Soil contamination around porphyry copper mines: an example from a semi-arid climate

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Abstract

Extraction and processing of disseminated metalliferous ores, porphyry copper in particular, results in significant tonnages of waste and can cause severe disturbances and contamination in natural ecosystems. This is particularly important in semi-arid climates where natural soils are often deprived of organic matter and nutrients. This study was conducted on seven sites around Sungun Copper Mine, northwest Iran. Soil texture, EC, pH, and concentrations of nutrients, organic matter, along with 16 metal and metalloids were measured in 94 soil samples. Results showed a gradient of contamination from low contamination in natural hillsides to high contamination in mine waste depositories, Waste Dump and Oxide Dump, alongside Pakhir and Sungun Rivers. Nutrient deficiency occurred in disturbed sites. The main contaminant point sources were Waste Dump, mine pit drainage, and Oxide Dump. The results of Non-metric multidimensional scaling ordination showed elevated Cd, Zn, Fe, Cu, Pb, As, Mo, Mn, Co, S concentrations, high EC, and higher sand percentage in the sites affected by mine waste and acid mine drainage. Geo-Accumulation and Potential Ecological Risk Indices indicated that Pakhir riverside, Sungun riverside and Oxide Dump have severe to moderate levels of environmental risks. Positive correlations between certain metal elements suggest common sources and similar reaction pathways, which may contribute to their similar geochemical behaviour in transport, deposition, and interdependence. Overall, the deficiency of organic matter and nutrients along with the soil sandy texture in contaminated sites of Sungun Copper Mine are the main limiting factors in managing metal mobility and soil remediation.

Keywords: Contamination indices, Heavy metals, Mine drainage, Oxide dump, Soil pollution, Waste dump

1. Introduction

As an important portion of ecosystems, soil is the source of water and essential nutrients for soil organisms and plants, and the geochemical sink or filter for contaminants (Bermudez et al., 2010; Terrones-Saeta et al., 2021). Dispersion of toxic metals due to the increasing rates of industrial cause soil contamination turn into worldwide environmental problems (Khalid et al., 2017; Adewumi et al., 2020; Sun et al., 2020). A potential source of metal and metalloid contaminants is mining operations, causing primary concern for environmental sustainability (Jung et al., 2001). Unlike many organic contaminants, some metals and metalloids accumulate in soil because they are environmentally persistent due to association with organic matter and/or sorption to mineral surfaces (Grafe & Naidu, 2008; Ahdy & Khaled, 2009). Soils in mined lands typically exhibit characteristics such as deficiencies in carbon compounds and nutrients, as well as elevated levels of toxic mineral elements (Rodríguez et al., 2009; Huang et al., 2012). Metalliferous mines are the source of significant amounts of metalliferous wastes, such as rock dumps, and wastes of mineral processing and smelters, leading to contamination and degradation in the vicinity of mine areas (Dinelli et al., 2001; Randelovic et al., 2014). Spatial heterogeneity of mine wastes, soil physicochemical properties and differences with natural geochemical background values are main aspects of soil contamination investigations on mined lands (El Azhari et al., 2017). Most mine wastes are not chemically or physically stable, and cause problems in surrounding environments as dust, erosion into surface waters, and through leaching and acid mine drainage (AMD) (Otte & Jacob, 2008). Acidic soil conditions (pH<7) and elevated metal and metalloid concentrations due to acid mine drainage exacerbate environmental problems by increasing the toxic elements bioavailability (Rezaie and Anderson, 2020). A crucial step in sustainable soil management, including improved soil quality of mined lands, is to determine the metal concentrations in soils in order to monitor their distribution and manage their effects and possible toxicities (Yun et al., 2016; Radi et al., 2023). However, the total concentrations, mobility, and toxicity of metal(loid) elements in soils depend on their specific chemical forms, as well as edaphic and environmental characteristics such as soil acidity or alkalinity, organic matter content, salt concentrations, and local climatic conditions (Chrastný et al., 2012; Zu et al., 2014; Mehrabi et al., 2015; Gabarron et al., 2017).

Open pit mines often have a significant environmental footprint as they remove large areas of vegetation cover, top soil, and overburden, and expose pit walls, waste rocks and mineral processing tailings to weathering and degradation processes (Karaca et al., 2018). Soil contamination and acid mine drainage cause considerable environmental degradation with severe impacts on communities and biodiversity (Kefeni et al., 2017). There are many reports of porphyry copper deposits for which mining activities have caused soil contamination in arid and semi-arid areas through elevated levels of some metal and metalloid elements. Soil contamination with high levels of Cd, Zn, Cu, Pb, Mn, Fe, and Al has been documented in various locations, such as around a copper mine in Peru (Bech et al., 1997) and in the Cu-Au-Hg district in Punitaqui, Chile, where the soil Cu concentration ranged from 95 to 230 µg/g (Oyarzún et al., 2001). In the Andacollo porphyry Cu district, soil exhibits high concentrations of Cu and Hg compared to reference baselines, as reported by Higueras et al. (2004). Also, Sulfate and copper enrichment have been reported at the El Teniente porphyry copper deposit in Chile (Kelm et al., 2009). Capillary forces in arid climates lead to upward transition and oxidizing conditions, resulting in the formation of water-soluble sulphates (Meza-Figueroa et al., 2009). In arid areas of Iran, such as the Darreh-Zereshk and Ali-Abad porphyry Cu deposits, hyrofracturing of ore-hosting deposits leads to the formation of quartz-sulfide veinlets (Zarasvandi et al., 2013). The Buchim mine in the Republic of Macedonia serves as an example of porphyry copper deposits, with Cu concentrations ranging from 94.8 to 1171 µg/g (Serafimovski et al., 2014). Despite numerous studies on soil contamination in different regions, little is known about such effects in semi-arid ecosystems in southwest Asia. Iran has three major copper deposits—Sarcheshmeh, Sungun, and Miduk, with Sungun representing a semi-arid climate (Saadmohammadi et al., 2011). While soil contamination studies have been undertaken for the Sarcheshmeh and Miduk areas (Khorasanipour & Aftabi, 2011; Moore et al., 2014), and water and sediment contamination in the Sungun Copper Mine have also been studied (Aghili et al., 2018), there is no published study about soil contamination around the Sungun copper mine. This study aims to (i) evaluate metal and metalloid contamination and other physicochemical properties of soils around the Sungun copper mine in Iran, (ii) determine contamination indices and gradients, and (iii) discuss the relationships between metal and metalloid elements and their association with soil physical and chemical characteristics.

2. Materials and Methods

2.1 Study area

Sungun Cu-Mo deposit in north-west of Iran, 120 km Northwest of Tabriz (46º 30′ E to 46º 52′ E, 38º 34′ N to 38º 45′) hosts Iran's second important and large copper mine (Fig. 1). The mineralization at Sungun Copper Mine is largely hosted by diorite/granodiorite and monzonite/quartz-monzonite porphyry (Hezarkhani, 2011; Moshefi et al., 2018). Original porphyritic texture through silicification process altered into heavily dense rock that is cut by quartz veins, which is together with pyrite, molybdenite and, to some extent chalcopyrite (Nabi Bidhendi et al., 2007). The soil taxonomic order of study area is rock outcrops with entisols. The approximate amounts of mined ore since the start of operation in 2006 is about 500 million tons (Hezarkhani et al., 1999). The elevation of the study area varies from 1645 m to 2700 m above sea level. Study area has a semi-arid climate with average evaporation from water surface of 836 mm, and annual mean precipitation is about 400 mm, also average monthly temperatures in summer and winter varies between 33°C and -20°C (Moore et al., 2011). Mining area is located near Arasbaran protected natural heritage and its vegetation mostly consists of Asteraceae, Poaceae, Fabaceae, Apiaceae, Brassicaceae, and Lamiaceae families (Ghorbani et al., 2018). Based on field observations and according to the potential of contamination, soil sampling performed in different areas surrounding Sungun copper mine (Figs 2 and 3). These were: (i) alongside Pakhir River which is in downstream of mine waste rocks and mine pit drainage; ii) alongside Sungun River that is not affected by mine drainages; (iii) Oxide Dump; (iv) Waste Rock Dump, (v) undisturbed hillside next to Waste Dump; (vi) natural hillside near mine pit; and (vii) natural hillside far away from the mine pit.

2.2 Soil sampling and laboratory analysis

Sample collection included 94 soil samples from 0–15 cm, and 15–30 cm depths. Topsoil samples were collected from 6 points in Waste Rock Dump, 5 points from Oxide Dump, 5 points alongside Sungun River, 7 points alongside Pakhir River, 9 points in hillside next to Waste Rock Dump, 5 points from hillside near mine pit and 10 points in the natural hillsides far from mine pit. A mixture of four soil samples in each point were used to form a single composite sample that were kept in Clear Polyethylene bags until chemical analysis. The soil samples were dried in room temperature then grinded and sieved through <0.2 mm (80 mesh). Then, the fraction of soil samples < 0.2 mm were used for physicochemical characterization. Topsoil samples pH and electrical conductivity (EC) were measured in a 1:2.5 (w/v) soil/water suspension by Adwa (AD 1000) pH meter and ELMETRON (CC 501) conductivity meter, respectively (Kabala et al., 2016). Organic matter (OM) and total nitrogen (N) were determined using titration and colorimetric method (Walkley and Black, 1934) and Kjeldahl method (Bremner and Mulvaney, 1982). The calcium carbonate ($CaCO_3$) contents were determined by titration with hydrochloric acid (HCl). Potassium (K) concentrations were measured by flame photometry (Colwell, 1963). Topsoil available phosphorus (P) concentration were measured by Olsen method (Olsen et al., 1954). Soil particle size analysis was conducted by Hydrometer (Bouyoucos, 1951). The Aqua regia digestion method was applied to prepare a multi-element analysis solution from soil samples, utilizing nitric acid (HNO3) and hydrochloric acid (HCl) at a 1:3 ratio under a heating system (Yun et al., 2016). Total concentrations of aluminium (Al), arsenic (As), calcium (Ca), Cerium (Ce), cobalt (Co), chromium (Cr), cupper (Cu), iron (Fe), magnesium (Mg), manganese (Mn), Molybdenum (Mo), nickel (Ni), cadmium (Cd), lead (Pb), Sulfur (S) and zinc (Zn) in soil were measured by inductively coupled plasma atomic emission spectrometry (ICP-OES, Varian 735, Margesin & Schinner, 2005). The detection limit for measured elements were Al= 100, As= 0.5, Ca= 100, Ce= 1, Co= 1, Cr= 1, Cu= 1, Fe= 100, Mg= 100, Mn= 5, Mo= 0.5, Ni= 1, Cd= 0.1, Pb= 1, S=50 and Zn= 1 ppm. All analyses were conducted at the accredited (ISO 17025) and certified (ISO9001:2008) Zarazma laboratory (http://zarazma.com/en).

2.3 Contamination indices

Geo-Accumulation index (Igeo) was calculated using Igeo = $\log_2$ [Cn/1.5Bn] (Muller, 1969). Cn is the metal (n) concentration, Bn is the background value of the corresponding metal (n) in soil (Kabata-Pendias, 2011), and because of lithological variations and variations in background data the factor 1.5 is added to the equation. The Geo-Accumulation index have seven classes (Table 1).

Contamination factor (C_f^i for short) evaluates soil potential contamination by a single toxic metal(loid) as follow (Weissmannová and Pavlovský, 2017):

$$C_f^i = C_{surface}^i / T_{reference}^i$$

In this equation C_f^i is pollution coefficient for a certain toxic metal(loid) that is determinant of contamination in study area but is not efficient to show the hazards and ecological effects of metal elements. $C_{surface}^i$ and $T_{reference}^i$ are measured and background concentration of toxic metal(loid) in soil suggested by Kabata-Pendias (2011).

Also the ecological risk factor (E_f^i) is used to assess the potential ecological risk of a given contaminant, as measured by $E_f^i = C_f^i * T_f^i$ (Hakanson, 1980) where T_f^i and C_f^i are toxic-response and contamination factors, respectively. Hakanson (1980) and Xu et al. (2008) reported the T_f^i values of toxic metal(loid)s as: Cd=30, As=10, Cu=Pb=Ni=5, Cr=2, Zn=1, Mn=1 and Co=5. Then the potential ecological risk index (RI) was calculated by RI= $\sum E_f^i$ (Table 2).

2.4 Statistical Analysis

The R statistical computing language was used for conducting statistical analysis (R Core Team, 2022). Shapiro–Wilk W test, dedicated to checking normality and then log-transformation performed on non-normal distributed data before analyses. Two-way analysis of variance (ANOVA) performed to determine the depth and the sites effects on physical, chemical and mineralogical characteristics of soil samples. Duncan multiple range test was applied to compare means of soil data in different sites. Relationships between pair-wise distances of sites and soil properties and dissimilarities (ordination space) analysed by NMDS (Non-metric Multidimensional Scaling) method. Then, sites were shown with ellipses on the gradient of soil contamination. To evaluate the association between soil characteristics and levels of metals and metalloids Spearman's correlation coefficient was used.

3. Results

3.1 Soil properties and contaminant concentrations

The results of two-way ANOVA showed that soil measured parameters only affected by differentiation of sampling sites and not by soil depth or the interaction of sampling sites and soil depths. Majority of the soil properties and metal and metalloid elements concentrations showed significant differences among studied sites (Table 3 and 4). The soil pH varied from slightly acidic (6.40) alongside Pakhir River (site 1 in Fig. 2) to neutral (7.01) in the Waste Rock Dump (site 4 in Fig. 2, Table 3). The soil samples collected alongside Pakhir River and Oxide Dump (site 3 in Fig. 2) exhibited significantly elevated levels of electrical conductivities (0.75 and 0.65 mS cm^{-1}). In contrast, the Waste Dump and natural hillsides had low EC values (Table 3). Soil samples from all sites had higher sand content specifically samples from Pakhir River and Sungun River showed elevated sand levels (91.45% and 87.0%), along with the lowest silt (7.71% and 9.72%) and clay (0.27% and 1.81%) contents, respectively. The greatest amount of clay was found in Oxide Dump (15.20) and natural hillside far from the mine pit (15.18) (Table 3). The Oxide Dump and three natural hillsides were found to have high silt content (24.52%, 19.75%, 27.73%, and 18.53%, respectively). Natural hillside located a few meters from mine pit showed the highest levels of calcium carbonate, while no significant differences were found among the other sites (Table 3). The N-P-K contents indicated that natural hillsides had greater levels of N (0.54%, 0.34%, and 0.36%) and K (1.87, 1.75 and 1.62 µg g^{-1}) compared with those of other sites in the mining area (Table 3). In contrast, Sungun riverside and Oxide Dump had higher P concentrations than other sites (5.31 and 6.95 µg g^{-1}). The soil organic matter contents ranged from 0.08% in waste dump to 5.84% in natural hillside located in the vicinity of Waste Dump. In mining affected sites (alongside Pakhir and Sungun River, Oxide Dump, and Waste Rocks), the organic matter content were significantly lower (0.47%, 0.51%, 0.55%, and 0.083%, respectively) than that in the natural hillsides (5.84%, 2.27% and 1.78%; Table 3).

Mean trace elements in the soils of different sites showed that Pakhir riverside and the natural hillside far away from the mine pit had the highest and lowest Co concentrations, respectively (18.8 µg g^{-1} and 11.6 µg g^{-1},; Table 4). There was no significant different between all the other studied sites. The three natural hillsides had significantly higher concentrations of Cr in the soil (46.3 µg g^{-1}, 44 µg g^{-1}, and 39.6 µg g^{-1}) compared to sites affected by mining activity. The concentrations of Cu (3141 µg g^{-1}), Fe (58708 µg g^{-1}), Zn (665 µg g^{-1}), Cd (0.66 µg g^{-1}), and Pb (945.8 µg g^{-1}) were highest at the Pakhir riverside (Table 4). Soil samples from two natural hillsides near the Waste Rock Dump and mine pits had higher Mg content (13138 µg g^{-1}) compared to other sites. The highest concentrations of Mo (69.1 µg g^{-1}) and Ce (70.3 µg g^{-1}) were found in Sungun riverside and Waste Dump, respectively (Table 4).

The maximum S concentrations were significantly found in the two riversides (Pakhir= 5838 µg g^{-1} and Sungun= 3694 µg g^{-1}) and Oxide Dump (4555 µg g^{-1}). The average As concentrations in the soil of the two riversides (35.3 and 22 µg g^{-1}) were significantly greater than that in all other sites. Elevated concentrations of Mn, Ni, Al, and Ca were recorded in natural hillsides, but they were not significantly different from those in some of the mining affected areas (Table 4).

3.2 Contamination gradient

The NMDS ordination of sampled plots, based on their soil properties, revealed that the arrangement of sites in the ordination space is strongly influenced by soil properties (Fig. 4). The ellipses of two riversides (S1 and S2) particularly Pakhir riverside are located in the upper right corner showing an association with the positive ends of two axes. These axes are mainly defined by Cd, Zn, Fe, Cu, Pb, As, Mo, Mn, Co, S concentrations, electrical conductivity, and sand content. In contrast, other sites (S4: Waste Rock Dump, S5: natural hillside close to Waste Rock Dump, S6: natural hillsides near the mine pit, S7: natural hillside far away from mine pit) are assembled together in the opposite direction associated with higher silt and clay contents, and higher K, Mg, N and organic matter concentrations, and higher pH. The Oxide Dump had a greater association with the positive end of axis 1 (As, Cu, Mo, P and Pb concentrations and electrical conductivity) and also the negative end of two axes (higher silt content, pH, and Mg, $CaCO_3$, K, and N concentrations). These result show a spatial gradient of soil contamination around the Sungun Copper Mine area.

3.3 Contamination indices

The Geo-Accumulation Index values for Cu (5.25) and Pb (5.39) indicated that the soil alongside Pakhir River was extremely contaminated (Table 1 and 5). The I_{geo} values for As showed strongly contamination of soil alongside Pakhir riverside (3.71) and Sungun riverside (3.02). Additionally, the average Geo-Accumulation Index for Mo (4.94) suggested that soil alongside Sungun River was extremely strongly contaminated by this trace element (Table 5). According to I_{geo} index values, elements such as As, Mo, Pb and Ce are sources of moderately to strongly contamination of soil in Waste Rock Dump (1.24, 1.42, 0.22, and 2.23) and Oxide Dump (2.28, 1.98, 1.67, and 1.86). Regarding all analyzed metal and metalloids elements Natural hillsides showed no contamination to moderate contamination rates (Table 5).

The ecological risk factor (E_f^i) showed that soil alongside Pakhir River had high levels of ecological risk especially for Cu (286), As (196), Cd (198), Pb (315) (Table 6). Alongside Sungun River, a high ecological risk factor was observed primarily due to As (122) (Table 6). Soil alongside Pakhir River and Sungun River had serious and sever potential ecological risk index, respectively (Table 7). A moderate potential ecological risk index was found for Oxide Dump and low-grade for other sites (Table 7).

3.4 Correlation matrix

The relationship between soil parameters showed some significant positive and negative correlations (Fig. 5), which is consistent with the results of NMDS ordination (Fig. 4). Soil pH had positive association with Ca and a negative relation with Cd, Fe, and Pb (Fig. 5). As, Cd, Ce, Co, Cu, Fe, Mn, Mo, Ni, Pb, S, and Zn concentrations were significantly associated with soil electrical conductivity (Fig. 5). Soil organic matter had a positive correlation with Al, Cr, and Mg concentrations but a negative relation with Ce, Cu, Mo, Pb, S, and Zn (Fig. 5). Except for Ca, other metal elements in soils were significantly correlated with each other (Fig. 5).

4. Discussion

Soil physicochemical characteristics are often used for assessing the soil quality. According to the results soil pH mean values were significantly different among the studied sites. The lowest pH was found alongside Pakhir River (6.40) and Oxide Dump (6.55) and the highest in Waste Rock Dump (7.01). This could have been caused by sulfide oxidation that leads to hydrogen ion release or dissolution of minerals that promote acidic pH (Sanderson et al., 2012; Aghili et al., 2018; Kulikova et al., 2019; Kavehei et al., 2021). In the Waste Rock Dump, the neutral range of pH indicates abundance of carbonates over sulfides along with the absence of AMD (Perlatti et al., 2015). As shown in NMDS ordination, the arrows of pH and $CaCO_3$ indicate that they are the main factors contributing to the separation of Waste Rock Dump from other sites, while Pakhir riverside, Sungun riverside, and Oxide Dump are mainly affected by S concentration. Lower content of calcium carbonate ($CaCO_3$), which can neutralise the acidity produced by the dissolution of sulphides (Likus-Cieślik et al., 2017; Candeias et al., 2015), was measured in the sites affected by mine drainage, particularly alongside Pakhir River, Sungun River, and Oxide Dump (Fig. 4 and Table 3). Reduction in soil acidity in result of higher $CaCO_3$ contents decrease trace element solubility and stimulate metal immobilization in soil (Khan & Jones, 2008). The Potential Ecological Risk Index (RI) values were higher in sites with elevated concentrations of metals, found along Pakhir River and Sungun River. This indicates that these areas serve as primary sources of environmental issues around Sungun Copper Mine (Weeks & Comber, 2005). The high values of RI and Igeo in this mining area indicate a substantial potential for influencing the ecological function of the adjacent ecosystems, notably the Arasbaran protected area. The transportation of metal(loides) through Pakhir and Sungun Rivers is likely to induce changes in hydrogeochemical processes, thereby contributing to the deterioration of water quality (Aghili et al., 2018; Punia & Singh, 2021). This underscores the significance of considering and addressing the potential environmental consequences associated with metal mobility in efforts to mitigate ecological impacts. Compared to the average copper concentration in earth crust (55 µg g^{-1}), soil in Pakhir riverside (3140 µg g-1), Sungun riverside (1044µg g-1) and oxide dump (288 µg g-1) was particularly contaminated with Cu due to mining and oxidation of mine wastes (Kabata-Pendias, 2011; Knabb et al., 2016). Mine drainage is the main cause of metal(liod) elements increasing in surface soil and surrounding water sources that leads to ecological risks (Lu et al., 2021). Moderate levels of ecological risks index, mainly related to Cd and Cu is also reported from Cyprus' old Agrokipia copper mine (Hadjipanagiotou et al., 2020). The CaCO3 content exhibited a negative correlation with trace metals such as Pb, Cd, As, Zn, and Cu. This correlation underscores the impact of carbonates on the decomposition and stabilization of toxic minerals in soils with elevated CaCO3 content. Carbonates, as a key soil geochemical characteristic, play a pivotal role in determining the soil's capacity to retain metal elements (Gasparatos et al., 2015; Taalab et al., 2019). According to the present findings, the main concern about Pakhir riverside, Sungun riverside, Oxide Dump and Waste Rocks in the long term is acid mine drainage (with the source of overbank flow and in-situ weathering and oxidation) that leads to mobility of metals/metalloids and release of sulfate, as Sungun porphyry deposit consist of anhydrites ($CaSO_4$) which leads to high S content (Kou et al., 2021).

Our findings revealed that sites disturbed by mining exhibited lower concentrations of N, K, Ca, and Mg. This suggests soil fertility degradation likely attributed to the loss of litter layers and organic materials (Närhi et al., 2012, Wang et al., 2021). We also found that concentrations of these nutrients were greater in the soil of natural hillsides, and there was a considerable correlation among N, K content with soil OM content, demonstrating their organic origin. Phosphorous concentrations were greater in alongside Sungun River and Oxide Dump, as Cu phosphates can be considered a possible reason (Abreu et al., 2008). Phosphate compounds are known to be effective in decreasing mobility of potential toxic minerals containing Pb, Cd, and Zn (Andrunik et al., 2020). Positive correlations were found between these elements and soil P concentration in both correlation analysis and NMDS.

The highest tolerable levels of soil contamination for plants include Co (10 µg g^{-1}), Cr (100 µg g^{-1}), Cu (100 µg g^{-1}), Fe (35000µg g^{-1}), Mn (550 µg g^{-1}), Ni (100 µg g^{-1}), Zn (300 µg g^{-1}), As (7 µg g^{-1}), Cd (5µg g^{-1}), Mo (10 µg g^{-1}), Pb (100 µg g^{-1}), Ce (10 µg g^{-1}) (Chon et al., 2011; Kabata-Pendias, 2011). Earlier, we identified the potential of native vegetation for phytostabilization in these sites (Alizadeh et al., 2022). In light of these values, our research provides evidence of pronounced soil contamination in areas impacted by mine drainage, particularly along two riversides and the Oxide Dump. This contrasts with findings from Europe's largest copper mine in Poland (Kabala et al., 2020) but aligns with data reported from Sarcheshmeh and Miduk Cu mines in arid regions of Iran (Khorasanipour & Aftabi, 2011; Moore et al., 2014), and a Cu mine in Malaysia (Ali et al., 2004). It is more probable to acid mine drainage occurrence in conditions that sulfidic, especially Fe-sulfides (FeS_2) or pyritic substances such as oxidised wastes and waste rocks are accumulated (Kavehei et al., 2021). Mine wastes because of physicochemical characteristics and geochemical transformations lead to release of H^+ and metal(loid)s with substantial environmental problems on mine water and vegetation covers that should be in priority of stabilization (Edraki et al., 2014). Sites affected by Acid Mine Drainage (AMD) showed strongly increased concentrations of the mined metal, Cu, along with some of other metals present in the mixed ore such as Fe and Al (Hajihashemi et al., 2023). Both mine pit and waste drainage contains elevated amounts of Cu and acid rock drainage creates toxic metal(loid) contaminated sites (Milu et al., 2002). The negative correlation of soil clay aggregates with Co, Fe, Mn, and Zn, as well the lower ranges of pH in contaminated sites, shows that the decrease in soil pH and lower Ca concentrations can reduce metal adsorption by soils. This is due to competition for negatively charged surfaces of clay minerals between H^+ and dissolved metals (De Matos et al., 2001; Pandey et al., 2007; Gonzalez-Fernandez et al., 2011; Sungur et al., 2014). The soil taxonomy in the study area is classified as entisol, distinguished by an elevated concentration of sand aggregates (Golia et al., 2019). Furthermore, NMDS plot indicated a correlation between most metals and metalloids (Co, Cd, Zn, Fe, Cu, Pb, As, and Mo) and increased sand content. This implies that larger soil particles are less effective in retaining metals (Çevik et al., 2009; Romzaykina et al., 2021). Fe and Cu contents of soil have positive association because Fe-oxides enhance fixation of metals, especially Cu (Arenas-Lago et al., 2014). The considerable positive correlations of metals including Cd, Cu, Fe, Mo, Pb, and Zn with S concentration, and also along with each other in the NMDS plot shows that these elements are mainly sulfide-associated (Kim et al., 2003; Alakangas et al., 2010).

There are three main rock types in Sungun deposit: Sungun porphyry with porphyry fabric and quartz monzonite, non-mineralized dykes, and Skarn, which is a metasomatic contact alteration and mineralization with Cretaceous limestone and marl (Talebi et al., 2015). Metal element correlation with each other also implies they had the same sources and similar weathering pathways, also suggesting similar origin, geochemical behaviours for the transport, deposition, and interdependence of these elements (Khalil et al., 2013; Lienard et al., 2014; Demkova et al., 2019; Antunovic et al., 2023). Also, mineral elements like Cu, Pb, and Zn regularly co-exist because of same process of geochemistry digenesis (Pu et al., 2010; Han et al., 2015). Sungun porphyry deposit and associated dykes consist of major oxides (SiO2, Al2O3, CaO, Fe2O3, K2O, MgO, MnO) and elements (As, Cd, Ce, Co, Cr, Cu, Mo, Ni, Pb, S and Zn) which contains Fe, Pb, Zn, and Cu mineralization (Kamali et al., 2018).

The three natural hillsides had lower ranges of chalcophile element concentrations than those in mine-affected sites. The data for three natural hillsides showed that those close to Waste Dump had greater concentrations of N and OM. This might be associated with the presence of nitrogen-fixing plant species from the Fabaceae family and higher vegetation cover that add litter material to surface soil (Ghorbani et al., 2018; Buta et al., 2019; Chen et al., 2020). Additionally, lower concentrations of metal elements were observed in a natural hillside far from the mine pit. This could be attributed to higher clay contents, as the clay fraction and organic matter in the soil are significant factors influencing metal sorption (Rieuwerts, 2007; Liu et al., 2022). Consequently, the concentration of metal(liod) in the soil and associated ecological risks are lower in natural hillsides (Gutiérrez et al., 2020). Fine grain soils commonly have elevated metal concentrations because of their greater surface to volume ratio, organic matter formation and enrichment, and Fe-Mn oxides (Jain et al., 2008; Redwan & Elhaddad, 2016).

5. Conclusions

This research explored the contamination levels of metal(loid) elements and their spatial distribution in a copper mine and its surrounding environments in a semi-arid region. Elevated values of metal contamination were observed across the entire mining area. Significant correlations between mineral element concentrations in soil samples helped identify the typical origins of contaminants in active mining areas. The concentrations of metals, Geo-Accumulation index (Igeo), and Potential Ecological Risk (RI) indices levels suggest that the Oxide Dump and Waste Rock Dump are the principal sources, and mine drainage causes the pollution to spread beyond the mine site particularly from Pakhir River. In terms of future remediation, there is a high potential of phytoremediation in this area (Alizadeh et al., 2022). However, the lack of organic matter, low soil nutrient status, and the sandy texture of highly polluted soils will pose challenges for controlling metal mobility.

6. Statements and Declarations

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Competing Interests

The authors have no relevant financial or non-financial interests to disclose.

Author Contributions

All authors contributed to the study's conception and design. Field sampling was performed by Arezo Alizadeh and Javad Motamedi. This work was supervised by Jamshid Ghorbani and the laboratory analyses were arranged by Ghorban Vahabzadeh. Antony van der Ent and Mansour Edraki helped with the development of the idea and interpretation of the results. All authors have read, understood, and have complied as applicable with the statement on "Ethical responsibilities of Authors" as found in the Instructions for Authors.

Ethical approval

No human or animal study was conducted during the present research.

Consent to participate

Not applicable.

Consent for publication

Not applicable.

Availability of data and materials

Data and materials will be provided if requested.

7. References

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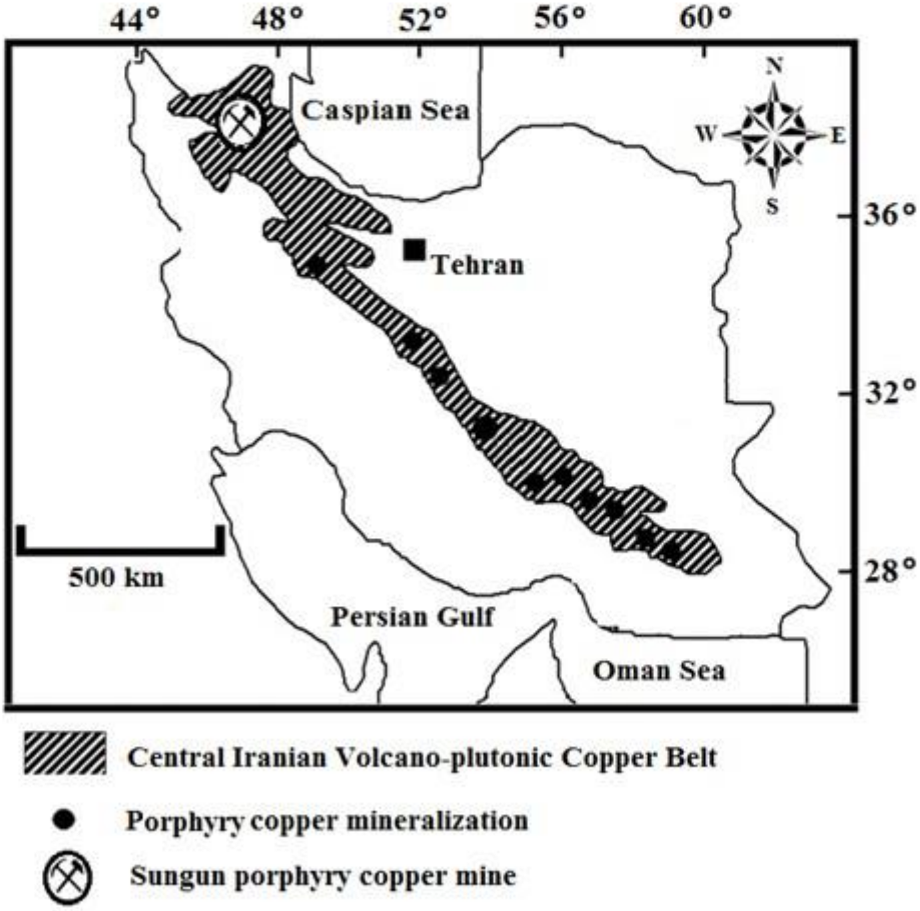

Fig. 1 Main porphyry copper deposits within the Central Iranian Volcano Plutonic Belt and the location of study area (Adapted from Zarasvandi et al. 2013)

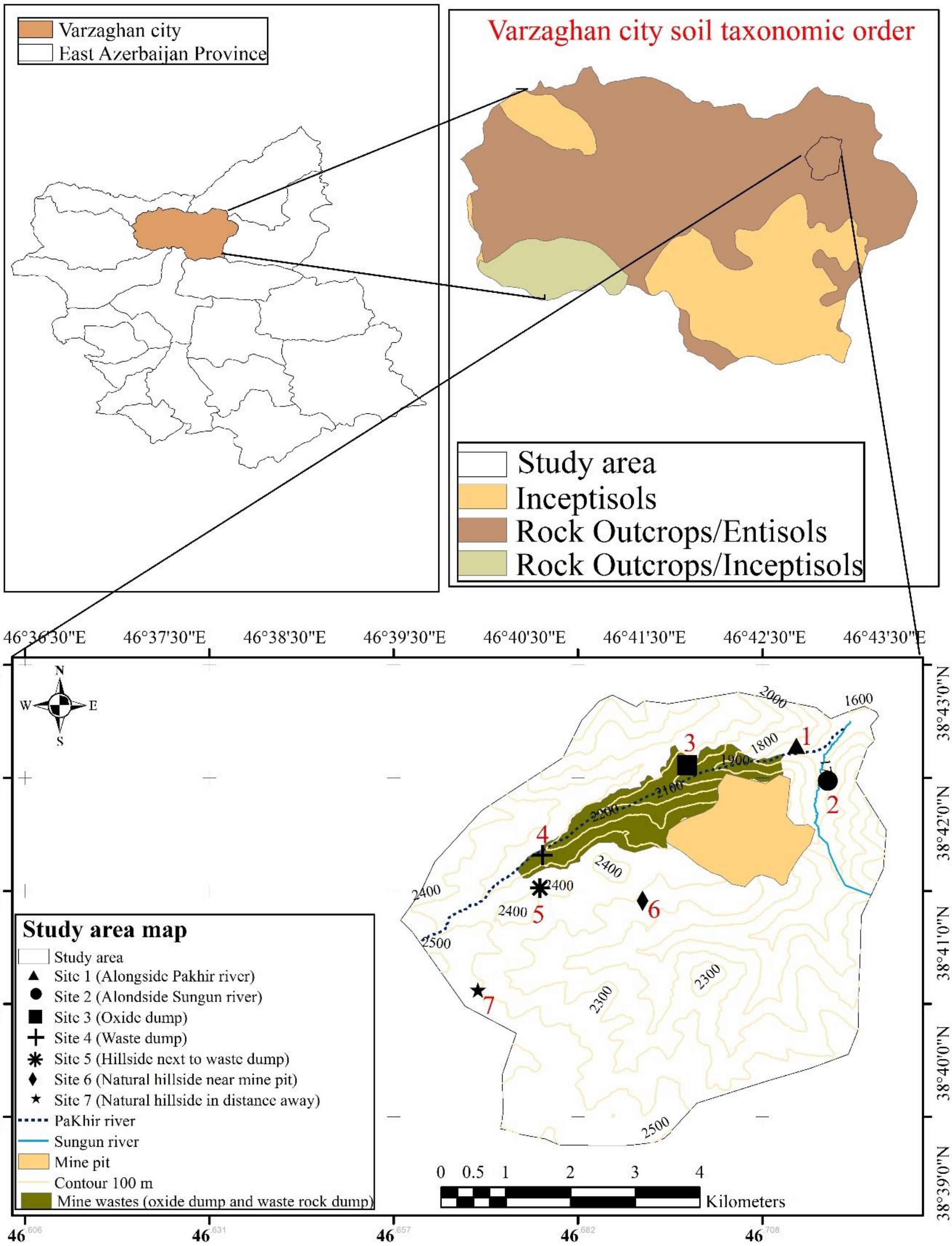

Fig. 2 Soil taxonomic order in the study area and the location of sampling sites within the Sungun Copper Mine, Iran

Fig. 3 Land cover of the **s**tudied sites in Sungun Copper Mine, Iran ((a): Waste Rock Dump and adjacent natural hillside, (b): joining point of Pakhir and Sungun Rivers, (c) and (d): natural hillsides). (Photo by Arezu Alizadeh)

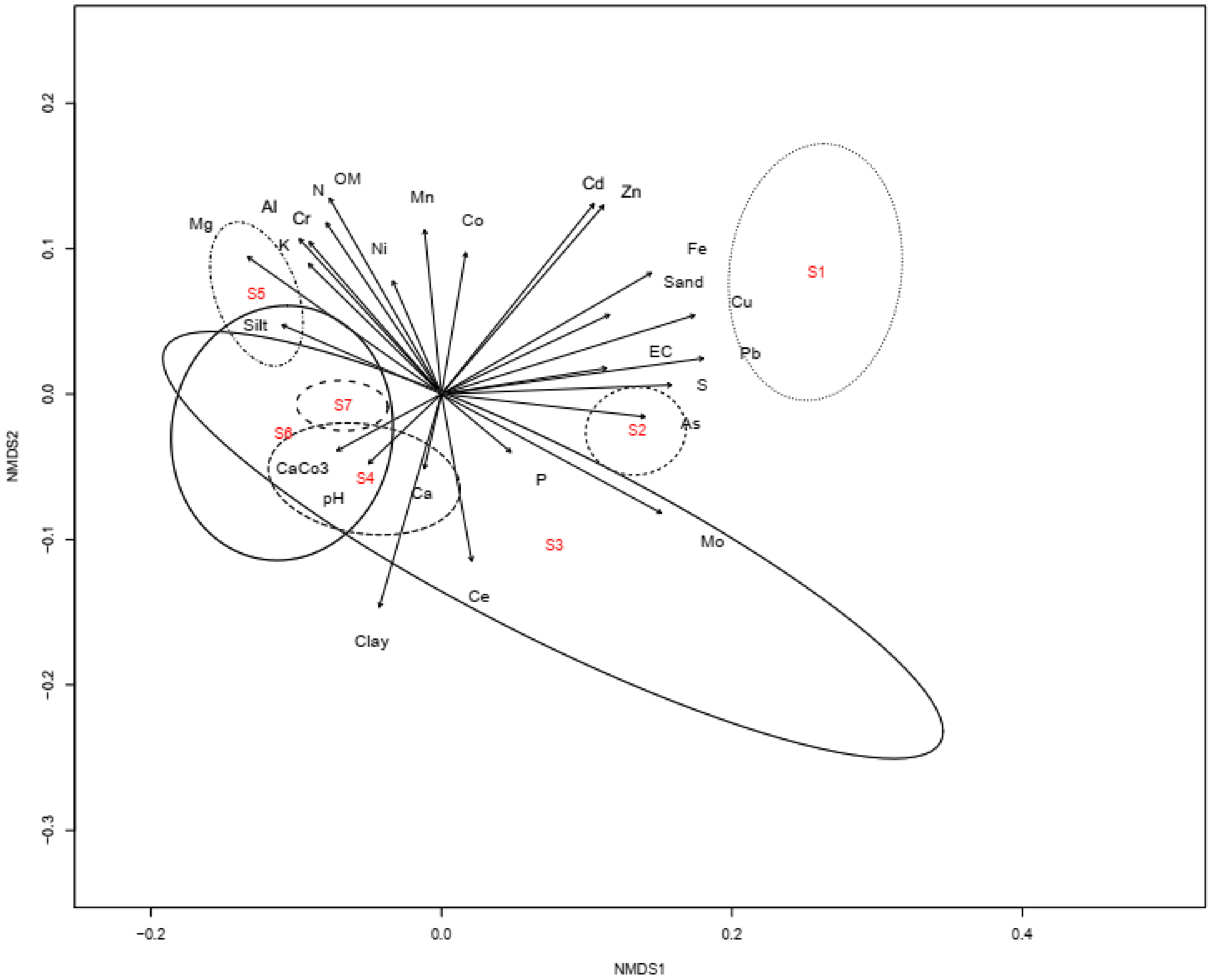

Fig. 4 NMDS ordination of studied sites and soil properties. Sites are shown with ellipses (S1: Pakhir riverside, S2: Sungun riverside, S3: Oxide Dump, S4: Waste Rock Dump, S5: natural hillside close to Waste Rock Dump, S6: natural hillsides near the mine pit, S7: natural hillside far away from mine pit) (Ordination stress>0.2)

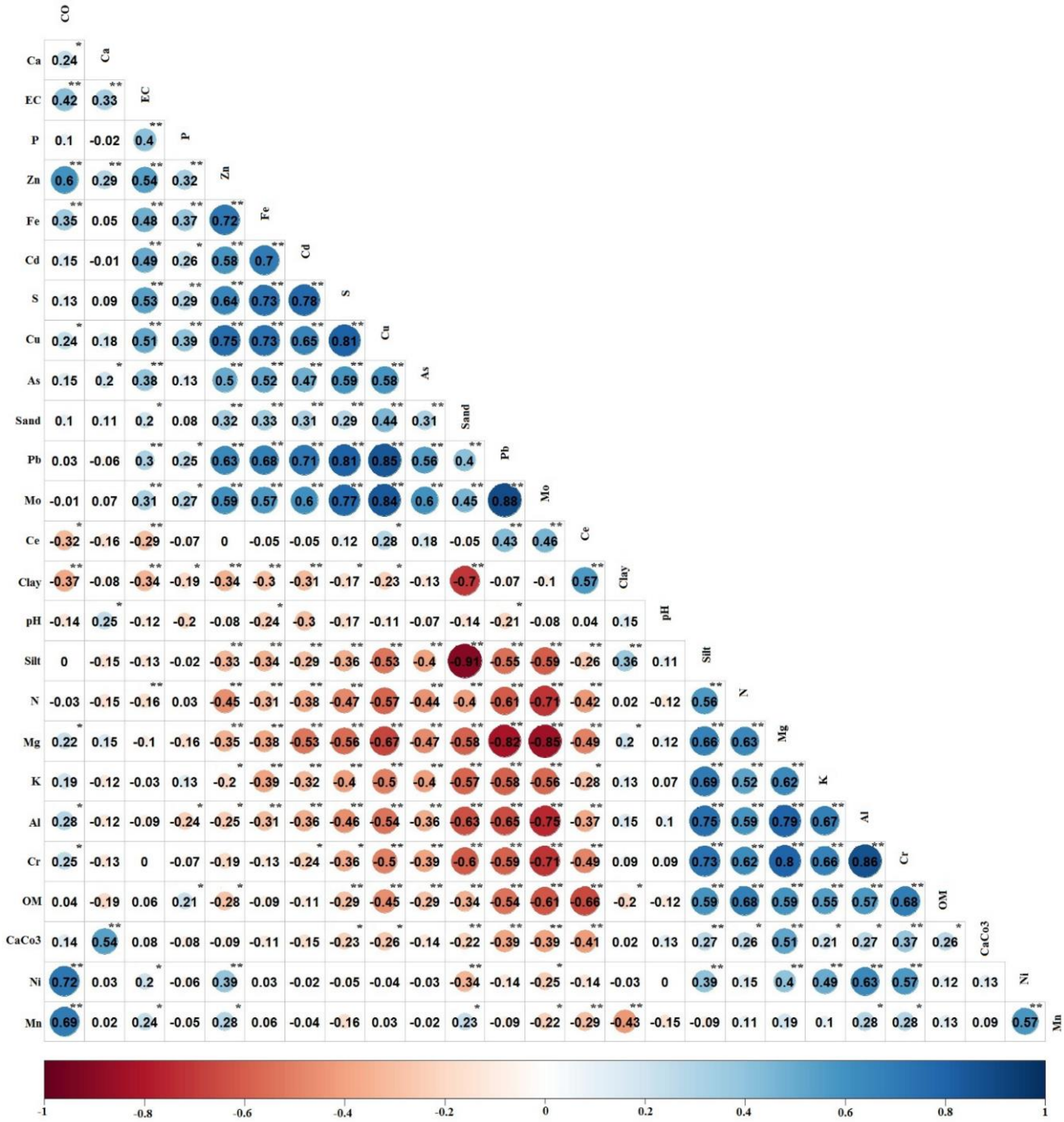

Fig. 5 Correlations of soil characteristics in Sungun Copper Mine, Iran.

Correlation is significant * at the 0.05>p>0.01, ** 0.01>p>0.001

Table 1 Contamination categories based on Geo-Accumulation Index (I_{geo})

Class	I_{geo}	Contamination categories
0	<0	Uncontaminated
1	0-1	Uncontaminated to moderately contaminated
2	1-2	Moderately contaminated
3	2-3	Moderately to strongly contaminated
4	3-4	Strongly contaminated
5	4-5	Strongly to extremely strongly contaminated
6	>5	Extremely contaminated

Table 2 Level of Ecological Risk Factor and Potential Ecological Risk

Ecological Risk Factor (E_f^i)	Level of single-factor contamination	Potential Ecological Risk Index (RI)	Level of Potential Ecological Risk
E_f^i<40	Low	RI<150	Low-grade
40≤E_f^i<80	Moderate	150≤RI<300	Moderate
80≤E_f^i<160	Higher	300≤RI<600	Severe
160≤E_f^i<320	High	600≤RI	Serious
320≤E_f^i	Serious		

Table 3 Means of soil physico-chemical characteristics in Sungun Copper Mine, Iran. Range of data are shown in parentheses. Means followed by different letter in each row are significantly different. Sites are shown in Fig. 2

Parameter	Pakhir River	Sungun River	Oxide Dump	Waste Dump	Hillside near Waste Dump	Hillside near pit	Hillside far from pit
pH	6.4[c]	6.9[ab]	6.55[bc]	7.01[a]	6.7[abc]	6.8[ab]	6.75[ab]
(Range)	(5.89–7.64)	(6.73–7.02)	(5.19–7.31)	(6.47–7.96)	(6.21–7.67)	(6.65–6.99)	(6.63–6.97)
EC (mS cm^{-1})	0.75[a]	0.48[bc]	0.65[ab]	0.3[d]	0.38[cd]	0.39[cd]	0.31[d]
(Range)	(0.36–2.38)	(0.33–0.96)	(0.28–1.52)	(0.23–0.38)	(0.27–0.47)	(0.32–0.56)	(0.26–0.37)
Clay (%)	0.26[d]	1.81[c]	15.2[a]	9.68[b]	1.52[c]	7.04[b]	15.8[a]
(Range)	(0–2)	(0–2)	(8–18)	(8–14)	(0–8)	(4–14)	(12–20)
Silt (%)	7.71[d]	9.72[cd]	18.5[ab]	14.2[bc]	24.5[a]	19.8[ab]	27.7[a]
(Range)	(5.92–18)	(2.64–18.6)	(12.6–28.6)	(1.36–26.6)	(18.5–32.8)	(0–44.2)	(23.3–31.2)
Sand (%)	91.4[a]	87.0[a]	64.5[c]	73.4[b]	72.5[b]	66.7[bc]	56.0[d]
(Range)	(82–94.1)	(79.7–95.4)	(49.4–79.4)	(63.4–86.6)	(63.4–81.2)	(49.9–92)	(48.9–62.7)
$CaCo_3$ (%)	1.55[b]	2.08[b]	1.56[b]	1.34[b]	2.21[b]	4.33[a]	1.84[b]
(Range)	(0–3)	(0.75–3.75)	(0–5.50)	(1–2)	(0.5–3.25)	(0.25–15.3)	(1–3)
K (µg g^{-1})	1.50[d]	1.56[cd]	1.65[bc]	1.62[bcd]	1.88[a]	1.75[ab]	1.62[bcd]
(Range)	(1.47–1.54)	(1.47–2.16)	(1.47–1.92)	(1.52–1.77)	(1.56–2.25)	(1.42–2.23)	(1.54–1.73)
N (%)	0.12[c]	0.07[c]	0.1[c]	0.13[c]	0.54[a]	0.34[b]	0.36[b]
(Range)	(0.03–0.52)	(0.03–0.14)	(0.07–0.14)	(0.03–0.59)	(0.11–0.91)	(0.14–0.7)	(0.1–0.76)
P (ppm)	3.77[abc]	5.31[ab]	6.946[a]	1.70[c]	3.66[abc]	3.27[bc]	2.82[bc]
(Range)	(0.21–11.1)	(2.73–9.03)	(0.42–78.8)	(0–3.15)	(0.84–10.9)	(0–14.7)	(1.26–4.62)
OM (%)	0.47[c]	0.51[c]	0.55[c]	0.08[d]	5.84[a]	2.27[b]	1.78[b]
(Range)	(0.21–0.71)	(0.34–0.84)	(0.23–2.02)	(0-17)	(3.33–8.61)	(0.37–5.80)	(1.38–2.40)

Table 4 Means of soil metal and metalloids elements in Sungun Copper Mine, Iran. Range of data are shown in parentheses. Means followed by different letter in each row are significantly different. Sites are shown in Fig. 2

Parameter ($\mu g\ g^{-1}$)	Pakhir River	Sungun River	Oxide Dump	Waste Dump	Hillside near Waste Dump	Hillside near pit	Hillside far from pit
Co	18.8^{a}	14.3^{b}	14.2^{b}	14.5^{b}	15.5^{b}	15.4^{b}	11.5^{c}
(Range)	(16–30)	(13–17)	(7–18)	(12–18)	(14–17)	(12–19)	(10–15)
Cr	27.2^{cd}	22.8^{d}	33.8^{bc}	23.7^{d}	46.3^{a}	44.0^{a}	39.6^{ab}
(Range)	(24–31)	(21–24)	(17–61)	(15–36)	(34–146)	(25–73)	(35–49)
Cu	3140^{a}	1040^{b}	288^{c}	83.5^{d}	47.9^{e}	79.3^{d}	78.7^{d}
(Range)	(1560–21800)	(823–1300)	(51–1140)	(58–120)	(30–127)	(43–206)	(61–128)
Fe	58700^{a}	38800^{b}	39400^{b}	27800^{e}	30800^{de}	32500^{cd}	34300^{c}
(Range)	(50300–67400)	(34100–42400)	(28400–80200)	(24900–30100)	(29000–33600)	(26300-36000)	(32000–36800)
Mn	961^{a}	489^{c}	436^{c}	671^{b}	779^{ab}	736^{ab}	446^{c}
(Range)	(722–1220)	(421–594)	(300–823)	(532–819)	(672–913)	(647–970)	(340–584)
Ni	40.7^{ab}	26.9^{c}	38.9^{ab}	38.2^{ab}	37.9^{ab}	42.8^{a}	32.6^{bc}
(Range)	(34–58)	(25–30)	(17–64)	(29–59)	(32–75)	(22–76)	(26–45)
Zn	665^{a}	122^{b}	112^{b}	81.3^{c}	77.5^{c}	83.6^{c}	76.1^{c}
(Range)	(391–902)	(103–141)	(62–231)	(70–91)	(70–90)	(63–110)	(61–87)
Al	17100^{c}	10900^{d}	19100^{ab}	18400^{bc}	24400^{a}	24900^{a}	22400^{ab}
(Range)	(13100–35100)	(9230–11300)	(9910–39600)	(12000–23100)	(21500–27400)	(15700–37100)	(19400–27700)
Ca	9360^{c}	15383.7^{ab}	11200^{bc}	8650^{c}	7670^{cd}	18400^{a}	6970^{d}
(Range)	(6540–12800)	(9760–19700)	(2110–28900)	(6270–15000)	(5410–9710)	(6470–50200)	(4330–6970)
Mg	6270^{c}	6310^{c}	8430^{b}	7250^{bc}	13100^{a}	12500^{a}	8680^{b}
(Range)	(5250–7400)	(5800–6940)	(1330–14700)	(4570–10100)	(11200–14800)	(8400–15800)	(7060–10600)
S	5840^{a}	3690^{a}	4560^{a}	660^{c}	578^{c}	513^{c}	1710^{b}
(Range)	(4080–11500)	(2830–4650)	(350-25300)	(246–1410)	(408–723)	(168–1250)	(1260–2470)

As	35.3^{a}	22.0^{ab}	13.1^{bc}	6.36^{cd}	3.33^{d}	5.91^{d}	4.91^{d}
(Range)	(4.3–107)	(3.4–55.9)	(2.3–160)	(1.5–14.8)	(1.6–20.1)	(1.4–15.3)	(1.6–10)
Cd	0.66^{a}	0.24^{b}	0.23^{bc}	0.19^{c}	0.2^{bc}	0.2^{bc}	0.24^{b}
(Range)	(0.4–1)	(0.22–0.27)	(0.19–0.28)	(0.17–0.21)	(0.18–0.23)	(0.17–0.23)	(0.2–0.27)
Mo	17.2^{b}	69.1^{a}	8.87^{c}	6.03^{c}	0.960^{e}	1.19^{d}	3.08^{d}
(Range)	(9–74.8)	(59.8–86.9)	(0.96–71.8)	(1.38–12.4)	(0.81–1.31)	(0.84–3)	(1.33–4.2)
Pb	946^{a}	98.8^{b}	71.6^{b}	26.2^{c}	6.45^{d}	8.88^{d}	67.2^{b}
(Range)	(521–1320)	(63–143)	(6–848)	(19–33)	(5–17)	(5–22)	(54–83)
Ce	48.0^{c}	47.6^{c}	54.6^{b}	70.3^{a}	35.6^{d}	49.8^{c}	50.8^{bc}
(Range)	(45–51)	(46–50)	(48–62)	(58–82)	(29–43)	(44–67)	(48–53)

Table 5 I_{geo} values for soil metal and metalloids elements in Sungun Copper Mine, Iran. Sites are shown in Fig. 2

Parameter	Pakhir River	Sungun River	Oxide Dump	Waste Dump	Hillside near Waste Dump	Hillside near pit	Hillside far from pit
I_{geo}Co	0.33	-0.07	-0.08	-0.05	0.04	0.04	-0.38
I_{geo}Cr	-2.46	-2.72	-2.15	-2.66	-1.70	-1.77	-1.92
I_{geo}Cu	5.25	3.66	1.80	0.02	-0.78	-0.06	-0.07
I_{geo}Fe	1.12	0.52	0.55	0.04	0.19	0.27	0.34
I_{geo}Mn	0.87	-0.10	-0.27	0.35	0.57	0.49	-0.23
I_{geo}Ni	0.44	-0.16	0.38	0.35	0.34	0.51	0.12
I_{geo}Zn	2.66	0.21	0.09	-0.37	-0.44	-0.33	-0.46
I_{geo}Al	0.19	-0.46	0.35	0.30	0.70	0.73	0.58
I_{geo}As	3.71	3.02	2.28	1.24	0.30	1.13	0.86
I_{geo}Cd	2.14	0.68	0.62	0.34	0.42	0.42	0.68
I_{geo}Mo	2.94	4.94	1.98	1.42	-1.23	-0.92	0.45
I_{geo}Pb	5.39	2.13	1.67	0.22	-1.80	-1.34	1.58
I_{geo}Ce	1.68	1.67	1.86	2.23	1.25	1.73	1.76

Table 6 The ecological risk factor (E_f^i) for soil metal and metalloids elements in Sungun Copper Mine, Iran. Sites are shown in Fig. 2

Parameter	Pakhir River	Sungun River	Oxide Dump	Waste Dump	Hillside near Waste Dump	Hillside near pit	Hillside far from pit
Co	9.42	7.17	7.12	7.25	7.74	7.70	5.76
Cr	0.540	0.460	0.68	0.47	0.93	0.88	0.79
Cu	286	94.9	26.1	7.59	4.35	7.21	7.15
Mn	2.74	1.40	1.25	1.92	2.23	2.10	1.28
Ni	12.2	8.08	11.7	11.5	11.4	12.8	9.77
Zn	9.50	1.74	1.60	1.16	1.11	1.19	1.09
As	196	122	72.7	35.3	18.5	32.8	27.3
Cd	198	72.0	69.0	57.0	60.0	60.0	72.0
Pb	315	32.9	23.9	8.74	2.15	2.96	22.4

Table 7 General level of Potential Ecological Risk Index in different sites of Sungun Copper Mine, Iran. Sites are shown in Fig. 2

RI	Pakhir River	Sungun River	Oxide Dump	Waste Dump	Hillside near Waste Dump	Hillside near pit	Hillside far from pit
Value	1029.35	340.77	213.99	130.93	108.38	127.72	147.50
Level of potential ecological risk	serious	severe	moderate	low-grade	low-grade	low-grade	low-grade