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ORIGINAL PAPER

Health risk assessment associated to heavy metal pollution levels in Mediterranean environment soils: a case study in the watershed of Sebkhet Ariana, Tunisia

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Abstract

We measured the concentrations of 9 heavy metals (Fe, Cd, Hg, Pb, Zn, Co, Cr, Cu, Ni) in the soils collected in the watershed of Sebkhet Ariana (Tunisia) and assessed health risks for residential adults and children. Also, we assessed their potential sources, contamination status, and ecological risks using pollution indicators such as the enrichment factor (EF), geoaccumulation index (I_{geo}), contamination factor (CF), pollution load index (PLI), ecological risk (RI), hazard index (HI), and carcinogenic risk index (CRI) to both children and adults. Heavy metal concentrations followed the order Fe > Zn > Pb > Cr > Ni > Cu > Co > Cd > Hg. Geoaccumulation index, contamination factor, and enrichment factor results indicated that watershed of Sebkhet Ariana was polluted with Cd, Hg, and Pb due to use of pesticides and fertilizers and industrial wastewater reuse. Also, the study region had "high potential ecological risk" for Cd, whereas "low potential ecological risk" for the other heavy metals. Factor and hierarchical cluster analyses revealed that Ni, Hg, and Cu were from anthropogenic sources; Cd, Cr, and Co from both anthropogenic and natural sources; while other heavy metals from natural sources. The hazard index and the carcinogenic risk of HMs in adults' group revealed an acceptable level; however, children's group faced a great chance of carcinogenic risk by Cr and Ni moreover non-carcinogenic risk due to high level of Co.

Keywords Heavy metals · Health risk assessment · Soil contamination · Pollution indices · Statistical analysis · Anthropogenic factors

Introduction

Heavy metal pollution in soil has been widely recognized as a serious environmental problem in the recent decades due to the rapid rise in urbanization and industrialization (Rodríguez Martín et al. 2015; Chabbi et al. 2020; Rodríguez Martín and Nanos 2016). Although naturally present in soils, excessive amounts of heavy metals in the soil environment resulting from human activities such as mining, smelting, electroplating, and other industrial activities, traffic, automobile exhausts, domestic waste pollution, and pesticides and fertilization in urban and agricultural soil may lead to a decline in soil quality and ultimately to the ecological safety of the affected areas via long-term, even at low concentration (Nanos et al. 2015; Jin et al. 2019; Chenghui et al. 2020; Kehuiet al. 2020; Sakizadeh and Rodríguez Martín 2021). Due to their contaminant effect, heavy metals have lately been the subject of many pieces of research (Ramos-Miras et al. 2014; Odumo et al. 2018; Chikaodili et al. 2020; Rastmanesh et al. 2020; Jawad et al. 2020; Varol et al. 2020; Tokatli and Ustaoglu 2020). Heavy metals in the soil are responsible for different diseases such as the human circulatory system and damage to the central nervous system, cancers, anemia, and gastrointestinal disorders caused by chronic and excessive exposure (Huang et al. 2014; Karimi et al. 2020). Understanding the risk

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assessment, many studies have been conducted on the human health risk assessment based on inhalation, ingestion, and dermal exposure (Baltas et al. 2020; Mirzaei et al. 2020; Varol 2020). Recently, various techniques are being investigated for the identification and apportioning of the potential sources of pollution, and numbers of pollution indices have been widely utilized for synergist effects and health risk assessment of heavy metals and developing pollution prevention strategies (Mazurek et al. 2019; Varol et al. 2020). Potential ecological risk index (RI), geoaccumulation index (I_{geo}), contamination factor (CF), pollution load index (PLI), and enrichment factor (EF) are among the most important and effective soil pollution and quality risk assessment indices (Aydi 2015; Varol et al 2020; Haghnazar et al. 2021; Tokatlı and Varol 2021; Mengjiao et al. 2021). Therefore, assessing non-carcinogenic and carcinogenic risks is a common method for investigating the effects of heavy metals on human health (Saleem et al. 2019; Adimalla 2020; Turhun and Eziz 2022).

The watershed of Sebkhet Ariana is an important coastal urban zone located on an alluvial plain in the capital of Tunisia, the most developed economic district in the country. Ariana region is the most important industrial and agricultural region of northeast Tunisia. In recent years, the soil quality in the region has declined dramatically owing to the anthropogenic activities such as urban activities and the use of fertilizers and pesticides (Aydi et al. 2013). In addition, industrial wastewaters are discharged into the irrigation canals in some regions (Mahmoudi et al. 2021). This may affect the quality of soils and has serious implications for human health. However, no heavy metal contamination and their health risk assessment studies emphasizing the soil pollution in the watershed of Sebkhet Ariana have been conducted up to the present time. This was the main motivation behind this research. Therefore, it is necessary to conduct a comprehensive study for assessing human health risks as well as environmental and ecological risks from heavy metals in the soils. In this regard, the goals of this research included (1) determination of certain heavy metals' (Fe, Cd, Co, Ni, Hg, Cr, Cu, Zn, and Pb) levels in the soil of the watershed of Sebkhet Ariana; (2) calculation of the degree of heavy metal pollution based on the relevant indices such as geoaccumulation indices (I_{geo}), enrichment factor (EF), contamination factor (CF), and pollution load index (PLI); (3) recognizing the main origins of pollution using multivariate statistical techniques; and (4) evaluating the ecological risk and potential human health hazards by means of the potential ecological risk formula as well as assessing the hazard quotient (HQ), hazard index (HI), and carcinogenic risk index (CRI) for the children and adults.

Materials and methods

The study area and soil sampling

The study area is located in the Northeast of Tunisia, the Sebkhet Ariana watershed, the subject of this study, occupying a strategic position at the regional and national levels. It is located on the eastern coast of the country in the governorates of Ariana and Tunis and north of the capital Tunis (Fig. 1). It is limited to the North and East by the Mediterranean, to the South by Lake of Tunis, and to the West by Jbel Naheli. The entire area covers about 140 km^2. The topographical context of the study area shows three types of reliefs. The hills delineate the watersheds of the North, West, and South coasts; the plain area occupies the central part and the East coastline.

Land use types of the Sebkhet Ariana watershed are agricultural, residential, industrial, and forest land. The climate in the study area is strongly affected by its positions on the southern side of the Mediterranean Sea and on the northern edge of Africa. The annual precipitation is about 460 mm and the annual temperature average is 19 °C (Aydi et al. 2013). The overall climate is of Mediterranean type.

For the purpose of the present investigation, soil samples were taken from nine stations (covering the Sebkhet Ariana watershed) at a depth of 0–10 cm from the top surface using a stainless steel grab. The samples were stored in nylon bags and brought to the laboratory for the determination of heavy metals.

Laboratory analysis

Soil samples were air-dried and sieved with a 2 mm grid sieve. The pH and the electrical conductivity (EC) of the sampled soils were measured by shaking an aliquot of soils in distilled water (10 g of dry soil in 25 mL of water) for 10 min. The suspension was left to stand for 10 min. The pH and the EC of the supernatant were measured using a pH meter (WTW Windaus pH 538 with combined electrode) and a multi-parameter conduct meter (WTW Windaus LF 538) for leachate samples.

To determine heavy metal content, the soil samples were air-dried, sieved to < 2 mm, and crushed manually in an agate mortar. In the powdered soil samples, the contents of nine heavy metals (Fe, Cd, Cr, Cu, Co, Ni, Pb, Zn, and Hg) were determined after a strong acid mineralization method using a mixture of 2 mL of HNO_3, 5 mL of $HClO_4$, and 20 mL of HF. The sample was then heated on a hot plate at 125° C to dry. Finally, it was transferred into a flask and diluted to 50 mL with 5 mL of HCl.

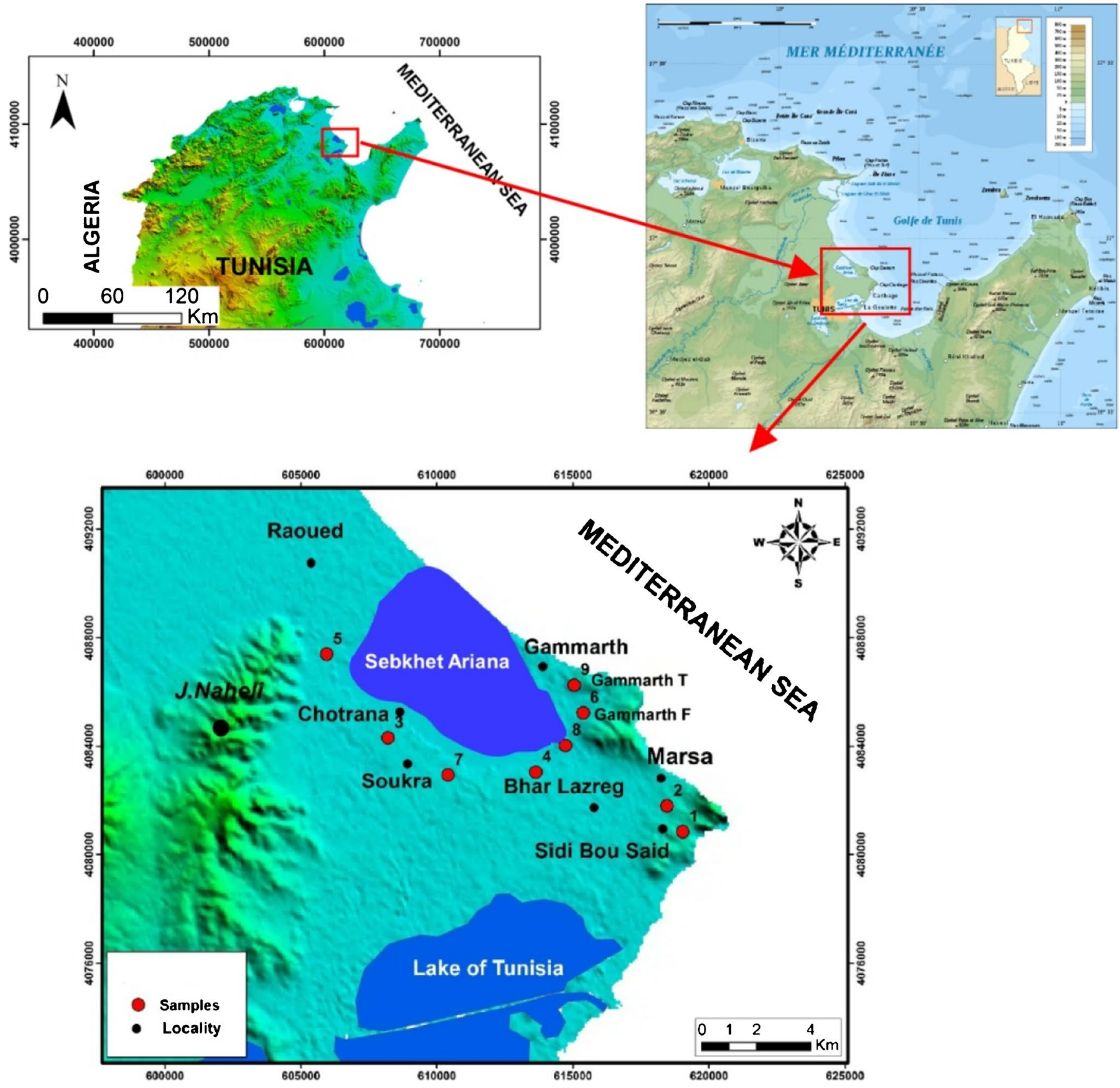

Fig. 1 Localization of the study area and sampling sites

Then, analyses were carried out using an inductively coupled plasma atomic emission spectrometer Ultima C (JobinYvon) at the chemistry laboratory of the National Office of Mines in Tunis. The operating conditions employed for ICP-AES determination were 1000 W RF power, 13 L/ min plasma flow, 2 L/min sheath gas flow, 0.02 L/ min nebulizer flow, and 1.5 mL/min sample uptake rate.

The limits of detection for examined trace metals were 0.05 mg/kg for Cd, 0.1 mg/ kg for Cu, and 0.5 mg/ kg for Fe, Cr, Ni, Co, Pb, Zn, and Hg. Duplicates were performed for each sample. The quality of the analytical procedure for the total heavy metal concentrations was

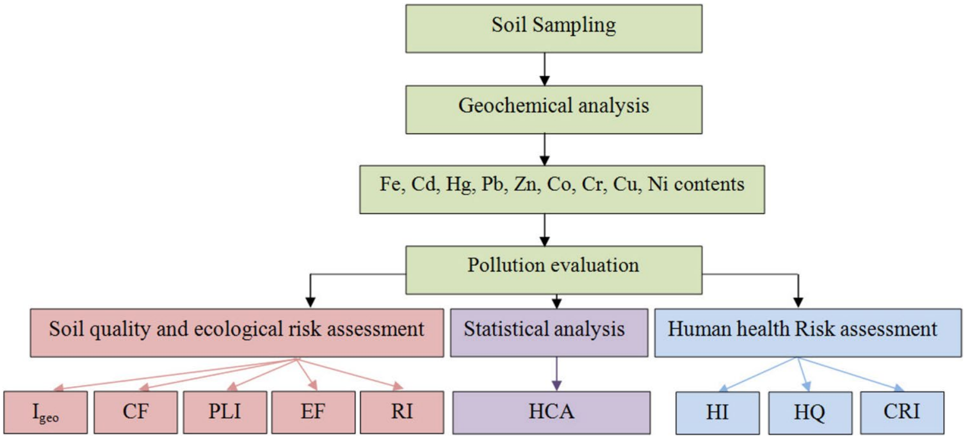

Fig. 2 Flowchart of the adopted methodology

checked by analyzing the stream soil reference samples (from the National Office of Mines, Tunisia) and nine replicate samples for which the relative standard deviations (%RSDs) were less than 10% for the heavy metals.

Assessment of soil quality

This methodology comprises the following main steps (Fig. 2):

1. Determine the levels of 9 heavy metals (Fe, Cd, Hg, Pb, Zn, Co, Cr, Cu, Ni) in the soil samples collected in the watershed of Sebkhet Ariana (Tunisia)
2. Recognize possible sources of heavy metals using statistical analyses
3. Evaluate contamination using pollution indices and ecological risk formulas such as contamination factor (CF), pollution load index (PLI), geoaccumulation index (I_{geo}), enrichment factor (EF), and potential ecological risk index (RI)
4. Evaluate potential human health hazards using the hazard quotient (HQ), hazard index (HI), and carcinogenic risk index (CRI) for the children and adults.

Contamination factor

The contamination factor (CF) is used to express the level of contamination (Aydi 2015; Ghannem et al. 2014).

In the version originally suggested by Hakanson (1980), the assessment of contamination was conducted through a reference of the elemental concentrations to preindustrial levels (Hakanson 1980).

$$\mathrm{CF_{metal}} = \frac{\mathrm{C_n}}{\mathrm{B_n}} \quad (1)$$

This parameter is expressed as follows:

where C_n = metal concentration in the soil sample; B_n = background value of that metal.

The following criteria are used to describe the values of the contamination factor: $\mathrm{CF_{metal}} < 1$, low contamination factor; $1 \le \mathrm{CF_{metal}} < 3$, moderate contamination factors; $3 \le \mathrm{CF_{metal}} < 6$, considerable contamination factors; and $\mathrm{CF_{metal}} \ge 6$, very high contamination factor (Ghannem et al. 2014). To calculate contamination indices, continental crustal and shale concentrations (Turekian and Wedepohl 1961) were chosen as the geochemical background for different heavy metals.

Pollution load index (PLI)

The pollution load index (PLI) was developed by Tomlinson et al. (1980) to evaluate the level of heavy metal pollution (Aydi 2015; Zarei et al. 2014) and it permits a comparison of pollution levels between sites and at different times.

The PLI is expressed as:

$$\mathrm{PLI} = \sqrt[n]{\mathrm{CF_2} \times \mathrm{CF_2} \times \mathrm{CF_3}} \quad (2)$$

where CF is the contamination factor; n is the number of metals.

The pollution load index can be classified as no pollution ($\mathrm{PLI} < 1$), moderate pollution ($1 < \mathrm{PLI} < 2$), heavy pollution ($2 < \mathrm{PLI} < 3$), and extremely heavy pollution ($3 > \mathrm{PLI}$).

Enrichment factor (EF)

EF is a useful tool to assess the degree of contamination (Chebbi et al. 2020; Debo et al. 2015) and to differentiate between anthropogenic and natural sources of heavy metal elements.

The EF is calculated according to the following equation:

$$EF = \frac{\left(C_n/C_{Fe}\right)_{sample}}{\left(C_n/C_{Fe}\right)_{background}} \quad (3)$$

where (C_n/C_{Fe}) sample is the heavy metal to immobile element ratio in the samples of interest and (C_n/C_{Fe}) background is the heavy metal to immobile element ratio in the selected reference sample.

In the present study, the continental crustal value of Fe was chosen as the background value.

Six categories are generally recognized: $EF \leq 1$ indicates background concentration; $1 < EF < 2$ indicates depletion to minimal enrichment; $2 < EF < 5$ is moderate enrichment; $5 < EF < 20$ is signification enrichment; $20 < EF < 40$ is very high enrichment; $EF > 40$ indicates extremely high.

Potential ecological risk index (RI)

It was used in order to assess the degree of environmental risk (Rastmanesh et al. 2020; Ramos-Miras et al. 2020) caused by a concentration of heavy metals in water and in air as well as in soil.

RI was obtained by the equation:

$RI = \sum Er^{\,i}$.

$Eri = Pi \times Tf^{\,i}$

$$P_i = \frac{C}{B} \quad (4)$$

P_i is the single pollution index of heavy metal using background data.

T_f^i is the standardized response coefficient for the toxicity of a single heavy metal.

If $RI < 150$, the ecological risk index is low; if $150 < RI < 300$, it is a moderate ecological risk; if $300 < RI < 600$, it is a high ecological risk; and if $RI > 600$, the ecological risk is very high.

Health risk assessment of heavy metals in soil

Heavy metals can harm human health through chronic accumulation in the human body via food intake. According to the different mechanisms by which heavy metal elements harm human health, they can be divided into non-carcinogens and carcinogens. In this survey, based on the recommendation of the US Environmental and Protection Agency US Epa (1989) was used to calculate the Non-carcinogenic risk by various pathways. These pathways are ingestion, inhalation, and dermal contact (Rastmanesh et al. 2020) using Eqs. (5–7).

Here, the ADI is the average daily intake of metal through consumption of contaminated soil and RFD is the reference dose.

$$\text{ADI ingestion} = \frac{C\text{ soil} \times \text{IngR} \times \text{ED} \times \text{EF}}{\text{BW} \times \text{AT}} \times 10^{-6} \quad (5)$$

$$\text{ADI inhalation} = \frac{C\text{soil} \times \text{InhR} \times \text{EF} \times \text{ED}}{\text{PEF} \times \text{BW} \times \text{AT}} \quad (6)$$

$$\text{ADI dermal} = \frac{C\text{soil} \times \text{SA} \times \text{AF} \times \text{ABS} \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}} \times 10^{-6} \quad (7)$$

Non-carcinogenic risks caused by the several pathways of heavy metals from soil can be determined based on the target hazard quotient HQ that is measured using Eq. 8 to assess the degree of toxicity (Varol et al. 2021):

$$\text{HQ} = \frac{\text{ADI}}{\text{RFD}} \quad (8)$$

The hazard index (HI) has been developed to measure the risk of carcinogenic health effects posed by heavy metals.

HI is the sum of three major pathways' hazard quotient as shown in Eq. 9:

$$\text{HI} = \sum \text{HQ} = \sum \frac{\text{ADIi}}{\text{RFDi}} \quad (9)$$

Carcinogenic risk index (CRI) is estimated using Eq. 10:

$$\text{Risk(CRI)} = \sum \text{ADIi} \times \text{SFi} \quad (10)$$

where the carcinogenicity slope factor (SF) is the probability of cancer per unit of exposure to metals.

The values of HI are classified into two categories. When $HI < 1$, it has no harmful effect on health, while $HI > 1$ means there is a potential for adverse effects on health (Jawad et al. 2020).

More, carcinogenic risk is considered as the possibility of an individual developing any type of cancer during a lifetime due to exposure to carcinogens (Chen et al. 2015). Furthermore, the CRI is considered negligible if the $CRI < 10^{-6}$, acceptable or tolerable if CRI is $10^{-6} < CRI < 10^{-4}$, and similarly considered high if the $CRI > 10^{-4}$ (Rastmanesh et al. 2020) (Table 1).

Results and discussion

Physicochemical properties and heavy metal contents

In general terms, the pH values of the soil are in the range of 8–8.7 with an average value of 8.3, indicating a slight alkalinity of the region of soil (Fig. 3). The electrical conductivity (EC) range is between 0.65 and 11.1 mS/cm with an average of 2.12 mS/cm indicating a low conductivity

Table 1 Parameters of the equations for the assessment exposure health risks of heavy metals in the soil of the watershed of Ariana

	Details	*Unit*	*Adult*	*Children*	*References*
IngR	Ingestion rate	mg/day	100	200	Adimalla and Haike (2018)
InhR	Inhalation rate	m^3/day	12.8	7.63	EPA (2001)
ED	Exposure duration	Year	24	6	EPA (2001)
EF	Exposure frequency	Day/year	350	350	Qing et al. (2015)
BW	Body weight	kg	55.9	15	Qing et al. (2015)
AT	Average time	days	ED*365	ED*365	EPA (1989)
SA	Exposed skin area	cm^2	4350	1600	EPA (2001)
PEF	Emission factor	m^3/kg	1.36E+09	1.36E+09	EPA (2001)
AF	Adherence factor	mg/cm^2-day	0.7	0.2	EPA (2001)
ABS	Dermal absorption factor	Unitless	0.001	0.001	Shi et al. (2011)
RFDing	Corresponding reference dose	mg/kg day	Cd = 1E − 03, Cr = 3E − 03, Cu = 4E − 02, Ni = 2E − 02, Pb = 3.5E − 03, Zn = 3E − 01, Hg = 3E − 04, Co = 2E − 2		Barraza et al. (2018)
RFDdermal		mg/kg day	Cd = 5E − 04, Cr = 3E − 03, Cu = 1.2E − 02, Ni = 2E − 02, Pb = 3.5E − 03, Zn = 3E − 01, Hg = 2.1E − 05, Co = 1.6E − 2		Barraza et al. (2018)
RFDinh		mg/kg day	Cd = 1E − 05, Cr = 2.86E − 05, Cu = 4E − 02, Pb = 3.52E − 03, Ni = 2.06E − 02, Zn = 6E − 02, Hg = 8.57E − 05, Co: 5.71E − 6		Adimalla and Haike (2018), EPA (2001)
SFing	Corresponding slope factor	(mg/kg)/day	Cd = 3.8E − 01, Cr = 5E − 01, Pb = 8.50E − 03, Ni = 9.1*10^{-1}		Barraza et al. (2018)
SFdermal		(mg/kg)/day	Cd = 1.58E − 01, Cr = 2.1 Ni = 4.55 Pb = 4.25E − 01		Barraza et al. (2018)
SFinh		(mg/kg)/day	Cd = 6.30E + 00 Cr = 4.10E + 01, Ni = 8.40E − 01, Pb = 4.20E − 02		Barraza et al. (2018)

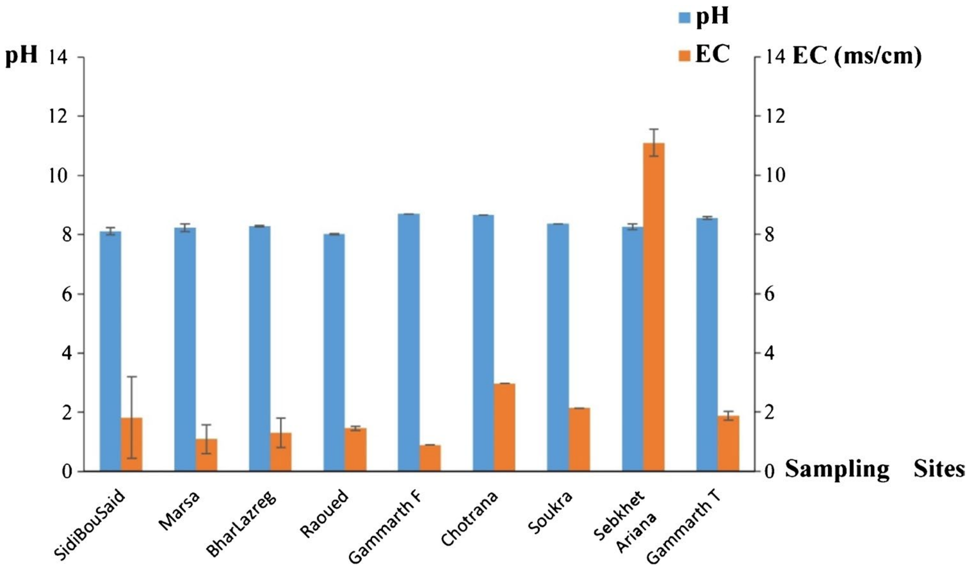

Fig. 3 pH and electrical conductivity (mS/cm) in soil samples

except for Chotrana with 11.1mS/cm. It may be linked to a dry climate due to a severe evaporation in the study area (Aydi et al. 2013).

The range of heavy metal concentrations in soils of the watershed of Sebkhet Ariana is provided in Table 2. Some of the mean values exceeded the soil background values of

Table 2 Comparison of heavy metal mean concentrations (mg/kg) in soil sampling sites in the study area with those recorded in other urban areas

	Cd	Co	Cr	Cu	Ni	Pb	Zn	Hg	References
Watershed of Sebkhet Ariana	1.2–7.8	3.2–18.4	7.9–92.3	4.4–32.2	7.6–33.8	8.5–97.6	25–267.2	0.5–1.3	This study
Harran Plain, Turkey		16	85	27	89	10.6	68		Varol et al. (2020)
Isfahan City, Iran	0.43	14.7	85.9	35.7	66.2	34.6	111.5		Esmaeili et al. (2014)
Ebro River Basin, Spain	0.41		20	17	19	17	57		Rodriguez et al. (2008)
Mouriki-Thiva, Greece		54	277	32	1591	24	67		Antibachi et al. (2012)
Daye City, China	1.41		60.7	105	25.8	43.7	159		Du et al. (2015)
Average crust	0.3	19	90	45	68	20	95	0.4	Turekian and Wedepohl (1961)

the Earth's crust such as Cd, Pb, Zn, Hg, and Cr indicating the influence of urbanization on urban soil pollution and that the pollutants' influence on the soil environment is serious. Cd, Pb, Hg, and Zn can be considered as the main pollutant of the environment because the concentration of this heavy metal in all samples was higher than the reference value (Fig. 4). The increase in heavy metal concentrations in some samples might be attributed to irrigation with contaminated water like in Raoued, Chotrana, Soukra, and BharLazreg, while a decrease might be due to the settling down of heavy metals in soils (Jawad et al. 2020).

In addition, mean concentrations of heavy metals in this study were compared with soils of other countries (Table 2). Co, Cr, Cu, and Ni concentrations in soils of the watershed of Sebkhet Ariana were lower than those in Harran Plain, Isfahan, Ebro River Basin, Mouriki-Thiva, and Daye City, while Cd, Pb, and Zn concentrations were higher than their corresponding worldwide average values.

Metal contamination levels

Geoaccumulation index (I_{geo})

The geoaccumulation index (Table 3) showed that all the samples could be considered as uncontaminated to moderately contaminated for Cr, Cu, Co, Zn, Pb, and Ni.

According to I_{geo} values of Cd, soils can be considered as extremely polluted (class 6) for Marsa, heavily to extremely contaminated (class 5) for Sidi Bou Said, Sebkha, and Soukra, heavily contaminated (class 4) for Gammarth T, moderately to heavily contaminated (class 3) for BharLazreg and Raoued, and moderately contaminated (class 2) for Chotrana and Gammarth F, and according to the contamination level of these heavy metals based on I_{geo} values of Hg, soils can be considered as moderately contaminated (class 2) for the great part of sampling sites.

Contamination factor and pollution load index

The CF and PLI (Table 4) are widely used to evaluate the degree of heavy metal pollution in the soils (Bhuiyan et al. 2010). The mean CF values for the metals in the study area follow the decreasing order Cd (29.33) > Hg (3.25) > Pb (1.9) > Zn (1.6) > Cr (1.31) > Co (0.97) > Cu (0.84) and Ni (0.05) demonstrated low contamination levels. The PLI mean values were found to be low in all the studied samples and varied between 0.2 and 0.8, indicating that the studied stations in the study area are in low pollution status considering the total of the studied metals, except in BharLazreg, PLI values was 1.2 inducing a moderate pollution. The CF values of Cd are very high in the samples studied, indicating that the soils in the watershed of Sebkhet Ariana are highly contaminated by this metal. Furthermore, the CF values of Hg were above 3 for some cases, showing a considerable contamination factors.

Enrichment factor (EF)

The enrichment factor (EF) in metals is widely used to assess the presence and intensity of anthropogenic contaminants relative to average natural abundance. Table 5 shows extremely high enrichment with Cd (EF > 50) in Soukra, Sebkhet Ariana, Marsa, and Sidi Bou Said; a significant enrichment with Hg, Zn, and Pb (5 < EF < 20) indicating the influences of anthropogenic sources (human, tourist activities, plastic waste, urbanizations, land use, and wastewater might be one of the most significant causes of different metals (Chaudhary et al. 2021). The soil had moderate enrichment in some cases with Zn (3 < EF < 5) and minor enrichment with Cr, Co, Cu, and Ni (EF < 3) which indicate that these metals are entirely from crustal materials or natural processes.

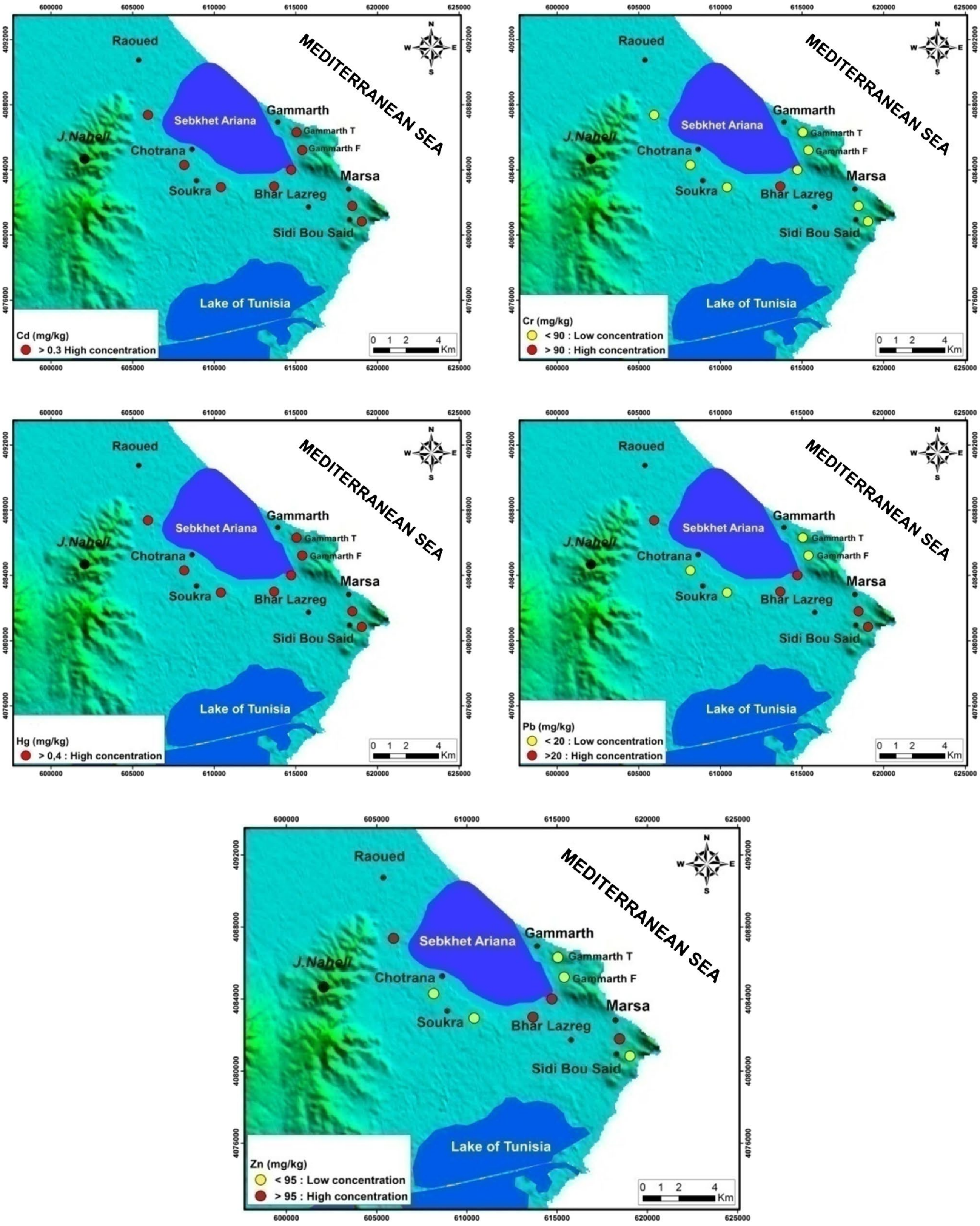

Fig. 4 Spatial distribution of soil Cd, Cr, Hg, Pb, and Zn concentrations (mg/kg) in the study area

Table 3 Geoaccumulation index for concentration of heavy metals soils of the study area

Site sampling	I_{geo}_Cd	I_{geo}_Co	I_{geo}_Cr	I_{geo}_Cu	I_{geo}_Hg	I_{geo}_Ni	I_{geo}_Pb	I_{geo}_Zn
Sidi Bou Said	4.07	−2.00	−2.45	−1.14	1.11	−2.75	0.26	−1.32
Marsa	5.07	−1.60	−2.10	−2.20	1.11	−2.37	0.18	−1.34
Chotrana	1.53	−1.76	−1.87	−2.20	0.26	−2.84	0.71	−1.36
BharLazreg	2.15	0.93	−1.10	−1.26	0.26	−1.97	0.99	0.54
Raoued	2.73	0.61	0.66	−1.07	0.26	−1.64	0.35	0.10
GammarthF	1.41	−2.39	−3.34	−3.69	0.26	−3.74	−1.57	−2.04
Sebkhet Ariana	4.11	−2.02	−2.35	−2.41	1.11	−2.61	0.68	−1.32
Soukra	4.18	−1.81	−2.33	−2.71	1.11	−2.46	0.88	−1.22
GammarthT	3.85	−3.06	−4.42	−4.00	1.11	−3.32	−1.96	−2.64

Table 4 Contamination factor values and pollution load indices of metals for different sampling sites

Site sampling	CF_Cd	CF_Co	CF_Cr	CF_Cu	CF_Hg	CF_Ni	CF_Pb	CF_Zn	PLI
Sidi Bou Said	25.30	0.37	0.27	0.67	3.25	0.01	1.20	0.60	**0.50**
Marsa	29.30	0.49	0.34	0.32	3.25	0.02	1.70	0.50	**0.60**
Chotrana	4.30	0.44	0.40	0.32	1.25	0.01	0.90	0.50	**0.40**
BharLazreg	8.60	0.86	1.31	0.84	1.25	0.05	3.40	1.10	**1.20**
Raoued	10	0.97	0.94	0.70	1.25	0.03	1.90	1.60	**0.80**
GammarthF	4	0.28	0.14	0.11	1.25	0.08	0.50	0.30	**0.20**
Sebkhet Ariana	26	0.36	0.29	0.28	3.25	0.01	0.90	0.60	**0.50**
Soukra	27.30	0.42	0.29	0.22	3.25	0.02	0.80	0.60	**0.50**
GammarthT	21.60	0.17	0.07	0.09	3.25	0.01	0.30	0.24	**0.20**

Table 5 Enrichment factor mean values of metals for sampling sites

Site sampling	EF_Cd	EF_Co	EF_Cr	EF_Cu	EF_Hg	EF_Ni	EF_Pb	EF_Zn
Sidi Bou Said	55.70	1.0	0.80	1.80	7.60	0.60	3.70	1.90
Marsa	60.40	0.90	0.70	1	6.30	0.60	12.70	7.30
Chotrana	10.80	1.10	1	0.80	3.10	0.50	2.20	1.40
BharLazreg	7.30	0.70	0.60	0.50	1.10	0.30	3	0.90
Raoued	9.70	0.90	0.90	0.60	1.20	0.50	1.40	1.30
Gammarth F	13.40	0.90	0.40	0.30	4.20	0.30	1.70	1.20
Sebkhet Ariana	66.60	0.90	0.70	0.70	8.30	0.60	2.30	1.50
Soukra	60.90	0.90	0.60	0.60	7.60	0.60	2.70	4
GammarthT	126.50	1.01	0.50	0.50	19.10	0.80	2.50	1.50

Ecological risk assessment (RI)

There is no universal concept of ecosystem health but the ecological risk assessment is the focus to provide basic information needed to determine if a release of hazardous substances to the environment presents a risk to human health or the environment. According to our results, the ecological risk index value (1054.6) in Marsa, in Soukra (826.7), and in Gammarth T (652.9) indicated that the urban soils of the study area could be classified as "at very high ecological risk" (Fig. 5). In this regard, Cd with the highest mean ecological risk index (679.1) confirmed the results achieved through the I_{geo} index. BharLazreg, Chotrana, and Raoued with RI values of 314.8, 337.9, and 324, respectively, have a considerable ecological risk for the environment, as agricultural soils of the study area; we concluded that the use of fertilizers and pesticides has a great impact on soil heavy metal concentrations (Ramos-Miras et al. 2020; Keshavarzi et al. 2021). However, the average RI value was less than 150 in Gammarth F, signifying low ecological risk. On the other hand, RI values in this study showed that Cd is the prime contaminant in the area, indicating that agricultural management is a potential source of metal accumulation in the study area.

Statistical methods were conducted to examine inter-correlation between metals in the soil samples and their

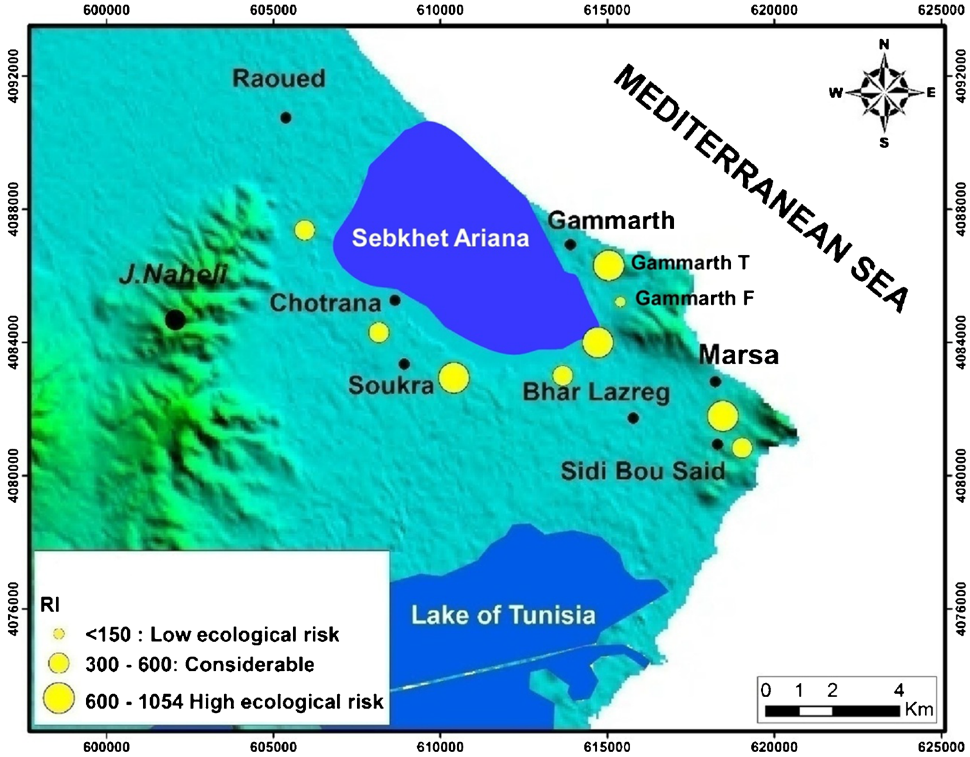

Fig. 5 Spatial level distribution of ecological risk index

Table 6 Correlation matrix between metals in the study area

Variables	Cd	Co	Cr	Cu	Ni	Pb	Zn	Hg
Cd	**1**							
Co	−0.29	**1**						
Cr	−0.24	**0.98**	**1**					
Cu	0.02	**0.83**	0.89	**1**				
Ni	0.06	**0.90**	**0.91**	0.82	**1**			
Pb	0.18	0.60	0.66	0.76	0.65	**1**		
Zn	0.31	0.57	0.59	0.65	0.63	**0.85**	**1**	
Hg	**0.93**	−0.58	−0.52	−0.26	−0.25	−0.12	0.02	**1**

possible origin. Pearson's correlation coefficient of the heavy metals and *p* values for statistical hypothesis testing are listed in Table 6. The matrix shows the strength of the linear relationships between each pair of variables.

The comparison among different metals and sampling points showed a significant correlation.

Cd appeared to be strongly positively correlated with Hg (correlation index: 0.932; $p < 0.05$). Similarly, Pb with Zn (correlation index: 0.853; $p < 0.05$) and Co with Cr (correlation index: 0.982, $p < 0.05$), Cu (correlation index: 0.835, $p < 0.05$), and Cr and Ni (correlation index: 0.91, $p < 0.05$) implied that the source of origin of metals was the same. In general, Cr was highly correlated with Ni (Mendoza-Grimón et al., 2014). Anthropic inputs of Cr and Ni in fertilizers, limestone, and manure are lower than the concentrations already present in the soil (Rodríguez Martín et al., 2006; Gil et al., 2018). Consequently, this suggests a lithogenic control over the distribution of Cr and Ni (Rodríguez et al., 2008; Nanos and Rodríguez Martín 2012). Other metals did not show any correlation with each other, indicating different sources of origin such as textile, detergents, tanneries, paints and dyes, plastic, pharmaceuticals, metallurgy, food and beverages, cement, lubricants, and auto-engineering (Ramos-Miras et al. 2020).

A negative correlation among the metals such as Cd with Cr and Co revealed that the input of these metals is not controlled by a single factor but rather by a combination of geochemical support phases and their mixed association (Aydi 2015).

The hierarchical clustering analysis (HCA) was applied to the sample soil quality data set to assess metal variables

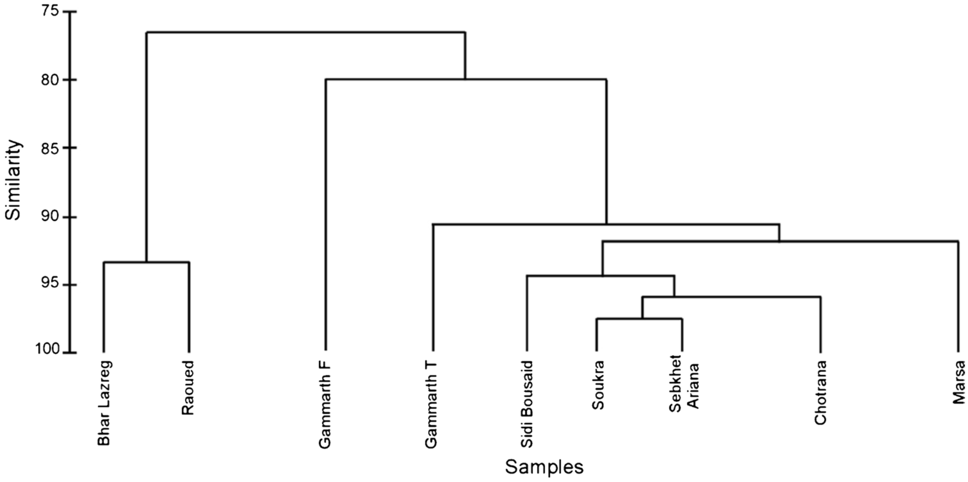

Fig. 6 Dendrogram showing clustering of heavy metal contents from sampling cities

to display a spatial sampling strategy (Deb Zhao et al. 2015).

The HCA dendrogram shows that the nine sites can be grouped into three statistically significant clusters (Fig. 6). Cluster 1 was associated with Marsa, Chotrana, Sidi Bou Said, Sebkhet Ariana, Soukra, and GammarthT. This observation was interesting, showing the cluster is at a relatively high pollution level.

Cluster 2 (BharLazreg and Raoued) is at a moderate pollution level.

Cluster 3 (Gammarth F) is at a level of relatively low pollution.

The clusters display a variable level of pollution obtained from anthropogenic sources.

Cluster 1 is located in an urban area characterized by high population density indicating the impact of man-made activities. BharLazreg and Raoued are farming areas; the use of agricultural waste may be the major reason for the contamination level. In contrast, Gammarth F is a forest, which has lower sources of pollution.

Health risk assessment

The health risk assessment is the process to estimate the nature and probability of adverse health effects in humans who may be exposed to contaminated environmental media or be in contact with the pollutant. The non-carcinogenic, hazard quotient (HQ).

According to the USEPA (2015), HQ values less than or equal to 1 are considered non-toxic, while an HQ value higher than one can pose considerable health effects. In this study, the calculated HQ values of heavy metals showed a variation among different metals and sampling points. Among the studied samples, Sidi Bou Said, Chotrana,

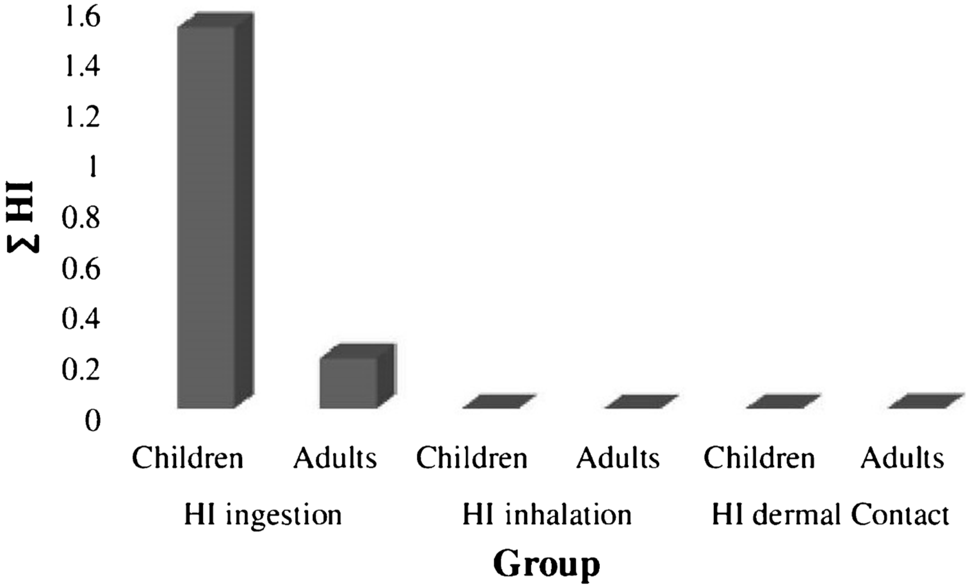

Fig. 7 The mean values of HI in the two groups studied via three pathways (ingestion, inhalation, and dermal)

BharLazreg, and Raoued showed high HQ value (HQ > 1), which is the indication of serious health hazards for the consuming population present in the study area. For instance, the highest HQ values were reported for Co with a value of 2.365. However, the HQ values for the other heavy metals were less than 1 and can be assumed as within the safe limits having no substantial health effects.

The hazard index (HI) of eight heavy metals through three potential exposure pathways (ingestion, inhalation, and dermal contact) for children and adults was estimated, and the results of the hazard index are mentioned in Fig. 7.

According to Fig. 7, results show that the HI dermal contact and HI inhalation values are quite lower than HI ingestion for both adults and children.

The trend of non-carcinogenic risk for both groups was in the order of ingestion > dermal > inhalation.

The average HI ingestion values indicate that the risk of non-carcinogenicity of heavy metals poses a greater threat to children's health than to adults'.

The HI ingestion values for all heavy metals in the children group ranged from 0.56 to 2.88, with the mean ∑HI ingestion values was 1.50, indicating that collective impacts of 8 heavy metals induced possible risk of non-carcinogenicity in all cases, except Gammarth F and Gammarth T, accurately by Cobalt with HI values higher than one in some cases such as in Raoued (HI = 2.36).

However, in adults' group, the HI ingestion for all heavy metals ranged between 0.07and 0.38 with the mean HI ingestion values being around 0.20, which did not exceed the international standards.

On the other hand, for inhalation and dermal pathways, all samples have their HI < 1 with the average values of ∑HI through inhalation, and dermal contact in the two groups studied was 6.16E – 04 for children and 2.81E – 04 for adults and 1.27E – 03 for children and 3.26E – 03 for adults' group, respectively, indicating that they did not pose a non-carcinogenic threat to human health both for children's and adults' group.

In general, hand-finger sucking is considered one of the crucial exposure pathways of soil metals in children. Children are more sensitive to a certain amount of toxin and, probably, ingest a considerable amount of soil, inadvertently (Chen et al. 2015; Khelifi et al. 2021).

The carcinogenic health risk in terms of CRI (carcinogenic risk) of Cd, Pb, Ni, and Cr through three potential exposure pathways for children and adults was computed in the soil samples, and results of CRI are presented in Fig. 8.

The CRI was only calculated for these metals. SF (slope factor) of Hg, Cu, Co, and Zn was not available in any database, so the CRI was not applicable for them.

According to Fig. 7, in both the adult and children's groups, the average values of ∑CRI through ingestion, inhalation, and dermal in the two groups studied were 4.69E – 04 for children and 6.15E – 05 for adults, 5.63E – 07 for children and 2.53E – 07 for adults, and 3.51E – 06 for children and 8.98E – 06 for adults' group, respectively.

The trend of carcinogenic risk for both groups was in the order of ingestion > dermal > inhalation.

The average values of ∑ CRI for inhalation and dermal pathways show quite lower than the tolerable limit of 1.00E – 04, which indicates no significant health effects to the local residents (adults and children) in the study area, while in children's group, the average value of ∑ CRI ingestion was higher than the acceptable level (1×10^{-4}).

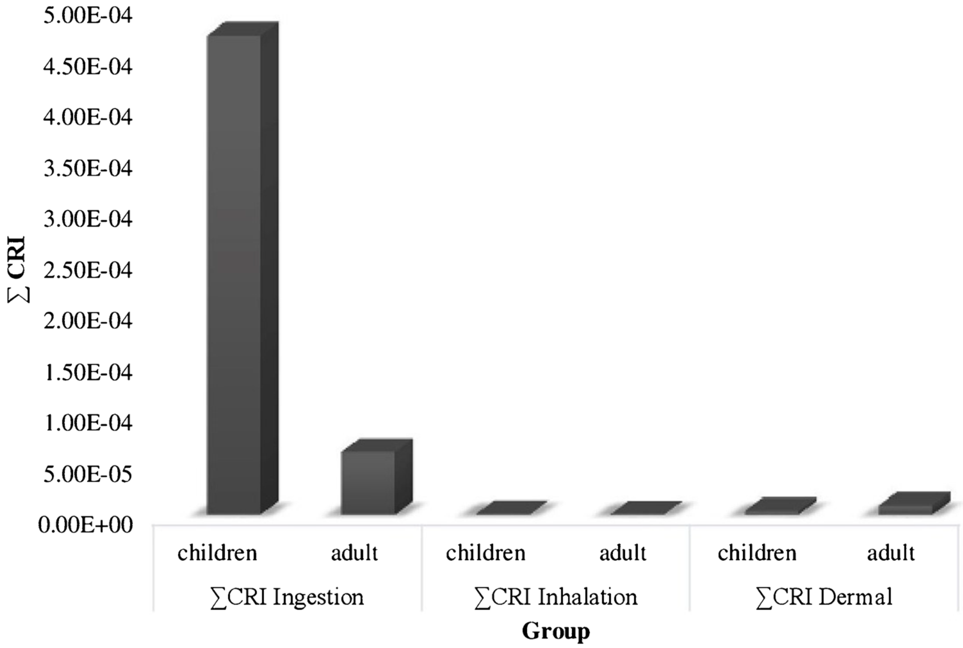

Fig. 8 The mean values of CRI from exposure to selected heavy metals on adults and children from all sites via three pathways (ingestion, inhalation, and dermal)

Specifically, for children's group, results of CRI ingestion of Cr and Ni values were above the 1.00E − 04 in all sampling sites except Gammarth F, suggesting that soil pollution of these metals has shown significant carcinogenic lifetime health risks on local residents in our study area. Particularly, Cr should be paid more attention to the potential occurrence of cancer risk to the local residents in the urban regions of the study area. Because Cr is a metal linked to cancer pathogenesis (Bwatanglang et al. 2019). Lung cancer is one of the effects of Cr on human health (Jaishankar et al. 2014).

Chromium is a naturally occurring element while it may enter the soil environment by means of some anthropogenic activities.

The main anthropogenic source for this metal is linked to industrial applications including plastic packaging and electroplating operations (Khelefi et al. 2021).

The comparison of the carcinogenic risk among the two groups shows that the health of children is more threatened. Based on the CRI ingestion values, children are more vulnerable through the ingestion pathway due to the higher intake of soil through their hands and mouth.

It is noticed from the analysis of carcinogenic health risk that ingestion is the foremost exposure pathway that can harm adults' and children's health in the urban cities of the Sebkhet Ariana watershed.

Conclusion

The purpose of this survey is to evaluate the degree of soil pollution, ecological risk by heavy metals (Cd, Hg, Pb, Zn, Co, Cr, Cu, Ni), and health risk assessment in the watershed of Sebkhet Ariana (Tunisia) influenced by anthropogenic activities using pollution indicators such as the enrichment factor (EF), geoaccumulation index (I_{geo}), contamination factor (CF), pollution load index (PLI), ecological risk (RI), hazard index (HI), and carcinogenic risk index (CRI) to both children and adults.

The main conclusions drawn from the present survey are given as follows:

1. Some of the mean values exceeded the soil background values of the Earth's crust such as Cd, Pb, Zn, Hg, and Cr indicating the influence of urbanization on urban soil pollution. The increase in heavy metal concentrations in some samples such as Raoued, Chotrana, and BharLazregmight be attributed to irrigation with contaminated water.

   The high concentration of Cr and Pb can be attributed to its anthropogenic origin in solid waste.
2. The mean levels of the heavy metals were followed in the order of Zn > Pb > Cr > Ni > Cu > Co > Cd > Hg.
3. Based on the results from Igeo values of heavy metals, the soil in the study area was frequently classified into uncontaminated to moderately polluted group for all heavy metals except Igeo values of Cd which soils can be considered as extremely polluted in Marsa.
4. The CF values of Cd and Hg are very high and considerable contamination factors, respectively, in the samples studied and the PLI mean values were found to be low in all the studied samples except BharLazreg.
5. The results of EF show that using the Fe concentration in the continental shale as a normalizer produces higher average EF values for Cd, Hg, Zn, and Pb indicating the influences of man-made sources (tourist activities, plastic waste, urbanization, and wastewater).
6. The ecological risk index value (1054.6) in Marsa, (826.7) in Soukra, and (652.9) in Gammarth T indicated that the urban soils of the study area were classified as very high ecological risk. It may be linked to agricultural management especially using fertilizers and pesticides.
7. Multivariate statistical analysis (Pearson's correlation coefficient and HCA) outlined that the metallic accumulation in the soils of the study area was related to lithological/geological origin and anthropogenic impacts.
8. The outcomes of HQ, HI, and CRI stressed out that heavy metals would not pose a significant health risk when adults are exposed to the soil in the study area, while non-cancerogenic health risk for children was considered as a collective effect of heavy metals (THI > 1) and the risk of the carcinogenic impact of Cr and Ni, with CRI values of ingestion pathway above the permissible limits in some cases.

For the first time, the study provided data for the soil quality of the region, which is helpful in making a remediation plan for heavy metal–affected soils. Therefore, it is recommended to monitor the level of macro- and micro-element contamination risk in soils of the watershed of Sebkhet Ariana, and the potential health hazards are recommended. In addition, routine monitoring programs should be conducted in the Sebkhet Ariana sustainable agricultural area management and long-term protection of water quality from further deterioration.

Data availability Not applicable.

Declarations

Ethics approval and consent to participate Not applicable.

Consent for publication Not applicable.

Conflict of interest The authors declare that they have no competing interests.

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