Investigation of heavy metal contamination and ecological and health risks in farmland soils from southeastern phosphate plateaus of Khouribga (Morocco)

Ahmed Barakat[1,*], Rida Khellouk[1], Widad Ennaji[1], Hassan Mosaid[1]

[1]Geomatique, Georesources and Environment Laboratory, Faculty of Sciences and Techniques, Sultan Moulay Slimane University, Morocco

*correspondending author e-mail: a.barakat@usms.ma

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Abstract. The present study was conducted in the SE area of phosphate plateaus (Khouribga) located in central Morocco. It attempted to assess the heavy metal (HM) (Cd, Cr, Cu, Pb, Zn) contamination in the farmland soils and their potential ecological hazard and non-non-carcinogenic risks using various pollution indices, magnetic susceptibility (MS), and Geographical Information System (GIS) methods. A total of 41 soil samples were collected and analyzed for pH, electrical conductivity (EC), grain-size, organic matter (OM), calcium carbonate ($CaCO_3$), and MS and HM elements. The results showed a mean dominance order of Zn>Cr>Cu>Pb>Cd where mean concentrations of HMs, except Pb, exceeded their local background and Food and Agriculture Organization (FAO) and World Health Organization (WHO) permissible guidelines. The values of geo-accumulation index (Igeo), nemerow pollution index (*PI*), and pollution load index (*PLI*) revealed significant high level of HM contamination in soils. The MS values showed a spatial distribution pattern similar to those of HMs, attesting the ability of the MS method for mapping the contaminated soils. Agricultural and mining activities and geologic materials were the main sources of HM accumulation. According to the potential ecological risk index (*RI*) (195.93<*RI*<1092.53), the soil samples had moderate (65.85%) to high ecological (34.15%) risk. The hazard index (*HI*) showed that adults and children are not exposed to non-carcinogenic risk from the studied HMs, apart from two soil samples where Cd posed health risks to children compared to the other studied HMs. The statistical results revealed that soils are polluted by anthropogenic activities. Accordingly, effective agricultural practices that respect the environment, including the reduction of inputs as fertilizers, pesticides, herbicides, and fungicides should be required to guarantee the safety of cropland and the residents in the studied area. Hence, the findings from this study provided some useful information for soil pollution control and management in the study area.

Key words: agricultural soil, heavy metals, integrated contamination, pollution indices, magnetic susceptibility, ecological and health risks.

1. Introduction

Soil is an important component of ecosystems, which serves as a filter for pollutants released into the environment, and detoxifies it by absorbing or degrading different types of contaminants, including heavy metals. Pollution with heavy metals (HMs) became one of the main sources of soil pollution worldwide (Zhou et al., 2015), and consequently the most widespread form of degradation of soil quality and health (Rubalingeswari et al., 2021). Nonbiodegradable and extremely toxic even at low concentrations, the HMs in soil originate from natural (soil/rock parent materials) and anthropogenic sources. The anthropogenic contamination may be derived from heterogeneous activities as industrial, mining, chemical fertilizers, waste animal manure, composting, wastewater irrigation, waste disposals, vehicular emissions, aerosol deposition (Barakat et al., 2020; El Baghdadi et al., 2012). So, agricultural soil contamination by HMs could affect ecosystems and human health, due to their easy transfer into water bodies and vegetation and thus their potential to end up inside the human body.

To characterize the HM pollution, geochemical methods and non-destructive magnetic techniques are increasingly applied since they seem promising in monitoring the HM accumulation in soils and identifying their potential origins. Because of its efficiency and rapidity, magnetic measurement has become a widely used tool for HM pollution assessment of soils, knowing that magnetic particles can absorb certain HMs (Cao et al., 2015). In numerous recent studies (Ayoubi et al., 2018; Canbay et al., 2010; Liu et al., 2016), it is stated that the soil magnetometry integrated geochemical analyses can be helpful to assess the HM contamination and to discriminate between geogenic and anthropogenic contribution and contamination of soils. What is more, many environmental indices, such as contamination geo-accumulation index (I_{geo}), pollution load index (*PLI*), and nemerow pollution index (*PI*), are proposed and widely employed to assess the HM enrichment in soils and their sources (Mukherjee et al., 2020), These indices combined with statistical analysis provided supplementary information on the sources of heavy metals in soils. In addition, as the contamination of soils and crops by HMs can damage ecosystems and the health of nearby residents, the potential ecological risk index (*RI*) and the hazard index (*HI*) have been applied successfully and widely (Barakat et al., 2019a; Gujre et al., 2021). Geographic Information Systems (GIS) also became widely applied to check and visualize the spatial distribution of HM pollutants in soils and their mutual relationships (Ennaji et al., 2020; Javaid et al., 2020).

In the past few decades, as Morocco has experienced rapid economic growth that depends on industry, mineral resources, and especially on agriculture, soil quality degradation, notably by HM pollution, has become a main ecological concern (Barakat et al., 2019a; Nassiri et al., 2021; Oumenskou et al., 2018). Because the HM accumulation in soils might reduce the soil fertility and crop production, and might also be transferred through inhalation, ingestion, and dermal absorption to the human body that could pose risks to the health and safety of persons (Barakat, 2020; Barakat et al., 2019b; Hilali et al., 2020). Many studies investigated the environmental effect of HM contamination of farmland soils in various Moroccan regions and found HM contents were above the local background levels and the Food and Agriculture Organization (FAO) and World Health Organization (WHO) permissible guidelines. These studies reported that the HM accumulation in the studied regions is linked to anthropogenic sources as farmland practices, intensive agriculture, wastewater irrigation by irrigation by untreated wastewater, urbanization, and mining and industrial activities. However, to develop a successful soil pollution control plan in Morocco, investigation of heavy metal pollution of soils in agricultural areas remains important to obtain reliable information on concentrations, spatial distributions, and sources of HMs.

Because of rapid urbanization, industrialization, and intensive agriculture, some studies examined the HM contamination in Tadla plain (Morocco) on urban soil (Barakat, 2020; Barakat et al., 2019b; Hilali et al., 2020), irrigated agricultural soil, and river sediments using physical, geochemical, and magnetic methods, and reported a significant increase in HM concentrations compared to background values. However, no study of HM enrichment in non-irrigated soil has been conducted. It is needed therefore to explore the HM concentrations and their possible enrichment at the non-irrigated farmlands in the southeastern phosphates plateaus (Khouribga, Morocco), especially since the study area is also experiencing strong open-pit mining phosphates. Thus, this research aimed to 1) understand the contamination of soils by HMs (Cd, Cr, Cu, Pb, and Zn) from southeastern phosphates plateaus using various environmental indices; 2) check the possible sources of HMs by exploring the relationships among HMs and indices; and 3) evaluate the ecological hazard and non-non-carcinogenic risks within the local residents through ingestion.

2. Materials and Methods

2.1.Study area

The study area straddles the border between Fkih Ben Saleh and Khouribga provinces located in central Morocco. It is situated within 32°32' to 33° 01'N latitude and 6°26' to 6°15W longitude, with an area of ~1541 km^2. The mean elevation of the area ranges from 440 to 882 m.a.s.l. It is in a semi-arid climate zone, with a rainy season from November to March and a dry season from April to October. The average annual temperature varies from 0°C to 38°C, the mean annual rainfall is 350 mm and the mean annual evapotranspiration level is 1800 mm.

The main land-use type of the study area is farmland (70%). Particularly, the largest proportion of farmland is utilized for rain-fed seasonal crops (Bour area) dominated by cereals

(42.7%), and for grazing. In the last two decades, the largest extent of irrigated agriculture and livestock activities and opencast phosphate mining has been witnessed, which represent the potential sources of metal pollution of the agricultural soils in the study area.

The soils of the study site are diverse due to the nature of the source rocks pedogenesis (Khellouk et al., 2019). They are generally shallow and developed on sedimentary phosphate Cretaceous-Eocene deposits of the Ouled Abdoun basin. The soil types are dominantly classified as crude mineral soil, poorly developed soil, calcimagnesic soil, isohumic soil, brunified soil, and iron sesquioxide soil (Khellouk et al., 2019).

2.2.Sampling and analysis

A number of 41 samples of topsoil (0–10 cm) were randomly taken from different sites covering the entire study area over a period from March to April 2020 (Fig. 1). The locations of sampling sites were recorded using a portable global positioning system (GPS), and the soil samples were stored separately in labeled polyethylene bags and transported to the laboratory. In the laboratory, all soil samples were air-dried, crushed, sieved through a 2 mm stainless steel sieve, and stored at room temperature before further physicochemical analyses. The physicochemical parameters, namely pH, electrical conductivity (EC), grain-size, organic matter (OM), calcium carbonate ($CaCO_3$), and magnetic susceptibility (MS) were analyzed.

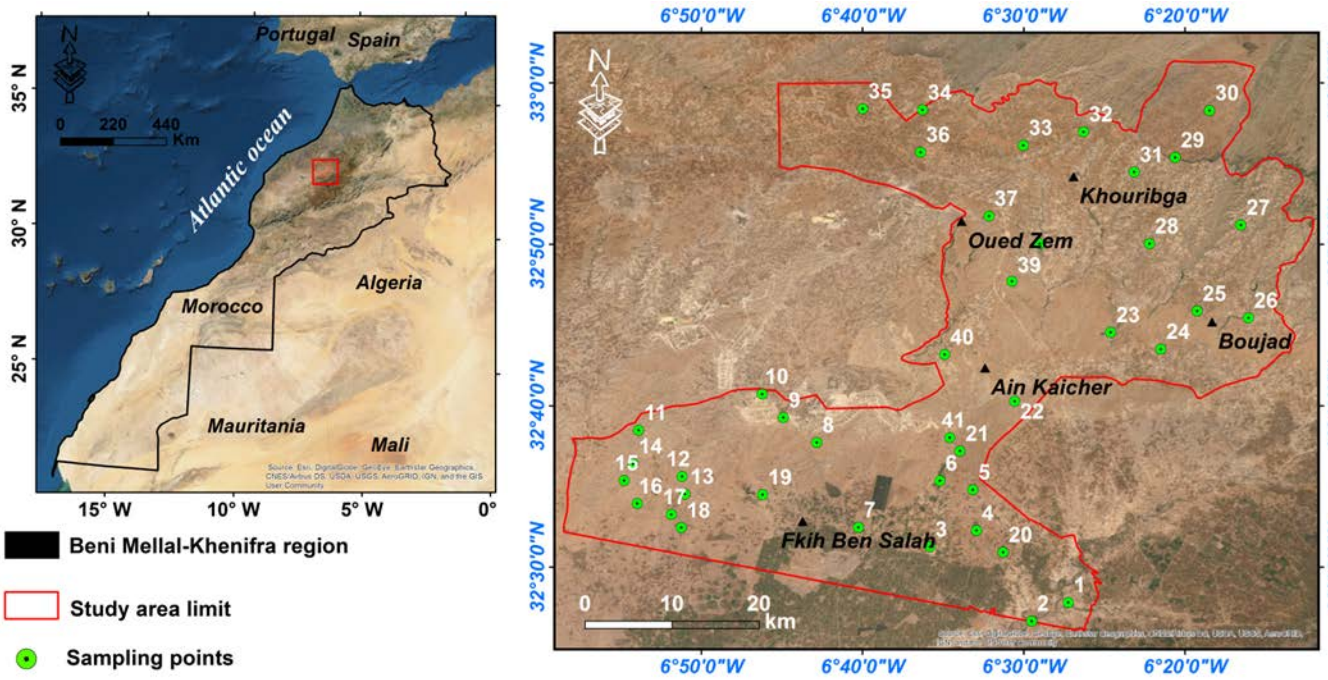

Figure 1. Location map of the study area and sampling stations.

The soil pH and EC were measured in deionized water (1:2.5 soil: distilled water mixture) using a pH/conductivity meter (Thermo Scientific Orion 4-Star Plus). The particle size analysis was realized using Robinson's pipette method. The OM content was calculated by igniting the dried soil sample in a muffle furnace maintained at 550°C for 4 h. The $CaCO_3$ content was calculated by calcination of the dried soil sample during 2 h at 930°C. The magnetic susceptibility (MS) was measured at least three times for each soil sample, by employing Bartington MS2B sensor operating at both low (470 Hz) and high (4700 Hz) frequencies. The frequency-dependent susceptibility (χ_{FD}) is computed by the application of Eq. (1) (Suresh et al., 2011):

$$\chi_{FD}\% = [(\chi_{LF} - \chi_{HF})/\chi_{LF}]x100 \qquad (1)$$

where, χ_{LF} and χ_{HF} represent low-frequency MS and high-frequency MS measured at 470 and 4700 Hz, respectively.

The concentrations of heavy metals, namely, Cd, Cr, Cu, Pb, and Zn, were determined using inductively coupled plasma atomic emission spectroscopy (ICP-AES) in the Division of Technical Support Units for Scientific Research of the Center National Scientific and Technical Research (CNRST, Morocco). A mass of 1 g of each sample was digested with a mixture (1:5 [v/v]) of 10 mL HNO_3 and 10 ml of HCl. The final filtered extract of each sample was diluted to 100 ml with distilled water and kept at 4°C until its ICP analysis.

The frontier of the study area was outlined using images of Landsat 7 ETM (http:// www.gscloud.cn), and the spatial distribution maps of the analyzed parameters were generated in ArcGIS 10.2 software by adopting the inverse distance weight (IDW) method. The analysis data were examined through basic descriptive statistics, using Excel software. The coefficient of variation (CV) and Pearson's correlation (R^2) were used to explore the spatial distributions of HM concentrations and to identify their sources. Primarily, the Shapiro-Wilk test was computed using raw HM and physicochemical parameter data to evaluate the data sampling adequacy for correlation analysis. Principal component analysis (PCA) was also employed to identify the source of the pollutant using SPSS 16.0 statistical software. The evaluation of the soil HM contamination in the study area was performed based on some environmental indices described in the next section, and the comparison with the local background concentrations (Oumenskou et al., 2018) and other worldwide references (Chiroma et al., 2014; Kamunda et al., 2016; Taylor, 1964). The spatial pattern of environmental index data was analyzed using GIS environment.

2.3.Assessment of heavy metal contamination

To check the contamination by HMs (Cd, Cr, Cu, Pb and Zn) in our study area, the geoaccumulation index (I_{geo}), pollution load index (*PLI*), and nemerow pollution index (*PI*), were employed due to their accurately estimating the anthropogenic pollution level of soils, as demonstrated by many relevant types of research (Barakat et al., 2020; El Baghdadi et al., 2012; Ennaji et al., 2020; Mirzaei et al., 2020; Oumenskou et al., 2018).

The I_{geo} index was proposed by {Müller, 1979 #2824} to evaluate the anthropogenic heavy metal contamination in soils by comparing metal content in the sample with the geochemical background. I_{geo} is calculated as expressed in Eq. (2):

$$I_{geo} = log_2\,(C_i/1.5B_i) \qquad (2)$$

where C_i is the obtained metal concentration, B_i is the background concentration of analyzed metal, and 1.5 factor is used to attenuate lithogenic variations in background concentrations. The <I_{geo} values are classified into seven classes: unpolluted (I_{geo}≤0), slightly polluted (0<I_{geo}≤1), moderately polluted (1<I_{geo}≤2), moderately to severely polluted (2<I_{geo}≤3), severely polluted (3<I_{geo}≤4), extremely severely polluted (4<I_{geo}≤5), and extremely polluted (I_{geo}>5).

The *PLI* index proposed by Tomlinson et al. (1980) and successfully used by several authors for evaluating quantitatively the integrated HM pollution status of soils, was computed based on the contamination factor of the metals (*CF*) as stated below:

$$CF = C_i/B_i \qquad (3)$$

According to Hakanson (1980), the *CF* values were interpreted as: *CF*<1 "low contamination", 1<*CF*<3 "moderate contamination", 3<*CF*<6 "considerable contamination"', and *CF*>6 "very high contamination".

$$PLI = (CF_1 x CF_2 x \ldots . x CF_n)^{1/n} \qquad (4)$$

where *n* is the number of determined heavy metals. The *PLI* values classify soil samples as: no polluted (*PLI*<1); and very high deteriorated (*PLI*>1).

The *PI* index, commonly applied to assess the degree of contamination in a soil environment (Ogunkunle & Fatoba, 2013; Qing et al., 2015), was calculated according to the equations below:

$$PI = \sqrt{\frac{\left(\frac{1}{n}\sum_{i-1}^{n} P_i\right)^2 + P_{i\,max}^2}{n}} \qquad (5)$$

$$P_i = C_i/B_i \qquad (6)$$

Where *PI* is the single pollution index of a particular HM, $P_{i\,max}$ is the maximum value of the single pollution index of all heavy metals; C_i and B_i represent respectively the measured contents and the European Union (EU) guideline (Kamunda et al., 2016) of particular metal (*i*); *n* is the number of studied HMs. The PI index indicates the level of HM contamination in soil as follows (Zhong et al., 2010): ≤0.1: clean, ≤1: warning limit, ≤2: slight pollution, ≤3: moderate pollution, and >3: heavy pollution.

The potential ecological risk index (*RI*) was introduced as efficacy means of evaluating the heavy metal toxicity status (Hakanson, 1980). The RI index is calculated as expressed below:

$$RI = \sum E_r^i \qquad (7)$$

$$E_r^i = T_i x CF \qquad (8)$$

Where E_r^i is the monomial potential ecological risk of a definite heavy metal, and T_i is the metal toxic factor for a given heavy metal. The T_i values used in this study, Cd = 30; Cu = Pb = Ni = 5, Cr = 2, and Zn = 1, were proposed by Hakanson (1980). The E_r^n values of <40, 40-80, 80-160, 160-320, and ≥320, are interpreted as low, moderate, considerable, high, and very high risk, respectively (Hakanson, 1980). The *RI* index is generally grouped into the following levels according to Hakanson (1980): *RI*<150 low risk, 150≤*RI*<300 "moderate risk", 300≤*RI*<600 "considerable risk", and *RI*≥600 "high risk" (Hakanson, 1980).

To evaluate the health risk of HM pollutants, including both non-carcinogenic effects on adults and children, a health risk quotient (HQ_i) is employed. Moreover, the total hazard index (*HI*) representing the comprehensive non-carcinogenic risk posed by all analyzed HMs was calculated by adding the *HQ* of each element as described by Eqs (9), (10) and (11).

$$ADD_i = \frac{C_i x IR x EF x ED x CF}{BW x AT} \qquad (9)$$

$$HQ_i = \frac{ADD}{RfD_i} \qquad (10)$$

$$HI = \sum_{i=1}^{n} HQ_i \qquad (11)$$

where ADD_i represent the average daily intake (mg kg^{-1} day^{-1}) of HMs through soil particle ingestion, C_i is the concentration of heavy metal *i* in soil (mg kg^{-1}), *IR* is the ingestion rate of soil/dust for children (200 mg day^{-1}) and for adult (100 mg day^{-1}) (EPA, U.S., 2011) , *EF* is the exposure frequency (350 days $year^{-1}$), *ED* is the exposure duration (6 years for children and 30 years for adults), *BW* is the body weight of the exposed individual (15 kg for children and 70 kg for adult), *AT* is the exposure period for non-carcinogenic effects (2190 days for children and 10950 days for adult), and *CF* is the conversion factor (10^{-6} kg mg^{-1}), and *RfD* representing the means reference oral dose, is 0.001, 0.003, 0.04, 0.0035, 0.3 and 0.7 mg day kg^{-1} for Cd, Cr, Cu, Pb, Zn and Fe, respectively (Leung et al., 2008). *HQ* or *HI* <1 suggests no potential health risk associated with HM accumulation, while HQ or HI >1 represents a significant non-carcinogenic health risk due to heavy metal presence.

3. Results and discussion

3.1.Physicochemical parameter and heavy metal contents

A soil capacity to adsorb HM varies by its physicochemical composition.. So, soil physical chemistry (e.g.: pH, EC, $CaCO_3$, OM, grain-size, and MS) may impact the mobility and distribution of heavy metal in the soil (Dragović et al., 2008). In the present study, the basic descriptive statistics of physicochemical parameters and HM contents of analyzed soils are summarized in Tables 1 and 2 and Figure 2.

The pH value ranged from 7.7 to 8.7, with a mean of 8.1. This indicated that the studied soils are alkaline, due to the calcareous nature of the soils. This soil alkalinity mainly caused a reduction of the HM retention and mobility in soils (Tian et al., 2017). The pH values exhibited a clear gradual decrease from the south to the north of the study area. The recorded EC values ranged between 118.4 (site 15) and 665.0 µS/cm (site 39), with an average of 282.8 µS/cm. These EC values were under the salinity edge (<40000 µS/cm) for agricultural soils according to Jahn et al. (2006). The $CaCO_3$ contents recorded at the various sampling sites varied from 12.6 (site 7) to 65.4% (site 27) with an average of 27.3%, suggesting that the studied soils are predominantly calcareous. The spatial variability showed a clear abundance of $CaCO_3$ in the southeast than the north of the study area. This corroborated thus the pH results. The OM contents measured in sampled soils were within the range of 0% (sites 4 and 18) - 15.8% (site 10), except those of site 24 that was 20%. The grain-size of soils controls the degree at which water circulates through the soil, and influences the magnetic sensitivity and heavy metal content (Lu et al., 2011). The average clay, silt and sand contents recorded at the study area falls within the range of 13.4, 34.2 and 52.3%, respectively. According to the USDA soil particle size classification, the studied soil samples belong to clay loam and loam clay textural classes.

The magnetic sensitivity values of the sampled soils measured varied between 11.55 10^{-8} m^3 kg^{-1} (S29) to 442.8 10^{-8} m^3 kg^{-1} (S25) and from 10.4 10^{-8} m^3 kg^{-1} (S29) to 397.6 10^{-8} m^3 kg^{-1} (S25) for the low (χ_{LF}) and high (χ_{HF}) frequency magnetic sensitivity, respectively. The coefficient of variations (CV) of χ_{LF} and χ_{HF} are 50.7% and 50.3% respectively, indicating that the spatial differences of soil MS in the studied area are significant.

Furthermore, based on the classification by Gautam et al. (2004), the χ_{FD} is medium (22.5%) to high (77.5%) in soil samples. The frequency-dependent susceptibility (χ_{FD}) calculated for all the samples showed values varying from 5.4 to 15%, with a mean of 10.8%. The CV was 18.6% (<50%), indicating a low degree of variations of χ_{FD}among sampling sites. According to Dearing et al. (1996) classification, slightly higher to higher χ_{FD} values occurred at 95% of the studied samples (Table 1), suggesting that the dominance of the contribution of the SP grains at more than 75% and the coarser multidomain magnetic (MD) grains and perhaps a single domain (PSD). However, ultrafine superparamagnetic (SP) particles were poorly represented. According to the findings reported by Liu et al. (2016), which suggested that magnetic minerals are present as MD and PS grains, as significantly larger particles, than pedogenic SP grains in soil, the results of the present study attested that the significant magnetic susceptibility was linked in part to anthropogenic activities.

Table 1. Summary statistics of soil parameters and of the magnetic susceptibilities (χ) of studied soils.

Statistics	Texture			pH	EC	OM	$CaCO_3$	χ_{fd}	χ_{hf}	χ_{lf}-χ_{hf}
	Sand (%)	Silt (%)	Clay (%)		(µS/cm)	(%)	(%)	(%)	10^{-8} m^3 kg^{-1}	
Min	5.6	34.1	0.3	7.7	118.4	0.0	0.0	5.4	11.6	1.1
Max	42.7	94.1	43.6	8.7	665.0	20.0	28.3	15.0	442.8	47.9
Mean	17.5	48.6	33.9	8.1	282.8	9.8	10.1	10.5	157.5	17.9
SD	8.1	8.8	8.1	0.2	148.1	4.1	8.2	2.0	79.8	10.6
CV (%)	46.3	18.1	23.8	2.4	52.4	41.3	80.7	18.6	50.7	59.5

From the Table 2, the measured HMs and Fe contents ranged as follows: Cd, 0.00-30.11 mg kg^{-1} with a mean of 8.60 mg kg^{-1}; Pb, 5.92-19.13 mg/kg with a mean of 9.51 mg kg^{-1}; Zn, 37.42-166.66 mg kg^{-1} with a mean of 87.21 mg kg^{-1}; Cu, 6.17-48.46 mg kg^{-1} with a mean of 24.90 mg kg^{-1}; Cr, 6.09-249.42 mg kg^{-1} with a mean of 77.07 mg kg^{-1}; and Fe, 12397.39-49668.10 mg kg^{-1} with average of 24,596.46 mg kg^{-1}. The observed HM contents are higher than those in other parts of the world (Table 2). Most of the HM concentrations decrease in the order of Zn> Cr> Cu> Pb> Cd. The CVs were 50.07%, 69.36%, 40.52%, 37.10%, and 39.42% for Cd, Cr, Cu, Pb, and Zn, respectively. The significant CVs values of all HMs, particularly the CVs of Cd and Cr that are above 50%, indicated that the spatial distributions of metals in studied soil samples are non-homogeneous and the HM concentrations in studied soil samples are influenced by anthropogenic activities.

To evaluate HM contamination in agricultural soil samples, comparative studies with local background elements and other world HM reference values are presented in Table 2. The mean Cu concentration for investigated soils was relatively lower than the local background and the world references, although higher values were noted in about 29% of soil samples. Cd, Zn and Cr were found to be the most predominant element in the study area and showed high concentrations than the local and regional reference values. The abnormal concentrations were observed in more than 80% of samples, mostly located in the northern parts of the study area, and can be related to anthropogenic activities such as phosphate mining and P fertilizers. Smolders and Mertens (2013) suggested a value of 1 mg kg^{-1} as the maximum limit for Cd in uncontaminated soil. However, according to Garrett et al. (2010) attributing large Cd concentrations (up to 900 mg kg^{-1}) in Jamaican soils to dispersal guano deposits, the high Cd values found in the study area is attributed, nonetheless in part, to the dispersal of phosphate rock formations. Chambionnat (1963) analyzed Cr in the raw and processed phosphates in Morocco. The results showed higher Cr contents ranging between 39 and 170 mg kg^{-1}, supporting thus that the abnormal Cr level in the study area could be associated with atmospheric deposition of Cr produced by the phosphate mining and by the use of P-fertilizers. The significant Zn contents compared to local and world references revealed that all sites were contaminated from anthropogenic sources. The Zn accumulations, as well as those of Cd and Cu, in agricultural soils, are commonly due to the abrasion of agricultural tools and engines, the combustion reactions, and the excessive use of chemical fertilizers and fungicides. Fe also showed higher concentrations in the study area, which might be linked to the lithological nature of parent rock materials, meanwhile phosphate sedimentary rocks can contain elevated Fe.

Table 2. Descriptive data of HM concentrations (mg/kg) in studied soil samples (n=41). Contents of Fe are in percent.

Samples	**Concentrations**						
	Cd	Cr	Cu	Pb	Zn	Fe	
Min	37.42	5.22	6.09	6.17	5.92	1.24	
Max	166.66	30.11	249.42	48.46	19.13	4.97	
Mean	87.21	8.60	77.07	24.90	9.51	2.46	
SD	34.38	4.31	53.45	10.09	3.53	0.82	
CV (%)	39.42	50.07	69.36	40.52	37.10	33.50	
Local Background	0.85	25.21	31.40	32.54	43.76	1.21	Oumenskou et al. (2018)
Earth's soil	0.11	84	26	29	60	3.2	Taylor (1964)
Earth's crust	0.6	100	50	14	75	4.1	Taylor (1964)
FAO/WHO	3	100	100	100	300	2.78	Chiroma et al. (2014)
EU	3	150	140	300	300		Kamunda et al. (2016)

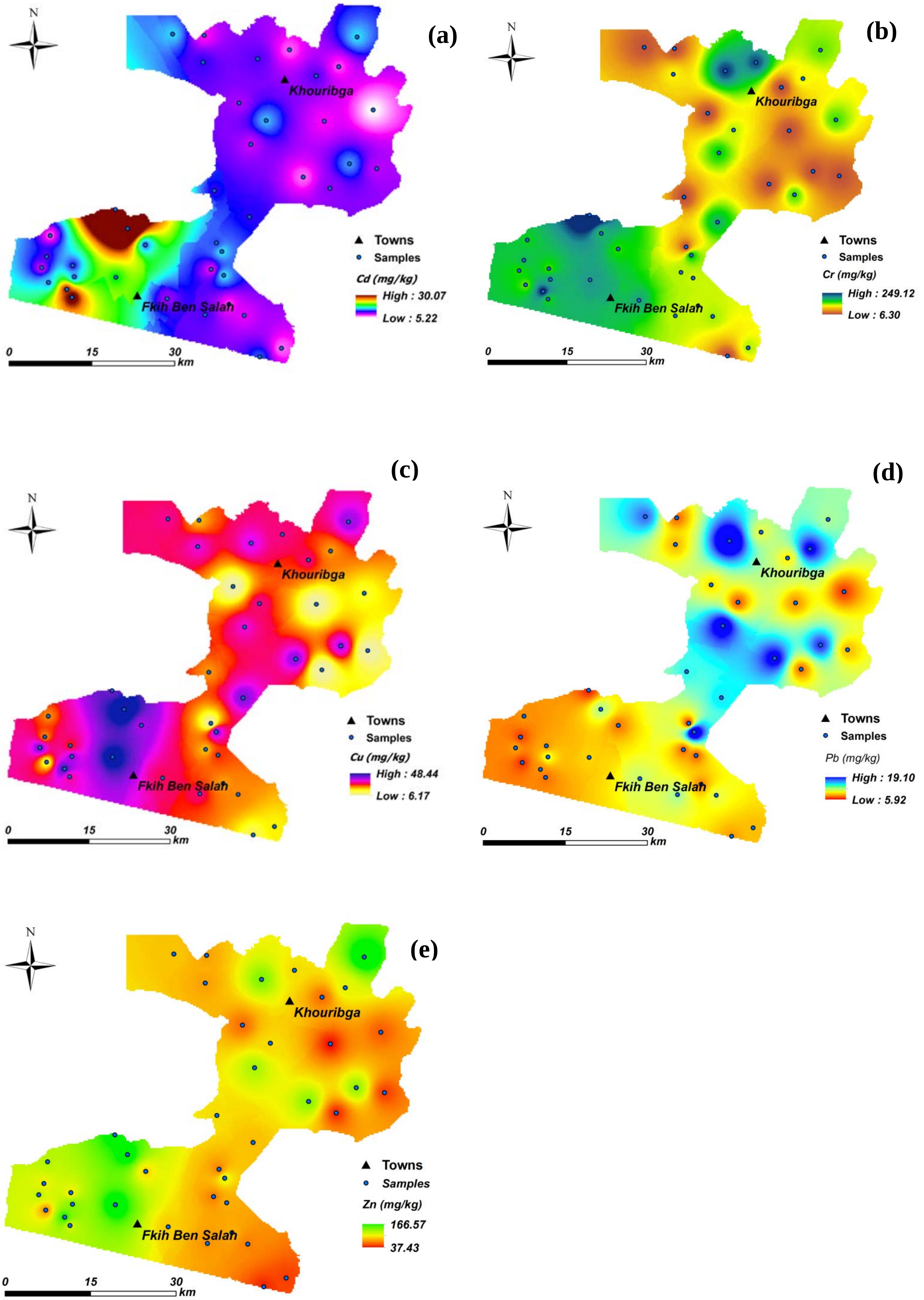
(a)
N
Khouribga
Fkih Ben Salah
Towns
Samples
Cd (mg/kg)
High : 30.07
Low : 5.22
0
15
30
km
(b)
N
Khouribga
Fkih Ben Salah
Towns
Samples
Cr (mg/kg)
High : 249.12
Low : 6.30
0
15
30
km
(c)
N
Khouribga
Fkih Ben Salah
Towns
Samples
Cu (mg/kg)
High : 48.44
Low : 6.17
0
15
30
km
(d)
N
Khouribga
Fkih Ben Salah
Towns
Samples
Pb (mg/kg)
High : 19.10
Low : 5.92
0
15
30
km
(e)
N
Khouribga
Fkih Ben Salah
Towns
Samples
Zn (mg/kg)
166.57
37.43
0
15
30
km

Figure 2. Spatial distributions of HM concentrations in soils of the study area.

3.2. HM contamination assessment

Based on their potential in the assessment of HM contamination in soil and sediments, a few environmental indices were used in the present study, namely *Igeo*, *PLI*, and *PI*. The local geochemical background values of the analyzed heavy metals were used to compute *Igeo* and *PLI* indices, while the EU guidelines (Kamunda et al., 2016), relatively higher to FAO and WHO limits (Jahn et al., 2006), were employed to calculate the PI index. The results are summarized in Table 3.

The *Igeo* values of Cd, Cr, Cu, Pb, and Zn are in the range of 2.03 to 4.56, -2.63 to 2.72, -2.93 to 0.04, -3.04 to -1.35, and -0.81 to 1.34, respectively (Table 3). It was observed that that 100%, 30%, and 100% of the samples were moderately to extremely polluted, slightly to severely polluted, slightly to moderately polluted in terms of the *Igeo* of Cd, of Cr, and Zn, respectively. The *Igeo* values of Cu and Pb indicated that studied soils are unpolluted to slightly polluted.

Table 3. Descriptive statistics of I_{geo} values.

	I_{geo}				
	Cd	Cr	Cu	Pb	Zn
Min	2.03	-2.63	-2.93	-3.04	-0.81
Max	4.56	2.72	0.04	-1.35	1.34
Mean	2.66	0.37	-1.08	-2.44	0.30
SD	0.47	1.72	0.75	0.47	0.57
CV (%)	17.69	468.73	-70.10	-19.16	188.54

The HM pollution status in studied soil samples was also evaluated using *PI* and *PLI* methods (Figs 3 and 4). The obtained *PI* values in comparison with EU Guidelines (Kamunda et al., 2016) varied from 1.32 to 7.43 with a mean of 2.14. It indicated slightly, moderately and heavily polluted at 65.85%, 24.39% and 9.76% of all samples (Fig. 4). The *PLI* values obtained in comparison with the local background values ranged from 0.67 to 3.29 with a mean of 1.82. The majority of the sampling sites (85.37%) exhibited very high deteriorated status, and about 14.63% of the soil samples showed no polluted status. Soil HM pollution status using *PLI* index was previously investigated in the Tadla plain forming the southerly limit of the watershed area (Barakat et al., 2020; Ennaji et al., 2020; Hilali et al., 2020; Oumenskou et al., 2018). They are higher, indicating that the pollution status of HMs is very high deteriorated. Elements such as Cd, Pb, and Cr were the major contributors to the HM pollution in this region. These various *PLI* values of these different studies have been associated with natural and anthropogenic origins. The soil and geologic characteristics, agricultural practices (mechanization, fertilization, fungicides, and pesticides), and industrial activities have been considered as the causes of the HM pollution in the region.

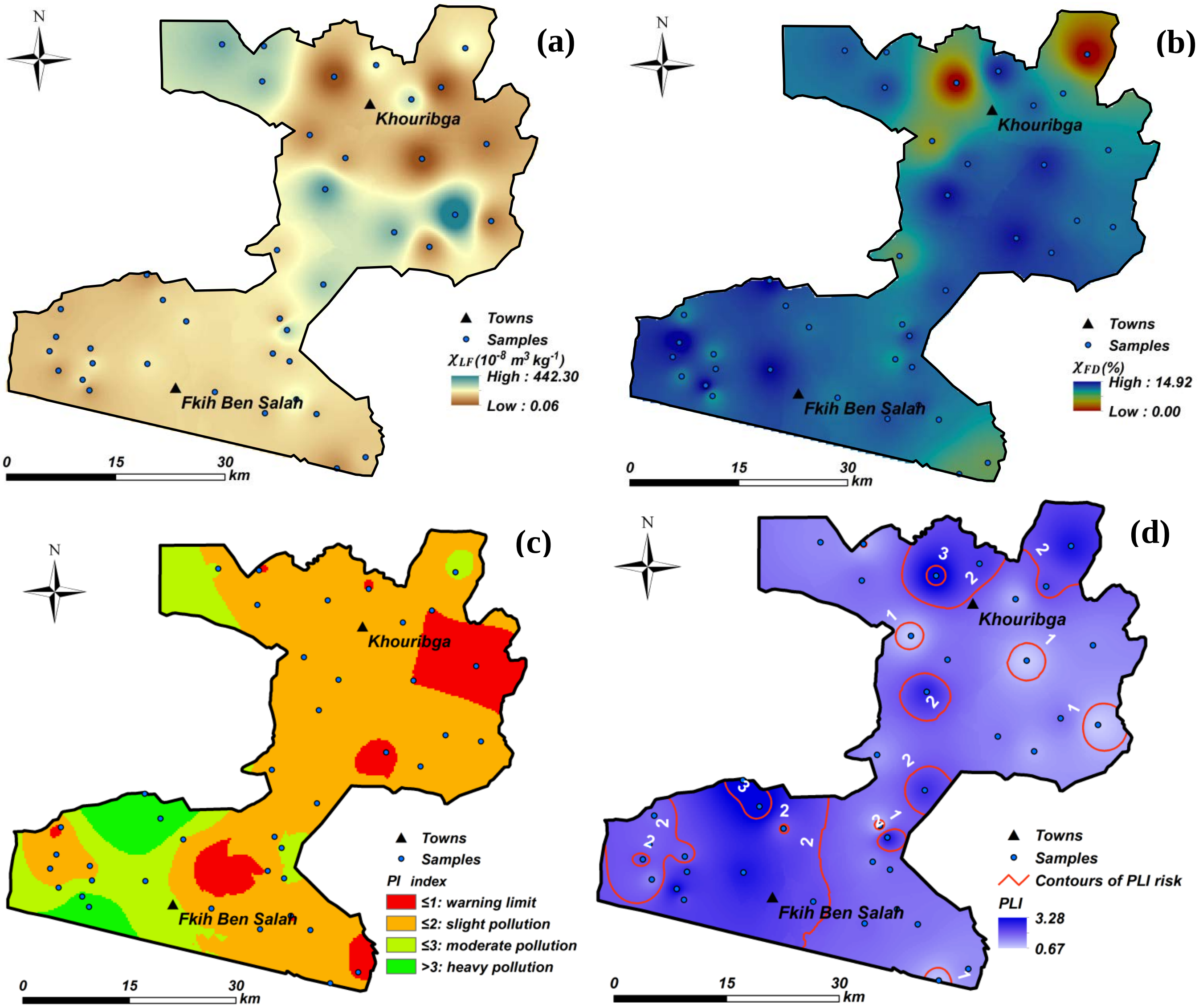

Figure 3. Maps of values of χ_{LF} (a), χ_{FD} (b), *PI* (c) and *PLI* (d).

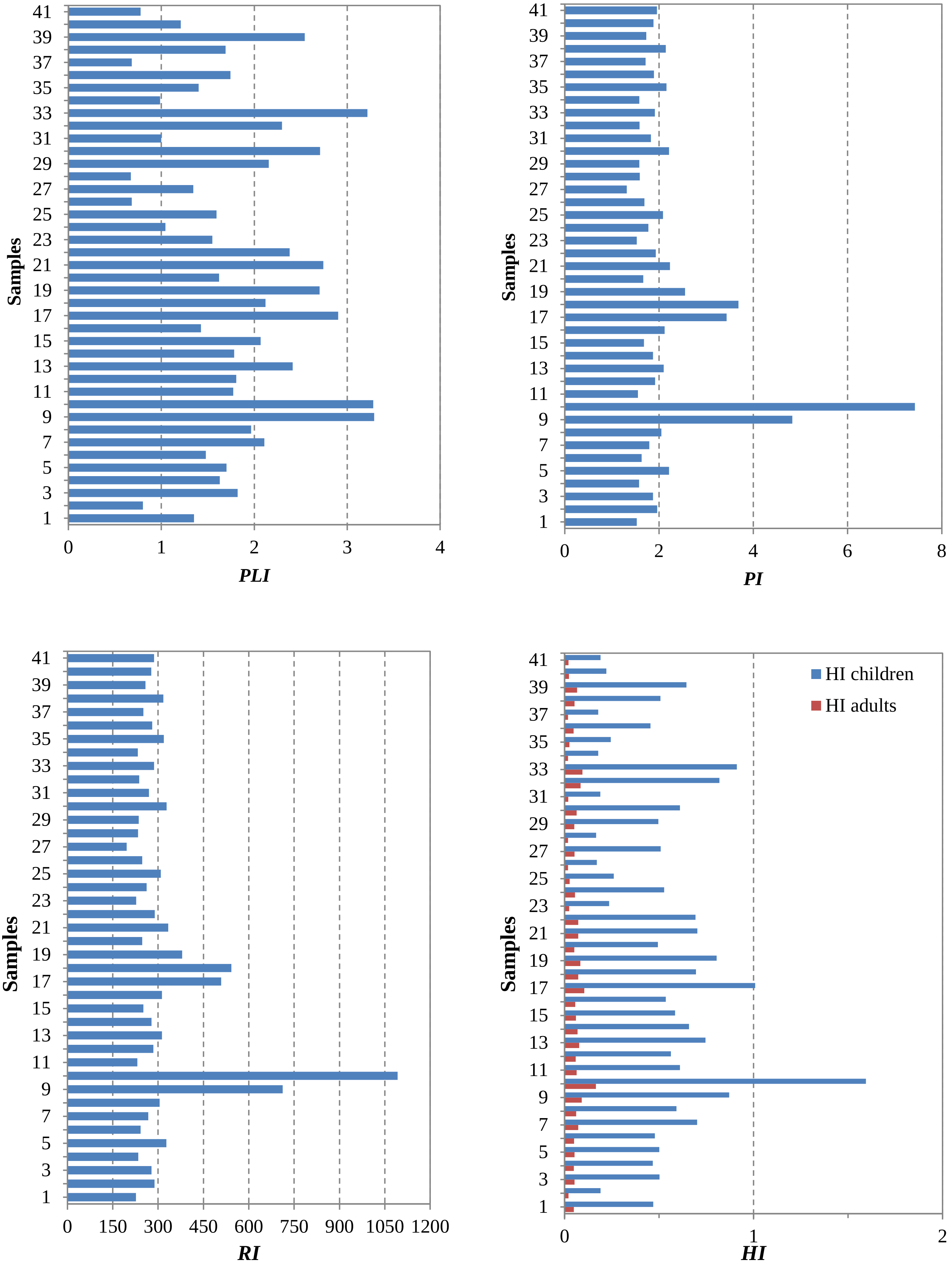

Figure 4. Values of the *PLI*, *PI*, *RI* and *HI* indices in the studied soils.

Assessment of ecological and health risks

The E_r was computed of all the analyzed HM ordered generally as follows: Cd>Cr> Cu>Zn>Pb. Cd was the dominant contributor to the ecological risks. The E_r values of all metals, except Cd, were lower than 40, confirming that these elements do not pose unacceptable ecological risks (Table 4). The ecological risk evaluated by *RI* index (195.93<RI<1092.53) showed that 34.15% of all soil samples had considerable to high ecological risk, whereas this risk was moderate at 65.85% of the sampling sites. All analyzed HM showing contents more than the local reference contents appear to contribute to the RI values, with varying degrees.

The non-carcinogenic health risk associated with the HM ingestion in studied soils were investigated using HQ and HI indices. The results summarized in Table 4 showed the *HQ* of HMs for non-carcinogenic risk in the studied agricultural soils shown as follows: *HQ*Cr > *HQ*Cd > *HQ*Zn > *HQ*Pb > *HQ*Cu. The HQ and HI values for adults were less than 1, indicating that adults were at risk of non-carcinogenic effects from HM ingestion exposure in the study area. For children, the *HQ* and *H*I, except the *HQ*Cr and HI in samples S10 and S17, are less than 1. The *HQ*Cr and *HI* in samples S10 and S17 more than 1 suggests that the child population would be exposed to non-carcinogenic health risk due to heavy metal exposure to soil. Although most samples showed the lowest levels of *HQ* and HI, suggesting that the intake of HMs in studied soils is safe for the children inhabitants, the *HQ*Cr and *HI* in samples S10 and S17 indicated the accumulated metals in soils would pose non-carcinogenic health risks to the local children. Similar observations have previously been reported in the region by Barakat et al. (2019a) in the suburban soils of Beni Mellal city.

Table 4. Descriptive data of E_r^i and *RI* for ecological hazard and *HQ* and *HI* for non-carcinogenic risk of HMs in studied soil samples (n=41).

	E_r^i					***RI***
	Cd	Cr	Cu	Pb	Zn	
Min	184.24	0.48	0.98	0.91	0.86	195.93
Max	1062.71	19.79	7.72	2.94	3.81	1092.53
Mean	303.60	6.11	3.97	1.46	1.99	317.13
SD	152.00	4.24	1.61	0.54	0.79	155.49
CV (%)	50.07	69.36	40.52	37.10	39.42	49.03
Children	**HQ**					***HI***
Min	6.67E-02	2.71E-02	2.06E-03	2.26E-02	1.66E-02	1.67E-01
Max	3.85E-01	1.11E+00	1.62E-02	7.29E-02	7.41E-02	1.59E+00
Mean	1.10E-01	3.43E-01	8.30E-03	3.62E-02	3.88E-02	5.36E-01
SD	5.51E-02	2.38E-01	3.36E-03	1.34E-02	1.53E-02	2.83E-01
CV (%)	5.01E+01	6.94E+01	4.05E+01	3.71E+01	3.94E+01	5.28E+01
Adult						
Min	7.15E-03	2.78E-03	2.11E-04	2.32E-03	1.71E-03	1.75E-02
Max	4.12E-02	1.14E-01	1.66E-03	7.49E-03	7.61E-03	1.66E-01
Mean	1.18E-02	3.52E-02	8.53E-04	3.72E-03	3.98E-03	5.55E-02
SD	5.90E-03	2.44E-02	3.46E-04	1.38E-03	1.57E-03	2.93E-02

CV (%)	5.01E+01	6.94E+01	4.05E+01	3.71E+01	3.94E+01	5.27E+01

3.3.Statistical analyses

Statistical analyses of the HM contents, MS data, and physicochemical parameters were carried out, and the correlation coefficients were obtained to evaluate the relationship between and MS data, HM contents, and physicochemical parameters in the topsoil samples. Before proceeding with the statistical tests, the compatibility of the distributions with the normality assumption was verified by the Shapiro-Wilk test. The normality test ($p> 0.05$) results showed that pH, OM, Cu, χ_{FD}, χ_{LF}, and *PLI* followed a normal distribution, and all other parameters did have a non-normal distribution. Therefore, the logarithmic transformation of the data was performed to approximately stabilize variances in the correlation analysis.

The correlation coefficient (R^2) values indicated a clear relationship between the heavy metals. The results showed that Fe exhibited a significant positive correlation (+0.75) with Pb that displayed normal concentrations, suggesting the same pollution source. Moreover, except Cd that displayed a negative correlation with Fe (-0.14) and Pb (-0.11), linear correlations showed significant positive correlations between all trace elements in soil samples, among which Pb seemed not influenced by human activities. This proved that Zn, Cr, Cu, Zn, and Cd contamination was ascribed to both soil parent materials and anthropogenic sources related to phosphate mining and agricultural activities. The coefficient of variation (CV) values of HMs in soil samples varying between 33.5 and 69.36% (Table 5), indicating moderate to high variations of HM contents in the study area. CV values of Zn, Cu, Pb, and Fe that were 39.42, 40.52, 37.10, and 33.50 respectively, indicated that human activities had moderate effects on the HM accumulation in studied soils. The highest CV values of 69.36 and 50.07% of Cr and Cd contents, respectively, indicated the greatest variation of Cr and Cd contents that would be influenced by external factors including mining and agricultural activities, especially the intense application of phosphate fertilizer as well documented in the literature (Atafar et al., 2010). Several local studies revealed major contributions of the anthropogenic sources to polluting farmlands with heavy metals. For instance, Cd, Cr, Zn, Cu, and Pb pollution has been reported in urban soils along the main roads in Beni-Mellal City, Morocco (El Baghdadi et al., 2012), in suburban soils irrigated by wastewater of Beni-Mellal City (Morocco) (Barakat et al., 2019a), and in agricultural soils of Tadla plain (Morocco) (Ennaji et al., 2020; Oumenskou et al., 2018). These studies have attributed these increases in HM levels to the traffic density, urbanization, growth of industry, and intensive agriculture.

As well documented in the literature, the HM accumulation in soils is largely dependent on the soil parameters. Ennaji et al. (2020) reported the same relationship between CaCO3 and HM accumulation in agricultural soils of NE part of Tadla plain, Morocco. Even though they are low, the positive linear interdependence between HMs-Clay and HMs-OM indicated that soil OM and texture played a role in determining the availability and mobility of HMs in soil. These results are consistent with those obtained by some authors (Barakat et al., 2020; El Baghdadi et al., 2012; Ennaji et al., 2020; Oumenskou et al., 2018) for local agricultural soils. Moreover, Zhao et al. (2020) reported the relationship between HM (Cd, Cu, Zn, Ni, and Cr) contents and soil properties of soils. In the present study, only weak correlation relationships were found between HM contents and studied soil physical parameters (pH, EC, clay, and OM) (Table 5). There is also a significant negative correlation between HMs and $CaCO_3$, suggesting that HMs are not linked to carbonaceous soil material. Therefore, the lack of a close correlation between HMs and the soil physical parameters suggested that HMS in studied soils originated especially from anthropogenic activities.

Correlation between MS and HM concentrations widely used as an indicator of HM pollution was also analyzed for the soil samples. As shown in Table 5, the results revealed that χ_{FD} and χ_{LF} are positively correlated with the HM contents, except Cd and Cr that exhibited a negative relationship with χ_{LF}. A good positive correlation (R^2=0.88) was also observed between χ_{LF} and χ_{LF}-χ_{HF}, suggesting that the studied soil contains a single population of magnetic minerals. Moreover, a significant positive correlation was observed between HMs and MS suggested that the Fe oxide minerals are the predominant magnetic carrier in the sampled soils. The significant correlation of Fe with HM contents confirmed therefore that the Fe oxide minerals facilitate the HM accumulation in studied soils. The HMs could be integrated into the structures and/or adsorbed onto surfaces of iron oxide minerals (Cornell & Schwertmann, 2003). The correlation between HM and MS has been reported in some research works on the region. El Baghdadi et al. (2012) observed for urban soils on Beni-Mellal city (Morocco) a strong correlation between MS and Pb, Cu, and Zn contents attributed to traffic density. El Hamzaoui et al. (2020) showed for agricultural soils of irrigated Tadla perimeter (Morocco)a very weak correlation between MS and Cu, Fe and Zn high levels coinciding with areas of intense activity; Sugar Plant, poultry intestines washing plant, and intensively cultivated land.

The relationships of HM (Cd, Cr, Cu, Pb, and Zn) concentrations with *PI*, *PLI*, *RI*, and *HI* in different soil samples were also studied, and are illustrated in Table 5. Furthermost of the indices showed significant ($p<0.05$) correlations with each other, and with HM contents. Except for Pb and Fe, the PI had substantial relationships with all studied indices, notably for Cd and Cr in which its lowest R^2 value was 0.54. Meanwhile, there are significant positive correlations between HMs and *PLI* that are above 0.44, except for Pb. Similarly, significant positive correlations occurred between HMs, apart from Pb, and *RI*, and *HI* indices. According to R^2 values (>0.5), Cd and Cu are the most important ecological risk, and Cr, Zn, and Cu are the most important health risk in the studied soils. There are also positive and significant relationships between MS and *PLI*, RI, and *HI* indices, especially with χ_{FD} (R^2>0.47), suggesting that the rapid and cost-effective magnetic techniques could provide valuable information about the soil metal enrichment.

Table 5. Correlation coefficients between HM concentrations, physical parameters, χ_{FD}, *PI, PLI, RI, and HI* in studied soil samples (n = 41).

Variables	Silt	Clay	SOM	CaCO3	Zn	Cd	Cr	Cu	Pb	Fe	χ_{FD} (%)	χ_{LF} (10^{-8} m^3 kg^{-1})	*PI*	*PLI*	*RI*	*HI* children
Zn	0.10	-0.11	0.17	**-0.78**	**1.00**											
Cd	0.13	-0.20	0.07	-0.20	**0.42**	**1.00**										
Cr	0.02	-0.06	0.09	**-0.56**	**0.58**	0.28	**1.00**									
Cu	0.04	-0.06	0.20	**-0.74**	**0.82**	**0.51**	**0.54**	**1.00**								
Pb	-0.13	0.13	0.18	**-0.49**	**0.33**	-0.06	-0.02	**0.40**	**1.00**							
Fe	0.06	0.11	0.14	**-0.82**	**0.62**	-0.01	0.26	**0.60**	**0.67**	**1.00**						
χ_{FD} (%)	0.10	-0.15	**0.58**	**-0.41**	**0.46**	0.19	**0.46**	**0.59**	0.13	**0.35**	**1.00**					
χ_{LF} (10^{-8} m^3 kg^{-1})	0.15	-0.08	0.27	**-0.49**	**0.39**	0.11	0.04	**0.58**	**0.51**	**0.70**	**0.37**	**1.00**				
PI	0.12	-0.21	0.10	-0.24	**0.45**	**1.00**	**0.34**	**0.54**	-0.04	0.02	0.23	0.13	**1.00**			
PLI	0.02	0.02	0.05	**-0.73**	**0.81**	**0.44**	**0.86**	**0.80**	0.23	**0.51**	**0.48**	0.25	**0.49**	**1.00**		
RI	0.12	-0.17	0.10	-0.29	**0.48**	**0.99**	**0.38**	**0.58**	-0.01	0.07	0.26	0.15	**1.00**	**0.54**	**1.00**	
***HI* children**	0.02	-0.02	0.10	**-0.64**	**0.70**	**0.40**	**0.97**	**0.65**	0.07	**0.34**	**0.47**	0.10	**0.46**	**0.93**	**0.50**	**1.00**
***HI* adults**	0.01	-0.03	0.11	**-0.64**	**0.70**	**0.41**	**0.97**	**0.65**	0.07	**0.34**	**0.47**	0.10	**0.46**	**0.93**	**0.51**	**1.00**

For further assessment of the relationships between HMs and to identify the HM source pollution in the study area, PCA technique classifying variables highly correlated into the same class is employed. The PCA results of HM concentrations are shown in Table 6. Five principal components with eigenvalues greater than 1 were extracted by varimax rotation.

The first component had moderate to high positive loadings on *RI*, Fe, *PL*I, χ_{FD}, *HI*, Pb, Zn, and Cr with a total variance of 21.37%, indicating that the PLI and ecological and non-carcinogenic risk levels are more sensitive and vulnerable to Cr, Zn, and Pb. The second component accounted for 18.29% with a high positive loading of Fe, Cu, Zn, Pb and *PLI*, while the third component was strongly to moderate loaded in Cd, PI, RI, Pb, *PLI*, and *HI* with a variance of 57.56%. Considering the significant negative loadings of $CaCO_3$ observed in these three components and the slight loadings of other soil physical parameters, the HM contamination in these components may not be of natural origin, but it would be largely

Table 6. Varimax rotated component matrix of physical and HM data and environmental indices.

	PC1	PC2	PC3	PC4	PC5
Sand	0.04	-0.24	0.12	0.37	-0.64
Silt	0.00	0.15	0.10	-0.06	0.92
Clay	0.17	0.02	-0.09	-0.21	-0.75
pH	0.10	-0.21	-0.08	-0.88	0.11
EC	-0.18	-0.09	-0.05	0.82	-0.19
OM	0.08	-0.07	0.06	0.68	0.07
$CaCO_3$	-0.33	-0.79	-0.29	0.04	-0.01
Cd	0.20	0.03	0.97	0.03	0.05
Cr	0.93	0.08	0.12	-0.17	-0.18
Zn	0.48	0.68	0.29	0.10	0.11
Cu	-0.13	0.83	-0.15	0.08	0.14
Pb	0.42	0.65	0.45	0.11	0.01
Fe	0.26	0.88	-0.17	0.08	0.16
χ_{FD}	0.64	0.10	0.05	0.38	0.32
χ_{LF}	0.02	0.33	-0.03	0.65	0.22
PLI	0.74	0.58	0.31	-0.02	-0.02
PI	0.23	0.04	0.97	0.03	0.04
RI	0.24	0.05	0.96	0.03	0.04
HI children	0.90	0.20	0.30	-0.10	-0.13
HI adults	0.90	0.20	0.30	-0.10	-0.13

attributed to anthropogenic activities like discussed above (agriculture, livestock and phosphate mining). The fourth and five components accounted respectively 13.66% and

10.79% of total variance due to high positive lodgings of EC, OM, Silt and MS. The significant loadings of soil properties meant that these components may be associated with soil-forming processes. Also, the close association of soil properties with MS suggested that the geologic materials contributed with HM to the enrichment of magnetic particles in the studied soils.

In summary, the results of the statistical analyses indicated that soils from southeastern phosphates plateaus of Khouribga (Morocco) are polluted by anthropogenic activities.

4. Conclusion

In the present study, 41 soil samples from agricultural soils from southeastern phosphates plateaus of Khouribga (Morocco) had been analyzed for assessing HM concentration, spatial enrichment, sources apportionment, ecological risk and health diseases by combining various indices, namely I_{geo}, *PI, PLI, RI, HI* and MS and GIS methods.

The mean contents of Cd, Cr, Cu, and Zn were below the local background values at about 100%, 73.17%, 29.27%, and 92.68% of the soil samples, respectively. They decrease in the order of Zn> Cr> Cu> Pb> Cd. According to the relatively high CV values, especially of Cr and Cd, the HM contents in farmland soils are influenced by external factors including mining and agricultural activities. A weak correlation relationship was observed between HM contents and studied soil physical parameters (pH, EC, clay, and OM). There is also a significant negative correlation between HMs and $CaCO_3$, suggesting that HMs are not linked to carbonaceous soil material. The I_{geo} values of most of the metals were >1 in many sampling sites suggesting that HMs have accumulated in the surface soil. The *PI* and *PLI* indices confirmed that the studied soils were polluted by HMs to different levels. The χ_{LF} and χ_{FD} values often showed a similar spatial pattern of HM and *PLI* distributions. The slightly higher to higher χ_{FD} values observed at 95% of the studied samples, indicated a dominance of the MD grains than the SP particles, and therefore suggested that magnetic minerals might originate from anthropogenic activities rather than pedogenesis. Furthermore, the HM source identification was analyzed by combining the pollution indices to coefficients of correlation (R^2) and variation (CV) analysis. The sources of HMs are both from natural materials and anthropogenic activities. Human-induced activities, including agriculture and dust from phosphate mining, were the main sources of pollution in the study area. Adopted to assess the ecological risk of HM, the *RI* index with values of 195.93-1092.53 revealed that the studied agricultural soils are moderately to highly polluted. Furthermore, regarding the results of *HI* index, there is no potential non-carcinogenesis risk of soil ingestion for adults, but more attention is required to avoid the acceptable level of non-carcinogenic hazard to children in the study area.

Finally, whether agricultural practices in the study area seeking to increase production continue, they will lead to severe HM pollution that could have negative effects on the environment and human health. However, continuous HM assessing appears to be critical for sustainable management and conservation of soils. After comparison between HM contents, pollutant indices, and MS, the *P*LI and MS seemed accurate and effective for identifying the HM sources.

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