Spatial Distribution of Heavy Metals in Al-Zarqa, Jordan

Reem Dabaibeh

Department Physics and Applied Sciences, Faculty of Engineering Technology, Al-Balqa' Applied University, Al-Salt, Jordan

* ***Corresponding author:***

tel: +962-798903008
email: reem.dabaibeh@bau.edu.jo

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Abstract: *Al-Zarqa is experiencing challenges in industry conversion and extensive urbanization. The environmental quality of soil in the Al-Zarqa region was analyzed by Spatial analysis for the identification of sources and estimation of the concentration of heavy metals, which helped in the assessment of soil quality and heavy metal pollution. The reason for the elevation of heavy metal pollution is increased urbanization, industrialization, traffic, oil refinery emissions, and mixed anthropogenic sources in that region. The main objective of this research was to assess the ecological impact of heavy metal pollutants in the Al-Zarqa region. The concentrations of Cd, Cr, Cu, Mn, Ni, Pb, Zn, and Fe were estimated and compared with the existing literature. The distribution pattern of each metal was identified by spatial distribution analysis. Results revealed that the concentration of Cd, Cr, and Ni) was high, while the concentration of Pb, Zn, and Cu was lower than the maximum allowed limits. Factor analysis identified the potential sources of heavy metals in the investigated area, and spatial distribution showed the geographical distribution of the heavy metals over the study area. It was concluded that identification of the potential sources of pollutants along with their geographical variations was more beneficial than only considering the individual point concentrations.*

Keywords: *heavy metals; soil pollution; Al-Zarqa; genotoxicity*

■ INTRODUCTION

Heavy metal pollution is one of the biggest challenges of the present century. Heavy metals cause negative impacts on aquatic and terrestrial organisms. These pollutants are also affecting the economy of the farming community [1]. Since the shortage of freshwater is a great concern in Jordan [2], the heavy metals available in drinking water can be considered as a threat to human health. The oncogenic risk of such metals has been proven in several studies [3]. The environmental occurrence of heavy metals is also causing adversarial effects on human health [4]. Heavy metal poisoning is identified as an important underlying factor for several diseases. These metallic elements have a relatively higher density than water [5].

Metal pollutants are one of the major environmental concerns across the globe [6]. Their presence could pose risks and hazards to humans and their trace levels in the environment are regulated by various laws and various authorities in each country [7]. Industrial, agricultural, and domestic development have also increased human exposure to heavy metals. Although these metals are naturally occurring elements in soil, major sources of metals in the environment include anthropogenic activities, industrial waste, agricultural practices, pharmaceuticals, and metal based industry operations [8]. Environmental pollution by heavy metals is considered dangerous and is particularly hazardous to human health [9]. Mineral rock weathering is also a well-identified source of metal pollutants in the environment [10].

Many studies have identified that the concentration of hazardous metals in topsoil is persistently increasing. Yaseen and Al-Hawari [11] discussed that there is a momentous increase in the total contents of metals in soils in areas experiencing increased industrialization. The major soil contaminants include Pb, Cd, Zn, and Cu, which are released into the environment through mechanical abrasion [12]. Soil contamination with metal ions is also increased through metal corrosion, soil erosion, and

other natural phenomena. Industrial sources such as oil refineries, petroleum combustion, power-plants, and recycling and waste treatment plants are also important contributors of these metals in the environment [2]. Metals such as cobalt (Co), copper (Cu), chromium (Cr), iron (Fe), manganese (Mn), molybdenum (Mo), nickel (Ni), selenium (Se), and zinc (Zn) are considered as vital nutrients for biochemical activities. Therefore, agricultural practices are also contributing to soil contamination of metals [4].

Heavy metal ions are toxic as they can gather in the human body and affect the central nervous system. Heavy metal poisoning is a serious health issue that causes physiological and neurological health problems [13]. The most hazardous metals to human health include Hg, Pb, Cd, Au, Pt, Ag, Bi, As, Se, V, Cr, and Ti [12]. Metal ions can interact with cell components such as DNA and cause carcinogenesis. Metals are also causing oxidative stress in the human body, which results in the carcinogenicity of these metals [11]. The level of exposure to heavy metals decides the degree of toxicity and their carcinogenic effects. High exposure to these metals may result in multiple organ failure and life-threatening situations [13].

Al-Zarqa has been experiencing rapid and uncontrolled urbanization and industrialization, resulting in the increase of environmental problems in this region. Heavy metals, which pose a potential health risk for human beings, are ubiquitous pollutants in urban dust because of the increase in urban population and intensity of anthropogenic activities [14].

Jordan Petroleum Refinery Company (JPRC), situated in Al-Zarqa, is a primary source of pollution and other environmental problems [15]. Al-Hussein thermal power stations, Russeifa dump, and Es-ssammra wastewater treatment complex are also important sources of pollutants in the environment [16]. The purpose of this study is to assess the key sources of pollutants in Al-Zarqa and compare the metal contamination in this region. The heavy metal contents in the soil were identified, and their concentration was measured.

Naser [10] studied the sources of contamination in soil and found that the metals naturally occur in the soil as a result of rock weathering. Metals occur in trace quantities in the environment and are often not toxic. Gao and Chen [5] highlighted that urbanization in various areas could disturb and accelerate the natural geochemical cycle of metals and results in their amassing in soil. According to Li et al. [1], anthropogenic sources are also important contributors to metal pollutants in soil. To assess the level of anthropogenic contamination of soil, it is necessary to conduct special monitoring studies. The issue of anthropogenic soil pollution caused by emissions from coal-fired thermal power stations has been studied worldwide, especially in countries where coal mining is an important industry [17]. They further added that metal mine tailings, petrochemicals, fertilizers, pesticides, landfilling with metal compounds, lead-based paints are also important sources of metal deposition in soil.

El-Hasan and Lataifeh [2] studied the impact of essential micronutrients in soil. Their findings revealed that micronutrients, including Co, Cu, Fe, Mn, Mo, Ni, and Zn, are crucial for plants and are frequently used in fertilizers. Several metals, which are essential as nutrients in lower concentrations (i.e. Zn, Cu, Fe, and Mn) can be considered harmful for plant growth if the levels of these elements in soil and atmosphere are increased [18]. The fertilizers used to supply these nutrients to soil frequently contain trace amounts of metals, including Cd and Pb. Some phosphate base fertilizers also contain impurities of F and Hg. The excessive use of these fertilizers also contributes to the accumulation of metals in soil. Yaseen and Al-Hawari [11] also presented similar findings and revealed that more than 10% of the pesticides and fungicides commonly used in agriculture are based on compounds that contain Cu, Hg, Mn, Pb, or Zn. These compounds are also an important source of soil contamination.

Municipal and industrial wastewater is an extensively discussed source of soil contamination. Wang et al. [6] found that industrial waste and effluents are commonly discharged over land. Their findings revealed that more than 20 million hectares of land received wastewater annually around the globe. Meanwhile, Al-Khashman [12] found that in developing countries, around 50% of the irrigation is based on

wastewater supply. The negative impact of heavy metal accumulation in soils includes the decrease of the yield and quality of crops, and also the quality of the atmospheric and aquatic environments. Generally, irrigation with wastewater elevates the total and available heavy metal concentrations in soils [19]. Even if the wastewater contains relatively low amounts of heavy metals, enduring exposure to this kind of wastewater can result in significant amassing of metal pollutants in soil.

Al-Taani et al. [20] studied the impact of urbanization on the heavy metals contamination in soil. The study identified that urbanization affected the ecological functions and increased Zn accumulation in soil. According to Massadeh et al. [21], sewage sludge, livestock manures, vehicle exhaust, industrial emission, and agricultural practices have become major environmental concerns worldwide. The extent of urbanization and increased population can significantly affect the inherent function of soil and facilitate the accumulation of polluted contents, as discussed by Zhang et al. [4].

According to Al-Najjar et al. [15], the natural concentration of trace elements in soil plays a critical role in controlling the effect of human activities on the soil. Heavy metals in urban soils have been widely studied due to their ubiquity, toxicity, and persistence [22]. The amassing of metals in topsoil is contributed through several environmental and human activities. Massadeh et al. [21] studied the urban population index map of different regions, and their findings revealed that in urban areas, the soil is found to be enriched in heavy metals. Soil pollution by heavy metals in industrialized areas is an on-going challenge.

Spatial analysis is frequently used to quantify the quality of soil and to assess the presence of contamination in soil. According to Al-Taani et al. [20], spatial analysis is an important predictive approach to assess the impact of urbanization on soil quality and the presence of heavy metals. The accuracy of heavy metal spatial distribution maps is critical for risk control [23]. The spatial distribution of metal ions helps in the assessment of the concentration of these pollutants in soil. According to Shan et al. [8], the rising values of these metals indicate toxicity and soil pollution. Alloway [24] suggests that heavy metals are toxic and not easy to decompose naturally. These metals are transferred from soil to the environment through water, plants, and contaminated foods.

Coal mining industries generate large volumes of gangue wastes, from which a considerable amount of toxic HMs could be released during weathering of the waste under the joint effects of water, microorganisms, vegetation, sunlight radiation, and heat. These hazardous substances enter the ecosystem by a variety of pathways where they could be detrimental to crops and animals, and might be taken up by humans through direct contact (ingestion, dermal absorption, and inhalation) or food chains [25].

Abderahman and Abu-Rukah [16] showed that heavy metal toxicity in the soil gradually increased through anthropogenic processes, including waste dumping in landfills, waste incineration, and smokestack emission. They also stated that the US environmental protection agency had identified seven metals as the priority control pollutants known for their hazardous effects on human health. These metals include nickel, zinc, copper, lead, mercury, arsenic, cadmium, and chromium. Al-Khashman [12] identified that contamination of these metal pollutants in the soil is a serious and rapidly growing problem in regions that are experiencing industrial development and urban expansion.

According to Li et al. [13], the level of topsoil contamination can be assessed through their spatial distribution. They defined spatial analysis as a geographical process that involves the topological, geometrical, and geographical properties of soil. It is based on the analysis of the pattern of human behaviors and their spatial expression. Al-Khashman [26] highlighted that spatial analysis is used to identify location-oriented problems. Digital mapping and spatial analysis are commonly used to characterize the soil quality and assess the spatial distribution of metals. Jaradat et al. [27] performed the comparison of non-restructured soil and restructured soil through the spatial distribution of heavy metals. The spatial variability of soil properties includes variations in soil moisture, physical and chemical properties, etc.

Moreover, temporal and spatial variations of heavy metals relate to both natural variability of soils and human activities, thus to be considered as powerful tracers for monitoring the impact of human activity [28]. Their findings revealed that territorial and human factors subsidize the gathering of various pollutants in soil, including heavy metals.

Al-Khashman [26] studied the extent of contamination in topsoil and spatial distribution of metals. Their findings indicated that the concentrations of Pb, Cu, Cr, Zn, and Cd are higher in areas near industries. They identified that the accretion of these metals was higher in areas near cement factories. Al-Khashman [12] studied the heavy-metals concentration in the urban samples of topsoil from Jordan. According to their findings, Zn and Pb had high concentrations, while Cd and Ni had lower concentrations. It was further concluded that large scale industrialization is a major factor for soil degradation and heavy metal pollution.

Al-Khashman [26] studied the extent of pollution in Karak Industrial estate, Jordan. Their findings revealed that the soil in the studied area was polluted with metals, including Fe, Cu, Zn, and Pb. They also found that the concentration of these metals exceeded the limits in topsoil, but their concentration was found to be decreased in lower soil. They concluded that the main sources of these pollutants were the nearby industrial places. El-Hasan and Lataifeh [2] extended the discussion by analyzing the topsoil composition in the Nepal region. They analyzed the amounts of eight trace metals, including Cd, Cr, Cu, Fe, Mn, Pb, V, and Zn. Their findings suggested that Pb and Zn were greater in concentration near the metropolitan regions while Cr, Fe, and V were found in exceeding concentrations in controlled soil.

Banat et al. [29] performed the spatial pattern study of metal pollutants in central Jordan. They identified that Cd, Pb, and Hg were found to be in high concentrations in urban areas. They also identified the major sources of these pollutants to be the cement industry, fertilizers, and vehicle emission. These metals were found in both residual and active phases. Al-Khashman and Shawabkeh [30] studied the distribution analysis of Pb, Zn, Cd, and Cr in Southern Jordan near industrial areas. Their results highlighted that Pb, Zn, and Cd were in higher concentration in areas near the cement manufacturing plant. The study further stated that anthropogenic activities are the major contributors to the pollutants in urban soil.

Soil, an important environmental medium, is exposed to a number of pollutants including toxic heavy metals by various natural and anthropogenic activities [19]. Extensive urbanization and industrialization lead to the accretion of toxic metals in topsoil. The current study is aimed to assess the sources of contamination and amounts of heavy metals in the Al-Zarqa region of Jordan.

■ EXPERIMENTAL SECTION

Materials

Analytical reagent-grade of acids, bases, nitrate salts of metals, and other chemicals obtained from Merck (Darmstadt, Germany) were used as received. All solutions were prepared in double deionized water (DDW). All plastics and glassware were cleaned by soaking in HNO_3 solution (10% v/v) and then rinsed with DDW before use. The stock standard solutions of analytes (1000 mg L^{-1}) were prepared from analytical grade nitrate salts of the analytes. The working standard solutions were prepared by appropriate dilution of the stock solutions. ATP was purchased from Sigma-Aldrich [31].

Instrumentation

A Shimadzu model AA-680 atomic absorption spectrometer (Japan) with a hollow cathode lamp as a radiation source and a deuterium background corrector at respective wavelengths (using an air-acetylene flame) was used for metal ions determination in standard and sample solutions. All operating parameters were those recommended by the manufacturer. A Metrohm 691 pH/ion meter (Buchs, Switzerland) supplied with a combined glass-calomel electrode was used for the pH adjustments. The size and morphology of GO were observed by TEM using a CM120 microscope (Philips, Netherlands). Raman spectra of GO were prepared using a SENTERRA microscope (BRUKER, Germany). FT-IR spectra were taken on a BRUKER VECTOR 22 Spectrometer. The flow of the sample and eluent

through the column was adjusted using a 10 roller peristaltic pump (Ultrateck Labs Co. Iran) [31].

Procedure

Studied area

Al-Zarqa is situated in the northeast part of Jordan, at the basin of the Zarqa River, Fig. 1. It has a total area of 4586 km^2; about 4074 km^2 of which is within Jordan and 512 km^2 is within Syria [32].The city is located northeast of Amman and has a population of more than 640,000 inhabitants. The city is experiencing extensive industrialization and urbanization since the past decades. The Al Zarqa Channelized Stream is considered to be one of the most polluted regions in Jordan due to massive discharge and accumulation of pollutants over the years [33]. The city is situated near the international Amman-Baghdad highway. It is also the main industrial center of Jordan, and more than 50% of the country's industries are situated in this city. Jordan oil refinery plants, leather and garment factories, new agricultural and pharmaceutical factories represent the recent industrial development in this region. The increased industrial development has facilitated the growth of the city's population. The increased rate of population growth and industries has caused serious environmental problems in this region.

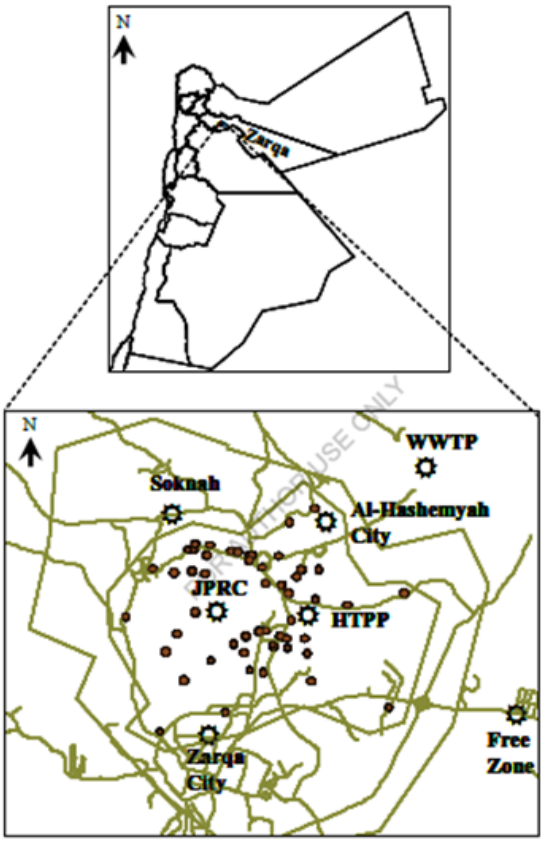

Fig 1. Jordan map shows the Al-Zarqa city

Sampling strategy

About one kilometer was included for acquiring the soil samples from the studied area. Different samples were collected with the help of an auger and were transferred into labeled polyethylene bags. The sample bags were labeled according to the sampling area, date, and weight of the collected samples. The samples were also documented with clear details to avoid any mixing between samples.

Sample processing

The pre-treatment of samples was performed by cleaning all glassware with tap water, detergent, and distilled water. Then the glassware was soaked in 10% HNO_3 (v/v) for one night. Finally, all glassware was cleaned and rinsed with deionized water. The collected samples were spread out to separate the extra materials such as grass, stones, mosses, and roots. All materials sized above 2 cm were separated from the samples using physical procedures. Afterwards, the samples were air-dried at room temperature for two weeks by placing them in a clean area. Later, the dried sections were sieved through 2 mm mesh; the attained sections were then further divided into three labeled polyethylene bags. These samples were stored at 4 °C in a refrigerator until it was time to perform the chemical analysis.

Chemical analysis

pH. The pH of all collected soil samples was measured by making a 1:1 ratio of soil and deionized water mixture (w/v). A potentiometric approach was used to measure the pH following the EPA standard analysis method 9045D [20]. The measured pH values were reported as pH (H_2O) at 25 °C.

Cation exchange capacity (CEC). An excess of sodium acetate solution mixed with soil samples resulted in the exchange of sodium cations with matrix cations. The mixture was then rinsed with isopropyl alcohol. Ammonium acetate solution was later added, which exchanged the adsorbed sodium with ammonium ion. This sample was then embedded in each batch (8 samples). In addition, a blank was embedded within

every three successive batches. The concentration of the released Na^+ was determined by atomic emission spectroscopy at 589 nm wavelength. The results were shown in centimoles of positive charge per kilogram of soil (cmol [+]/kg); (cmol Na = {(weight of Na in the sample/ atomic weight of Na) × 100}).

Total organic carbon (TOC). A 0.15 mm sieve was used to sieve soil samples, and about 0.5 g of soil that passed the sieve was weighed to the nearest of 0.0001 g and placed in a digestion tube (250 mL). Then, 15 mL of the digestion solution (0.066 M $K_2Cr_2O_7$ and 9 M H_2SO_4) was poured into the digestion tube and boiled gently over a hot plate for 45 min. The digested samples were then allowed to cool before adding 50 mL H_2O, 5 mL 85% H_3PO_4, and four drops of the indicator (*o*-Phenanthroline). Finally, the solution was titrated against ferrous ammonium sulfate with the endpoint color change from green to reddish-brown. The quality of the analysis was monitored by triplicate analysis of a known sample, and then it was embedded with each batch (10 samples). In addition, two unheated blanks and two heated blanks were also analyzed with each batch. Before titration, ferrous ammonium sulfate solution was standardized against $K_2Cr_2O_7$. The TOC of the samples was reported as a percentage of TOC.

Metals. About 0.5 g of each air-dried and sieved samples were taken and placed in a Teflon beaker. Around 15 mL of HNO_3, 4 mL of HF, and 4 mL of $HClO_4$ were added to the sample filled beakers, and the attained mixtures were then boiled in a sand bath at 170 ± 20 °C. The boiling was done in closed beakers until the samples were completely dissolved. Then the covers were removed to allow the solution to evaporate and become dry. Afterwards, 5 mL of HNO_3 was dropped into the samples and evaporated until 2 mL was left. Then the mixture was diluted with deionized water. The obtained mixtures were kept in polyethylene bottles. All samples were again labeled and documented in sample preparation sheets.

Flame atomic absorption spectrometry analysis was performed for all obtained samples, certified reference materials, reagent blanks, and method blanks. An air-acetylene flame was utilized in the spectrometer to attain the details of present metals. The stock solution of 1000 ppm was obtained, and different successive dilutions were prepared to attain the calibration graphs. The graphs were analyzed to identify the linear relationship. Table 1 presents all-instrumental parameters used in this experiment.

Method validation

The accurate analysis of metal contamination requires comparison with blank measurements. The contamination of metals was detected by measuring the sample to blank ratios. Blank samples or acid samples were prepared and treated like samples. The effects of used acids and reagents were calculated. The sample to blank ratios of all metals except Cd were estimated to be larger than 10, while the sample to blank ratio of Cd was measured around 9. These findings suggest that blank subtraction has no significant impact on the observed concentrations of metals. The limits of detection of the measured metals were also calculated, and the results indicate that measured elements were higher than the LOD, Table 2.

The limits of detection (LOD) of the identified metals in the samples were measured as the concentration

Table 1. The instrumental parameters used in this study

Element	λ (nm)	Air flow (L/min)	Acetylene flow (L/min)	Slit width (nm)	Lamp current (mA)
Pb	217.0	3.5	1.5	1.0	10.0
Cr	357.9	4.0	2.5	0.2	7.0
Cd	228.8	3.5	1.5	0.5	4.0
Zn	213.9	3.5	1.5	1.0	5.0
Cu	324.8	3.5	1.5	0.5	4.0
Ni	232.0	3.5	1.5	0.2	4.0
Mn	279.5	3.5	1.5	0.2	5.0
Fe	386.0	3.5	1.5	0.2	5.0

Table 2. Sample to blank ratio and detection limits of elements

Elements	Average concentration (mg/L)	Average blank concentration (mg/L)	Sample to blank ratio	LOD (mg/L)
Cu	1.113	0.015	74	0.013
Ni	4.140	0.119	35	0.053
Zn	2.582	0.180	20	0.004
Mn	3.437	0.029	119	0.011
Pb	2.501	0.040	63	0.031
Cd[a]	0.037	0.0048	9	0.008
Cr	0.883	0.043	21	0.040
Fe	396	0.27	1467	1.25

[a]: Blank levels were below the detection limits. Therefore, half of the detection limit was used to calculate the sample to blank ratio

that produces three times the SD at a particular wavelength. The LOD was calculated for each sample with the help of a blank sample. The calculated values were much higher than the Limits of detection. A periodic examination of Standard Reference Materials (SRM) was performed to assure the accuracy of the results. The SRM was collected from the National Institute of Standards and Technology. The results of the SRM were found to be fairly close to the certified values. Table 3 represents the list of SRMs used in this study, and Table 4 shows the accuracy and precision based on three standard reference materials for FAAS analysis.

Method of heavy metal pollution assessment

This study utilized extensive field research, and it is derivative in nature. The collected soil samples were examined for the presence of metals. Geostatistical methods were used for the mapping of metal concentrations. The correlation analysis was used for principal components analysis of the collected soil samples.

■ RESULTS AND DISCUSSION

Chemical Characteristics of Soil

The descriptive analyses of the chemical properties of the topsoil samples collected from Al-Zarqa city are presented in Table 5. The studied chemical properties include pH, total organic carbon (%TOC), and cation exchange capacity (CEC). A conventional pH scale 1–14 was used to measure the pH of soil at 25 °C. The % TOC indicates the total organic carbon per gram of soil. CEC indicates the centimoles of the sodium cation per kilogram

Table 3. Standard reference materials used in the study

SRM code	Material
SRM-1646aS	Estuarine sediments
SRM-1633b	Trace elements in coal fly ash
SRM-2702	Inorganics in marine sediments

Table 4. Accuracy and precision based on three standard reference materials for FAAS analysis

Accuracy and precision based on three standard reference materials for FAAS analyses						
Elements	SRM-1646a		SRM-1633b		SRM-2702	
	Found ± SD	Certified ±SD	Found ± SD	Certified ± SD	Found ± SD	Certified ± SD
Cu	10.53 ± 2.01	10.01 ±0.34	107.0 ± 3.34	112.8 ± 2.6	102.2 ± 3.77	
Cr	45.2 ± 3.5	40.9 ± 1.9	352 ± 54	352 ± 22	202	198.2 ± 4.7
Ni	24.4 ± 2.3	23	146.1 ± 12.9	102.6 ± 1.8	78.3 ± 4.8	75.4 ± 1.5
Pb	12.9 ± 4.6	11.7 ± 1.2	73.7 ± 9.4	68.2 ± 1.1	135.6 ± 16.7	132.8 ± 1.1
Cd	0.18 ± 0.18	0.15 ± 0.01	2.43 ± 0.29	0.784 ± 0.01	0.99 ± 0.22	0.82 ± 0.01
Zn	55.0 ± 2.9	48.9 ± 1.6	214.0 ± 2.7	210	479.2 ± 1.4	485.3 ± 4.2
Mn	236.0 ± 10.9	234.5 ± 2.8	133.9 ± 1.5	131.8 ± 1.7	1685 ± 64	1757 ± 58
[a]Fe	2.15 ± 0.19	2.00 ± 0.04	7.70 ± 0.53	7.78 ± 0.23	7.81 ± 0.28	7.91 ± 0.24

Note: all concentrations are in μg/g except for Fe in (w/w) percent

of soil (cmol [Na^+]/kg), as presented in Table 5.

The results indicate that the pH of all studied samples was between 7.0–8.2, with an average of 7.7. These findings suggest that the soil was neutral to slightly alkaline in nature. pH is an important chemical property that plays a decisive role in the determination of the behavior of different chemical components of soil samples such as metals. Acidic pH is more favorable for the mobility of metal ions, while alkaline pH usually limits the bioavailability of metal ions [6].

The %TOC values ranged between 0.2–3.6%, while the average of %TOC for all soil samples was 0.8%. These findings revealed that the distribution patterns of the organic matter in the studied area were irregular. This indicates the variable distributions of the vegetation in the studied region. Abderahman and Abu-Rukah [12] suggested that the extent of organic matter significantly affects the absorption of the heavy metals in soil due to the cation exchange characteristic of organic matter. Therefore, the varied distribution of organic matter also resulted in the varied distribution of heavy-metals throughout the investigated area.

The CEC ranged from 14.46 to 42.9 cmol [Na^+]/kg with an average of 28.5 cmol [Na^+]/kg. It is also evident that the pH and amount of organic matter have a direct effect on CEC values of soil, and an increase in these values can also increase the CEC values [17].

Measured Concentrations of Metals

The concentrations of seven metals, Cd, Cr, Cu, Ni, Mn, Zn, and Pb, were measured in mg/kg of the dry weight. The concentration of Fe was measured in w/w or mass to mass ratio of the dry soil. The results revealed that the measured concentrations of all metals were significantly higher than their detection limits. The values of the metals concentrations, their mean, standard deviations (SD), minimum and maximum concentration are presented in Table 6. It is evident that the concentrations of the metals in topsoil are log-normally distributed rather than normally distributed.

There are several factors that can influence the concentration of metals in soil. The important factors which can alter the concentration of heavy metals in soils include pH, the ion-exchange capacity of the organic matter, wind directions, nature, and composition of the soil [21]. The distribution and mobility of metal ions are also varied according to the mentioned soil properties. The findings of the current study indicate that there was a lack of correlation between the pH, CEC, and % TOC of the soil samples and the concentrations of the heavy metals. The results of this study show strong agreement with Khashman and Shawabkeh [19].

Comparison with the Literature

The studied literature and the findings of the present study were matched to analyze the extent of the soil contamination by the studied metals. This comparison increased the understanding of the extent of soil pollution by metals in the investigated area. The metals that include Pb, Cd, Cr, Zn, Al, Fe, and Mn, are mostly discussed in the literature as heavy metal pollutants

Table 5. Chemical characteristics of soil

	Average	SD	Median	Min	Max
% TOC	0.8	0.6	0.6	0.2	3.6
CEC	28.5	6.6	27.8	14.6	42.9

Table 6. The concentration of metals in the studied area

Metals	Unit	Mean	SD	Median	G. Mean	Min	Max
Cd	mg/kg	6.6	5.2	4.8	5.5	2.3	27.1
Cr	mg/kg	88.2	29.4	84.0	83.8	41.6	183.1
Cu	mg/kg	21.7	8.8	19.10	20.8	9.2	57.1
Mn	mg/kg	492.2	164.3	502	459.0	79.2	932.4
Ni	mg/kg	113.1	128.5	62.2	81.7	41.9	600.5
Pb	mg/kg	58.9	30.6	64.0	47.8	5.7	166.7
Zn	mg/kg	122.0	71.2	109.9	105.9	27.8	356.2
Fe	% (w/w)	1.8	1.0	1.6	1.6	0.2	4.8

in soil [17,21]. The literature also indicated that these metals are the more frequently occurring metal pollutants in urban soil [22-23]. The comparison of the metal concentrations in soil in the present study and the reported metal concentrations in literature are presented in Table 7.

The result indicates that Cd concentrations in the studied areas ranged from 2.3 to 27.1 mg/kg, with a mean concentration of 6.6 (mg/kg). This suggests that the mean concentration of Cd in the current study is around 30 times higher than that was found in the proximity of an oil refinery in Spain. This concentration is also higher than what had been found in the Ankara region [18,23]. The obtained Cd concentration in the current research validates the previously measured concentrations of Cd in this region which was 7.0 mg/kg [25].

The Cr concentration ranged from 41.6 to 183.1 mg/kg, with a mean concentration of 88.2 mg/kg. Comparison with previous studies shows that this measured concentration is almost 5 times higher than the Cr concentration measured in the proximity of an oil refinery in Spain and 4 times higher than the calculated values for the nearby area of JPRC [26]. The comparison also revealed that the measured values of Cr concentration in this study were in close agreement with the results of Banat et al. [18] and significantly lower than the Cr concentration in the Ankara region as measured by Yay et al. [23].

The calculated values of Cu concentration in the current study ranged from 9.2 to 57.1 mg/kg, with a mean concentration of 21.7 mg/kg. The comparison analysis revealed that the Cu values found in this study are much lower than the previously stated concentration by Momani et al. [26]. These values are also lower than the reported Cu concentration for the Ankara region [23]. However, the means of concentrations of these results are still higher than the values measured by Al-Khashman [16].

The Ni concentrations in this study ranged from 41.9 to 600.5 mg/kg, with a mean concentration of 113.1 mg/kg. The comparison analyses revealed that the mean concentration of Ni in the current study is 40 times greater than the previously measured Ni concentration by Al-Khashman [16], but lower than 121.5 mg/kg, which is the value reported earlier in JPRC by Al-Shatnawi [25].

The Pb concentrations in the current study ranged from 5.7 to 166.7 mg/kg, with a mean concentration of 58.9 mg/kg. This value is slightly lower than the value calculated by Banat et al. [18]. However, the average concentration of Pb is significantly greater than the formerly reported values in other literature [16,24]. The values reported by Yay et al. [23] for the Ankara region were 3 times greater than the results of the present research.

The measured concentrations of Zn in this study ranged from 27.8 to 356.23 mg/kg with a mean value of 122.0 mg/kg. This mean value is in close agreement with the values reported by Al-Shatnawi [25], which was 127.5 mg/kg. However, when compared to the results obtained by Banat et al. [18] and Yay et al. [23] the measured concentrations of Zn in the current study are lower.

Table 7. Comparison of metal concentrations with literature

Metals	Results of this study	JPRC[a]	Cement[b]	Ind. Estate	Oil refinery	Ankara[e]
Cd	6.6	1.2	5.0		0.2	2.2
Cr	88.2	23.0	83.9		16.5	284.0
Cu	21.7	28.0		11.3		90.0
Mn	492.2				268.9	1100.0
Ni	113.1			4.2		81.0
Pb	58.9	21.0	62.2	11.2	37.8	189.0
Zn	122.0	104.0	146.9	13.1		181.0
Fe	18200	3755		43		858000

[a] [26]; [b] [18]; [c] [16]; [d] [23-24]; [e] [22]

Probability Distribution

The Gaussian Distribution is a well-recognized technique for factor analysis and geostatistical analysis [27]. These techniques help in attaining the approximately normal data while the non-normality is apparent in the collected data. The shape and symmetry of distribution are measured by analyzing the Kurtosis and skewness of the distribution. The normal distribution is considered to be symmetric and has a zero value for its skewness. In this study, the obtained values of metals and values of soil properties were not normally distributed. Therefore, these values were normalized via logarithmic transformation to normalize the positively skewed distribution of heavy metal concentrations [28]. The Kolmogorov-Smirnov (K-S) test for normality was performed. The findings of this test are presented in Table 8.

The raw data showed positively skewed values for the obtained concentrations of all metals except for Mn. This result suggests that there are various processes such as mineralization and other pollutants that affect the concentration of these metals. The value of the K-S test for normality of Cr, Cu, Fe, Mn, and Pb ($p > 0.05$) indicates that their concentrations can be normalized. However, the values of Pb and Ni did not pass the K-S test for normality. The log-normal transformation was used to minimize data skewness. Fig. 2 illustrates the comparison of the skewed distribution of data before and after the log-transformation of the data.

Sources Identification

A factor analysis (FA) was conducted for the identification of the minimum number of common factors that have the potential to impact the variance of heavy metal concentration in soil. There is no specific rule for the identification of factors and deciding how many factors should be included in FA. However, the common rule is to either keep factors with eigenvalues higher than 1.0 or keep all interpretable factors. The results of probability distribution and log transferred data were then analyzed under factor analysis, and then a varimax (orthogonal) rotation was also applied.

The factor analysis of the metals data set revealed that only the first three factors have eigenvalues higher than 1. The scree-test results also indicated that the first three factors are meaningful for source identification. The findings suggest that the first factor (FA1) explains 45.8% of the total variance. It was highly loaded with Zn (0.94) and Pb (0.94) and moderately loaded by Cu (0.71) and Cr (0.64). Since Pb, Cu, and Zn are identified as traffic markers by several authors, FA1 was assigned as a trafficking factor. The results showed that the second factor (FA2) explains 20.3% of the total variance. It was highly loaded with Ni (0.90) and moderately loaded by Fe (0.73). Moreover, Cr was distributed between FA1 and FA2. Ni has been identified as an important marker of oil combustion; therefore, this factor was assigned to oil refinery emission.

The third factor (FA3) (FA3) was highly loaded with Cd (0.85) and moderately loaded with Mn and Fe. Mn had high loadings on the first and third factors, 0.59 and 0.64, respectively, but was even higher on the third factor. Moreover, Fe (0.46) had comparatively significant loading in FA3. Fe and Mn are well recognized to be linked with industrial processes. The presence of Cd, along

Table 8. Shape statistics and results of the Kolmogorov-Smirnov (K-S) test for the raw and log-normally transformed data

Metal	Raw data			Log-normal transformed data		
	Skewness	Kurtosis	K-S p	Skewness	Kurtosis	K-S p
Cd	2.7	7.5	0.00	1.2	1.6	0.14
Cr	1.2	2.1	0.12	0.05	0.3	0.69
Cu	1.9	5.21	0.12	0.42	0.8	0.74
Fe	0.8	0.4	0.37	-0.15	-0.5	0.97
Mn	0.14	0.9	0.97	-1.8	5.45	0.24
Ni	2.7	6.5	0.00	1.6	1.45	0.00
Pb	0.37	1.6	0.39	-1.3	0.8	0.00
Zn	1.8	3.4	0.04	-0.02	0.5	0.79

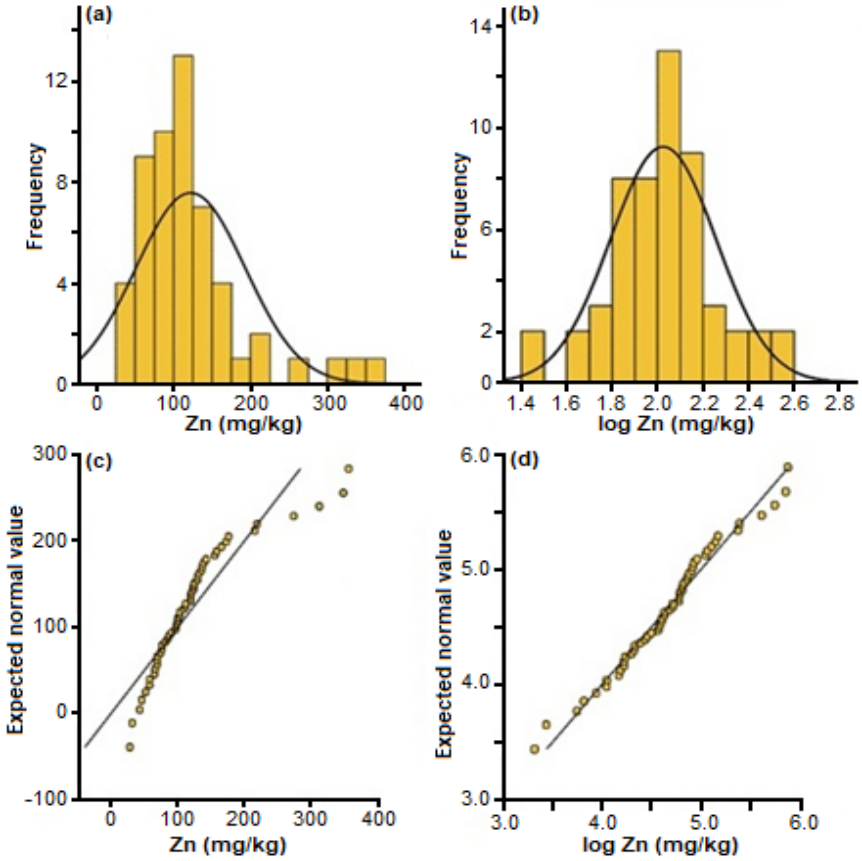

Fig 2. (a) Histogram of Zn concentrations in soil samples showing a skewed distribution, (b) Histogram of the log-transformed Zn concentrations showing normal distribution. (c) Q-Q plot of Zn concentrations showing a deviation from linearity, (d) Q-Q plot of the log-transformed Zn concentrations

Table 9. Correlation matrix of metals

	Cd	Cr	Cu	Fe	Mn	Ni	Pb
Cr	**0.05**						
Cu	0.05	0.53					
Fe	-0.33	0.61	0.29				
Mn	-0.24	0.47	0.35	0.43			
Ni	-0.13	0.42	0.20	0.52	0.11		
Pb	0.20	0.57	0.57	0.25	0.45	0.20	
Zn	0.20	0.57	0.57	0.25	0.45	0.20	1.00

Table 10. Factor loadings from factor analysis after rotation for the maximum variance

Elements	FA1	FA2	FA3	Communalities
Zn	**0.94**			0.90
Pb	**0.94**			0.90
Cu	**0.71**			0.54
Cr	**0.64**	**0.57**		0.74
Ni		**0.90**		0.82
Fe		**0.73**	0.46	0.79
Cd			**0.85**	0.80
Mn	**0.59**		**0.64**	0.76
Variance	45.8%	20.3%	12.1%	78.2%
Total Variance Explained, %				78.2

with these metals, indicates the occurrence of anthropogenic activities. Zarqa city has an industrial zone with four steel smelters [29]. Therefore, FA3 was assigned to mixed anthropogenic factors. The findings of factor analysis are presented in Table 9 and Table 10.

The outcomes of the factor analysis revealed that the chief sources of metals in topsoil include anthropogenic processes, oil burning, and emission from an oil refinery and steel industries.

The Spatial Distribution Analysis

The results obtained in this study present significant matching with earlier documented distribution patterns of Pb and Zn concentrations in Al-Zarqa city. The geochemical map shows that the distribution of the concentrations of these metals was lower than the maximum allowable limits in other countries, including Germany, UK, Japan, and Canada. It was also identified that the distribution of Cu was close to Pb and Zn concentrations. Factor analysis confirmed these results; the first factor was highly loaded by Pb and Zn and moderately loaded by Cu and Cr. The results indicate that these heavy metals are uniformly distributed in the studied areas, and there were no major hotspots present for these elements as shown in Fig. 3. This homogeneous distribution pattern indicates that the sources for their emission are mobile sources. Therefore, automobile and vehicle emission was a major source for these elements.

The geochemical map showed that the studied area was significantly polluted with Cd as its measured concentration crossed the max allowable limits in the UK, Germany, Poland, and Australia (2 mg/kg). Moreover, a major hotspot was also identified in the new Al-Zarqa city with a Cd concentration of around 27.1 mg/kg. The other studied areas were less polluted while the northern outskirts were identified to have the lowest concentration of Cd (< 5.6 mg/kg). The same area was also identified for Cu hotspot as shown in Fig. 4. This distribution pattern indicates that there were Cd and Cu emitting sources present in the area. The topographic map

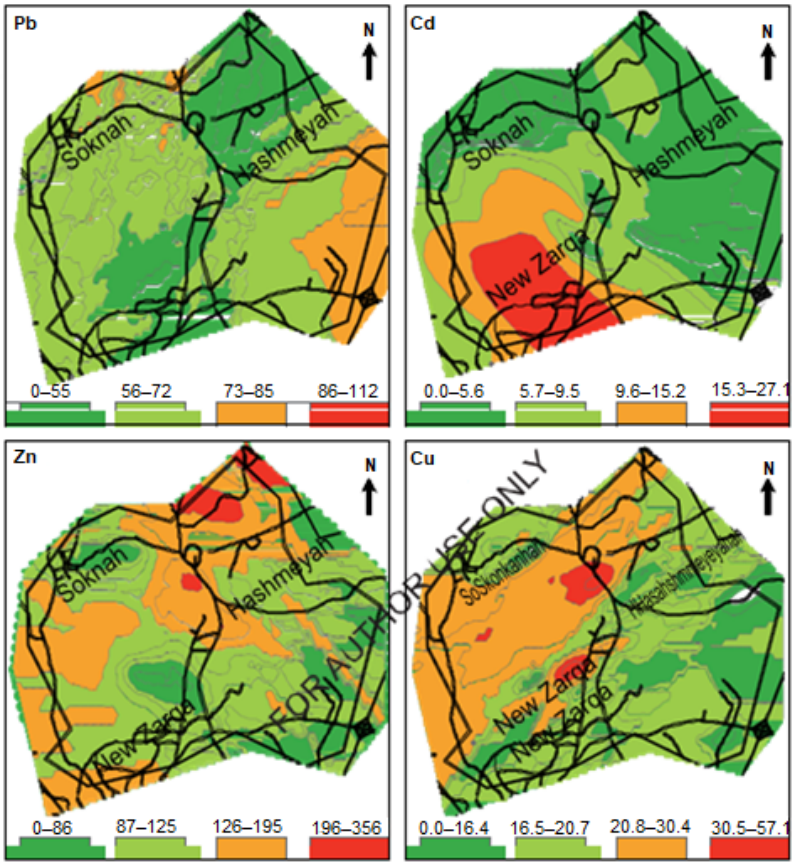

Fig 3. Spatial distribution of metals (Pb, Cd, Zn, Cu)

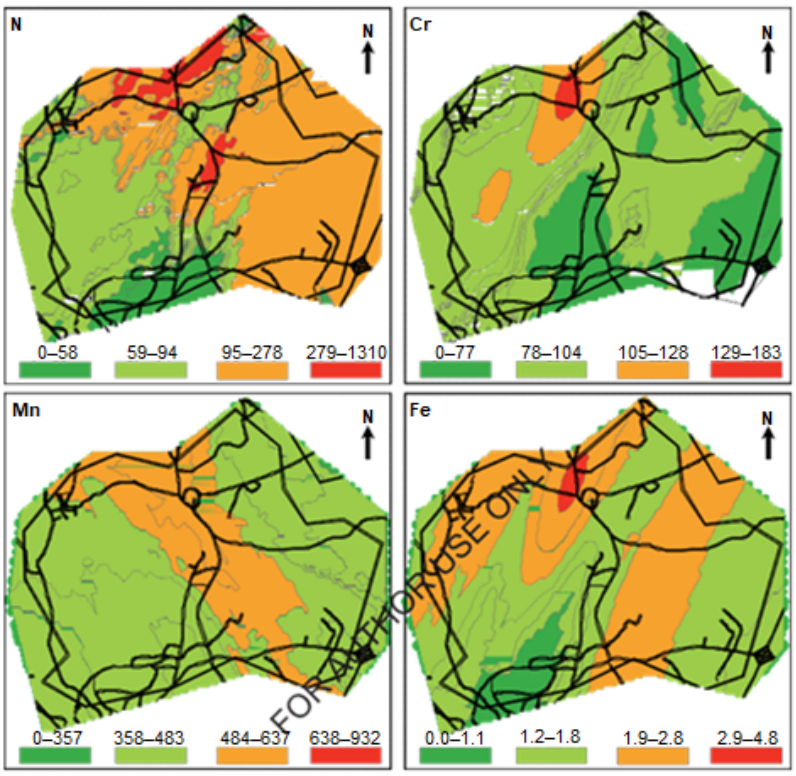

Fig 4. Spatial distribution of metals (Mn, Fe, Cr, Ni)

of this region suggests the presence of several industrial constructions; however, the nature of these industries was not identified.

These findings also revealed that the studied area was considerably polluted with Ni as its measured concentration was more than the maximum allowable limits in Australia, Poland, Canada, Japan, and Germany. The major hotspots for Ni concentration were seen in the east and north of the JPRC. There was no significant information about the distribution of industrial activities available; therefore, any correlation between the hotspot and their sources was difficult to identify. However, it is believed that the burning of oil was the major source of Ni concentration.

The findings also revealed that the studied area was significantly polluted with Cr, and its concentration was identified to be higher than the maximum allowable limits in the UK and Canada. However, the south-eastern area had a lower concentration of Cr, while the North-western area had a higher concentration. More hotspots were found in JPRC nearby locations. The factor analysis further confirmed that JPRC was the key source for the emission of these metals.

The Geochemical maps of Fe and Mn revealed that the greater concentration of these two elements was clustered and not scattered. The widespread crust contribution of these elements attributes to their significantly higher concentrations. One major hotspot for higher concentrations of these metals was identified in the middle of the northern region of the studied area. The existence of human activities and the steel industry in that region was identified as the primary source of these elements. However, to validate this result, more information about these locations and meteorological data is needed.

■ CONCLUSION

Our results indicate that there is a higher degree of heavy metal pollution in Al-Zarqa city. The average values of the measured concentrations of identified metals were as follows; Cd (6.6), Cr (88.2), Cu (21.7), Ni (113.1), Pb (58.9), and Zn (122.0) mg/kg. The major

sources for the contribution of these elements in the studied area include traffic, emission from oil-refineries, and mixed anthropogenic sources. The distribution pattern of these metals and the pollution state in Al-Zarqa city were assessed by comparing the metal's concentrations with the maximum allowable limits (MAL) in different countries. Results showed that the average concentration of Cd was higher than the MAL in Australia, Poland, UK, and Germany. While the average concentration of Cr was higher than the MAL in Canada and the UK. Meanwhile, the average concentration of Cu was lower than the MAL in Australia, Canada, Poland, Japan, UK, and Germany. In addition, the average concentration of Ni was higher than the MAL in Australia, Canada, Poland, Japan, UK, and Germany. While the average concentration of Pb and Zn was lower than the MAL in Australia, UK, Canada, Poland, Japan, and Germany.

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