



Heavy metal contents of contaminated soils and ecological risk assessment in abandoned copper mine harbor in Yedidalga, Northern Cyprus

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Received: 10 October 2017 / Accepted: 12 May 2018 / Published online: 18 May 2018
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Abstract

Total concentrations of heavy metals in the soil from Yedidalga mine harbor, one of the biggest copper export facilities in Cyprus having produced large amounts of mine wastes, were measured to determine the total contents of Cu, Pb, Cr, Cd and Zn. The study also evaluated the level of contamination and assessed the potential ecological risk posed by heavy metals. Soil samples were collected and heavy metal contents were determined using atomic absorption spectrophotometer (AAS). Several quantitative indices were utilized to assess the soil pollution status. Results revealed that comparatively all heavy metals exceeded the background values. The peak values were observed in the soils from the locations close to the Yedidalga farming lands. Geo-accumulation index (I_{geo}) showed that Cu had the highest contamination ($I_{geo} > 3$) among the other heavy metals. Pearson correlation and cluster analysis identified that Cd, Cr and Pb had a relationship, besides the significant correlation between Cu and Zn, the presence of which was mainly related to the mine wastes. Pollution load index (PLI) graded the overall studied area as moderately–heavily contaminated level. Potential ecological risk analysis manifested that the entire ecological risk level indicated that 55.6% of sampling locations exceeded 300 (RI > 300). These study results suggest that pollution control measures must be implemented. The main cause of accumulation of these metals is found to be associated with the presence of dispersed mine wastes in Yedidalga mine harbor.

Keywords Heavy metals · Soil contamination · Pollution assessment · Ecological risk · Yedidalga mine harbor

Introduction

Soil plays an essential role in the global environment and acts as an ultimate reservoir for numerous pollutants (Adriano 2001). Among pollutants, heavy metals are of serious concern worldwide and are considered to be the most hazardous pollutant groups, especially due to their toxicity, ecological risks, non-degradable nature, biogeochemical recycling nature and environmental persistence (Soylak

et al. 2005; Nobil et al. 2010; Zhang et al. 2016). Contamination of soils by trace metals have been a major concern to agricultural products and human health due to their possible transfer into the food chain. In general, heavy metals in soils may possibly be derived either from the weathering of soils (geogenic) or could be induced as a result of various contributions from anthropogenic activities, such as traffic emissions, industrial activities, mining and metallurgical operations and metropolitan wastes (Adriano 2001; Hu et al. 2013). Mining and smelting facilities have been found to be the most significant sources of a variety of toxic and potentially dangerous elements in the surrounding environments, resulting in the contamination of the soils. Particularly, mining operations have severe environmental impacts on the soils and cause persistent toxicological problems for the surrounding environment even after centuries (Zornoza et al. 2012; Khalil et al. 2013).

Hence, accumulation of heavy metal contents in surface soils, due to continued exposure become high and provide perceptible metal contamination that can be equal

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to or higher than the natural environment input in various geochemical forms (Eisler 2004; Hu et al. 2013). This can be associated with environmental quality, particularly in agricultural soils close to mining areas. Consequently, this may not only result in soil contamination, but also adversely affect agricultural productivity and safety, creating potential health risks to the inhabitants (El Hamiani et al. 2010; Moore et al. 2011). Thus, determination of the soil pollution status and assessment of soil heavy metal contamination in abandoned mining sites become a significant issue (Loska et al. 2004).

Lefke district in Northern Cyprus has been an area of severe mining and has been exploited for long periods in search of copper, gold and iron mineralization. Copper–iron mining activities and ore-processing started in the area in the early 1900s and continued until the mid-1970s. Huge quantities of Cu- and Fe sulfide ores were produced between 1930 and 1970s during the operation period predominantly around Lefke (Constantinou 1994). These extensive copper mining operations in Lefke district were massively carried out at the main Gemikonađı site located 50–100 m from the Mediterranean sea coast and used as a port termed Yedidalga mine harbor in the village of Yedidalga (Yükselen 2002). The land area that was subjected to potential contamination has resulted in large heaps of abandoned mine wastes, covering approximately 2000 hectares (Cohen 2002). After abandoning the mining operations in 1974, tons of mine waste (tailings, low grade ore) estimated to be approximately 10 million tons were left behind (Akün 2008). These uncovered mine waste and residual heaps have not undergone any kind of environmental treatment, which also distorted the landscape (Yükselen 2002). These constitute extensive contaminations and the mine wastes left in the area gave way to a permanent heavy metal (Cu, Cd, Pb, Cr, Zn) pollution in the sea and soil environment, which resulted in a serious threat to agricultural land and extended risk inputs to nearby surface waters and ecosystem (Pyatt 2001; Gücel et al. 2009). Nowadays, the Yedidalga harbor area covers a large thick layer of soil with gray and black colors, due to the input of mine wastes. In fact, this comprises a sign of reflection of soil pollution by metals present in mine wastes and transformed the area into a potentially contaminated area. This situation creates serious environmental risks related to agricultural land and surface waters and may finally cause potential health risks to local inhabitants (Zornoza et al. 2012). Thus, it became a critical issue to describe the current situation of heavy metal contamination status and the level of their contents in the surface soil, to suggest worthwhile remediation approaches. However, little information exists on the current status of the harbor area and there are no previous studies in the current area. Being the first research study at this location, it was thus vital to

assess the total concentration of heavy metals and their ecological risks upon agricultural land and ecosystem.

The main objective of the current study was thus to determine the total concentration of selected metals (Cu, Cd, Pb, Cr, and Zn) in topsoil of Yedidalga mine harbor in Northern Cyprus for the first time and to assess the status of their potential hazards to the environment. Different pollution indices: geoaccumulation index (I_{geo}), pollution load index (PLI), potential ecological risk factor (E_r^i), and potential ecological risk index (RI) were used to evaluate the contamination and risks associated with heavy metals in the soil. Multivariate analysis (PCA—principal component analysis; CA—cluster analysis) and correlation matrix were also applied to identify the association between metals and their possible contributing sources.

Materials and methods

Study area

This study was carried out at an old abandoned mine harbor in Yedidalga village in Cyprus. Yedidalga is located at a latitude of 35°08'39.22"N and longitude of 32°48'28.96"E, approximately 40 km west of Nicosia, 3.8 km north of central Lefke and 1.2 km north-west of Gemikonađı (35°08'34"N–32°48'58"E) in the Lefke district (Fig. 1). It displays Mediterranean climate: hot, relatively dry, with rainfall mostly during November to March. The geological setting of the wide area encompasses Troodos Ophiolite Complex and is in compliance with the geological characteristics, consisting of an elongated dome resulting from uplift caused by emplacement of the central serpentinite diapir (Christodoulou 1959). The major land surrounding the harbor is mostly used for agriculture and the surrounding is densely inhabited. There were a number of docks for mine exportation used by some American and English companies in the region. Besides, Yedidalga mine harbor was one of the biggest copper and gold export facilities in North Cyprus, which operated until 1974. After cessation of the mining operations and finally closing down of the harbor facilities in 1974, tons of mine wastes were left behind at the mining sites and exportation harbors. Such huge quantities of mine wastes had resulted in a legacy of abandoned mine wastes at Yedidalga mine harbor, without any environmental treatment (Baycu et al. 2015). Thus, a vast quantity of mine wastes, seriously contaminated by trace metals (Cu, Cd, Pb, Cr, and Zn), could be easily transported from the source area by being exposed to wind and other environmental factors to nearby agricultural lands and the sea.

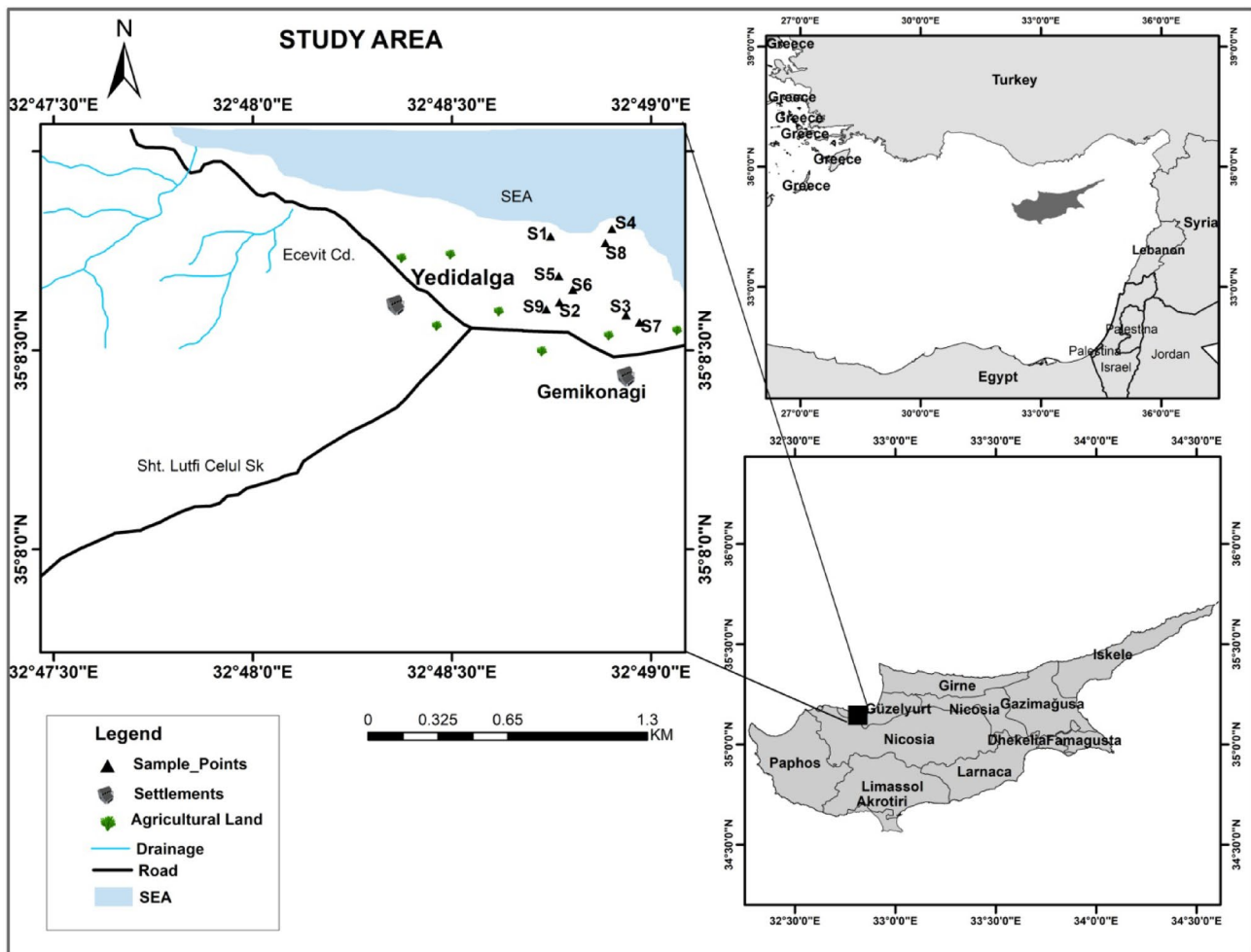


Fig. 1 Map of the study area and sampling locations

Soil sampling and analytical methods

Sampling and preparations

Soil pollutants such as heavy metals display seasonal variations (Suthar et al. 2009). The soil sampling was carried out during the dry period in 2016, collecting a total of 27 representative samples from 9 different sampling locations, 50 m apart from each other, covering the entire harbor area via a grid system designed. During sampling, GPS devices were utilized for recording the exact coordinates of the sampling locations at Yedidalga mine harbor (Fig. 1). According to soil sampling strategy guidelines (Desaules et al. 2001), sampling was carried out in each of these representative locations, equally sub-divided into three and taken at three equivalent points within a 2 m diameter circle. These composite samples were retrieved by employing a spiral auger of 2.5 cm diameter at a depth of 0–20 cm. Besides, further soil samples were collected from another location, to serve

as the control area (unpolluted) which is 40 km away from the abandoned mine area. Right after collection, the samples were placed in polyethylene bags, transported to the laboratory and were air-dried at room temperature for 5 days, homogenized and then sieved through a < 2 mm sieve to remove stones, tree leaves, and coarse debris. Because of the affinity of trace metals toward fine-grained soil components, the samples were pulverized gently with agate pestle and mortar, and less than 63 μm fractions were stored in self-sealing polyethylene bags, to be ready for extraction and acid digestion methods.

Analysis and quality assurance

Soil samples were air-dried and sieved through < 2 mm nylon sieves. The soil pH was measured by a pH meter (InoLab pH 7110-WTW) through a mixing process (1:2 w/v soil water suspension). Organic matter content (OM) was estimated by the mass differential method at 550 °C

in a furnace. Eutech Cond-600 (1:5 w/v soil–water suspensions) detected the electrical conductivity (EC) of soils. All investigations were carried out on the representative sample in triplicate.

To determine the total element concentrations of soil samples (< 63 µm fraction), closed digestion system was employed for the samples. 1 g of each soil sample was digested in a microwave unit (BERGHOF speedwave MWS-2, Germany) using strong acids according to the EPA 3050B method (USEPA 1996). Aqua regia + HF digestion (method B), in conformance with the EN13656 (CEN 2002) technique, was employed in this work. Weighed samples were placed in 100-mL Teflon vessels and a freshly prepared mixture of 2 mL HNO₃ (65%) + 6 mL HCl (37%) and 2 mL HF (40%) was added to the sample, manually shaken for 1 min, and then digested for 40 min. Each digested solution was filtered through a 0.8 µm Millipore membrane, taken into the 25 mL flask and diluted with triply Milli-Q water. A blank digest was carried out in a similar way with an absence of soil sample. The heavy metal (HM) detection was performed by Atomic Absorption Spectrophotometer (Shimadzu AA-7000 Series Model, FAAS). To insure the accuracy of the method and validate the precision, standard reference material (SRM, GBW 08303) for polluted farmland soil was analyzed for corresponding elements. The accuracy of the certified and observed values of the metals is given in Table 1. The detection limit of each selected element was defined and expressed as three times the standard deviation (3 × SD) of ten blanks concentrations. The detection limit values of the observed elements in AAS were found to be 0.12 µg/L for Cu, 0.01 µg/L for Cd, 0.43 µg/L for Pb, 0.07 µg/L for Cr, and 0.02 µg/L for Zn. The instrumental settings and the measurements of metal absorptions were in compliance with the recommended conditions in the user manual. Dilution of the standard stock solutions (Merck) of 1000 mg/L was realized for each trace metal (Pb, Cd, Cr, Cu, and Zn) calibration. Milli-Q water and analytical-grade reagents (Suprapur, Merck) were utilized for all analyses. All the glassware and plastic materials were soaked in 10% (v/v) of diluted HNO₃ overnight for cleaning and then rinsed with Milli-Q water. Quality control protocol approved by the US Environmental Protection Agency (EPA) for heavy

Table 1 The observed and certified values and percentage recovery of elemental concentrations in SRM (GBW 08303 polluted farmland soil) as average ± SD

Element	Observed value (µg/g)	Certified value (µg/g)	Recovery (%)
Cd	1.15 ± 0.05	1.21	95
Cu	114 ± 5.9	110.2	103
Zn	263 ± 13	261.1	100.7
Cr	109 ± 5.4	112.3	97
Pb	74 ± 2.1	73.1	101

metal analysis was employed as a part of quality control. All elemental analyses were performed in triplicate ($n = 3$) and the concentrations were calculated on dry weight (mg/kg dw) basis. According to data calculations so as to ensure the quality of results, the measurement accuracy of concentrations and errors were estimated at ± 5% for all selected elements.

Quantification of soil pollution

Soil pollution status is often assessed by comparing metal concentrations with the relevant geochemical background values or related environmental guidelines. There are various indices for evaluating the degree of HM contamination, and among those indices the index of geoaccumulation (I_{geo}), single contamination factor (C_f^i), and pollution load index (PLI) were used in this study. Geoaccumulation index (I_{geo}) is a tool that enables the evaluation of contamination by comparing the recent and pre-industrial concentrations of the metals in the earth crust (Srinivasa Gowd et al. 2010; Müller 1969). It is calculated using the following mathematical equation (Eq. 1):

$$I_{\text{geo}} = \log_2 \left[\frac{C_n}{1.5 \times B_n} \right], \quad (1)$$

where C_n is the concentration of the measured metal in the soil (mg/kg) and B_n is the geochemical background value of the corresponding metal in the Earth's crust (mg/kg). The factor '1.5' is a constant value, used to detect and analyze the very tiny anthropogenic influences (Fei et al. 2011). The background reference value used in the current study is based on the average continental crust of worldwide soils (Kabata-Pendias 2011). The I_{geo} values proposed by Muller (1969) were interpreted as: $I_{\text{geo}} \leq 0$ —uncontaminated; $0 < I_{\text{geo}} \leq 1$ —uncontaminated to moderately contaminated; $1 < I_{\text{geo}} \leq 2$ —moderately contaminated; $2 < I_{\text{geo}} \leq 3$ —moderately to heavily contaminated; $3 < I_{\text{geo}} \leq 4$ —heavily contaminated; $4 < I_{\text{geo}} \leq 5$ —heavily to extremely contaminated; $I_{\text{geo}} > 5$ —extremely contaminated.

The single contamination factor (C_f^i) was calculated by the mathematical formula (Eq. 2) to assess the degree of contamination in soil.

$$C_f^i = (C_n^i / B_n^i), \quad (2)$$

where C_n^i is a measured concentration of metal i in the soil sample (mg/kg) and B_n^i is the background value of same metal i in the soil (Kabata-Pendias 2011).

The pollution load index (PLI) of the metals proposed by Tomlinson et al. (1980) was calculated by the following formula (Eq. 3):

$$\text{PLI} = (C_{f_1} \times C_{f_2} \times C_{f_3} \times \dots \times C_{f_n})^{1/n}, \quad (3)$$

where n is the number of parameters (i.e., 5 elements) and C_f^i is the single contamination factor of each metal. The

results of the PLI values related to the quality of soil (Singh et al. 2003; Zhang et al. 2011) are shown in Table 2.

Potential ecological risk index (RI), initially introduced by Hakanson (1980), was employed to estimate the degree of metal pollution in soil. RI represents the sensitivity of different biological communities to toxic metals and illustrates the potential eco-risks caused by heavy metals. The integrated RI is calculated as the sum of all single ecological risk factors of heavy metals in soils and is determined as follows (Eqs. 4 and 5):

$$RI = \sum_{i=1}^n E_r^i, \tag{4}$$

where E_r^i is the monomial ecological risk factor of metal i , and T_r^i is the toxic-response factor of metal i according to the toxicity of heavy metals and the response of the environment.

$$E_r^i = T_r^i \times \frac{C_n^i}{C_b^i}. \tag{5}$$

Based on the report of Hakanson (1980), the toxic factor for Cu, Pb is 5, 30 for Cd, 2 for Cr, and 1 for Zn (Yu et al. 2012). C_n^i is the measured concentration of metal i and C_b^i is the background value (Kabata-Pendias 2011) of metal i . The higher the values of E_r^i and RI, the higher are the eco-risks of metals. The categorization of potential ecological risk indices and the corresponding risk grades is summarized in Table 2.

All statistical analysis was performed using SPSS v.20 with assisting OriginPro 9.0 software. GIS mapping technique was used to illustrate the spatial distribution of the PLI and RI in the study locations using ArcGIS 10.4. In addition, Pearson’s correlation, principal component analysis (PCA), and cluster analysis (CA) were also performed. In

the PCA, the principal components were calculated by using varimax-normalized rotation method on the analytical data to estimate the sources. Before execution of cluster analysis, raw data were standardized by means of the Z score (with a mean of 0 and standard variation of 1). The Euclidean distances for similarities were calculated by the following formula (Guan et al. 2014):

$$\begin{cases} S_{ik} = (I_{Ni}RI_i) \\ S_{jk} = (I_{Nj}RI_j) \\ d_{ij} = \sqrt{(I_{Ni} - I_{Nj})^2 + (RI_i - RI_j)^2} \end{cases}. \tag{6}$$

After the hierarchical dendrogram was constructed, totally five heavy metals were then divided into clusters.

Results and discussion

Soil properties

The physicochemical results of the soil samples are listed in Table 3. In general, the total soil samples were widely exposed to distinct soil characteristics. Based on USDA (1993) for soil classification, the pH values across the soils in the entire study area showed a wide range as follows: (1) extremely acidic (3.5–4.4) soils: S1, S4, S5, S7, S8, and S9; (2) strongly acidic (5.1–5.5) soil: S6; and (3) moderately acidic (5.6–6.0) soil: S2. Several factors can influence the acidity of the soil. Within the scope of this study, the acid mine waste drainage from the mine is accepted to be one of the factors responsible for this. Moreover, the chemical reactions related with the ore-processing activities possibly altered the physiochemical properties of the soil (Monaci et al. 2011), while sulfides in mine tailings possibly reacted with water vapor to generate acid waters leading to solubilizing of heavy metals (Moreno and Nerretneiks 2006).

Table 2 Class indices of potential ecological risk index (RI), potential ecological risk factor (E_r^i), contamination factor (C_f^i), and pollution load index (PLI) in relation to soil quality

RI	Risk degree ^a	E_r^i	Pollution degree ^a	C_f^i	Contaminated level ^a	PLI	Soil quality ^b
RI < 150	Low risk	$E_r^i < 40$	Low	$C_f < 1$	Low C_f	0	Background concentration
150 ≤ RI < 300	Moderate risk	$40 \leq E_r^i < 80$	Moderate	$1 \leq C_f < 3$	Moderate C_f	0–1	Unpolluted
300 ≤ RI < 600	Considerable risk	$80 \leq E_r^i < 160$	Considerable	$3 \leq C_f < 6$	Considerable C_f	1–2	Unpolluted to moderately polluted
RI ≥ 600	Very high risk	$160 \leq E_r^i < 320$ $E_r^i \geq 320$	High risk Very high	$C_f \geq 6$	High C_f	2–3 3–4 4–5 > 5	Moderately polluted Moderately to highly polluted Highly polluted Very highly polluted

^aHakanson (1980)

^bSingh et al. (2003), Zhang et al. (2011)

Table 3 Physicochemical properties of the soil samples from the study area

Sample site	pH (H ₂ O)	EC (mS/cm)	OM (%)	Clay (%)	Silt (%)	Sand (%)	USDA texture class ^a
S1	3.41	2.66	0.83	12.21	53.09	34.70	SiL
S2	5.63	2.72	3.36	23.74	19.13	57.13	SCL
S3	4.90	1.41	2.79	22.14	59.23	18.63	SiL
S4	3.89	0.96	1.34	13.97	18.22	67.81	SL
S5	2.12	0.88	3.51	19.15	60.13	20.72	SiL
S6	5.26	0.74	4.06	21.39	26.75	51.86	SCL
S7	1.63	1.47	2.49	17.92	60.73	21.75	CL
S8	3.15	0.70	1.98	24.31	12.28	63.41	SCL
S9	2.47	0.39	0.97	10.54	64.73	24.73	SCL
Control	7.61	0.27	4.78	25.34	30.71	43.95	L

OM organic matter, EC electrical conductivity, SCL sand–clay–loam, SiL silt–loam, CL clay–loam, L loam

^aUSDA (1993)

Besides, neutral pH mean values determined in control soil samples strengthened the above explanation for the acidity of the soil samples analyzed. The average organic matter contents (OM), on the other hand, ranged from 0.83 to 4.06% in the overall study locations. The organic matter contents in harbor soils are found to be much lower than the average organic content in the control soil. In fact, the differences in organic matter reveals a reduction of the organic content in soils due to soil disruptions caused by mine wastes (West and Post 2002; Mccauley et al. 2017). However, some sampling locations (e.g., S2, S5, and S6) showed a normal content (West and Post 2002) because of a slight vegetation cover in some parts in the vicinity. Widely varying high electrical conductivity (EC) values were observed at

the samples. According to USDA triangle for soil classification, the soil samples from the study area were classified as silty-loam and sand–clay–loam, while the soil at the control location was classified as loam.

Total concentrations of heavy metals and spatial variations

The results of statistical analysis (mean, range, standard deviation, skewness, and kurtosis) for heavy metals in the study soils are summarized in Table 4. To make possible the evaluation and comparison measurements, the following are utilized together: the average control values (ACVs) of metals in the current study, Turkish Soil Pollution Control

Table 4 Summary statistics of elemental concentrations (dry-wt) in soil samples under the study area and some reference values (mg/kg)

	Cu	Pb	Cr	Cd	Zn
Min	154.6	41.2	5.7	1.9	90.4
Mean ± SEM ^c	208.4 ± 27.6	119.4 ± 20.6	18.38 ± 3.1	6.19 ± 0.97	144.2 ± 27.64
Max	421.3	208.3	32.9	10.3	357.1
Standard deviation	82.9	61.9	9.2	2.9	82.9
Skewness	2.6	0.2	0.08	− 0.36	2.59
Kurtosis	7.2	− 1.05	− 0.98	− 1.3	7.15
Average control values	50.6	38.2	4.3	1.2	81.7
Clean soil (TSPCR) ^a	50	100	10	1	300
Polluted soil (TSPCR) ^a	100	150	250	5	500
Dutch standard target values	36	85	100	0.8	140
Limit of environment in the European Union (EEA)	2–100	2–200	1–1000	0.01–0.70	10–300
Limit of world unpolluted soil ^b	13–24	22–44	12–83	0.37–0.78	45–100

Mean of three determinations

^aTurkish Soil Pollution Control Regulations TSPCR (2000)

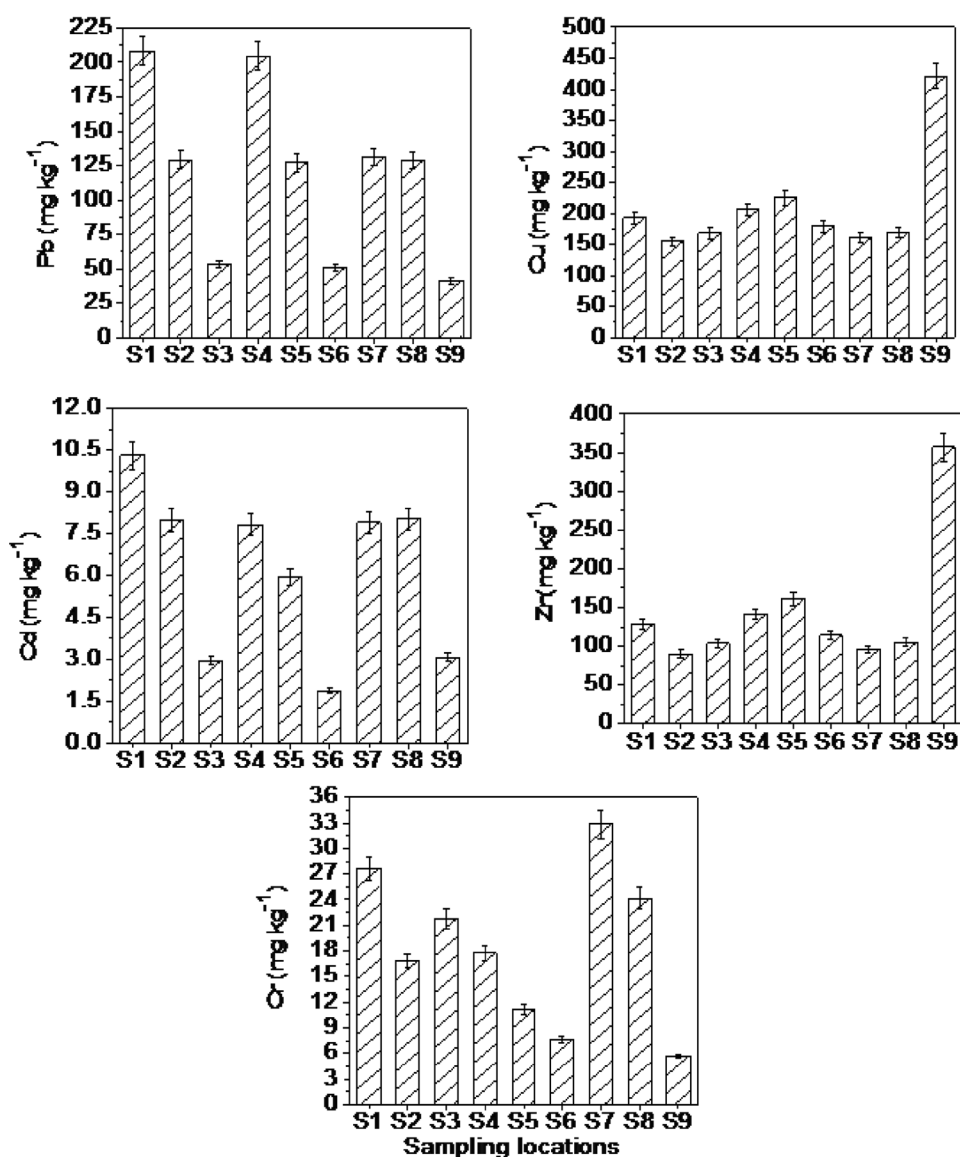
^bMean ranges of world unpolluted soil Fergusson (1990), Kabata-Pendias and Pendias (1992)

^cStandard error of the mean (SEM)

Regulation (TSPCR 2000), Dutch target soil guideline (VROM 2000), standard threshold limits of European Union soils (EEA 1999), and world unpolluted soils (Fergusson 1990; Kabata-Pendias and Pendias 1992). The total concentrations of the observed elements (Cu, Pb, Cr, Cd, and Zn) in the soil samples widely varied (Fig. 2). The average values (in mg/kg) were as follows: Cu, 208.4; Pb 119.4; Cr, 18.38; Cd, 6.19; and Zn, 144.2. In addition to these mean values, total metal concentration ranges were 154.6–421.3 for Cu, 41.2–208.3 for Pb, 5.7–32.9 for Cr, 1.9–10.3 for Cd, and 90.4–357.1 mg/kg for Zn. As Fig. 2 shows, the distribution of heavy metal concentrations in the soil of the sampling locations followed the order Pb > Cu > Zn > Cr > Cd at the location S1, and Cu > Pb > Zn > Cr > Cd at the locations S2, S4, S7, and S8, while the order was Cu > Zn > Pb > Cr > Cd at the locations S3, S5, S6, and S9. Among

the accumulated elements from the soils, Cu, Pb, and Zn were found to be dominating. In fact, the soil polluted by heavy metals as a result of the continuous weathering of mine waste produced by the ore-processing activities over half a century was found to display values much higher than the corresponding average control values (ACVs). Except for Cr, almost the entire metal content values exceeded the world’s unpolluted soil limits. Apart from Cu and Cd, the mean concentration of the examined elements was lower than the limit of ‘polluted soil’ established by the Turkish Soil Pollution Control Regulations (TSPCR 2000). However, by utilizing Dutch standard limits (VROM 2000), which are based on a broad and expansive investigation of studies for both ecotoxicological and human effects from soil contaminants, it can be said that relatively 99.9% of soil samples were highly contaminated by Cu and Cd. On the other hand,

Fig. 2 Distribution of heavy metal concentrations in the soil of the Yedidalga study area



66.7% samples were contaminated by Pb and 33.3% for Zn were considered as contaminated. In fact, among the selected metals, the detected values significantly exceeded the corresponding limit background values (Table 4). Unquestionably, Cu has the highest content levels in the studied soils, about fourfold higher than the limit values of soils suggested by the European Union of Environment (EEA 1999) and 8.4 times more than the 'safe' limit of Turkish soil (TSPCR 2000). On the other hand, the metal concentrations of the current study were relatively lower than the previous studies' results on soil metal contents in other sites of Cyprus soil near mining areas, such as CMC-abandoned copper mine in Lefke, North Cyprus (Baycu et al. 2015) and Gemikonađi (Xeros) copper and gold mine, North Cyprus (Yükselen 2002).

However, some of the metal values measured in this study were higher than those measured in other areas of the region such as Mount Olympus in Troodos area, Cyprus (Pyatt 1999), as well as than those in some international soils: gold mine in Wadi El-Allaqi, Egypt (Rashed 2010), 'and Mn mining' in Drama District, Northern Greece (Sofianska and Michailidis 2016). Based on the current study results, it can be observed that Yedidalga soil was highly contaminated by Cu, Cd, Zn, Pb, and Cr. Comparatively, Cu (421.3 mg/kg), possessing the maximum concentration in the overall measured metals, was almost 2.4 times lower than the Cu content (1023 mg/kg) of the CMC-abandoned copper mine area in North Cyprus (Baycu et al. 2015). However, this amount was observed to exceed the concentration of Cu (3.1 mg/kg) measured in Egypt soil in the Wadi El-Allaqi gold mine area (Rashed 2010). The same behavior was observed for Cr, which ranged between 5.7 and 32.9 mg/kg, being lower than the contents of Cr (40 mg/kg) in the soil of Gemikonađi-Xeros in North Cyprus (Yükselen 2002). However, Cd was lower than that in the findings (Cd 73.6 mg/kg) of Baycu et al. (2015) in the CMC copper mining area. Moreover, Pb presence seems to be highly significant and 4.7 times higher than the international soil values (world unpolluted soil). In fact, the value obtained for Pb displayed contamination signals to a certain extent and possesses the capability of changing the quality of water and other ecosystems. Although metals such as Cd and Pb are categorized as two of the most toxic and harmful elements for human beings that have no essential biological functions, they are further considered as heavy metals of public concern (Tchernitchin et al. 1998). The mean value of Cd exceeded almost all the limit references (Table 4). Indeed, there is developing environmental worries about cadmium being among those eco-toxic heavy metals exhibiting hazardous impacts on soil health, plants, and other living things. Further, chronic accumulation of Cd in the human body can cause health complexity for kidneys, induce osteotoxicity, and damage the immune system (Wilson and Pyatt 2007; Volpe et al. 2009). This heavy metal is also known to possess a triggering effect

on breast cancer. Thus, based on the current study observations, the distribution of heavy metals and their concentration differences in the soil can influence several parameters. Based on Table 2, low pH, low clay, and higher silt and sand content values enhance the transportation of soluble metals to other areas via acid mine drainage. Moreover, as numerous authors have reported (Burt et al. 2003; Conesa et al. 2006), mine dust and other particulate matters from mining plants are possibly transported by the wind. This in turn can be regarded as another crucial factor influencing the spread of metal-induced pollution.

In a different way, under superficial conditions of the environment and continuous weathering of mine waste, metals such as Cu, Pb, and Zn are often found in the nature in the form of sulfides, which are rapidly oxidized by air. Thus, they are separated from sulfur and a huge quantity of various metal pollutants escape from the source (mine waste and tailings) and move into the soil in the surrounding environment (He et al. 2005; Teng et al. 2010). Hence, the highest values in soils for the measured elements were observed in the sampling locations close to Yedidalga agricultural lands. Therefore, without controlling the continuous spread of metal contaminants and remediating the probable consequent mine pollution for the past several decades, the soil in Yedidalga vicinity and the surrounding local environment may influence agricultural products and negatively affect human health.

Overview of pollution indices

I_{geo} is the quantitative measure of the contamination index in the soils. Any increment in the current I_{geo} result levels verifies the anthropogenic nature. As shown in Table 5 and Fig. 3, apart from Cr which was uncontaminated for the entire sampling locations, the I_{geo} values for the majority of sampling locations (55.6%) were moderately to heavily contaminated by Cd (S1, S2, S4, S7, S8), 22.2% of the sampling locations by Pb (S1, S4), and 11% of the sampling locations (S9) moderately by Zn. However, around 78% of the sampling locations (S1, S4, S5, S6, S9) were classified as heavily contaminated, while 11% (S9) were extremely contaminated by Cu. The maximum I_{geo} value determined was 4.32 for Cu, followed by 2.64 for Cd, 2.47 for Pb, and 1.94 for Zn. Therefore, the accumulation of these trace metals is associated with the presence of dispersed mine wastes in Yedidalga mine harbor.

The single pollution factor and overall pollution load in the soil of each sampling locations are presented in Table 6 and Fig. 4a. Based on the result of the calculated C_f values for Cu, Cd, Pb, Cr, and Zn, wide variations of Cu and Cd values were found. The average Cu values exceeded the maximum standard values more than twice, as given in Table 2. The entire locations displayed high contamination with Cu,

Table 5 Geocontamination index (I_{geo}) and the level of different heavy metals in the study soils

Locations	Cu	Pb	Cr	Cd	Zn
s1	3.2	2.47	- 1.18	2.64	0.47
s2	2.88	1.78	- 1.9	2.27	- 0.04
s3	3.0	0.51	- 1.53	0.85	0.16
s4	3.29	2.44	- 1.8	2.24	0.60
s5	3.41	1.76	- 2.5	1.84	0.78
s6	3.08	0.45	- 3.04	0.17	0.30
s7	2.93	1.8	- 0.93	2.29	0.04
s8	3.01	1.77	- 1.38	2.27	0.18
s9	4.32	0.14	- 3.47	0.8	1.94
Min to max	2.88 to 4.33	0.14 to 2.47	- 3.47 to - 0.94	0.17 to 2.64	- 0.04 to 1.94
Average	3.24	1.46	- 1.97	1.72	0.49
Contamination level	Heavily contaminated	Moderately contaminated	Uncontaminated	Moderately contaminated	Uncontaminated/moderately contaminated

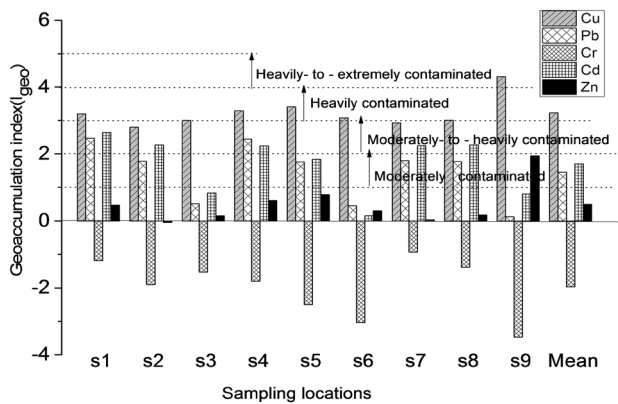


Fig. 3 Geoaccumulation index (I_{geo}) of the selected metals at the different locations in the Yedidalga soils

followed by 56% of the study locations which showed high contamination with Cd, followed by 22% of the study locations which showed high contamination with Pb, while 11% of the sample locations were classified as considerably contaminated by Zn and Cr showed lower contamination. The highest values of metal contaminations were encountered at the north and south-western part of the harbor, close to the farming area and sea coasts. These values suggested that the Yedidalga soil was extensively influenced by mining activities, indicating exceptionally extreme concentrations of Cu.

The overall assessment of metal pollution load at each of the locations was based on PLI. The GIS-based spatial distribution of PLI values (Fig. 4a) illustrated that serious metal pollution load values were encountered at locations close to the seashore and dispersed extensively near agricultural

Table 6 Single pollution factor (C_f^i) and pollution load index (PLI) in the soil of the study area

Sample location	Single pollution factor indices for heavy metals (C_f^i)					Pollution load indices for heavy metals (PLI)
	Cu	Pb	Cr	Cd	Zn	
s1	13.79	8.33	0.66	9.37	2.07	4.30
s2	11.05	5.16	0.40	7.25	1.45	2.99
s3	12.0	2.13	0.52	2.67	1.67	2.26
s4	14.68	8.17	0.43	7.1	2.27	3.83
s5	16.05	5.08	0.27	5.38	2.57	3.13
s6	12.77	2.05	0.18	1.68	1.85	1.71
s7	11.45	5.24	0.78	7.18	1.55	3.49
s8	11.45	5.15	0.57	7.28	1.70	3.34
s9	30.09	1.65	0.13	2.78	5.76	2.54
Min-max	11.05-30.09	1.65-8.33	0.13-0.78	1.68-9.37	1.46-5.76	1.71-4.3
Average	14.81	4.77	0.44	5.63	2.32	3.06
Pollution status	High	Considerable	Low	Considerable	Moderate	Moderately to highly polluted

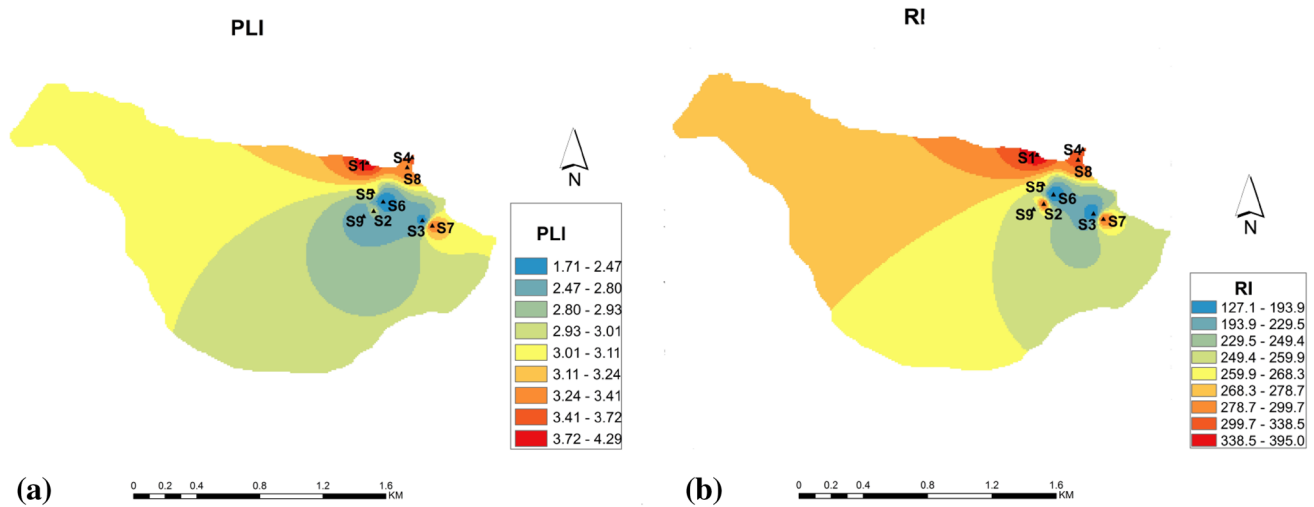


Fig. 4 Spatial distribution of **a** pollution load index (PLI) and **b** potential ecological risk index (RI) in soils in the Yedidalga study area

areas, indicating that the area received a massive amount of mine waste from the mine operations in Yedidalga harbor area. Owing to pollution load index (PLI) classification (Table 2), about 33% of the sampling locations were classified as moderately polluted with metals, 56% of the sampling locations were uncontaminated to highly polluted, and 11% close to the sea coast were highly polluted. Therefore, based on general pollution status, the area of Yedidalga harbor close to the farming areas is graded as highly polluted, and it was observed that Cu, Cd, and Pb contributed most to the level PLI of the vicinity soils, while there was moderate soil contamination by Zn and lower levels of Cr influenced the soil pollution.

Sources identification of heavy metals

Table 7 shows the inter-elemental relationships of heavy metals in the soil matrix and provides interesting information on the sources and pathways of the metals. According to the Pearson correlation analysis, very strong significant

positive relationships existed between Cu and Zn ($r = 1.000$) and Cd and Pb ($r = 0.907$) at the $P < 0.01$ significance level. Moderate positive correlation was also detected between Cd and Cr with $r = 0.686$, at $P < 0.05$ level. The significantly correlated coefficients between metals could result from the same sources of pollution (Li and Feng 2012). The statistical results also indicated that EC values showed positive correlation with soluble metals (e.g., Pb and Cr), suggesting the possibility of change related to the alternation process (Li and Feng 2012).

To expose the analysis results better, PCA was also used for this study to further identify the sources of metal pollutants. Table 8 shows the efficiency of the PCA method and the first four factor loadings of metals and their variances. The PC1, which accounted for 66.37% of the total variance, was positively dominated by Cr, Cd, and Pb, respectively. Cr and Pb were not as high as Cr in PC3 and Pb in PC4. However, PC1 showed a significant relationship and their average concentrations were higher than the corresponding background contents. Therefore, it suggested that PC1 findings

Table 7 Pearson’s correlation coefficients among heavy metal contents, pH, OM, and EC values in the soil samples

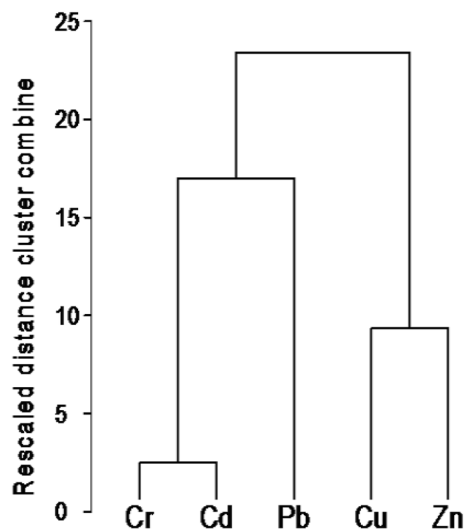
Items	pH	OM	EC	Cu	Pb	Cr	Cd	Zn	
pH	1								
OM	0.391	1							
EC	0.346	- 0.024	1						
Cu	- 0.382	- 0.470	- 0.481	1					
Pb	- 0.172	- 0.419	0.481	- 0.363	1				
Cr	- 0.236	- 0.268	0.494	- 0.594	0.534	1			
Cd	- 0.251	- 0.418	0.600		- 0.375	0.907 ^a	0.686 ^b	1	
Zn	- 0.382	- 0.470	- 0.481		1.000 ^a	- 0.363	- 0.594	- 0.375	1

^aCorrelation is significant at the 0.01 level (2-tailed)

^bCorrelation is significant at the 0.05 level (2-tailed)

Table 8 Results of principal component analysis (PCA) for sources of selected metals in the soils

	PC 1	PC 2	PC 3	PC 4
Eigenvalue	3.32	1.22	0.4	0.065
% Total variance	66.37	24.34	7.99	1.30
% Cumulative variance	66.37	90.71	98.7	100.0
Cu	-0.452	0.502	0.204	0.051
Pb	0.423	0.499	-0.4454	0.612
Cr	0.461	0.045	0.847	0.261
Cd	0.448	0.495	-0.041	-0.744
Zn	-0.452	0.502	0.204	0.051

**Fig. 5** Dendrograms of five different elements produced by hierarchical clustering

show the influence of mine activities in the region. Cu and Zn were highly correlated, demonstrating moderate positive factor loadings in PC2 with 24.34% of the total variance. Some studies showed that Cu was classically anthropogenically influenced (McMartin et al. 2002). PC3 accounted for 7.99% of the total variance dominated by strong positive loading of Cr, having the highest factor (0.847) in the overall PC loadings, whereas PC4 explained 1.3% of the total variance also indicating strong positive loading for Pb. Thus, within the scope of this study, the method can as well be identified as a further tracer of anthropogenic sources.

Hierarchical cluster analysis was also carried out to further divide the clusters of metal pollutants to identify the possible resources. The results were illustrated in a hierarchical dendrogram (Fig. 5). The degree of the association between heavy metals was determined by a distance cluster: the lower the value on the distance cluster, the more significant was the association (Fan-xin et al. 2016). Three different clustering can be identified from Fig. 5. The first cluster

includes Cd and Cr with a low distance cluster criterion (less than 5), indicating that Cd and Cr have a relationship. It was allied with the second cluster containing Pb, with their presence mainly associated with mining operations and atmospheric depositions (Peng et al. 2014). The third cluster including Cu and Zn was also significantly influenced by mining activities. Cu, which indeed resulted from the deterioration of mine waste, also was found at the highest concentration in the Yedidalga harbor soils (Table 4). Obviously, the results obtained are acceptably in conformance with the indications of descriptive statistics, cluster analysis, and correlation analysis (PC).

Potential ecological risk analysis

The single ecological risk indices and potential ecological toxicity response indices of soil heavy metals in Yedidalga harbor area are shown in Table 9 and Fig. 4b. The ecological risk (E_r^i) values of Cr and Zn in all locations were lower than 40, which indicated that these heavy metals posed low ecological risk to soils. The main elements causing potential ecological risks were Cu ($40 \leq E_r^i < 80$) and Cd ($E_r^i > 160$), indicating moderate to high ecological risk. Pb demonstrated low eco-risk on the average. However, two locations (S1, S4) indicated moderate ecological risk. Among the metals displaying the highest potential ecological risk, Cd exhibited serious ecological risk ($E_r^i > 160$), especially at locations S1 and S8. In addition, the elevated values of Cd and Cu in the study area were probably the result of mineral activities in the harbor. Generally, high ecological risk grades for Cu, Cd, and Pb were identified at the sample locations close to the farming areas.

Computed RI indices showed that 56% of the sampling locations posed considerable potential ecological risk, while 44% were classified as low-to-moderate risk ($RI < 300$) and 33% had $150 \leq RI < 300$ values. Based on GIS-mapped spatial distribution, higher RI values were found in sampling locations (S1, S2, S4, S7, and S8) close to the Mediterranean Sea coast (50–100 m) and Yedidalga farming lands in the vicinity of the study area and relatively showed similar distribution as PLI (Figs. 1, 4b). Hence, soils around the obviously contaminated areas are more vulnerable for further potential ecological risks and eventually may create serious health problems to the local habitants. Therefore, this information concerning the polluted area of Yedidalga harbor soil enforces the need of essential intervention to reduce risks associated with heavy metals.

Table 9 Potential ecological risk factor (E_r^i) and potential ecological risk index (RI) of heavy metals in the Yedidalga harbor soil

Sample location	Ecological risk indices for single heavy metal (E_r^i)					Potential ecological risk (toxicity response) indices for heavy metals (RI)	Risk grade
	Cu	Pb	Cr	Cd	Zn		
s1	68.95	41.65	1.31	281.10	2.08	395.09	Considerable
s2	55.25	25.82	0.80	217.55	1.46	300.88	Considerable
s3	60.01	10.68	1.03	80.25	1.67	153.64	Moderate
s4	73.4	40.88	0.85	213.03	2.28	330.44	Considerable
s5	80.23	25.40	0.53	161.40	2.59	270.15	Moderate
s6	63.85	10.26	0.36	50.40	1.85	126.72	Low
s7	57.24	26.21	1.56	215.4	1.55	301.96	Considerable
s8	60.57	25.75	1.15	218.49	1.70	307.67	Considerable
s9	150.45	8.24	0.26	83.71	5.76	248.43	Moderate
Min-max	55.25–150.45	8.25–41.65	0.26–1.56	50.4–281.1	1.46–5.76	126.72–395.09	
Average	74.4	23.9	0.87	169.1	2.33	270.55	Moderate

Conclusions

The results of the current study illustrate the impact of former mine harbor activities on the abundance of heavy metal in the soils of the Yedidalga harbor area. The average contents of Cu, Cd, Pb and Zn were detected to be far higher than those of international threshold values, while Cr was moderately low. Specially, this was valid for the sampling locations which were at a close proximity to the Yedidalga farming lands and Mediterranean Sea coast. The results of the correlation matrix and multivariate analyses indicated that the heavy metals in the soils were mainly present due to anthropogenic sources. These soils were significantly affected by pollutants associated with mine waste from Yedidalga mine harbor. In the vicinity of Yedidalga mine harbor, pollution assessment methods including I_{geo} , PLI, and C_f classify the soils as moderately to heavily contaminated with Cu, Cd, Zn, and Pb, but not Cr. According to RI results, these soils display moderate to considerable potential ecological risk to the environment, generated mainly by Cd, Cu, and Pb. Therefore, immediate intervention measures are recommended to control heavy metal pollution problems in the area, and developing a proper pollution protection and management strategy is suggested.

Acknowledgements This study was materially supported by the Environmental Research Center of Cyprus International University. However, the study did not receive any specific grant from funding agencies.

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