Potential soil pollution by heavy metals in Kurdistan region, western Iran: the impact of ultramafic bedrock

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Abstract

Ultramafic rocks of the ophiolitic complexes are prone to alteration and degradation, and therefore, ease of mobility of heavy metals such as nickel (Ni), chromium (Cr), cobalt (Co), lead (Pb), and vanadium (V), resulting in environmental hazards. The ultramafic rocks of the ophiolitic realm of Kurdistan province, west of Iran, show field and compositional evidence for such hazardous conditions. The ultramafic rocks are extremely rich in Mg, Fe, Ni, Cr, and Co and they are severely altered and decomposed, resulting in the formation of Serpentine minerals including chrysotile (white asbestos). Comparison of the heavy metal concentrations with standard data indicates that the samples are enriched in Ni, Cr, Co, and V. Natural processes such as alteration and anthropogenic factors such as mining, facilitated the release and mobility of these heavy metals. For reactive contaminant species, attenuation of the pollutant transport occurs by various processes including chemical precipitation, sorption, redox reactions and changes in pH.

Keywords: *Pollution, Heavy Metals, Ultramafic Bedrock, Alteration, Kurdistan.*

Introduction

Today, environmental issue is one of the crucial challenges facing humanity (Jha, 2010; Singh *et al.,* 2016). The beginning of the geochemical cycle (i.e., introduction of elements to various geochemical processes) is the generation of magma in a mantle source region. The elements which are formed in this way, enter the environmental cycle through weathering and alteration, and may be added to water, soil, air, and finally, the biotic cycles (Smedley & Kinniburgh, 2002; among many others). Therefore, determining the chemical composition of magmatic rocks as a starting point for environmental studies in regions with high magmatic activities, is crucial.

The ultramafic and mafic parts of ophiolites are usually enriched in some potentially-dangerous elements and heavy metals such as Ni, Cr, Co, V, and Pb (Bini & Bech, 2014; Cheng *et al.,* 2011; Hseu & Iizuka 2013; Hseu *et al.,* 2015; Rajapaksha *et al.,* 2012; Rinklebe *et al.,* 2016; Vithanage *et al.,* 2014). Since generation of ophiolite is mostly involved with the sea-floor spreading processes, the ophiolites commonly accommodate some heavy metals and hazardous elements such as As, Hg, Pb, Sb, Se, Mo, Sc, and Cu through submarine hydrothermal activities. This would be in addition to elements such as Ni, Cr, Co and V which have remarkable high concentrations in these rocks. In this study, the rocks from the Kurdistan ophiolites are investigated with reference to their environmental impact. As the ophiolitic complexes are the significant bedrocks for agricultural soils in Kurdistan region, western Iran, chemical composition of the ophiolitic bedrock may indirectly affect the quality of agricultural products in the region. Besides, dissolution of most of the elements and their transfer in aqueous media which can generate toxic species via chemical reactions should be in mind. Therefore, the study of changes in the concentration of chemical elements in these rocks can reveal the possibility of elemental contamination, its possible source in the region and importance of transport of the heavy metals to the food chain.

Geological setting

Ophiolites of Upper Cretaceous age (Shafaii-Moghadam & Stern, 2015) are an important feature of the Kurdistan region of western Iran and are present from Penjween to Kamyaran (Fig. 1), for more than 90 km along the Zagros Crush Zone (ZCZ, Wells, 1969). This ophiolite suite is termed “Kurdistan Ophiolite” (KO) (Shafaii-Moghadam & Stern, 2015) and can be subdivided into Chour-Nagel ophiolite Complex (CNC) and serpentinized alpine peridotites. CNC occurs as an ~ 23 km-long, NW-trending zone, parallel to the Zagros main thrust, and is surrounded by metamorphic rocks of the Sanandaj-Sirjan Zone (SSZ) (Fig. 1B), and pre-ophiolite sedimentary rocks and sheared, serpentinized ultramafic rocks of ZCZ. The Chour-Nagel segment of the KO exhibits a complete ophiolitic sequence, and despite the “crushed” nature of the ZCZ, the CNC seems to be authoctonous, showing an intact structure of the oceanic crust and the overlying sedimentary rocks. This authoctonous part is locally thrusted over the lower crustal segments of the ophiolite (ultramafic cumulates), mantle peridotites, and metamorphic rocks. Since the footwall rocks are strongly mylonitized at the contact, it is concluded that the ductile behavior of the peridotites and metapelites facilitated the dislocations in a limited distance.

Orderly from bottom to top the Kurdistan ophioltes consist of mantle peridotites, ultramafic cumulates, gabbroic rocks, sheeted dike complexes, and extrusive rocks. The gabbroic rocks occur at the base of the structurally-intact segment of the CNC. Generally, coarse-grained cumulate gabbros pass upward into microgabbros and sheeted dikes through medium-grained gabbros. The microgabbros pass transitionally upward into sheeted dikes.

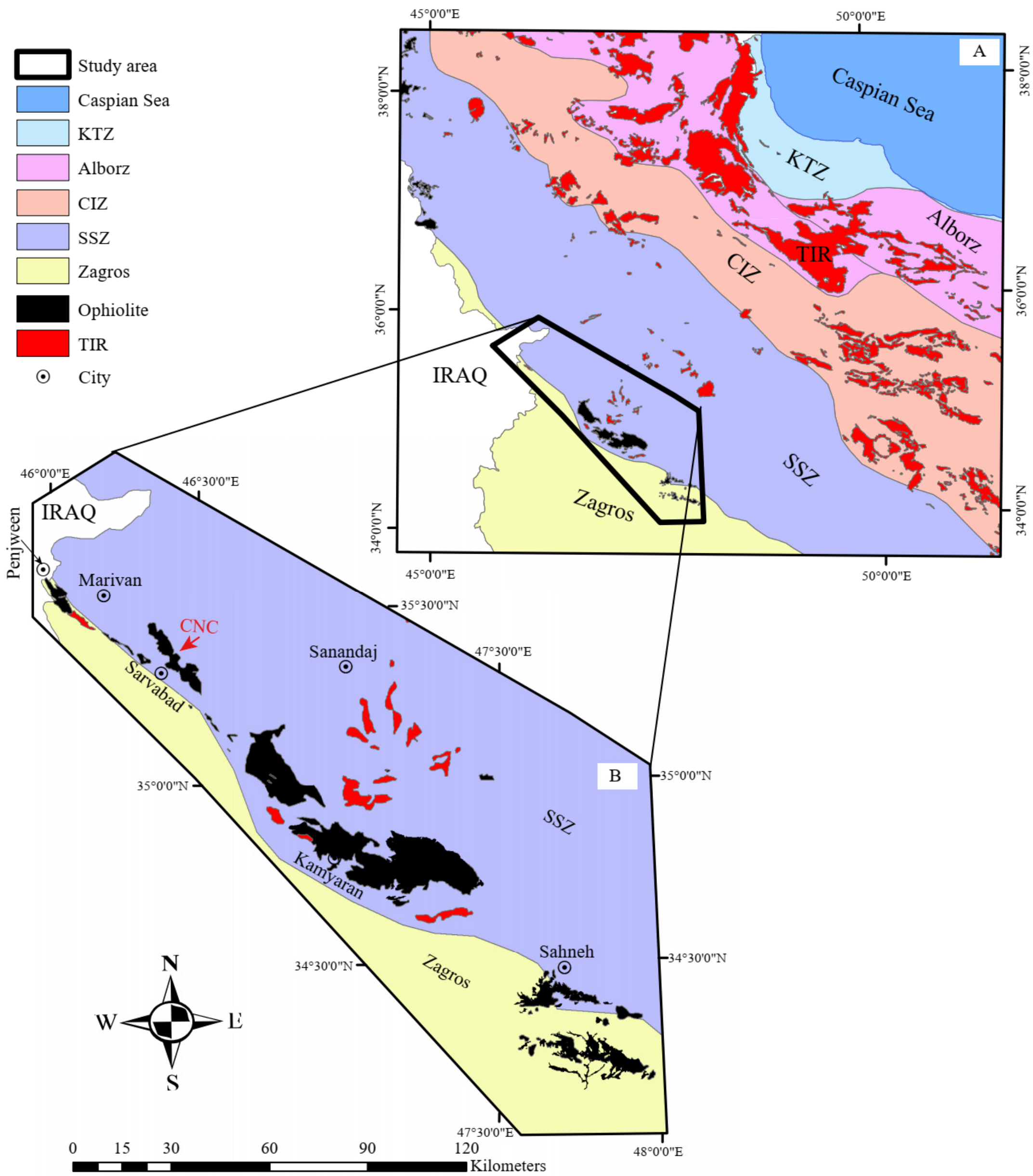

Figure 1. A) Tectonic and structural map of Iran (modified after Aghanabati 1998, 2005; Alavi 1991); B) Distribution of ophiolites in the study area (Ali et al., 2012; Shafaii-Moghadam & Stern, 2015; with modifications). Abbreviations: KTZ, Khazar-Talesh-Ziveh structural zone; CIZ, Central Iranian zone; SSZ, Sanandaj-Sirjan magmatic-metamorphic zone; TIR, Tertiary Igneous Rocks.

The sheeted dike complex is the most widespread ophiolitic unit exposed in the ophiolites of the region (Fig. 2A). The sheeted dikes are vertical and usually parallel, with thickness up to 2 m (Fig. 2B). Both the lower and upper contacts of the sheeted dike swarms are gradational over meters. Sheeted dike-gabbro transition zone displays mutually intrusive relationships between dikes and the lower gabbros. The contact between the sheeted dikes and the extrusive sequence is more complex, because dikes penetrate into various levels of pillow lava unit (Fig. 2B). The extrusive sequence is generally located in the uppermost levels of the CNC (Fig. 2C), and consists of pillow lava with minor massive lava flows.

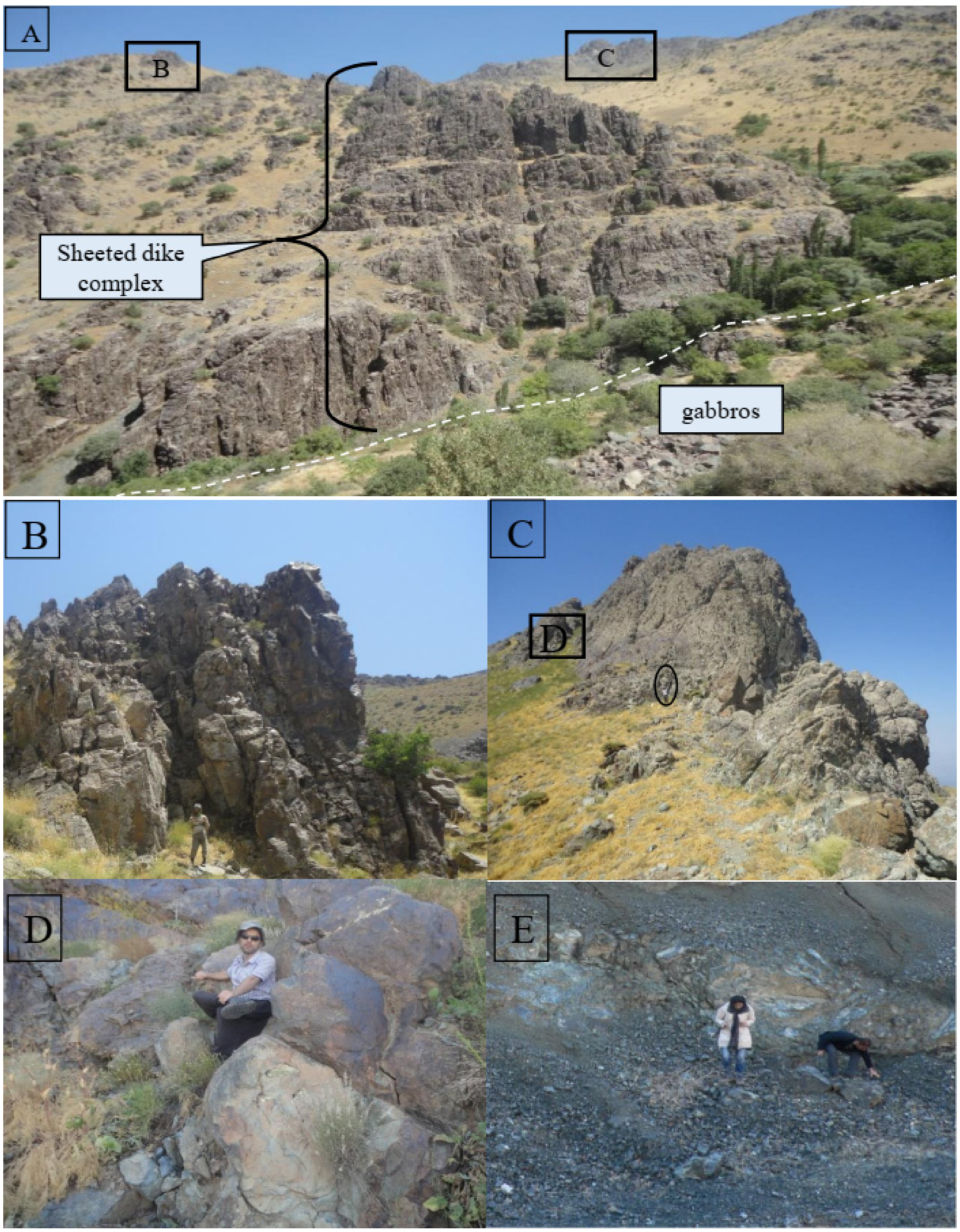

Figure 2. Outcrop view of the A) sheeted dikes complex (SDC), B) complex transitional zone between the SDC and the extrusive sequence, C) the extrusive sequence, D) Pillow lavas and E) ultramafic rocks.

Pillow lavas display variable size and shape. They have bun-shapes with a maximum diameter of 2 m (Fig. 2D). Both the sepentinized mantle peridotites and the ultramafic cumulates occur as small to medium outcrops in low-angle tectonic contact with the gabbroic rocks (Fig. 2E). They include harzburgite, and minor dunite and lherzolite (Fig. 2E and 3).

As mentioned above, the severity of degradation in the ultramafic rocks is greater than other rocks (Fig. 2E and 3). Petrographic studies also confirm severe alteration of the ultramafic rocks (Fig. 4).

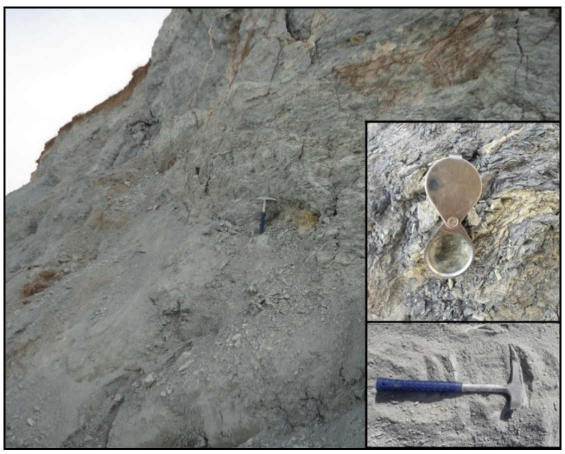

Figure 3. Outcrops of the ultramafic rocks of the Kurdistan ophiolites, which has become completely powdered due to high alteration and degradation. The upper left image shows a close-up of a serpentine vein.

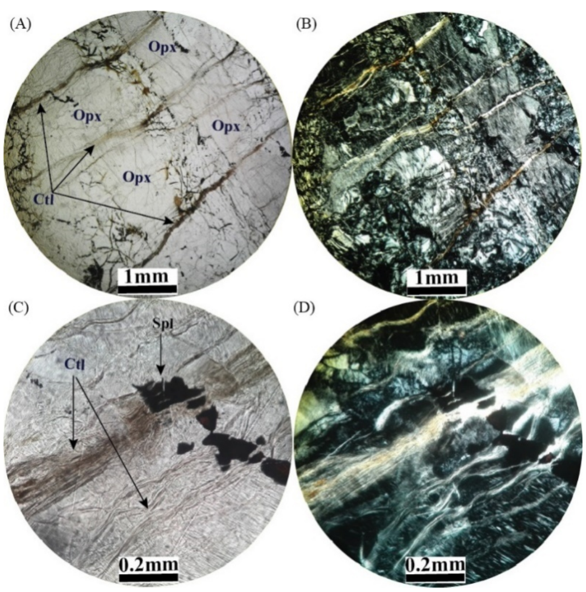

Figure 4. Development of the chrysotile (asbestos mineral) as vein and veinlet in ultramafic rocks. A and C in PPL; B and D in the XPL. Mineral abbreviations: Spinel (Spl), Chrysotile (Ctl), Orthopyroxene (Opx).

The alteration can be important from an environmental standpoint, because it forms secondary minerals, facilitates and intensifies the mobility of elements and their entry into soil and groundwater resources. Microscopic examination shows that antigorite and Lizardite are the main serpentine minerals. Chrysotile (white asbestos) is abundant as vein and veinlet in serpentinized samples (Fig. 4).

Material and Methods

To investigate the mineralogical and chemical changes, more than 300 samples of ultramafic, mafic and felsic rocks from Kurdistan ophiolites were collected. Mineral chemistry data for serpentine minerals were obtained using an Electron Probe Microanalyzer (EPMA), at the Department of Earth Sciences, University of Toronto (Table 1). After preparation and study of thin sections, the samples were powdered, and have been analyzed at the Met-Solve Analytical Lab Services Inc., Vancouver, Canada (Table 2). Major element analyses were carried out using fused disc-X-Ray Fluorescence (XRF) method. All trace elements, other than Ni, Co, Sc and Pb for which inductively coupled plasma optical emission spectrometry (ICP-OES) was used, were determined by inductively coupled plasma mass spectrometry (ICP-MS) method using two acid (HNO_3-HF) digestion. To ensure maximum dissolution of refractory elements such as transitional metals (TMEs) and high-field-strength elements (HFSEs), fused discs (using lithium metaborate flux) were prepared before digestion, and sealed vials were used. Six simultaneous analyses of an in-house standard, TD-1 (Dunn & Stringer, 1990), as unknown, have been used to evaluate analytical accuracy and precision. Accuracies ([(measured-real)/real × 100]; Jenner, 1996) are better than1 6.5% for major elements, 11% for LILEs, 15% for HFSEs, 6% for REEs, and 15% for TMs (transitional metals). 2δ standard deviation on statistical mean (Jeffrey, 1975) was used as criterion for precision of analyses. Reproducibilities evaluated in this way were better than 0.08% for major elements, 7.5% for LILEs, 7% for HFSEs, 4% for REEs and TMs

Discussion

Hazardous material in ophiolitic terrains

Some metals which are abundant in ophiolitic realms, may be potentially toxic and hazardous to public health.

Table 1. Representative microprobe analyses of major oxides (wt. %) in serpentine group minerals of ultramafic rocks.

Sample	US-1	US-2	US-3	US-4	US-5	US-6	US-7	US-8	US-9	US-10	US-11	US-12	US13
SiO_2	34.90	41.57	42.91	41.29	43.69	42.72	42.87	41.85	41.45	41.45	41.97	41.18	43.29
TiO_2	0.04	0.69	0.95	0.03	0.03	0.00	0.00	0.30	1.18	1.37	0.37	1.58	0.05
Al_2O_3	13.87	2.67	2.05	0.50	0.95	0.16	0.38	0.70	2.55	2.42	1.32	2.35	0.17
Fe_2O_3	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
FeO	4.52	4.28	3.89	6.44	3.37	2.89	4.27	3.47	2.13	1.97	1.55	2.05	6.88
MnO	0.15	0.12	0.15	0.08	0.15	0.03	0.08	0.10	0.02	0.07	0.06	0.09	0.14
MgO	33.60	39.34	39.51	41.34	39.30	41.38	41.14	39.05	39.23	39.24	41.31	38.64	44.28
CaO	0.02	0.08	0.10	0.04	0.05	0.06	0.07	0.08	0.11	0.08	0.06	0.07	0.08
Na_2O	0.03	0.03	0.00	0.00	0.02	0.00	0.00	0.06	0.00	0.02	0.00	0.00	0.00
K_2O	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Cr_2O_3	0.29	0.21	0.31	0.06	0.44	0.00	0.02	0.11	1.10	1.06	0.39	1.01	0.04
Total	87.42	88.99	89.88	89.76	88.00	87.24	88.83	85.72	87.77	87.68	87.02	86.97	94.91
Structural formula based on the 7Oxygens													
Si	1.64	1.92	1.96	1.89	2.02	1.99	1.98	1.99	1.92	1.92	1.96	1.93	1.87
Ti	0.00	0.02	0.03	0.00	0.00	0.00	0.00	0.01	0.04	0.05	0.01	0.06	0.00
Al	0.77	0.15	0.11	0.03	0.05	0.01	0.02	0.04	0.14	0.13	0.07	0.13	0.01
Fe^{+3}	0.00	0.00	0.00	0.18	0.00	0.00	0.02	0.00	0.00	0.00	0.00	0.00	0.24
Fe^{+2}	0.18	0.17	0.15	0.07	0.13	0.11	0.14	0.14	0.08	0.08	0.06	0.08	0.01
Mn	0.01	0.00	0.01	0.00	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01
Mg	2.36	2.71	2.68	2.83	2.71	2.88	2.83	2.77	2.71	2.71	2.87	2.70	2.85
Ca	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.00	0.01	0.00	0.00	0.00	0.00
Na	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.00
K	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
Cr	0.01	0.01	0.01	0.00	0.02	0.00	0.00	0.00	0.04	0.04	0.01	0.04	0.00
Total	4.97	4.98	4.95	5.00	4.94	5.00	5.00	4.98	4.95	4.94	4.99	4.93	5.00

Table 2. Representative trace element (mg/kg) concentration of the ultramafic rocks from Marivan-Kamyaran ophiolite complex.

Sample	U-1	U-2	U-3	U-4	U-5	U-6	U-7	U-8	U-9	U-10
Pb	9	3	3	6	3	6	3	6	3	3
V	40	55	62	68	45	81	63	41	60	61
Co	152	144	135	131	148	119	126	126	132	137
Cr	1938	2794	3557	3271	3120	3131	2521	4013	2630	3156
Ni	3465	3420	3573	2569	3678	2826	2924	3449	3114	3410

Elements such as Ni, Cr, Co, V, Fe, Ti, and Sc are enriched in ultramafic and mafic rocks and may be released upon weathering and alteration. Some hazardous elements like Hg, As, Mo, Cd, Cu, and Pb may be added to the ophiolitic rocks through submarine hydrothermal activities. Among the hazardous elements, As, Cd, Pb, Hg, and Cr are particularly harmful (Skei, 1978). As, Cd, Pb, and Cr^{6+} are all toxic and carcinogenic (Fergusson, 1990). Sb, Sc, Se, and Cu are toxic at higher concentrations (Merian, 1991). Hg may cause the nervous system defective, and V has a negative role in lung defect (Snyder, 1999).

Furthermore, asbestos minerals occur in ophiolites (Skinner, 2003; Vignaroli *et al.,* 2014). Presence of asbestos mineral groups in these rocks is remarkable in terms of environmental studies and a part of researches on the environmental health hazards link with these minerals (Vignaroli *et al.,* 2014). Asbestos causes disorders such as asbestosis and various types of cancers in living organisms (e.g., Bhattacharjee & Paul, 2016; Fornero *et al.,* 2009; Wu *et al.,* 2014).

Concentration of heavy metals in soil

Alteration increases orderly from top to the bottom of the ophiolite column. This may be largely due to the increasing instability of the minerals of the ultramafic rocks at surface conditions, which together with tectonic movements and weathering, exacerbate the generation of soil. Hence, the ophiolitic rocks, especially the ultramafic segment, can play a more important role in soil production than other rock types in the region. Consequently, concentration of the elements in the ultramafic rocks is reflected in the composition of the related soil, as in the Kurdistan region soils are underlain by mafic and ultramafic bedrock. This bedrock is Serpentinized due to alteration of major secondary minerals in ultramafic rocks (D'antonio & Kristensen, 2004). Microprobe analyses of serpentines (Table 1), consistent with microscopic observations, show that in addition to lizardite and antigorite, also chrysotile is a remarkable in the studied ultramafic rocks of Kurdistan region (Fig. 5).

Many elements, such as arsenic (As), selenium (Se), cadmium (Cd), Mercury (Hg), molybdenum (Mo), Pb, Ni, Cr, Co and V may be dangerous from an environmental point of view (Reeves *et al.,* 2007; Shanker *et al.,* 2005). However, in ophiolitic complexes heavy metals such as Ni, Cr, Co, Pb, and V should be considered as more important (e.g., Oze 2003; Reeves *et al.,* 2007). It is also documented by several authors that Hg, Mo, Cd, and As are commonly added to the seafloor via hydrothermal activities (e.g., de Ronde *et al.,* 2005). Various studies on heavy metals indicate that if the concentration of such elements in soil is higher than the permissible environmental standards, it can be toxic to organisms (e.g., Reeves *et al.,* 2007; Shanker *et al.,* 2005). As shown in Figure 6, concentrations of Ni, Co, and Cr in ultramafic samples are higher than the standard values; while, lead is lower than the standard values (Kabata-Pendias, 2007; MEF, 2007; Ander *et al.,* 2013). Enrichment of Ni approximately 100 times the permissible value, Co about 10 times, and Cr more than 10 times, in ultramafic rocks is a serious environmental hazards for this region (Fig. 5). Because of relative immobility of most transitional metals in aqueous systems (Shervais, 1982; Stumm & Morgan, 1996), minor leaching of these elements have been reported during weathering and alteration of ophiolitic rocks (Gillis & Thompson, 1993). Therefore, the soils which are formed from this type of protolith have a high potential for contamination with heavy metals, particularly Ni, Cr, and Co.

Immobility of elements which leads to their concentration in the soil depends strongly to pH (Perelman, 1986). In oxidizing conditions only Fe, Mn, and are concentrated in the soil. In reducing conditions, particularly when the ophiolitic bedrock contains sulfide minerals, in pH values between 4 and 5 and the pertinent soil would be rich in Mn, Co, Ni, Cu, Pb, Cd, and Cr, while in relatively acid to strongly basic conditions (pH = 6-10) elements such as Cr, Cu, and As have remarkable residence time in soil (Stumm & Morgan, 1996). Adsorption is a common and the most plausible mechanism of concentration of heavy metals in soils and depends on pH and the type of clay mineral (Stumm, 1992).

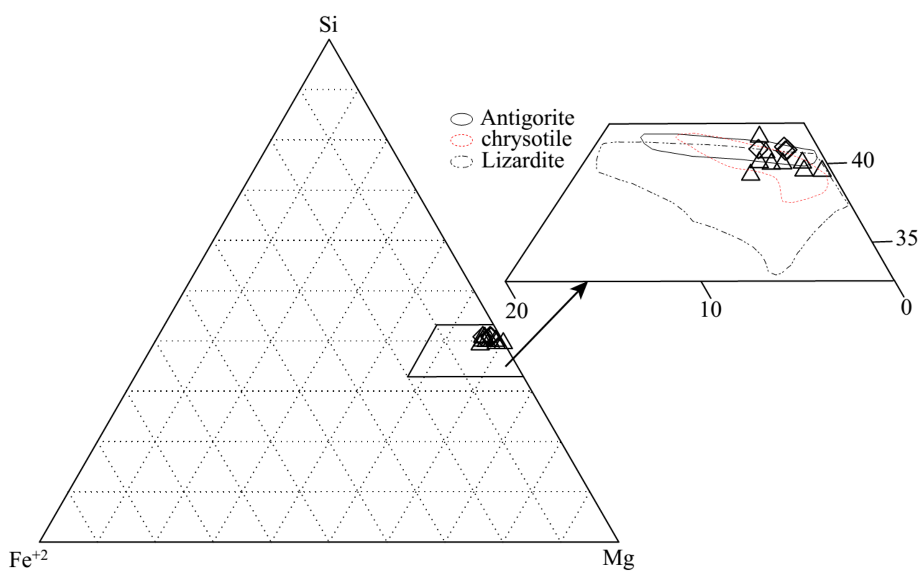

Figure 5. Composition of serpentine minerals in Fe-Si-Mg ternary diagram (D'antonio & Kristensen, 2004).

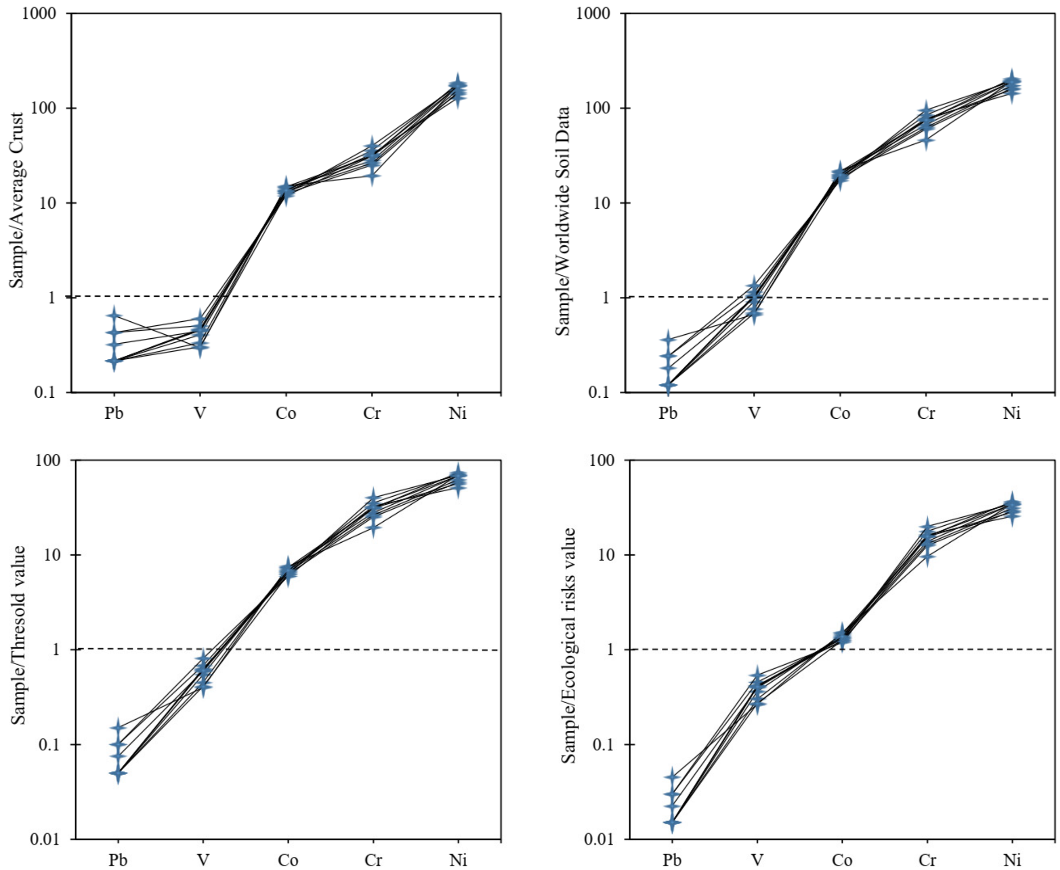

Figure 6. Comparison of heavy metal concentrations with average crust and global soil concentration (Kabata-Pendias, 2007; MEF), threshold and ecological risk values (MEF, 2007), indicating high Ni, Cr, and Co contents of the samples.

In neutral and basic conditions elements such as Cr, Cu, V, and Mo may be absorbed by clay absorbents. In acid conditions Sc, V, and As, and in weakly acid conditions a majority of heavy metals, Ni, Cd, Co, Pb, V, Zn, Cu, and As are readily absorbed by clay minerals (Stumm, 1992). In general, smectite and montmorillonite absorb V, Cr, Ti, Mn, Co, Ni, Cu, Zn, and Pb more readily, while Illite tends to absorb V, Ni, Co, Cr, Zn, Cu, and Pb, and vermiculite commonly absorbs Ti, Mn, and Fe (Fordyce *et al.,* 2000).

Progressive soil contamination via water-particle interaction

In terms of water pollution, mining and digging quarries in the ultramafic section of the ophiolite complex causes the entrance of surface water into these rocks through open pits (Fig. 7A). Water enhances the water-rock interaction process and facilitates release of various elements such as heavy metals, into the surface and groundwater bodies. In aqueous media some of the potentially-toxic elements may exist as free cations such as Co^{2+}, Cu^{2+}, Zn^{2+}, or they may be in the form of complex ions like V_4O_9, $Cr_2O_7^-$, H_2AsO_3 (Giller *et al*., 1998) In either case they are chemically active and they may get involved in hazardous reactions.

In aqueous solutions, metals, particularly as hydroxide, may be absorbed by clay minerals and fine-grained organic material and contaminate the soils. Absorption of heavy metals, like their mobility, depends on their ionic charge and ionic radius, z/r (Drever, 1988). For these reasons, Hg, Pb, Cd, Cu, Co, Fe, and Zn exist as soluble cations (White & Brantley, 1995) and some heavy metals including V, As, Cr, Se, and Mo are complex anions in aqueous media (James & Healy, 1972). In strongly reducing and acid conditions, Cd, Pb, Co, and Ni are immobile and, and therefore, precipitate. The residence time of Ni, Co, Cd, Zn, and Cu in medium pH values, and Cd, Cu, Zn and Mn in basic conditions, may be significant (Oelkers & Schott, 2009).

One of the obvious manifestations of the environmental significance of the studied ultramafic rocks, is the difference in the vegetation in areas with ultramafic basement rocks compare to the adjacent areas. Examining the satellite images reveal that in areas where the ultramafic sections are exposed, vegetation cover is very low or without vegetation, compared to adjacent areas (Fig. 7B). Vegetation in each region is influenced by a combination of different factors such as climate (temperature and humidity), seasonal variations, access to water, rock and soil types (Alexander *et al.,* 2007; Kumar & Maiti, 2013; Oze *et al.,* 2008; Roberts & Proctor, 2012). But changes in factors such as climate, seasonal variations and access to water on a limited scale (*i.e.*, the study area) are not effective enough to cause significant differences in vegetation. Hence, it could be stated that the type of rock and the resulting soil is probably a more important factor in making differences in vegetation in a limited scale (Fig. 7B).

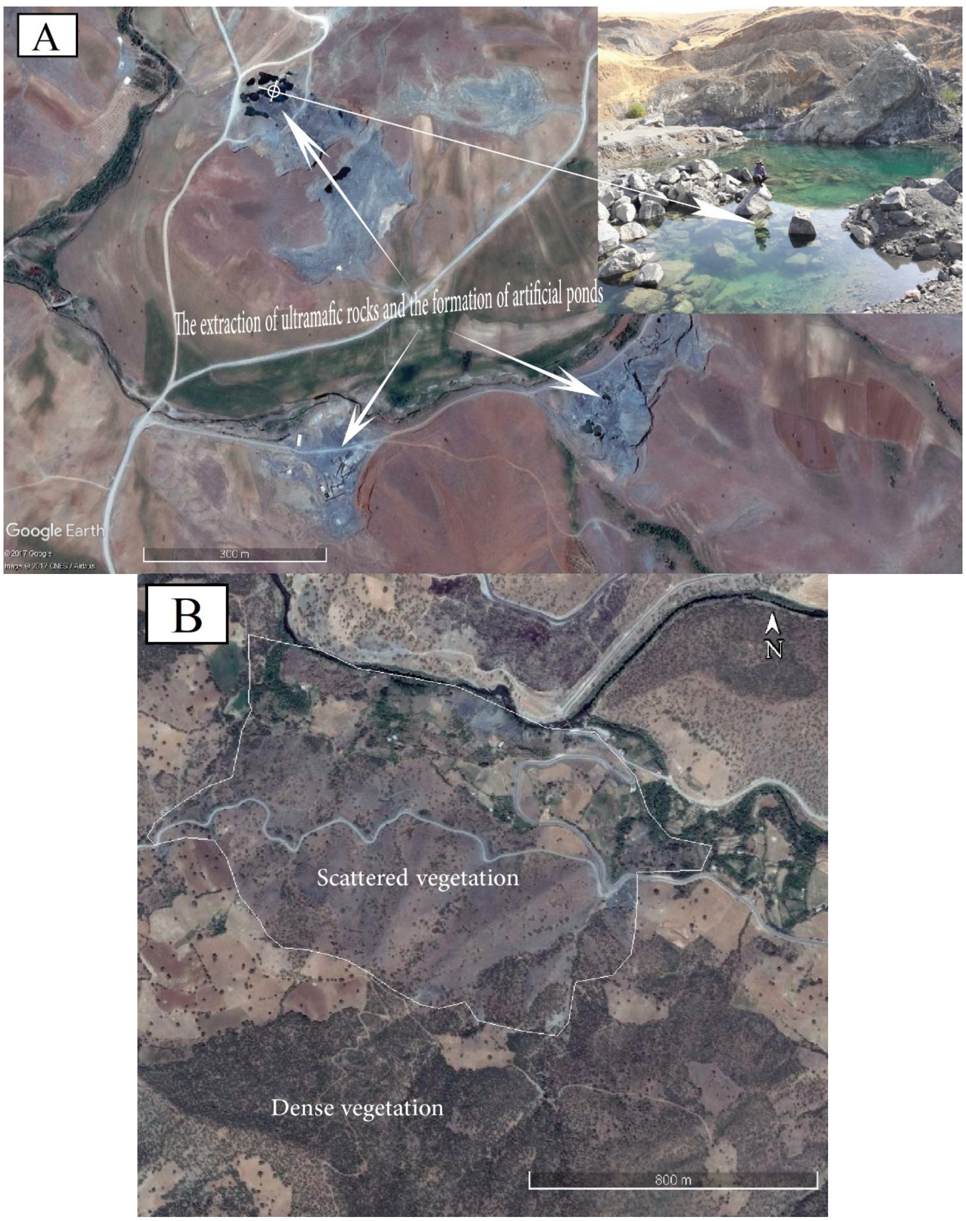

Figure 7. A) The formation of artificial ponds in quarries in ultramafic rocks; meteoric water-ultramafic rock interactions may cause release of heavy metals into the surface water and groundwater. B) Two contrasting vegetation types, scattered in soils with ultramafic bedrocks, and relatively dense in adjacent regions.

It is evidently shown that poor nutrient content, low Ca/Mg ratio, and higher concentrations of heavy metals are the main reasons for poorly distributed vegetation in ultramafic rocks and soils (e.g., Kumar & Maiti, 2013; Proctor, 2003). Furthermore, plant resistance to metallic stress is different (Ernst *et al.,* 1990; Fargašová, 2008; Freeman *et al.,* 2004; Shanker *et al.,* 2005). High concentrations of heavy metals such as Ni and Cr, however, weaken stem and root growth, deform various plants, and disrupt the natural formation of the plants (Ahmad & Ashraf, 2012). This may explain the weak and scatter vegetation over the ultramafic outcrops of the study area.

Slope instability related to serpentinization

From the perspective of natural hazards, instability of slope and landslide, is another aspect of environmental issues in the ultramafic part of the ophiolites. Extensive slope instabilities facilitate bedrock degradation, weathering and soil formation, and hence, may contribute to the soil pollution processes. In the study area, landslides mostly occur in areas where the ultramafic rocks are exposed. In transition of ultramafic rocks to serpentinite, about 12-15% of water is added to the rock (O'hanley, 1991). The addition of water will create a major difference in the mineralogical composition and consequently physical properties of the rocks, in such a way that serpentinites become prone to deformation and breakage. For this reason, serpentinites are susceptible to large fractures and landslides, and they create slopes with high instability and steepness (Alexander, 2007). Further alteration and transformation of serpentine minerals to clay minerals (Caillaud *et al.,* 2004; Deer *et al.,* 1996; Gaudin *et al.,* 2004) may enhance the instability of ultramafic rock associations (e.g. Alexander, 1988; Alexander, 2007; Dirven *et al.,* 1976; Hseu *et al.,* 2007; Lee *et al.,* 2003; Lee *et al.,* 2004; Schreier *et al.,* 1987).

Conclusion

Ultramafic rocks of the Kurdistan ophiolites are significantly altered and decomposed. Thick soil horizons have been developed over such altered, serpentinized bedrock. The soil is well-developed in the regions with ultramafic bedrock, probably as a consequence of higher weathering rates of these rocks. Generation of serpentinites contaning lizardite, antigorite, and chrysotile is indicative of water-rock interaction which may have caused release of hazardous metals to the overlying soils.

Comparison of heavy metal concentrations with standard data indicates that Ni, Co, and Cr are enriched in the serpentinized ultramafic bedrocks. Therefore, contamination of the soil which has originated due to weathering of such bedrock is readily plausible. Anthropogenic factors like mining and digging quarries may have facilitated the involvement of aqueous media, both groundwater and surface water, and may have contributed more to the environmental pollution. The lower-density of vegetation in the regions where ultramafic bedrocks are prevalent, compared to other region, may also confirm the effect of elemental contaminations on the vegetation in these areas.

The lack of adequate vegetation and the presence of secondary minerals such as serpentine and clay minerals are also important factors in the instability of slopes and the occurrence of numerous landslides in ultramafic bodies.

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