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Distribution patterns, health hazards, and multivariate assessment of contamination sources of As, Pb, Ni, Zn, and Fe in agricultural soils

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A B S T R A C T

Soil heavy metal contamination is a worldwide environmental concern that presents considerable risks to ecosystems, agricultural progress, and human health. This study aims to evaluate the potential environmental and health hazards linked to the presence of arsenic (As), lead (Pb), nickel (Ni), zinc (Zn), and iron (Fe) in agricultural soil in Al Majma'ah governorate, Saudi Arabia. The contamination factor (CF), pollutant load index (PLI), chronic daily intake (CDI), hazard index (HI), and total lifetime cancer risk (LCR) were calculated for 34 soil samples. The results from the CF and PLI analysis demonstrate that the examined soil has a low contamination factor and is free from heavy metal pollution. The average CDI values for adults and children exhibited the following descending order: Fe > Zn > Cu > Pb > As. The highest HI values observed in adults ranged from 0.0375 (Fe) to 0.00019 (Zn), but in children, the range was from 0.3497 (Fe) to 0.0018 (Zn). The hazard index values for heavy metal(loid)s (HMs) in the Al Majma'ah area were all below 1.0, suggesting that residents in the area are not exposed to a significant non-carcinogenic risk. The LCR values ranged from 8.37E−06 to 7.80E-05 for As in both adults and children, and from 7.50E−08 to 6.98E-07 for Pb. The findings indicated a level of risk that was deemed acceptable or tolerable, without any significant adverse health effects.

1. Introduction

Agricultural soil has a crucial role in preserving food safety and directly affects human health (Agyeman et al., 2021; Alarifi et al., 2023). In the last ten years, there has been a notable increase in industrial activity, rapid urbanization, and population expansion. Consequently, significant amounts of solid and liquid waste have been generated, leading to the dispersion of HMs into various environmental compartments. Consequently, there has been a substantial deterioration in the quality of water and soil, constituting a hazard to both marine organisms and human well-being (El-Sorogy and Al Khathlan, 2024; Alzahrani et al., 2024). Regrettably, the disposal of trash from residential and industrial sources has resulted in the contamination of soil with substantial quantities of hazardous substances, known as HMs. This contamination constitutes a threat to both animals and people (Mishra et al., 2019).

The presence of HMs in soils and crops is frequently influenced by both natural and human-induced influences. Agricultural soils may possess substantial levels of HMs due to natural processes such as rock weathering and volcanic activity. Human activities, particularly land modifications involving sewage sludge, livestock manure, wastewater irrigation, and the use of and fertilizers and insecticides are the principal contributors to the presence of HMs (Alharbi et al., 2024). Agriculture is significantly affected by the use of industrial waste for irrigation, resulting in increased levels of metal pollutants such as cadmium, lead, chromium, and arsenic in crops (Ilyas et al., 2019; Alharbi and El-Sorogy, 2021, 2023).

Heavy metal(loid)s are hazardous contaminants that present a substantial threat to human well-being. Accumulation of these substances in crops can lead to significant problems (Zhang et al., 2018). Children are particularly vulnerable to HMs due to their increased likelihood of exposure through many routes, including the placenta, nursing, hand-to-mouth contact, and higher rates of uptake compared to adults (Rahman et al., 2021). As, Cd, Pb, and Hg can reach the human body by ingestion, inhalation, and dermal absorption. Metal poisoning has well-documented detrimental consequences on the human body, including behavioral illnesses, damage to essential organs like the kidneys, liver, and lungs (Jaishankar et al., 2014).

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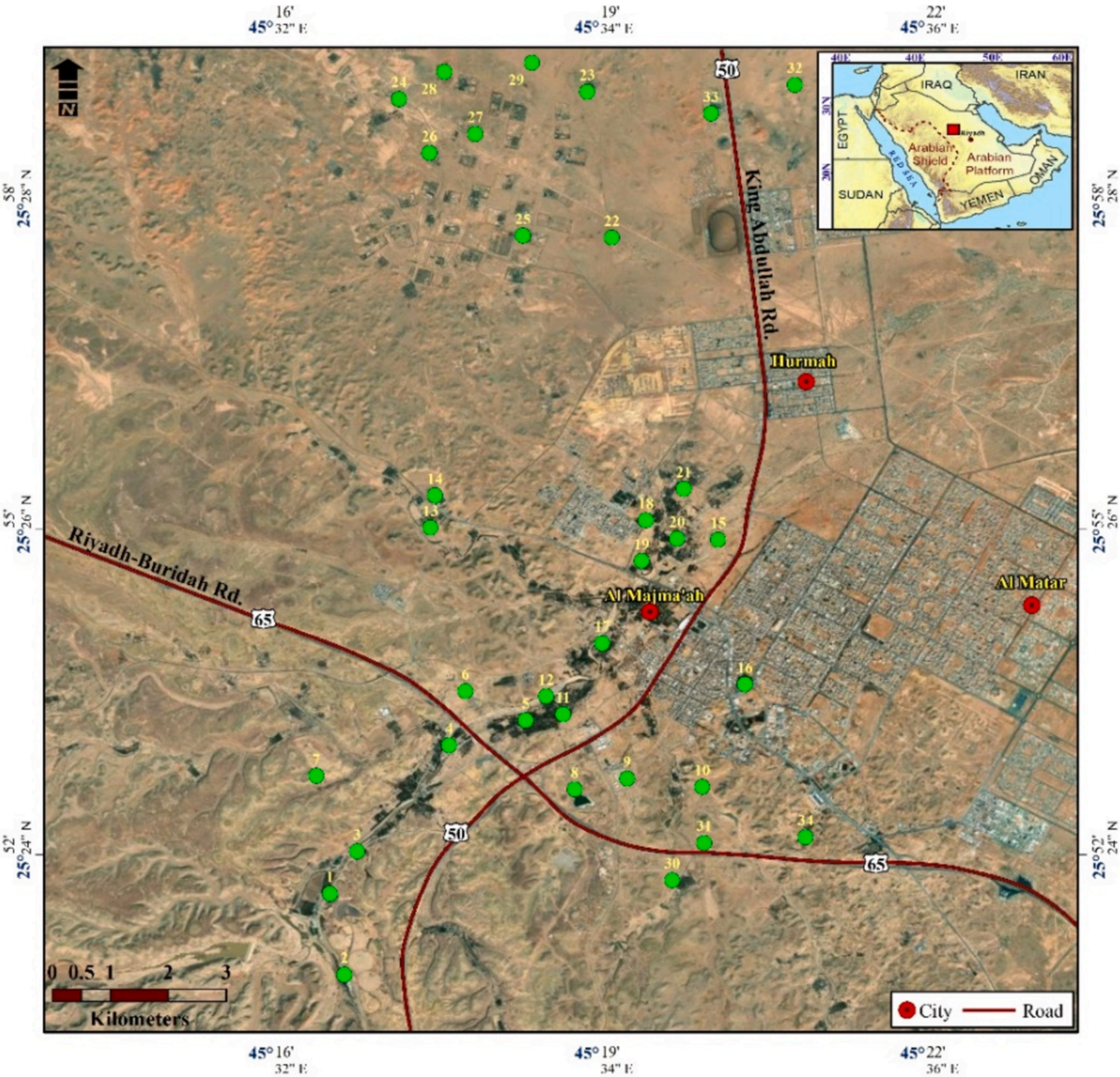

Fig. 1. Location map of the study area and sampling sites (Al-Kahtany, 2024).

Agriculture is a vital activity that holds a pivotal position in guaranteeing food security. The agricultural activities inside the Al Majma'ah governorate including the cultivation of date palms, vegetables, and various crops such as wheat, barley, and corn. Accumulation of HMs in Al Majma'ah soil was attributed to the chemical weathering and erosion of Jurassic to Quaternary sediments in the study area (Al-Kahtany, 2024). This study aimed to: (i) evaluate the environmental hazards linked to the presence of arsenic, lead, nickel, zinc, and iron in the soil of Al Majma'ah area, (ii) calculate the chronic daily intake levels of these HMs by ingestion, skin exposure, and inhalation for both children and adults, and (iii) evaluate the hazard index and total lifetime cancer risk linked to these levels.

2. Materials and methods

2.1. Study area and sampling

Al Majma'ah is situated around 180 km to the northwest of Riyadh, along the Riyadh-Sudair Al-Qassim Highway. The research region is situated within the geographic coordinates of N25°00′061 – N45°19′526 latitude and E26°03′375 – E45°20′116 longitude (Fig. 1). The studied area mostly comprises marine carbonates and siliciclastics of the Oxfordian to Quaternary age (Powers et al., 1966; Youssef and El-Sorogy, 2015; El-Sorogy and Al-Kahtany, 2015; El-Sorogy et al., 2016; Tawfik et al., 2016; Khalifa et al., 2021). The investigation entailed the collection of surface soil samples from 34 palm and citrus plantations. The specimens were collected at a depth of under 10 cm utilizing a rigid plastic hand trowel. At each location, a representative sample was formed by amalgamating three subsamples. Subsequently, the resulting mixture was tightly sealed in plastic bags and stored in a refrigerated

Table 1
Classification of the contamination and health indices (Hakanson,1980; Reimann and de Caritat, 2000; Miletic et al., 2023; USEPA, 2023).

CF	Cf < 1	Low contamination factor
	1 ≤ Cf < 3	Moderate contamination factor
	3 ≤ Cf < 6	Considerable contamination factor
	Cf ≥ 6	Very high contamination factor
PLI	PLI > 1	Polluted
	PLI = 1	Baseline levels of pollution
	PLI < 1	Not polluted
HI	HI < 1	The impact of HMs is insignificant
	HI > 1	HMs may have a harmful effect on health
LCR	LCR < 10^{-6}	No risk of developing carcinogenic diseases
	between 1×10^{-6} and 1×10^{-4}	Acceptable or tolerable carcinogenic risks
	LCR > 10^{-6}	The risk is unacceptable

container.

2.2. Analytical procedures

The soil samples were dried through exposure to the atmosphere, and subsequently meticulously purged of large stones and organic substances. Then, the material was pulverized using an agate mortar and pestle. The analysis of As, Pb, Ni, Zn, and Fe has conducted in the ALS Geochemistry Lab, Jeddah branch in Saudi Arabia, utilizing inductively coupled plasma-atomic emission spectrometry (ICP-AES). Approximately 0.50 g of each sample are subjected to digestion with aqua regia over 45 min within a graphite heating block, at temperatures between 60 and 120 degrees Celsius. The selected HMs are recognized for their susceptibility to environmental and human health hazards (Jaishankar et al., 2014; El-Sorogy and Al Khathlan, 2024).

The limit of detection (LOD) for the ICP-AES technique was established by calculating the concentration that is three times the standard deviation of blank solution measurements divided by the slope of the calibration curves for each element. This validation technique adheres to established methodologies (Papadoyannis and Samanidou, 2004). To guarantee the precision and dependability of the outcomes, many phases of QA/QC were executed throughout the examination of HMs.

2.3. Data analysis

The contamination factor (CF) and pollution load index (PLI) were employed to evaluate the degree of HM contamination in soil samples (Hakanson, 1980; Reimann and de Caritat, 2000; Neeraj et al., 2022). An assessment was carried out to determine the health hazards related to the consumption, inhalation, and skin exposure routes for both adults and children. This was done by calculating the chronic daily intake (CDI) for these three pathways (mg/kg/day). The hazard index (HI) and total lifetime cancer risk (LCR) were computed. Table 1 delineates the classification of the indices employed in this investigation. The equations are employed to assess environmental and health risks (Chonokhuu et al., 2019; Mondal et al., 2021).

$$CF = C_o/C_b LCR = \Sigma CancerRisk = Cancerrisk_{ing} + Cancerrisk_{derm} + Cancerrisk_{inh} \quad (1)$$

$$PLI = (CF_1 \times CF_2 \times CF_3 \times CF_4 \ldots \times CF_n)^{1/n} \quad (2)$$

$$CDI_{ing} = (Csoil \times IngR \times EF \times ED)/(BW \times AT) \times CF \quad (3)$$

$$CDI_{inh} = (Csoil \times InhR \times EF \times ED)/(PEF \times BW \times AT) \quad (4)$$

$$CDI_{derm} = (Csoil \times SA \times AFsoil \times ABS \times EF \times ED)/(BW \times AT) \times CF \quad (5)$$

$$HQ = CDI/RfD \quad (6)$$

$$HI = \Sigma HQ = HQ_{ing} + HQ_{derm} + HQ_{inh} \quad (7)$$

$$Cancerrisk = CDI \times CSF \quad (8)$$

$$LCR = \Sigma CancerRisk = Cancerrisk_{ing} + Cancerrisk_{derm} + Cancerrisk_{inh} \quad (9)$$

Co denotes the concentration of HM, whereas Cb is the typical background level of the HM. HQ refers to the hazard quotient, utilized to evaluate the potential dangers associated with exposure to specific compounds. Table S.1 delineates the exposure factors utilized to compute the chronic daily intake (CDI) for non-carcinogenic risk (IRIS, 2020; USEPA, 1989; Miletic et al., 2023). The Environmental Protection Agency (EPA) sets the reference dose (RfD) values for all examined HMs, specifically for ingestion (USEPA, 2023). Table S.2 is incomplete for Fe since there are no RfDinh and RfDderm values available for Fe, which can be ascribed to either discrepancy in the published data or the absence of reliable documentation linking back to the original study for the reference value (Miletic et al., 2023).

3. Results and Discussion

3.1. Distribution patterns and environmental hazards

The average concentrations of HMs in mg/kg of dry weight are arranged in descending order as follows (Table S. 3): Fe (19108), Zn (41.25), Ni (31.11), Pb (6.47), and As (4.07). Fig. 2 and Table S.3 demonstrate that the greatest HM values were recorded in S2 (Fe and As), S9 (Zn), S10 (As and Ni), and S11 (Pb). The minimal concentrations of HMs were seen in S1 (Ni), S6 (As and Pb), and S29 (Fe and Zn). The samples obtained from farms in mountainous regions exhibited elevated concentrations of Fe, particularly S2 and S3, whereas those collected from farms next to residential zones displayed greater levels of Pb, As, Zn, and Ni, namely S9 to S12, S14, and S18.

Several HMs have a vital role in essential nutritional activities, requiring little quantities, such as iron, nickel, and zinc (Häder et al., 2021). However, an excessive quantity of these HMs can pose severe health complications such as diabetes, renal and neurological diseases, and cardiovascular disorders (Neal and Guilarte, 2013; Abbaspour et al., 2014). The contamination factor (CF) serves as a metric for assessing the extent of contamination (Hakanson, 1980). The findings of the CF study indicated that all HMs found in the soil being studied had a contamination factor that was low, with average values below 1 (Table S. 4). The pollutant load index (PLI) was utilized to assess the overall contamination level of HMs at the sampling locations (Tomlinson et al., 1980; Chon et al., 1996). The study area exhibited PLI values between 0.10 and 0.52, with a mean of 0.28 (Table S. 4), suggesting that the soil is unpolluted.

Table 2 displays a correlation matrix that reveals a substantial positive connection among all examined HM pairs, suggesting analogous geochemical behavior for these heavy metals (Alharbi et al., 2024). Since the CF and PLI were below the contamination threshold, the HMs in the investigated soil might be derived from natural sources, primarily due to the chemical weathering of the surrounding Jurassic to recent sediments (Al-Kahtany, 2024). One principal component is derived from the principal component analysis (Table 3), exhibiting loadings for all HMs, hence supporting the correlation matrix of the geogenic source for the examined HMs. The sedimentary rocks of central Saudi Arabia host a variety of important metallic minerals, including the investigated HMs, like arsenopyrite (FeAsS), sphalerite (ZnS) associated with carbonate-hosted deposits, hematite (Fe_2O_3) and magnetite (Fe_3O_4) (Al-Ateeq et al., 2014; Liu et al., 2021).

3.2. Health risk assessment

3.2.1. Chronic daily intake (CDI)

The average CDI values for the ingestion pathway in adults are listed

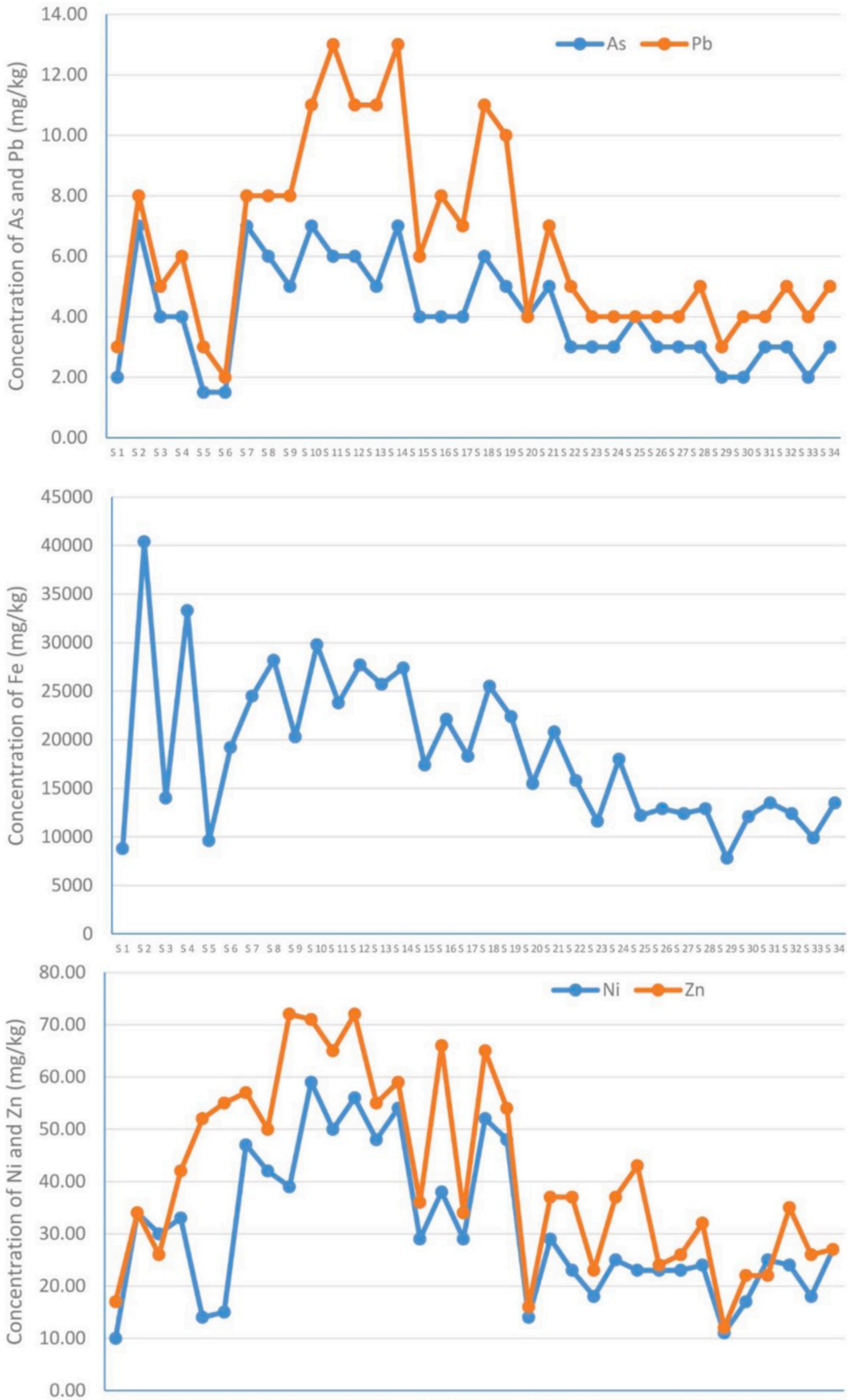

Fig. 2. Distribution of heavy metals throughout the study area.

Table 2
Correlation matrix for HMs in the investigated agricultural soils.

	As	Fe	Ni	Pb	Zn
As	1				
Fe	0.824**	1			
Ni	0.884**	0.758**	1		
Pb	0.868**	0.729**	0.949**	1	
Zn	0.625**	0.618**	0.803**	0.746**	1

**Correlation is significant at the 0.01 level (2-tailed).

Table 3
Loading matrix of the principal component and its explained total variance.

	Component PC1
As	0.928
Fe	0.863
Ni	0.970
Pb	0.949
Zn	0.829
% of Variance	82.70
Cumulative %	82. 70

in descending order as follows: Fe > Zn > Ni > Pb > As (Fig. 3, Table 4). To be more exact, the CDI values range from 5.575E to 06 (As) to 0.026 (Fe). The average concentrations of CDIing values in children exhibit a similar decreasing trend as observed in adults, with iron having the highest concentration, followed by zinc, nickel, lead, and arsenic. The CDI values vary from 5.20E to 05 (As) to 0.244 (Fe). Children who come into contact with soil through different means may have an increased concentration of CDI. This is because children are more vulnerable to exposure and have a greater ability to absorb HMs while engaging in outdoor play activities in soil, compared to adults (Häder et al., 2021).

3.2.2. Hazard quotient (HQ) and hazard index (HI)

The hierarchy of HQ values for adults and children is as follows: HQing > HQder > HQinh. The hierarchical values for the three pathways in children demonstrate a downward order of Fe > As > Pb > Ni > Zn. The HQ values for ingestion, dermal contact, and inhalation pathways range from 0.349 (Fe) to 0.0018 (Zn), 0.00070 (Fe) to 3.51E-06 (Zn), and 2.57E-06 (Fe) to 1.29E-08 (Zn), respectively. In adults, the HQ decreases in the same sequence as observed in children. The values for ingestion range from 0.037 (Fe) to 0.00019 (Zn), for dermal contact they range from 0.00015 (Fe) to 7.52E-07 (Zn), and for inhalation they range from 5.50E to 07 (Fe) to 2.77E-09 (Zn). The primary factor that influenced the HQ values was the method of intake, which accounted for 99.8 % in children and 99.6 % in adults.

The adults had HI values ranging from 0.0375 (Fe) to 0.00019 (Zn), whereas the children had values ranging from 0.3497 (Fe) to 0.0018 (Zn). The HI for HMs was notably higher in children compared to adults (Table S. 5). The findings suggested that the primary route of human exposure to HMs in the study region was through their consumption. The HI values for HMs in the research area were all less than 1.0. These findings indicate that individuals living in the Al Majma'ah region are not encountering any significant non-carcinogenic consequences (Bello et al., 2019; Tianv et al., 2020).

It is noteworthy that the HI for iron surpassed 0.2 in children individuals, underscoring the importance of safeguarding their health. Children, who are particularly vulnerable to the health impacts, are

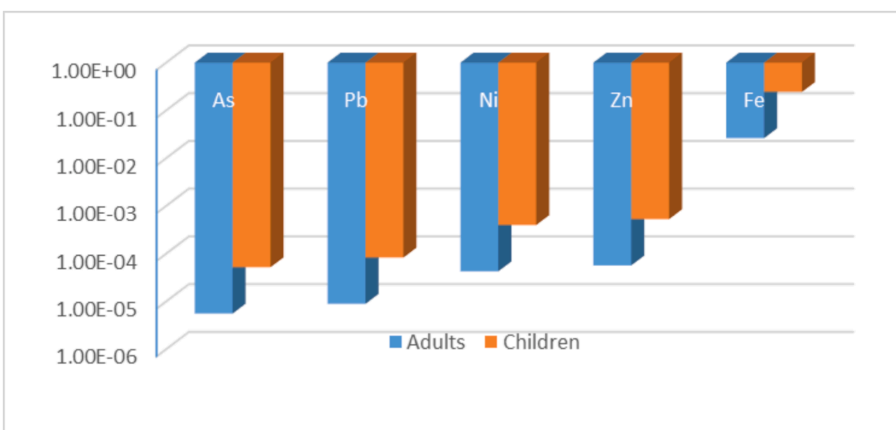

Fig. 3. Average CDI_{ing} values for adults and children in Al Majma'ah soil.

Table 4
The CDI, HQ and HI values for the non-carcinogenic risk in both adults and children.

HMs	**Adults**						
	$CDI_{Ing.}$	CDI_{Dermal}	$CDI_{Inhal.}$	$HQ_{Ing.}$	HQ_{Demal}	$HQ_{Inhal.}$	HI
As	5.57458E-06	2.22426E-08	8.19791E-11	0.018581938	7.41419E-05	2.73264E-07	0.018656353
Pb	8.86606E-06	3.53756E-08	1.30383E-10	0.002533159	1.01073E-05	3.72523E-08	0.002543304
Ni	4.2618E-05	1.70046E-07	6.26735E-10	0.002005386	8.50228E-06	2.9491E-08	0.002014267
Zn	5.65068E-05	2.25462E-07	8.30983E-10	0.000188356	7.51541E-07	2.76994E-09	0.00018911
Fe	0.026175799	–	–	0.037393999	0.000149202	5.49912E-07	0.037543751
HMs	**Children**						
	$CDI_{Ing.}$	CDI_{Dermal}	$CDI_{Inhal.}$	$HQ_{Ing.}$	HQ_{Demal}	$HQ_{Inhal.}$	Hi
As	5.20294E-05	1.03799E-07	3.82569E-10	0.173431422	0.000345996	1.27523E-06	0.173778693
Pb	8.27499E-05	1.65086E-07	6.08455E-10	0.023642821	4.71674E-05	1.73844E-07	0.023690162
Ni	0.000397768	7.93546E-07	2.92476E-09	0.019888382	3.96773E-05	1.46238E-07	0.019928205
Zn	0.000527397	1.05216E-06	3.87792E-09	0.001757991	3.50719E-06	1.29264E-08	0.001761511
Fe	0.244307458	–	–	0.349010654	0.000696276	2.56625E-06	0.349709497

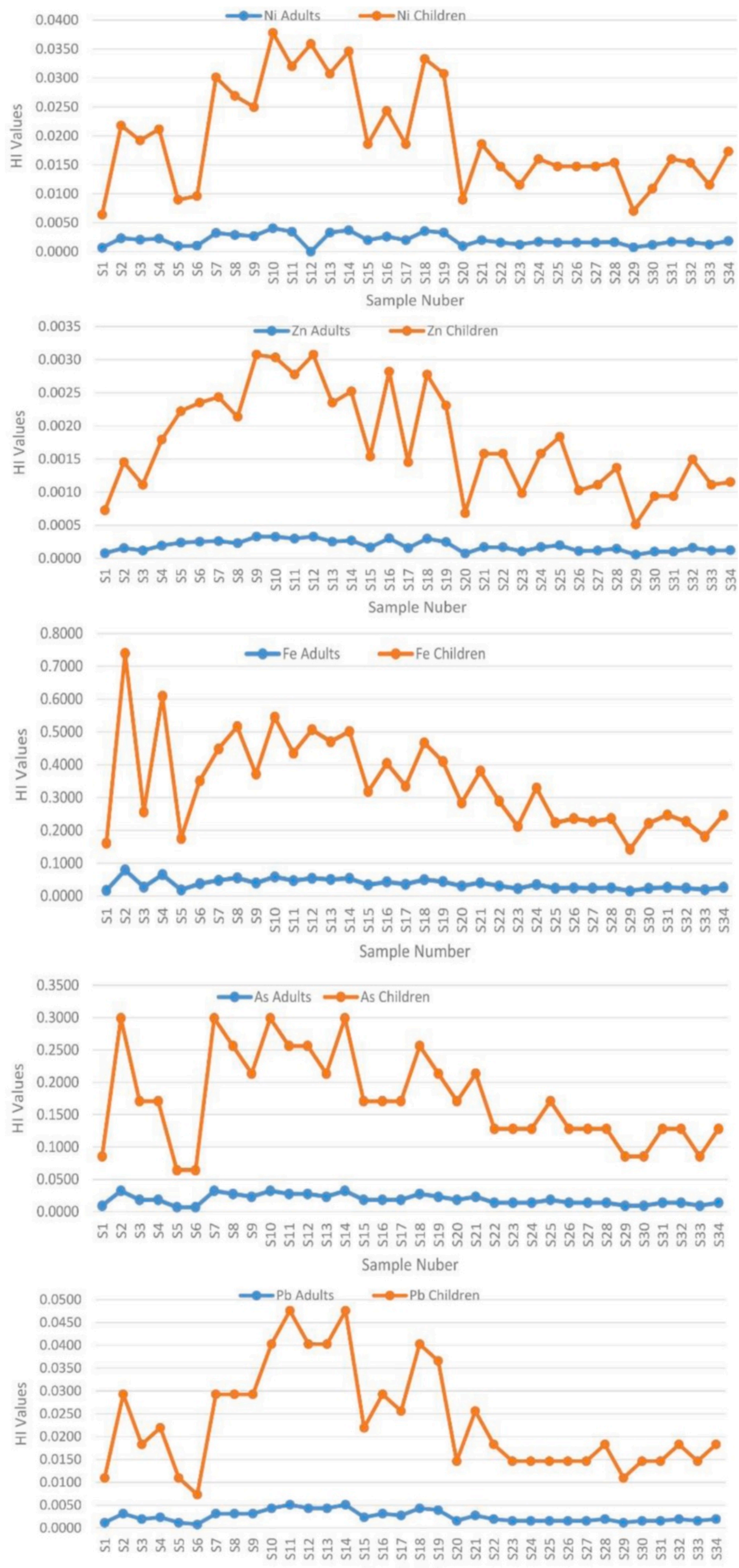

Fig. 4. Distribution of hazard index (HI) of heavy metal(loid)s per sampled location.

Table 5
Average CRs and LCR for HMs in the study area.

HMs	Adults			
	CR Ing.	CR Dermal	CR Inhal	LCR
As	8.34005E-06	3.32768E-08	1.22648E-10	8.37345E-06
Pb	7.46575E-08	–	1.0979E-12	7.47E-08
	Children			
As	7.78405E-05	1.55292E-07	5.72356E-10	7.79963E-05
Pb	6.96804E-07	–	5.12356E-12	6.97E-07

highly susceptible to HMs, potentially due to their oral and manual behaviors (Agyeman et al., 2021). The spatial distribution of the HI for HMs throughout the sample locations exhibited consistent areas of high concentration for both children and adults. The hotspots were specifically identified in the following locations: S9, S10, and S12 for zinc; S10, S12, and S14 for nickel; S2 and S4 for iron; S2, S7, S10, and S14 for arsenic; and S11 and S14 for lead (Fig. 4). Sample 2, taken from mountainous locations, showed higher HI values for Fe. Conversely, samples 9–12, 14, and 18, which were collected from regions adjacent to residential areas, had higher HI values for various HMs.

3.2.3. Carcinogenic risks (CRs) and total lifetime cancer risk (LCR)

The accumulation of HMs in the human body might result in detrimental health repercussions. Exposure to HMs throughout childhood has been linked to several health issues, including impaired respiratory function, cardiovascular disease, reproductive toxicity, cognitive impairments, and bone degradation (Wang et al., 2015; Madrigal et al., 2018; Agyeman et al., 2021). The study demonstrated that the cancer risks (CRs) linked to arsenic and lead in children were markedly greater than those in adults (Table 5 and Fig. 5). The average CR values for adults in the pathways of ingestion, dermal contact, and inhalation ranged from 7.46575E to 08 (Pb) to 8.34005E-06 (As), from 2.97884E to 10 (Pb) to 3.32768E-08 (As), and from 1.0979E to 12 (Pb) to 1.22648E-10 (As), respectively. The children's CRs varied between 6.97E and 07 (Pb) and 7.78E-05 (As), between 1.39E and 09 (Pb) and 1.55E-07 (As), and between 5.124E and 12 (Pb) and 5.72E-10 (As), respectively.

The lifetime cancer risk (LCR) values for arsenic and lead consistently showed higher levels in children compared to adults at all the tested sites (Table S. 6). The findings suggest that children individuals are more vulnerable to being exposed to hazardous materials, possibly due to their behavioral tendencies (Ayeman et al., 2021). The LCR values showed differences between adults and children, ranging from 8.37E to 06 to 7.80E-05 for As, and from 7.47E to 08 to 6.97E-07 for Pb (Table 5). The primary determinant of the LCR was the method of consumption, which accounted for 99.60 % in adults and 99.80 % in children across all two HMs.

The regional distribution of LCR for arsenic and lead across sampling locations displayed analogous patterns in both children and adults, with elevated values observed in children (Fig. 5). The LCR values for As and Pb were determined to be between 1×10^{-4} and 1×10^{-6}, or maybe below 1×10^{-6}. This implies no to acceptable risk of developing carcinogenic diseases (El-Sorogy and Al Khathlan, 2024; Alarifi et al., 2023).

4. Conclusions

1. The findings from the CF and PLI analyses indicated that the agricultural areas in the Al Majma'ah region exhibits a low contamination factor and is free from HM pollution. 2. The CDI values for adults through ingestion, dermal contact, and inhalation pathways varied between 0.0262 (Fe) and 5.57E-06 (As), 0.00010 (Fe) and 2.22E-08 (As), and 3.85E-07 (Fe) and 8.20E-11 (As), respectively. The children's CDI values ranged from 0.244 (Fe) to 5.20E-05 (As) for ingestion, 0.00049 (Fe) to 1.04E-07 (As) for dermal contact, and 1.80E-06 (Fe) to 3.83E-10 (As) for inhalation.

3. The HI values for HMs in the research area consistently remained below 1.0, indicating that residents in the Al Majma'ah region are not at a significantly non-carcinogenic risk. However, the HI value for Iron (Fe) exceeded 0.2 especially for children.

4. The LCR findings ranged from 1×10^{-4} to 1×10^{-6}, or were below 1×10^{-6}. This suggests that the analyzed samples do not present health risks to individuals.

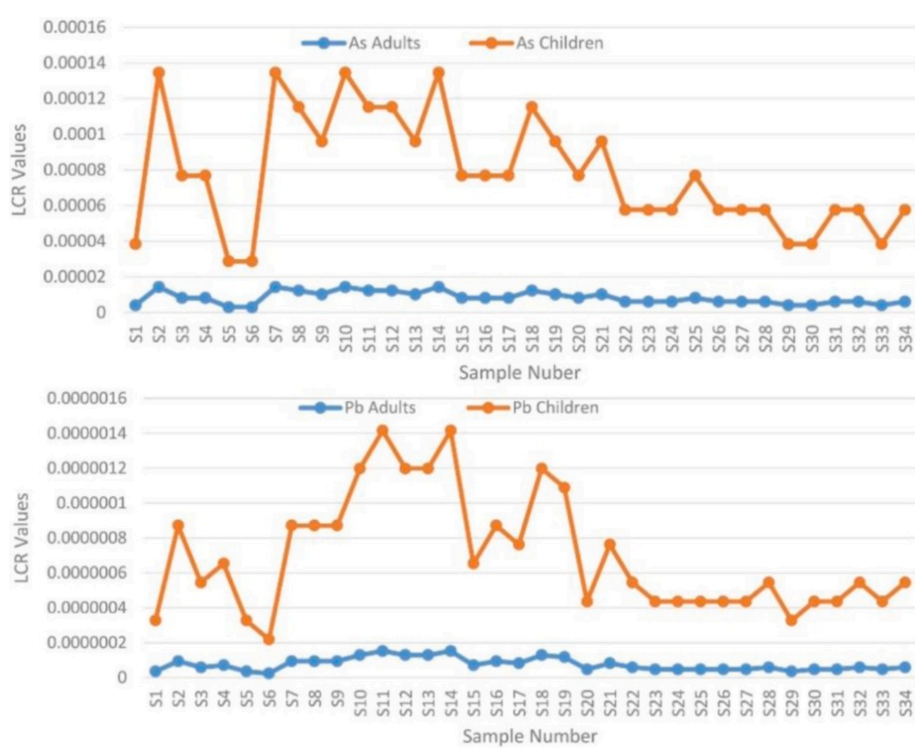

Fig. 5. Distribution of LCR for As and Pb per sampled location.

CRediT authorship contribution statement

Abdelbaset El-Sorogy: Writing – review & editing, Writing – original draft, Project administration, Methodology, Investigation. **Khaled Al-kahtany:** Writing – review & editing, Writing – original draft, Software, Funding acquisition. **Talal Alharbi:** Writing – review & editing, Writing – original draft, Software, Methodology. **Saad S. Alarifi:** Writing – review & editing, Writing – original draft, Methodology.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Ethical approval: The present study does not use or harm any animals and followed all the scientific ethics.

Appendix A. Supplementary material

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jksus.2024.103489.

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