

The occurrence of heavy metals in irrigated and non-irrigated arable soils, NW Albania

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Abstract The study analysed the content of heavy metals in surface soil and sediment samples from the Bregu i Matit Plain in NW Albania in relation to irrigation in order to evaluate the soil pollution and the potential risk to human health. Evaluation of soil pollution was performed using the enrichment factor and geo-accumulation index. Contents of cadmium, chromium and nickel of irrigated soils were significantly higher than those of non-irrigated soil, while contents of lead (in three of the irrigated locations), zinc and arsenic (in one of the irrigated locations) were significantly lower. Correlation analysis (CA) and principal component analysis (PCA) indicated that the primary source of the first three metals was irrigation, and the last three metals were originated from other anthropic sources, like the use of chemicals, etc. Enrichment factor (E_f) calculation showed that irrigated soils were most enriched in cadmium, chromium, copper and nickel. Index of geo-accumulation (I_{geo}) revealed that arable soils of Bregu i Matit are unpolluted to moderately polluted with cadmium, chromium, copper and zinc and moderately to strongly polluted with nickel and arsenic. The presence

of heavy metals in the studied soils indicates a potential risk of transfer of these elements in the food chain. Therefore, further studies on the speciation of heavy metals in the studied soils in order to evaluate their mobility are needed.

Keywords E_f · Heavy metal · I_{geo} · Irrigated soil · Non-irrigated soil · Albania

Introduction

Accumulation of heavy metals in soil may pose risks to human health through food chain. This issue has been a worldwide subject of environmental research (Felix-Henningsen et al. 2010; Jia et al. 2010; Aydinalp et al. 2010; Prabu 2009; Mukherjee and Mishra 2008; Salf et al. 2005).

The amount of urban and industrial waste water discharged and returned to continental water in Albania is about 0.3 km³/year (Kamizoulis et al. 2004), which is used for irrigation (Massoud et al. 2003). Although some studies have been conducted on heavy metal pollution of soils and crops in industrial and agricultural areas (Topi et al. 2012; Gjoka et al. 2002, 2011; Belalla et al. 2010; Lazo et al. 2003; Saraçi et al. 1995), arable soils irrigated with waste water have not yet been studied. Soils of the Bregu i Matit Plain are threatened by heavy metal pollution associated mainly with irrigation water supplied by a channel system which is fed by the adjacent Mati River. This river is

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polluted from effluents released from industrial units and settlements situated along the Mati basin (Gjoka et al. 2010). Accordingly, a risk of soil and crop pollution exists which is not yet assessed. A previous investigation has shown high levels of crop available elements such as copper, manganese and zinc in soil samples collected from this area (Gjoka 1994), but there have been no detailed studies for heavy metal pollution of soils and the transfer to crops. Therefore, this study aims to analyse the content of heavy metals such as cadmium, chromium, copper, nickel, lead, zinc and arsenic in irrigated and non-irrigated arable soils and in irrigation channel sediment from the Bregu i Matit Plain in NW Albania in order to evaluate the soil pollution and the potential risk to human health.

Materials and methods

Study area (41° 42' 47" N and 19° 39' 16" E) is located in the Bregu i Matit Plain, NW Albania. Geomorphology of the area represents a product of the alluvium sedimentation transported by the Mati River in the south and Drini River in the north. Detritic materials, which constitute this plain area, are derived from the eastern geological terrains which consist mainly of ophiolitic rocks (peridotite, pyroxenite, gabbro, plagiogranite, volcanic rocks) that are rich in Cr, Cu, Ni, Co, etc. In addition, the Triassic, Jurassic and Cretaceous limestones are widespread followed by molassic and flyschoidal formations. Soils in this area belong to Eutric Fluvisols and Haplic Cambisols (FAO 2006).

Four locations (villages) were selected for this study: location 1 without irrigation (L1-Pllane) was selected as control (plot planted to alfalfa), location 2 (L2-Gajush) and location 3 (L3-Shenkoll) are the more irrigated locations (plots are irrigated six to eight times per year and planted to maize and vegetables), and location 4 (L4-Rrile) is the less irrigated location (plots are irrigated three to four times per year and planted with wheat, sunflower, maize and fodder). According to a local expert, the irrigation in the study area is applied in the period 1966–1995, and the amount of water used for irrigation has been 600–800 m³/ha.

Fifteen surface soil samples (one from L1, four from L2, six from L3 and four from L4) and one sediment sample from irrigation channel were taken (Fig. 1). At each sampling point, five subsamples were taken and mixed together to obtain a composite sample. Samples were collected into polyethylene bags. Air-dried samples were passed through a stainless steel sieve with a 2-mm mesh. Finely ground samples were prepared by grinding a subsample of <2-mm soil to a fine powder in a ball mill. All samples were analysed for soil pH, carbonates, total carbon, total nitrogen, cation exchange capacity, texture and total contents of cadmium, chromium, nickel, lead, arsenic, copper and zinc in the labs of Institute of Soil Science and Soil Conservation of the Justus-Liebig University Giessen (Germany).

Soil pH was measured in a solution of soil and 0.01 M CaCl₂ at a ratio of 1:2.5, using a pH meter (DIN ISO 10390). Content of carbonates was determined by the gas volumetric method using a calcimeter (DIN 18129). Total carbon (Ct) and nitrogen (Nt) were determined by the gas chromatography using a CNS element analyzer (Heraeus). Cation exchange capacity (CEC_{pot}) as the sum of exchangeable cations was measured according to Mehlich (1948) in buffered BaCl₂-TEA solution (0.1 M, pH 8.2), percolation for 8 h, using an atomic adsorption spectrometer (Varian AA240FS) (as described by Schlichting et al. 1995). Texture was determined by sieving and pipetting method after destroying humus with H₂O₂ and carbonates with HCl and disaggregation with Na pyrophosphate (DIN 19683). Total contents of heavy metals were extracted with aqua regia (DIN ISO 11466) and measured by inductively coupled plasma–optical emission spectrometry (ICP–OES). Precision of the analyses was checked by analysing a certified reference sample (CRS).

Data analysis

Level of soil pollution with heavy metals is evaluated based on comparison with critical values by BBodSchV (1999), maximum permissible levels by the 86/278/EEC Directive (CEC 1986) and calculation of enrichment factor (E_f) (Yu et al. 2008) and geo-accumulation index (I_{geo}) (Ho et al. 2010). Enrichment factor is calculated using the formula

$$E_f = \text{Metal content in a given soil} / \text{Metal content in the earth's crust.} \quad (1)$$

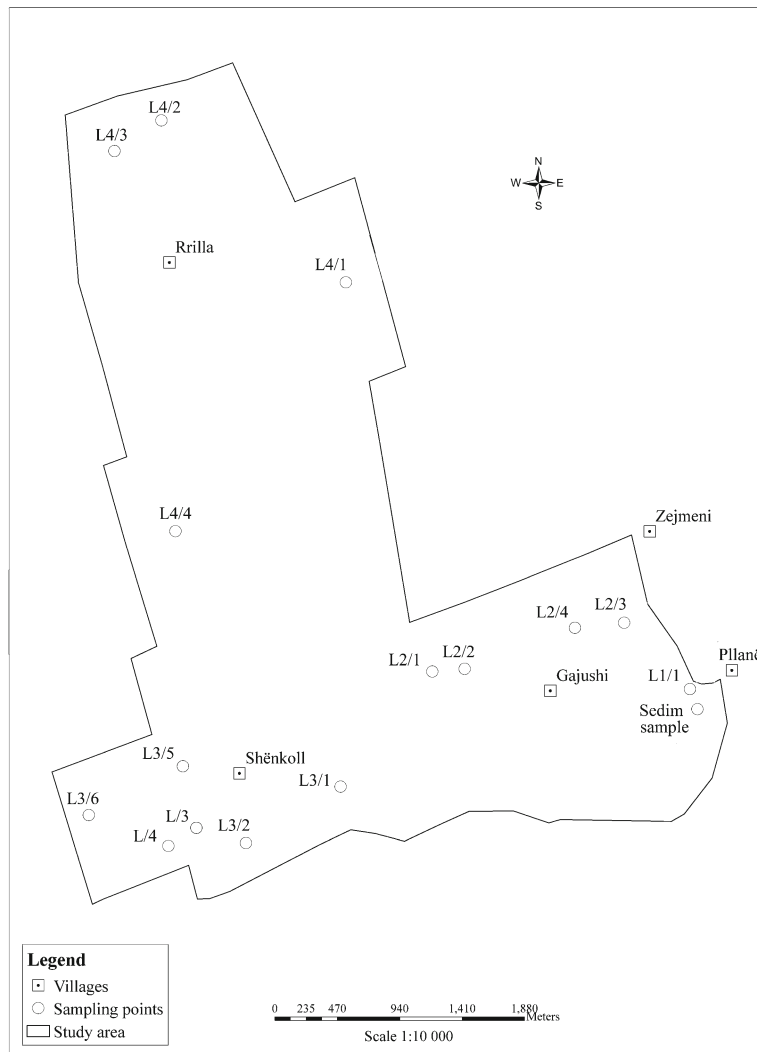


Fig. 1 Location of sampling points

In this study, contents of heavy metals (cadmium, chromium, copper, nickel, lead, zinc and arsenic) in the earth’s crust are taken from Rudnick and Gao (2005), while geo-accumulation index is calculated using the formula

$$I_{geo} = \log_2(\text{Metal content in a given soil/} \quad (2)$$

Metal content in the earth’s crust $\times 1.5$) where 1.5 is used to compensate for possible variations of the reference data due to lithogenic effects.

One-way analysis of variance (ANOVA) and least significant difference (LSD) test were used to find out statistical differences between sampling locations. The significant threshold was $p < 0.05$. Correlation analysis (CA) and principal component analysis (PCA) were used to investigate the role of

soil properties on heavy metal content of soils and their origin. Statistical analysis was conducted using SPSS 19.0.

Results and discussion

Soil properties

Selected properties of the studied soils are shown in Table 1. Soil pH was slightly acid to neutral. Its mean values ranged from 6.15 in L1 (control) to 7.44 in L3/1. This parameter plays an important role in the mobility of heavy metals in soils and their bioavailability to crops. Carbonate content was generally low, ranging from

Table 1 Selected properties of surface soils (0–25 cm) from the study area

Location/sampling point	pH (CaCl ₂)	CaCO ₃ (%)	OC (%)	Nt (%)	P-av. (mg/100 g)	K-av. (mg/100 g)	CEC (+ cmol kg ⁻¹)	Clay (%)
L1/1	6.15	0.8	1.23	0.16	1.54	4.82	17.46	22
L2/1	7.32	2.32	1.38	0.16	1.66	5.31	25.61	20.2
L2/2	7.41	1.86	1.17	0.13	4.65	3.06	27.36	22.6
L2/3	7.41	0.93	1.41	0.19	2.74	8.65	24.56	30.2
L2/4	7.42	0.89	1.45	0.17	19.37	6.13	28.50	32.6
L3/1	7.44	2.05	1.06	0.12	0.77	2.82	23.16	16.1
L3/2	7.30	0.84	0.97	0.10	1.93	1.64	23.16	16.4
L3/3	7.10	0.35	1.32	0.15	1.61	4.94	26.22	22.6
L3/4	6.72	0.35	1.02	0.11	6.51	2.64	23.44	14.7
L3/5	7.43	3.12	1.20	0.14	2.50	6.87	26.25	21.5
L3/6	7.14	0.91	1.06	0.11	2.31	4.70	25.62	14.3
L4/1	6.49	1.30	1.81	0.21	1.47	15	29.89	38.1
L4/2	6.28	0.80	1.59	0.18	1.18	5.21	34.70	48.7
L4/3	6.98	0.95	1.17	0.13	2.06	2.30	21.19	25
L4/4	7.32	3.41	1.46	0.16	2.84	3.98	22.04	26.6
Min	6.15	0.35	0.97	0.10	0.77	1.64	17.46	14.30
Max	7.44	3.41	1.81	0.21	19.05	15	34.70	48.70
Mean	7.06	1.39	1.29	0.15	3.52	5.20	25.28	24.77
SD	0.44	0.95	0.23	0.03	4.61	3.29	4.04	9.45
CV (%)	6.27	68.29	18.16	21.43	130.22	65.39	13.67	38.13
LSD ($p < 0.05$)	0.51	1.55	0.31	0.04	7.87	5.71	7.80	14.43

SD standard deviation, CV coefficient of variation, OC organic carbon

0.35 % in L3/3 and L3/4 to 3.41 % in L4/4. This probably reflects the effect of acidified irrigation water on the dissolution of carbonates, reducing their content in soil. Total contents of total carbon and nitrogen were medium and ranged from 0.97 and 0.10 % in L3/2 to 1.81 and 0.21 % in L4/1, respectively. These values are typical for soils of this area, indicating farming with little or no organic fertilizer. Generally, cambisols have higher contents of organic carbon and nitrogen than fluvisols.

Contents of plant available phosphorous and potassium were medium to high and ranged from 0.77 mg/100 g in L3/1 to 19.05 mg/100 g in L2/4 and from 1.64 mg/100 g in L3/2 to 15 mg/100 g in L4/1, respectively, depending on the clay content and pH. However, these levels of nutrients in the soil seem to be related to the application of chemical fertilizers (samples were taken from plots where plants are cultivated). The CEC_{pot} was medium to high and ranged from 217.46 cmol(+)kg⁻¹ in L1 to 34.70 cmol(+) kg⁻¹ in L4/2, reflecting the influence of humus and clay content, and perhaps the effect of irrigation on this parameter. Soils

with high CEC can adsorb larger amounts of heavy metals, avoiding their transfer into soil solution. The soils were generally loams or clay loams in texture. The clay content ranged from 14.3 % in L3/6 to 48.7 % in L4/2. There were no significant differences in soil properties between irrigated locations (as the average) and control location, except pH and CEC.

Distribution of heavy metals in surface soils

Mean contents of heavy metals in the studied soils are shown in Table 2. Maximum metal contents were obtained for cadmium (0.195 mg kg⁻¹), chromium (280.94 mg kg⁻¹) and nickel (464.6 mg kg⁻¹) in L4 (less irrigated location) and for copper (73.60 mg kg⁻¹), lead (17.35 mg kg⁻¹) and zinc (114.5 mg kg⁻¹) in L2 (more irrigated location). Cadmium in L4 occurs in contents significantly higher than control (non-irrigated location) while chromium and nickel in three the irrigated locations. Copper in irrigated locations occurs in contents higher, but not significantly than control. Increased

Table 2 Mean total contents (in milligrams per kilogram) of heavy metals in the studied soils

Location/sampling point	Cd	Cr	Cu	Ni	Pb	Zn	As
L1/1	0.12	128.64	54.91	179.22	16.28	102.82	7.33
L2/1	0.16	231.05	70.59	433.34	8.60	81.66	3.31
L2/2	0.16	236.76	73.60	437.17	9.21	86.50	3.46
L2/3	0.13	165.19	50.38	232.65	17.35	114.50	7.05
L2/4	0.16	207.00	72.44	332.66	14.47	110.03	5.83
L3/1	0.14	227.35	65.27	411.36	7.49	75.48	3.23
L3/2	0.16	227.02	61.59	413.61	7.30	72.47	3.09
L3/3	0.18	237.70	67.74	450.06	9.89	88.74	3.87
L3/4	0.13	227.83	73.05	387.21	6.74	80.03	3.16
L3/5	0.15	226.39	64.22	419.05	9.77	78.51	3.73
L3/6	0.13	222.82	68.73	374.61	7.27	81.84	2.51
L4/1	0.20	246.52	69.54	447.77	13.26	99.48	4.56
L4/2	0.18	280.94	70.96	464.60	14.54	95.80	4.71
L4/3	0.13	229.22	41.67	431.89	8.76	72.18	5.33
L4/4	0.18	191.58	39.77	328.81	12.37	80.60	9.28
Min	0.12	165.19	39.77	232.65	6.74	72.18	2.51
Max	0.19	280.94	73.60	464.60	17.35	114.50	9.28
Mean	0.16	225.52	63.54	397.48	10.50	86.99	4.51
SD	0.02	26.29	11.37	62.93	3.32	13.31	1.85
CV (%)	14.08	11.66	17.89	15.83	31.60	15.30	41.17
LSD ($p < 0.05$)	0.04	56.23	21.97	124.87	5.18	20.31	3.03
Irrigation channel/sediment sample	0.272	181.609	128.329	280.916	17.085	170.309	4.962

SD standard deviation, CV coefficient of variation

Table 3 Summary of correlation analysis between heavy metals and soil properties

Variable	Function	Correlation coefficient (R^2)
Cd/organic C	$y=0.0703x+0.0636$	0.471 ^a
Cd/clay	$y=0.0014x+0.1189$	0.315 ^a
Pb/organic C	$y=10.279x-2.3389$	0.460 ^b
Pb/clay	$y=0.2629x+4.3735$	0.495 ^b
Zn/organic C	$y=35.106x+42.873$	0.369 ^b
Zn/clay	$y=0.8346x+67.367$	0.343 ^a
Cu/Cr	$y=0.1625x+27.356$	0.268 ^a
Cr/Ni	$y=0.4118x+61.381$	0.917 ^c
Zn/Pb	$y=3.3779x+51.268$	0.784 ^c
Pb/As	$y=1.3586x+4.5058$	0.552 ^b
Cr/Cd	$y=813.19x+93.837$	0.297 ^a
Cd/Ni	$y=1,867.7x+95.307$	0.289 ^a

^a Correlation is significant at the 0.05 level (two-tailed)

^b Correlation is significant at the 0.01 level (two-tailed)

^c Correlation is significant at the 0.001 level (two-tailed)

contents of cadmium, nickel and chromium in irrigated locations is due to the presence of these metals in Mati River used for irrigation. In fact, Gjoka et al. (2010) and Marku et al. (2010) reported elevated contents of cadmium, nickel and chromium in sediments of this river. Heavy metals in the form of colloidal suspension can be transported to the soil via irrigation water and accumulate in the soil (Aydinalp et al. 2010). High contents of these metals in the sediment sample also support this assertion. Lead, zinc and arsenic in control location occur in contents significantly higher than those in irrigated location (L3). The presence of these metals in

Table 4 Eigenvalues and amount of variance of six factors

Component	Initial eigenvalues		
	Total	Percent of variance	Cumulative (%)
1	8,114.126	96.107	96.107
2	211.335	2.503	98.610

Table 5 Component matrix for heavy metals

Element	PC1
Ni	0.999
Cr	0.968
As	-0.665
Pb	-0.600
Zn	-0.538
Cd	0.537
Cu	0.455

L1 can be from natural sources and agricultural practices other than irrigation, as the use of chemicals, or from road traffic (L1 was roadside soil). Relative accumulation followed the order: nickel (121 %) > chromium (75 %) > cadmium (33 %) > arsenic (27 % at one sampling point) > copper (15 %) > zinc (11 and 7 % at two sampling points) > lead (7 % at one sampling point).

Controlling factors and origin of heavy metals

In order to investigate the role of soil properties on heavy metal contents by soils and to identify the origin of metals, correlation analysis was widely applied (Felix-Henningsen et al. 2010; Jia et al. 2010; Qiu 2010; Maldonado et al. 2008; Qishlaqi and Moore 2007; Bech et al. 2005; Kabata-Pendias 2001; Ritter and Rinefierd 1983). Results of correlation analysis (Table 3) indicate that cadmium, lead and zinc are significantly correlated with organic carbon and clay contents. Cadmium, chromium, copper and nickel indicate a significant correlation with CEC. This means that the variations in the heavy metal contents can be attributed to the variations in the organic matter and clay contents and CEC. Arsenic was not correlated with selected soil properties, suggesting anthropogenic sources for this metal such as the use of arsenic-based pesticides, atmospheric arsenic emissions from copper smelting (copper smelting plant in Laç) and road traffic.

Table 7 Interpretation of soil pollution with heavy metals

Index	Level of pollution
E_f	
≤ 1	No enrichment
$1 < E_f \leq 3$	Minimal enrichment
$3 < E_f \leq 5$	Moderate enrichment
$5 < E_f \leq 10$	Moderately severe enrichment
$10 < E_f \leq 25$	Severe enrichment
$25 < E_f \leq 50$	Very severe enrichment
$E_f > 50$	Extremely severe enrichment
I_{geo}	
< 0	Unpolluted
$0 \leq I_{geo} < 1$	Unpolluted to moderately polluted
$1 \leq I_{geo} < 2$	Moderately polluted
$2 \leq I_{geo} < 3$	Moderately to strongly polluted
$3 \leq I_{geo} < 4$	Strongly polluted
$4 \leq I_{geo} < 5$	Strongly to very strongly polluted
$I_{geo} \geq 5$	Very strongly polluted

Content of cadmium has a significant positive correlation with chromium ($R^2=0.297$, $p<0.05$) and nickel ($R^2=0.289$, $p<0.05$), indicating that these metals have a common origin. Chromium showed a significant correlation with copper ($R^2=0.268$, $p<0.05$), suggesting that copper at some locations has the similar origin with chromium. Content of lead also displayed a significant positive correlation with zinc ($R^2=0.784$, $p<0.001$) and lead with arsenic ($R^2=0.552$, $p<0.01$), indicating that the source of arsenic is different from zinc.

To identify the heavy metal sources, PCA was applied. PCA revealed two components that explained 98.61 % of the total variance and included all heavy metals (Tables 4 and 5). Variables with loading >0.4 were considered as significant.

Contents of nickel, chromium, cadmium and copper had positive loadings (>0.4), while arsenic, lead and

Table 6 Obtained and reference values (in milligrams per kilogram) for soil heavy metals

	Cd	Cr	Cu	Ni	Pb	Zn
Mean metal content in non-irrigated soil	0.12	128.64	54.91	179.22	16.28	102.82
Mean metal content in irrigated soils	0.16	225.08	63.00	395.51	10.91	88.23
86/278/EEC Directive (CEC 1986)	3.0	150	140	75	300	300
BBodSchV (1999)	1.0	60	40	50	70	150

Table 8 Mean contents, average earth’s crust (in milligrams per kilogram), E_f and I_{geo} of heavy metals in the studied soils

	Cd	Cr	Cu	Ni	Pb	Zn	As
Non-irrigated soil							
Mean	0.12	128.64	54.91	179.22	16.28	103.05	7.33
E_f	1.22	1.51	2.20	4.07	0.96	1.45	4.89
Category of pollution	II	II	II	III	I	II	III
I_{geo}	0.88	1.18	1.72	2.61	0.53	1.12	2.87
Category of pollution	II	III	III	IV	II	III	IV
Irrigated soils							
Mean	0.16	225.08	63.00	395.51	10.91	88.23	4.71
E_f	1.60	2.65	2.52	8.99	0.64	1.24	3.14
Category of pollution	II	II	II	IV	I	II	III
I_{geo}	0.09	0.82	0.75	2.58	-1.21	-0.27	1.06
Category of pollution	II	II	II	IV	I	I	III
Average earth’s crust (Rudnick and Gao 2005)	0.098	85	25	44	17	71	1.5

zinc had negative loadings (>0.5). This suggests that the soil contamination with nickel, chromium, cadmium and copper comes from the same source (irrigation), and contamination with arsenic, lead and zinc originates from other sources (underlying geology, the use of agrochemicals or road traffic).

Evaluation of soil pollution

Total mean contents of cadmium, chromium, copper, nickel, lead and zinc in this study were compared with reference values from different sources (Table 6). In irrigated soils, chromium and nickel exceeded the maximum permissible limits of the 86/278/EEC Directive by 150 and 527 %, respectively, while chromium, nickel and copper exceeded the critical values of the BBodSchV by 375, 791 and 158 %, respectively, posing

risks to humans as well as the environment. Continuous cultivation and regular absorption by plants possibly keep the contents of cadmium, lead and zinc in soil within the safe limits in irrigated soils.

Impact of irrigation on soil pollution with heavy metals is further evaluated using the E_f and I_{geo} . Level of soil pollution is determined based on the following categorization of these two quantitative indexes (Ho et al. 2010) (Table 7).

Table 8 and Figs. 2 and 3 show the E_f and I_{geo} values of heavy metal in soils along with the mean contents and average earth’s crust of these metals.

Enrichment factor of the heavy metals in non-irrigated soil showed that lead (0.96) has no enrichment; cadmium (1.22), chromium (1.51), copper (2.20) and zinc (1.45) have minimal enrichment, and arsenic (4.89) and nickel (4.07) have moderate enrichment. In irrigated soils, lead (0.64) has no enrichment; cadmium (1.60),

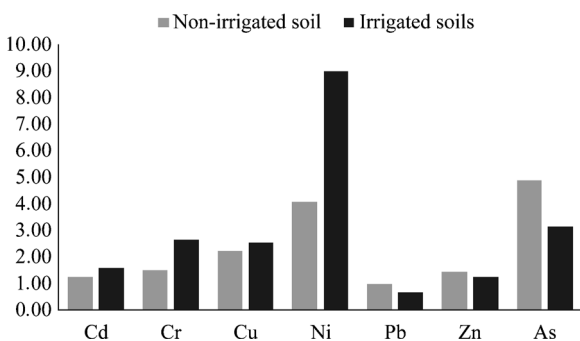


Fig. 2 Enrichment factor (E_f) of heavy metals

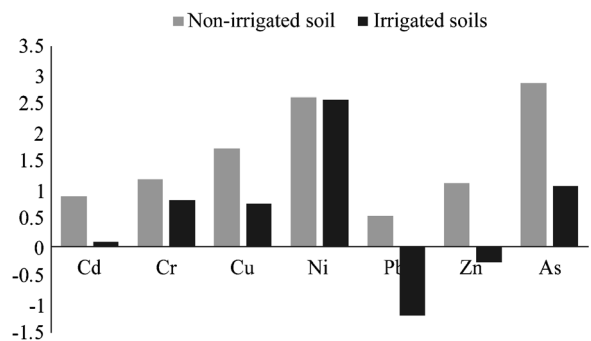


Fig. 3 Geo-accumulation index (I_{geo}) of heavy metals

chromium (2.65), copper (2.52) and zinc (1.24) have minimal enrichment; arsenic (3.14) has moderate enrichment, and nickel (8.99) has moderately severe enrichment. Enrichment values for cadmium, chromium, copper and nickel were significantly higher in irrigated soils than in non-irrigated soils, suggesting an increased contribution of anthropogenic sources for these metals.

Geo-accumulation index shows that the non-irrigated soil was unpolluted to moderately polluted with cadmium (0.88) and lead (0.53); moderately polluted with chromium (1.18), copper (1.72) and zinc (1.12); and moderately to strongly polluted with nickel (2.61) and arsenic (2.87). The irrigated soils were unpolluted with lead (−1.21) and zinc (−0.27); unpolluted to moderately polluted with cadmium (0.09), chromium (0.82) and copper (0.75); moderately polluted with arsenic (1.06); and moderately to strongly polluted with nickel (2.58). Discrepancies in the categories of soil pollution, determined through the use of E_f and I_{geo} can be explained with the different ways of calculating these two indexes.

Conclusions

Results indicated no significant differences in soil chemical–physical properties between sampling locations, except CEC and pH. The significantly higher contents of cadmium, chromium and nickel in the irrigated soils ($p < 0.05$) compared to those in non-irrigated soils indicated that the metal accumulation in the irrigated soils was heavy. Relative accumulation followed the order: nickel > chromium > cadmium > arsenic > copper > zinc > lead. Humus and clay contents play a significant role in heavy metal accumulation in soil. Mean chromium and nickel contents of irrigated soils exceeded the maximum permissible levels of the 86/278/EEC Directive, while chromium, nickel and copper contents exceeded the critical values of the BBodSchV. According to the enrichment factor, cadmium, chromium, copper and nickel contents were most enriched in irrigated soils, suggesting that the irrigation water is the main important source of these metals. This finding is confirmed by the correlation analysis and PCA. Data of geo-accumulation index indicated that arable soils of Bregu i Matit Plain are unpolluted to moderately polluted with cadmium, chromium, copper and zinc and moderately to strongly polluted with nickel and arsenic. Further investigation has to be done to determine the

speciation of heavy metals in the studied soils in order to evaluate their mobility.

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