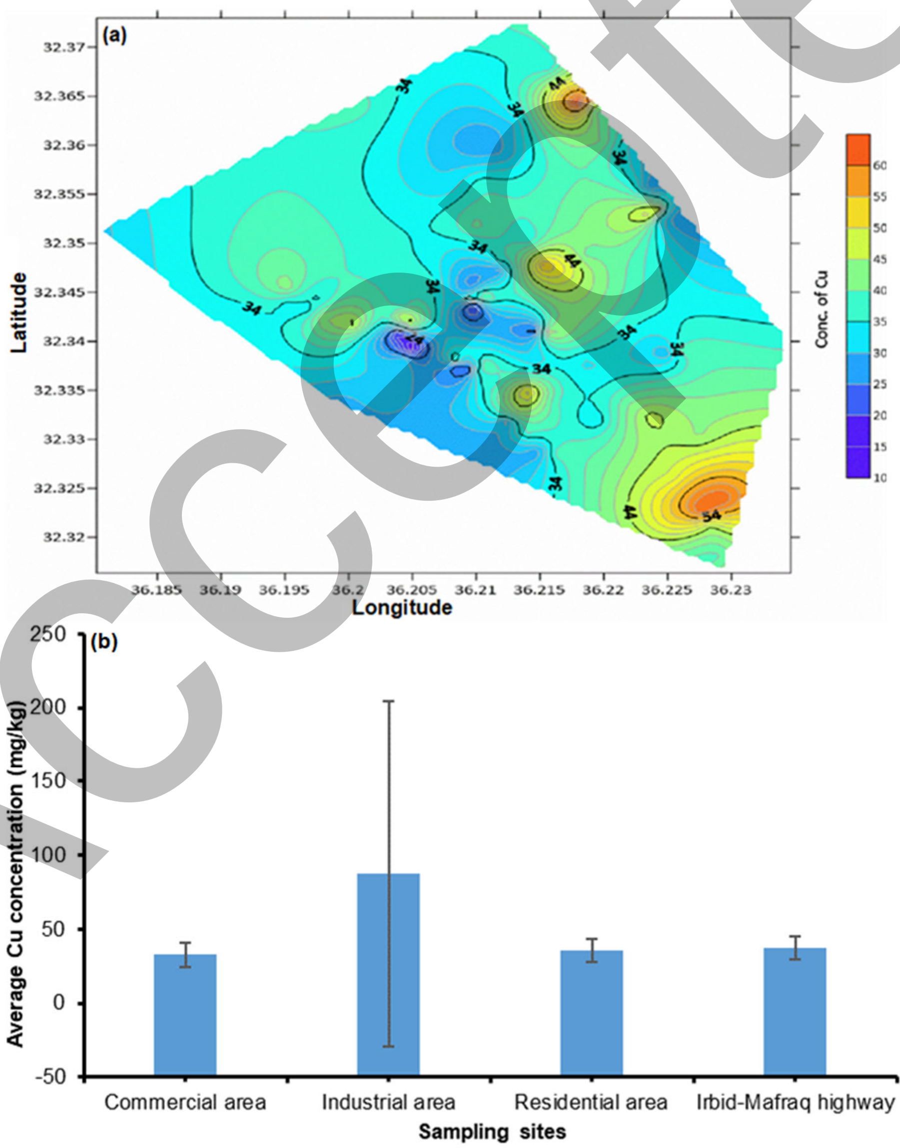


Fig 7. (a) Spatial distribution and (b) average concentration of Cu in the studied areas

India [43], Jordan, Petra city [31], and Jordan, Al-Zarqa city [8], but lower than those found in England [44], Egypt [9], and Turkey [7] as shown in Table 3.

Chrome

The average Cr concentrations in the analyzed soils ranged from 64.9 to 83.1 mg/kg with a mean of 72.9 mg/kg, as shown in Table 2 (for details refer to Tables S2–S5). The maximum Cr concentration permitted in the soil is 100 mg/kg according to the Dutch, EU, Nigeria, and FAO/WHO standards [33-35,39]. This implies that the average Cr concentrations obtained in this study were within the permissible limits for this metal. Results show that the highest Cr concentration was found at the Irbid-Mafraq highway with a mean of 83.1 mg/kg, which was also heavily polluted with Cd, Ni, Cu, Co, Pb, Fe, and Mn (Table S5, Fig. 8). This is because Cr is mainly derived from vehicle emissions, tire abrasion, vehicle lubricant wear, engine wear, and brake dust [5,11,36,45]. Other sources of Cr in urban soils reported in the literature include paints, corrosion of appliances, using chrome-plated house products, coal burning, combustion of fossil fuels, and chrome steel manufacturing [5,23,36,50]. The highest concentration of Cr is observed in the Irbid-Mafraq highway, followed by the industrial area, as highlighted by the red color in the map (Fig. 8(a)). The average Cr concentrations in the analyzed soils are in the

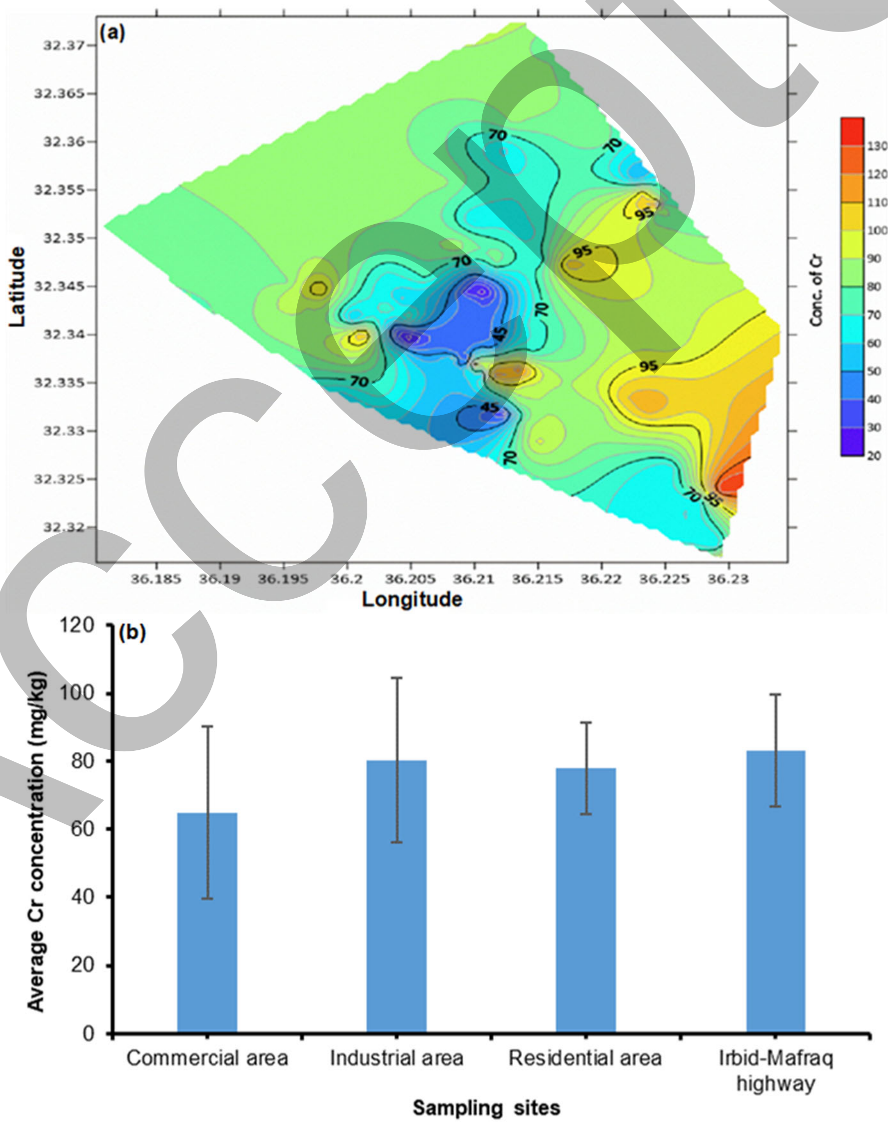


Fig 8. (a) Spatial distribution and (b) average concentration of Cr in the studied areas

following order: Irbid-Mafraq highway > industrial area > residential area > commercial area (Fig. 8(b)). The average Cr concentration in this study was higher than those found in India [43] but lower than those found in Egypt [9], Jordan, Al-Zarqa city [8], and Turkey [7], as shown in Table 3.

Manganese

The average Mn concentrations in the analyzed soils ranged from 431.8 to 576.5 mg/kg with a mean of 477.8 mg/kg, as shown in Table 2 (for details refer to Tables S2–S5). According to FAO/WHO standards, the maximum Mn concentration permitted in the soil is 2000 mg/kg [35], which confirms that the average Mn concentrations obtained in this study were within the permissible limit for this metal. Results show that the highest Mn concentration was found in the industrial area with a mean of 577 mg/kg, where Ni, Cu, Co, Pb, Fe, Zn, and Mn were also at maximum (Table S3 and Fig 9). The elevated levels of Mn are observed in the industrial area followed by the Irbid-Mafraq highway, as indicated by the red color in the map as shown by the legend on the right side of (Fig. 9(a)). The average Mn concentrations

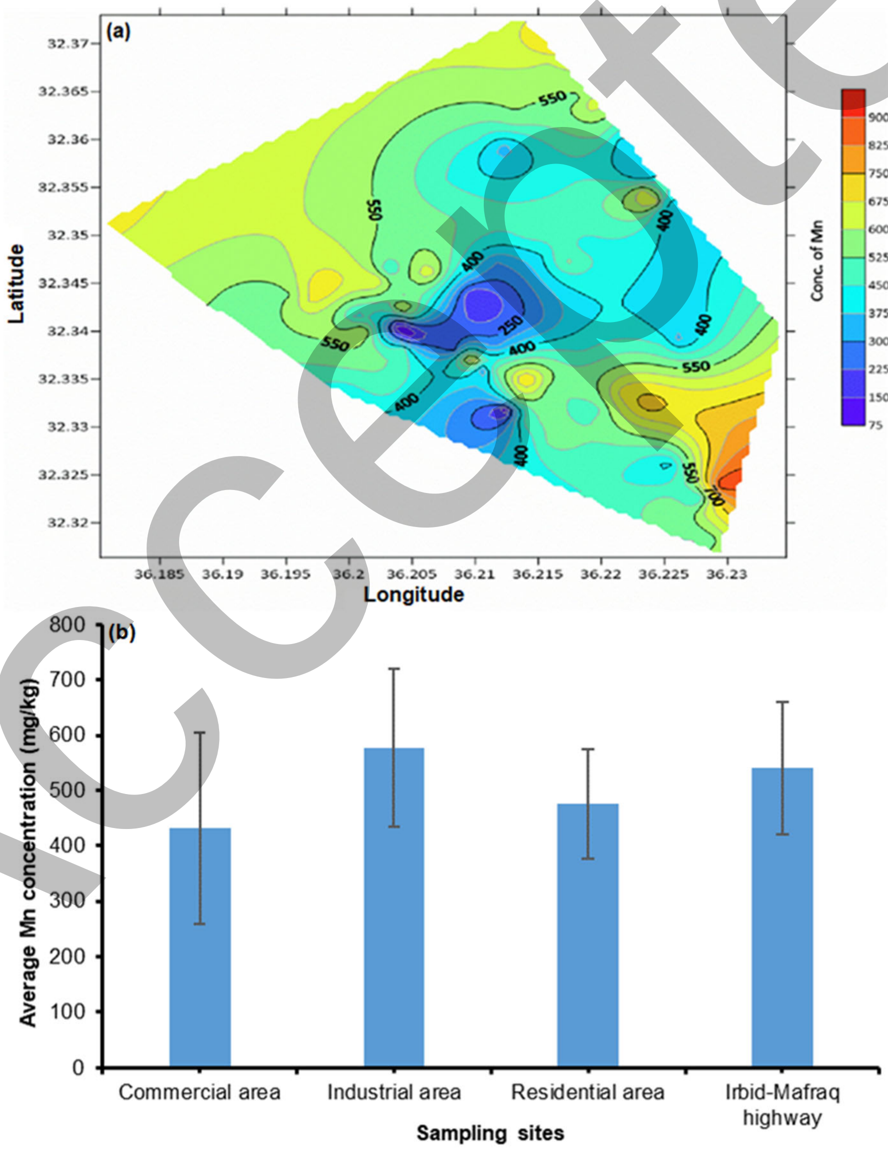


Fig 9. (a) Spatial distribution and (b) average concentration of Mn in the studied areas

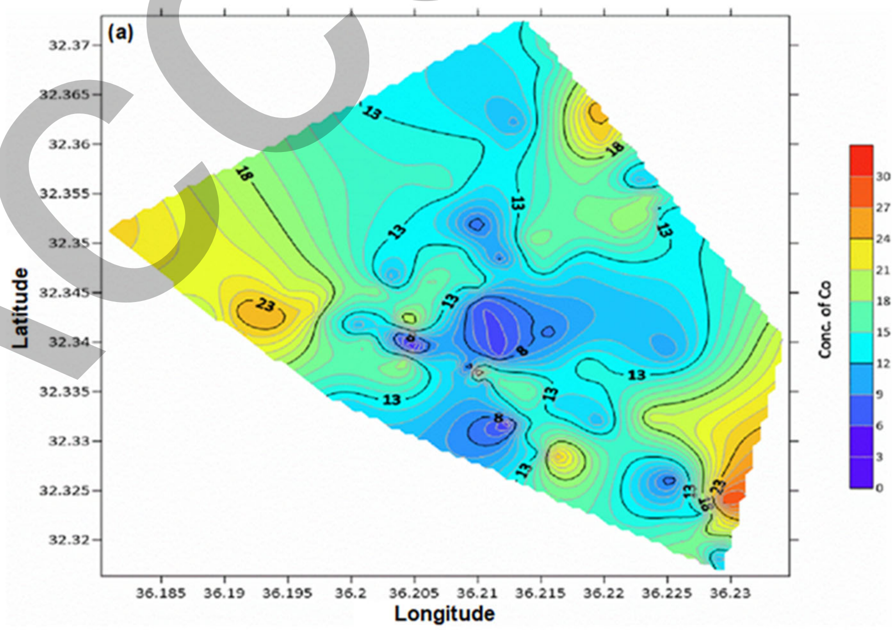
in the analyzed soils are in the following order: industrial area > Irbid-Mafraq highway > residential area > commercial area (Fig. 9(b)). We attribute the high Mn levels in the analyzed soils to vehicle emissions and using Ni-Mn batteries [5,11,51]. Other sources of Mn in urban soils reported in the literature include steel mills, glass industry, and smelting operations [52]. The average Mn concentration obtained in this study was much higher than those found in Pakistan [37] and India [43] but lower than those found in Turkey [7] and Jordan, Al-Zarqa city [8], as presented in Table 3.

Cobalt

Based on the results of this study, the average Co concentrations in the analyzed soils ranged from 13.2 to 16.7 mg/kg with a mean of 14.0 mg/kg (Table 2) (for details refer to Tables S2–S5). The maximum Co concentration permitted in the soil is 9.0, 50, and 20 mg/kg according to the Dutch, EU, and Nigeria standards, respectively [33-34,39]. This means that the average Co concentrations in the analyzed soils were found to be within the permissible limits set by Nigeria and EU standards but higher than the permissible limit set by Dutch standards. The highest Co concentration was found in the industrial area with a mean of 16.7 mg/kg, where Ni, Cu, Mn, Pb, Fe, Zn, and Mn were also at maximum (Table S3 and Fig. 10). This is expected because Co is derived from vehicle emissions, corrosion of batteries, oil leakage, and burning coal and oil [5,11,45,49]. The higher Co concentrations are indicated by the red color on the map, as shown by the legend on the right side of Fig. 10(a). The highest Co concentration was observed in the industrial area, followed by Irbid-Mafraq highway. Furthermore, it can be seen that all the studied areas exhibited similar spatial distribution patterns (Fig. 10(a)). The average Co concentrations in the analyzed soils are in the following order: industrial area > Irbid-Mafraq highway > residential area > commercial area (Fig. 10(b)). The average Co concentration determined in this study was higher than those found in Pakistan [37] but lower than those found in Turkey [7], as presented in Table 3.

Enrichment Factor

With the aim to evaluate whether the source of metal pollution in the analyzed roadside soils is due to geological or anthropogenic reasons, the enrichment factor (EF) for the studied metals was calculated and listed in Table 4. In the literature, the EF was measured by calculating the ratio of the concentration of the studied



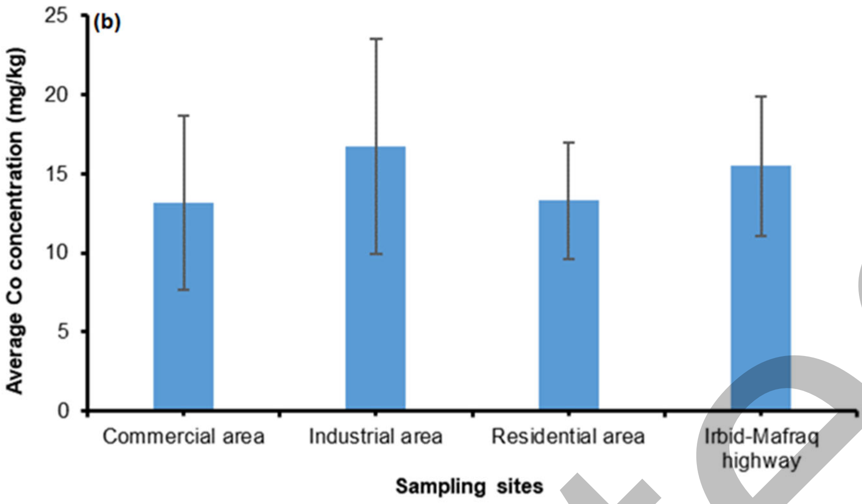


Fig 10. (a) Spatial distribution and (b) average concentration of Co in the studied areas

Table 4. Average values (±SD) of enrichment factor for Cd, Mn, Cr, Cu, Pb, Zn, Co, and Ni in the studied areas (commercial, industrial, residential, and Irbid-Mafraq highway)

Metal	Commercial area	Industrial area	Residential area	Irbid-Mafraq highway
Cu	9.0 (±12.2)	4.8 (±6.1)	7.2 (±9.2)	6.6 (±11)
Zn	11.7 (±16.1)	2.2 (±1.3)	6.8 (±9.5)	6.5 (±11.7)
Mn	4.5 (±5.7)	1.4 (±0.2)	3.9 (±4.9)	3.7 (±5.6)
Pb	9.8 (±13.6)	6.3 (±9.1)	8.3 (±11)	4.9 (±3.5)
Cd	57.0 (±45.3)	31.7 (±21.1)	107 (±191)	48.6 (±25)
Cr	13.1 (±18.4)	3.3 (±0.7)	12.7 (±17.3)	12.0 (±21.3)
Co	8.0 (±9.7)	2.3 (±0.7)	6.9 (±8.9)	6.3 (±9.9)
Ni	15.0 (±19.4)	3.6 (±0.8)	12.8 (±18.5)	12.5 (±22.3)

metal in the soils over the concentration of a crustal or background metal such as Fe or Al [11,31,53]. In the present study, the enrichment factor is calculated using the Eq. 3 [11]:

$$EF = \frac{[M]_{soil}/[Fe]_{soil}}{[M]_{crust}/[Fe]_{crust}} \quad (3)$$

where $[M]_{soil}$ is the concentration of metal in the roadside soil, $[Fe]_{soil}$ is the concentration of Fe (as a crustal metal) in the roadside soil, $[M]_{crust}$ is the concentration of metal in the earth crust, and $[Fe]_{crust}$ is the concentration of Fe in the earth crust. In the literature, several pollution classifications are based on the EF values. For example, Sutherland classified pollution levels into five categories based on the EF values: (1) minimal enrichment ($EF < 2$), (2) moderate enrichment ($2 < EF < 5$), (3) significant enrichment ($5 < EF < 20$), (4) very high enrichment ($20 < EF < 40$), and (5) extremely enrichment ($EF > 40$) [54]. Eid Alsbou and Al-Khashman have three pollution levels based on the EF values: (1) $EF < 10$ indicate low enrichment, (2) EF values ranging between 10 and 100 indicate medium enrichment and (3) $EF > 100$ indicate high enrichment [31].

In the present study, the average EF values for the studied metals were calculated in roadside soils and listed in Table 4 (for details, refer to Table S6). Results show that the high enrichments of the studied metals were obtained in the soils collected from the commercial and residential areas. These results can be attributed to the presence of different anthropogenic activities in these areas, such as traffic emissions, vehicle wear, lubricating oils, and abrasion of brake linings. The average EF values of the studied metals were found to decrease in the following order: Cd > Ni > Cr > Pb> Cu > Zn > Co > Mn (Table 4). In addition, results show that

Cd has a medium to high enrichment factor (ranging from 31.7 to 107) (Table 4). These results reveal that the source of Cd in the analyzed soils is attributed to anthropogenic sources such as old car tires, traffic emissions, and engine oil leakage [5,11,23,47]. In addition, the results show that Zn, Cd, Cr, and Ni have medium enrichment factors ($10 \leq EF \leq 100$) in all sampling sites, which confirms that the origin of these metals in the analyzed soils is due to different anthropogenic sources like traffic emissions and different industrial activities. Moreover, the results show that Mn, Cu, Co, and Pb have low enrichment ($EF < 10$), which means that the origin of these metals in the analyzed soils is due to geological sources (Table 4).

Statistical Analysis of Results (ANOVA)

Analysis of variance (ANOVA) was used to assess whether there was a significant difference in metal concentrations between soils collected from the different studied areas (commercial area, industrial area, residential area, and Irbid-Mafraq highway). From the statistical analysis of the data obtained in this study, the P values obtained for Cr, Cu, Fe, and Mn concentrations were found to be 6.6×10^{-3}, 8.1×10^{-4}, 2.1×10^{-3}, and 8.2×10^{-3}, respectively. These findings indicate a significant difference in metal concentrations between the analyzed roadside soils collected from the studied areas investigated in this study ($p < 0.05$). In addition, results show that the P-value for Cd, Co, Mn, Ni, Pb, and Zn were 0.0558, 0.1265, 0.0896, 0.2838, and 0.3434, respectively. These results clearly indicate no significant difference in metal concentrations between the roadside soils collected from the studied areas investigated in this study ($p > 0.05$). One possible explanation for the significant variation in metal concentrations found between the studied areas might be due to different anthropogenic sources like traffic density, industrial emissions, using of chemical-based fertilizers, tire rubber, or engine oil leakage [5,11].

■ CONCLUSION

The results of this study showed that the roadside soils collected from different sampling sites in Mafraq city were polluted with the studied heavy metals (Cd, Cr, Cu, Mn, Pb, Co, Fe, Ni, and Zn). The high concentrations of the studied metals found in the roadside soils are considered a serious risk to human health, the environment, and the atmosphere. The high concentrations of these metals in the analyzed roadside soils may be attributed to different anthropogenic sources like traffic emissions, industrial, commercial, and agricultural activities. The average concentrations of Fe, Mn, Cu, Co, Ni, and Pb in the industrial area are higher than in other studied areas. The average concentrations of Cr and Cd in the Irbid-Mafraq highway are higher than in other studied areas. The ANOVA results revealed significant differences between the studied areas for Cr, Cu, Fe, and Mn, but no significant differences for other studied metals. Spatial distribution results show that the highest concentrations of Ni, Cd, Co, Cr, Cu, and Mn were in the industrial area, Pb was highest in the residential area, and Zn was highest in the Irbid-Mafraq highway. The EF values for Mn, Cu, Pb, and Co indicate no enrichment, whereas Zn, Cr, and Ni show moderate enrichment in the studied areas. In addition, the EF values for Cd show high enrichment in most of the studied areas. According to the results obtained in this study, it is recommended that regular and comprehensive monitoring of the elevated levels of the studied metals in the analyzed roadside soils is encouraged to improve our understanding of the sources, distribution, and accumulation of these metals in the roadside soils in Mafraq city, and consequently, ensure the suitable management of human, industrial, agricultural, and traffic activities in this city. It is also recommended that better technologies be employed to decrease the amounts of fertilizers, metal-based pesticides, industrial effluents, waste disposal, and other pollutants that are thrown into the soil and water system.

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■ CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

■ AUTHOR CONTRIBUTIONS

Ahmed Al-Sarhan collected and prepared roadside soils for analysis. Idrees Faleh Al-Momani performed the FAAS analysis. "Ayat Allah" Al-Massaedh and Ahmed Al-Sarhan wrote the manuscript draft. All authors analyzed and discussed the data obtained during this study. All authors read and approved the final manuscript.

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