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Spatial variability of arsenic in relation with some soil forming factors

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Soil and water samples collected from Bijar area were analyzed in order to investigate arsenic contamination sources and their human risk potentiality assessment. Routine physical and chemical characteristics, iron oxides and arsenic contents were measured in 227 soil samples. Spatial variability of arsenic was calculated using inverse distance weighting (IDW) to assess the arsenic contamination potential. Furthermore, the relationships between soil parent material, physico-chemical characteristics and the spatial distribution of arsenic were analyzed. Topsoil arsenic concentration showed significant correlations with clay ($r = 0.77$), sand ($r = -0.45$), silt ($r = -0.48$), Fe oxides contents ($r = 0.65$) and cation exchange capacity ($r = 0.65$) statistically. The highest arsenic concentrations were observed in the map delineations with higher clay, Fe_2O_3 and cation exchange capacities. Parent materials derived from Fe-rich igneous (or pyroclastic) rocks and mudstone (arsenic source parent materials) composed of significantly higher arsenic contents, compared to the rest parent material types. Spatial variability analyses demonstrated that considerable extent of the studied soils was potentially contaminated by arsenic. The distribution map of arsenic contaminated areas was rasterized by IDW which confirmed that arsenic contamination in the studied area originated from some specific point sources (arsenic bearing parent materials) and spread over the lower positions by water pollution and erosion-deposition processes.

Key words: Parent materials, IDW, Physicochemical characteristics.

INTRODUCTION

Arsenic contamination as a heavy metal, not only directly affects soil physico-chemical properties, biological activity and nutrients availability, but also poses a serious threat to human health and environmental security by entering into food chains and joining to ground water (Shen and Chen, 2000). Arsenic has been recognized as a toxin and carcinogenic element to human (Hossain, 2006). New findings about the environmental and human toxicity of arsenic (Jain and Ali, 2000), combined with widespread arsenic contamination in Argentina, Bangladesh, Chile,

China, Mexico, India, Thailand, Taiwan and Vietnam (example, Meng et al., 2001; Plant et al., 2004), have sparked an increasing interest in the study of arsenic sources and how arsenic is released from the aquifer matrix.

Soil particle size distribution, organic matter, type and nature of constituent minerals, pH, redox potential, and competing ions have all been shown to influence arsenic concentration (Mahimairaja et al., 2005). The main factors affecting arsenic concentration in soils are rock composition and human activities such as mining, smelting, combustion of fossil fuels, pesticides and herbicides applications. The parent material is the most important factor affecting arsenic content (Chen et al., 2002). In general, the concentrations of heavy metals may show

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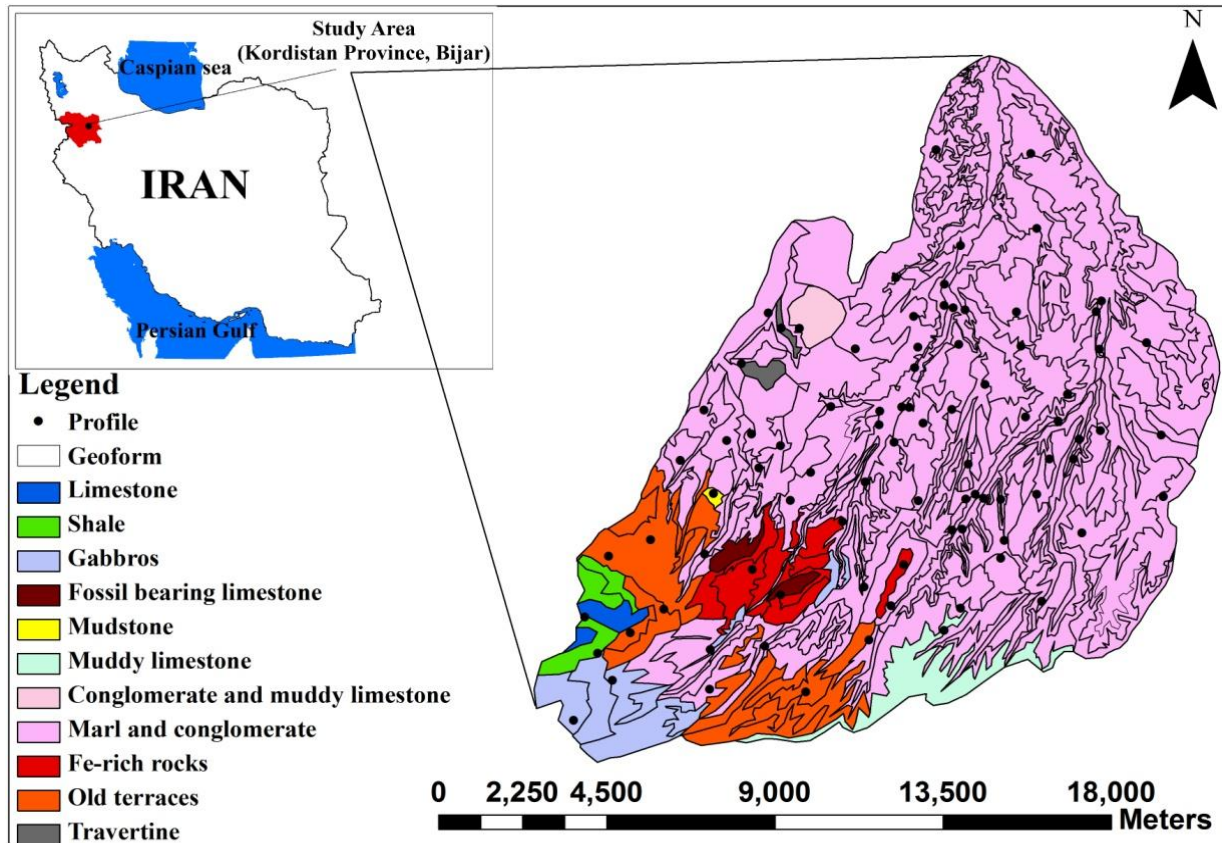


Figure 1. Geopedology map for the study area.

complex spatial patterns. Hence, to delineate the contaminated areas using limited sampling accurately, assessment of a heavy metal contamination would start by determination of its spatial distribution.

Geostatistics is a powerful interpolation tool for quantifying and reducing the uncertainties, as well as minimizing the investigation costs (Chunfa et al., 2008). It has been popularly applied in soil pollution investigation and mapping recently (Romic et al., 2007; McGraph et al., 2004). Information on heavy metals spatial distribution in soil is an important managerial and decision-making tool in contaminated areas for diagnosing the sources for arsenic in soils (Burak et al., 2010). Barati et al. (2010) and Mosaferi et al. (2005) indicated that gangrene is one of the most important illnesses observed in the Bijar county villages. Arsenic concentration in drinking water in the studied area is about 42 to 1500 $\mu\text{g/L}$ (Barati et al., 2010), which is rather high compared to provisional WHO guideline for drinking water (10 $\mu\text{g/L}$), (WHO, 2001). Therefore, people who live in these contaminated villages or even in neighboring villages are exposed to high risk of toxic elements, especially arsenic (Barati et al., 2010). The aims of this study were to identify the most effective parameters on the arsenic spatial distribution in the study area and use geostatistical approach to map the arsenic spatial distribution.

MATERIALS AND METHODS

Site description

The study area is located at west Bijar in Kurdistan Province, Iran (Figure 1). Total area, soil moisture and temperature regimes, are 20000 ha, Xeric and Mesic, respectively. Average annual rainfall is approximately 333.4 mm. Minimum and a maximum average annual temperature are 2.13 and 18.26°C, respectively. The majority of rocks are consisted of shale, travertine, marl, conglomerate, limestone, mudstone, muddy limestone, recent alluvial, old terraces, conglomerate and muddy limestone, fossil bearing limestone, Fe rich- rock and gabbros. The surveyed soils, developed mainly on quaternary alluvial, Miocene and Pliocene deposits, are characterized by a high degree of heterogeneity, ranging from fine to coarse textured Entisols, Mollisols and Inceptisols. Land use of the area is allocated to agriculture and rang uses. The great groups of studied soils are of: Xerorthents, Calcixerolls, Haploxerolls, Endoaquolls, Haploxerepts and Calcixerpts. Sampling points were selected based on the geopedologic map.

Geopedologic map and collection of soil samples

At first, topographic (1:25000), geology (1:100000) and land use (1:25000) maps were scanned, imported to the GIS and geo-referenced. The contour lines of the topographic map were digitized and DEM were created through "contour interpolation" function in ArcGIC using linear interpolation method. Then the slope map was produced using DEM. In addition, the geology map was digitized. Aerial photos of study area were interpreted according to the

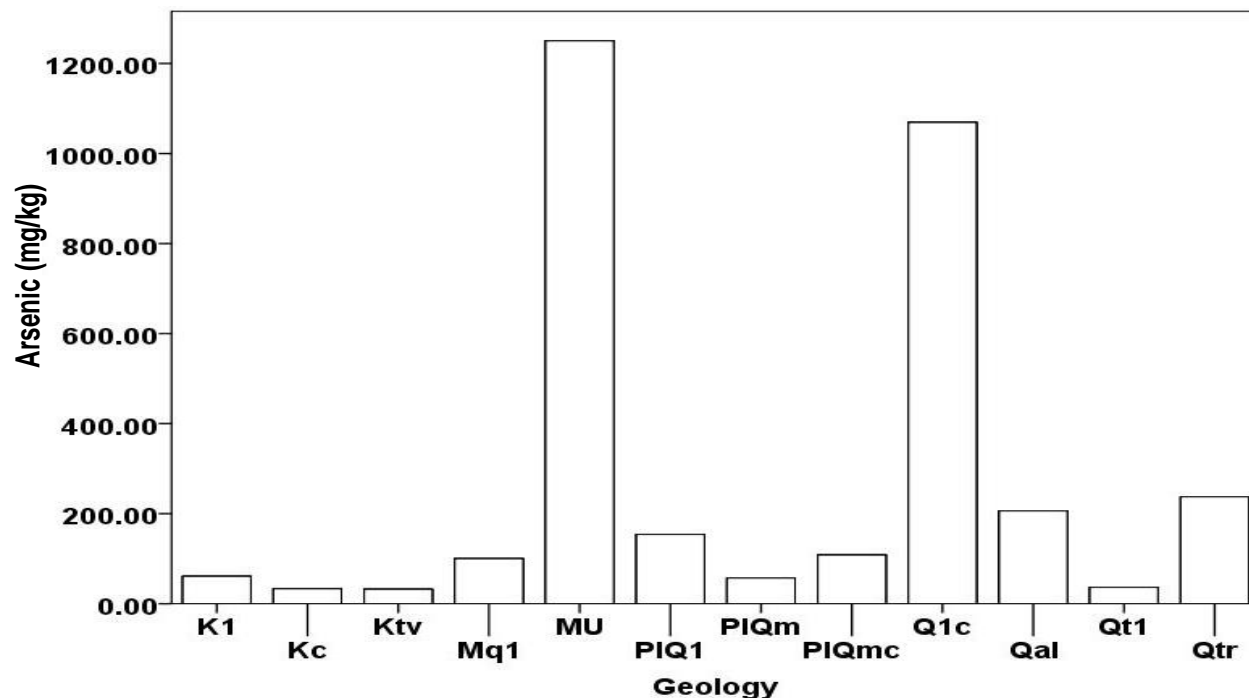


Figure 2. Arsenic concentrations mean (mg kg^{-1}) for the different parent materials. K1, Shale; Kc, limestone; Ktv, gabbros; Mq1, fossil bearing limestone; Mu, mudstone; PIQ1, muddy limestone; PIQm, conglomerate and muddy limestone; PIQmc, marl and conglomerate; Q1c, Fe rich- rock; Qal, recent alluvial; Qt1, old terraces; Qtr, travertine.

geopedological method of Zinck (1988). The aerial photos covering the study area at 1:55000 were scanned, imported into ARCGIS, geo-referenced with an ortho-correction to a horizontal precision of 3 to 15 m, using eight tie-points per photo (Rossiter and Hengl, 2003). Also, the geomorphic units which were interpreted and stratified with bases of Zinck (1988) on aerial photos (milars) were scanned and imported into ARCGIS and after ortho-georeferencing were subjected to on screen digitizing. Finally the geopedological map was produced.

Based on geopedologic map and purposive sketch, 88 representative profiles were selected for sampling. The profiles were excavated, described and classified according to the Soil Survey Manual (Soil Taxonomy, 1993) and Keys to Soil Taxonomy (Soil Survey Staff, 2010) respectively. From the genetic horizons, 227 samples were taken and sent for laboratory analysis. Soil samples from A horizon were grouped as topsoil, while those from B and C horizons were grouped as subsoil samples (Figure 1).

Laboratory analyses

All of the soil samples were air-dried and ground to pass through a 2-mm sieve. All the 227 representative samples (contains topsoil and subsoil) were analyzed for arsenic. The soil samples were digested by aqua regia with a mixture of nitric and hydrochloric acids according to the 3050B method of USEPA (USEPA, 1996). Soil arsenic was measured by graphite furnace atomic absorption spectrometry (GTA 110; Varian Spectra 220). Particle-size distribution was determined after the dissolution of CaCO_3 with 2 N HCl, and decomposition of organic matter with 30% H_2O_2 . After repeated washing for removal of salts, the soils were dispersed using sodium hexa metaphosphate, and the sand, silt and clay fraction were separated by sedimentation and determined by the pipette method (Richards, 1954). Alkaline-earth carbonate (lime) was measured by

acid neutralization (Richards, 1954). Organic carbon was measured by wet oxidation with chromic acid and back titrated with ferrous ammonium sulfate according to Black (1982). Soil pH was measured in saturation paste. Electrical conductivity was determined in the saturation paste extract (Richards, 1954). Cation exchange capacity (CEC) was determined using sodium acetate (NaOAc) at a pH of 8.2 (Chapman, 1965). Free Fe oxide was measured by sodium citrate and dithionate (Richards, 1954). Available phosphorus was measured according to Nelson (Olsen et al., 1954). Soluble anions were measured by titration method (Richards, 1954).

Spatial variability and statistical analysis

The data for soil as (total), soil Fe (sodium citrate and dithionate extractable) are shown as Krige map (Arc GIS version 9.2, Spatial Analyst module: Point Interpolation using IDW method). Statistical analyses of the data were performed with SPSS and MINITAB softwares. Statistical analyses of the studied soil properties were carried out on the samples prepared from topsoil and subsoil. Descriptive statistical analyses including mean, standard deviation, minimum, maximum, Skewness, Kurtosis, Pearson's correlation and mean comparison using Duncan's test were conducted using SPSS software.

RESULTS AND DISCUSSION

Relationship between spatial variability of soil arsenic with parent materials

Figure 2 shows the arsenic mean concentrations for different parent materials of the area. The Fe-rich igneous (or

Table 1. Grouping of parent materials according to arsenic (As) concentration mean by Duncan's test.

Landform	As (mg kg ⁻¹)
Travertine	237.43 ^b
Marl and conglomerate	109.09 ^d
Limestone	33.08 ^f
Gabbros	32.3 ^f
Muddy limestone	154.15 ^c
Mudstone	1250 ^a
Shale	60.74 ^e
Fe-rich rocks	1069 ^a
Alluvium	206.78 ^b
Old terrace	36.37 ^f
Fossil bearing limestone	169.94 ^c

Numbers with the same superscripted letter in each column are not statistically different.

pyroclastic) rocks and mudstone (arsenic source parent materials) are the most important source of the arsenic in studied area. These parent materials (arsenic source parent materials) have high arsenic. The arsenic of the parent materials (arsenic source parent materials) was weathered and transported with sediments and water along time. The transported arsenic from the mentioned parent materials was accumulated in the lower parts. The lower parts with alluvium conglomerate and muddy limestone, marl and conglomerate are affected by the transported sediments and water of Fe-rich rock and mudstone and their arsenic concentrations were increased and have high to medium arsenic content. In contrast, the other parts located at the upper with shale, limestone and gabbros parent materials have not been by the transported arsenic; therefore, they have lower arsenic concentration.

Muddy limestone and travertine have been reported as medium arsenic concentrations. Arsenic concentrations have been reported up to 490 mg kg⁻¹ in a mud rock (Smedley and Kinniburgh, 2002) and similar results were obtained for the mudstone bearing landforms in this study. Arsenic is often significant in hot spring deposits, but reports from travertine are few. It has been noted in a deposit at Vichy, France at a level of 1300 mg kg⁻¹. Arsenic is an important component of the heavily mineralized travertine known from Western Turkey (Bernasconi, where it is associated with several oxides and sulphides of antimony, example dussertite, scorodite and stibnite). Arsenic has also been reported from Italian deposits at levels up to 1600 mg kg⁻¹ (Allan, 2005). These results demonstrate that local trends in arsenic concentration are obviously detectable as related to parent materials.

Also, arsenic concentrations in igneous rocks are generally Low (average: 1.5 mg kg⁻¹). Volcanic glasses are only slightly higher with an average of around 5.9 mg kg⁻¹. Arsenic concentrations in metamorphic rocks tend to

reflect the concentrations in their igneous and sedimentary precursors. Most contain around 5 mg kg⁻¹ or less. The concentration of arsenic in sedimentary rocks is typically in the range of 5 to 10 mg kg⁻¹. Average sediments are enriched in arsenic relative to igneous rocks. Marine argillaceous deposits have higher concentrations than non-marine deposits. This may also be a reflection of the grain-size distributions, with potential for a higher proportion of fine material in offshore pelagic sediments as well as systematic differences in sulphur and pyrite contents. Some of the highest observed arsenic concentrations are found in ironstones and Fe-rich rocks. Collected data for ironstones from various parts of the world showed arsenic concentrations up to 800 mg kg⁻¹ in a chamosite-limonite oolite. Arsenic concentrations are reported up to 490 mg kg⁻¹ in a mud rock (Smedley and Kinniburgh, 2002). Boyle and Jonasson (1973) reported arsenic concentrations with values up to 400 mg kg⁻¹ and for Fe-rich rocks up to 2900 mg kg⁻¹. Findings of also Glenn and James Lester (2010) showed the arsenic values for samples from Oligocene wells had the highest percentage compared to the other constituents (Holocene, Pleistocene, Pliocene and Miocene).

The statistical analysis carried out using SPSS confirmed the accuracy of this grouping as shown in Table 1. The results of statistical comparison (Duncan's test) showed that the Fe-rich rock and mudstone (arsenic source parent materials) were significantly different regarding soil arsenic content than the other parent materials (Table 1). It showed that the Fe-rich rock and mudstone parent materials (arsenic source parent materials) have higher arsenic than the parent materials.

The topsoil and subsoil

Relationship between spatial variability of soil arsenic with Fe, Fe₂O₃ and clay and CEC

As indicated by the skewness coefficient (Table 2), arsenic distribution is strongly positively skewed, meaning that the data does not follow a normal distribution. In addition, arsenic maximum values were very high, with nine to ten standard deviations from the mean. However, more robust measures can be considered in order to reduce the influence of extreme values and skewness. The transformation of the variable arsenic overcomes these problems. Logarithmic transformation is applied in order to normalize positively skewed data sets. Figure 3 shows the spatial variability maps of topsoil and subsoil the soil arsenic. Both topsoil and subsoil arsenic values have been divided into ten groups shown as legends in the maps (Figure 3a and b). Figures 4 to 6 show the spatial variability maps of topsoil and subsoil for the clay, CEC and sodium citrate and dithionate extractable Fe₂O₃. The topsoil and subsoil values have been divided into five groups shown as legends in the maps.

Table 2. Descriptive statistics for topsoil and subsoil soil properties and arsenic concentration.

Parameter	pH	OC (%)	CaCO ₃ (%)	Clay (%)	Silt (%)	Sand (%)	Fe ₂ O ₃ (%)	CEC (cmol ⁺ /kg)	P (mg kg ⁻¹)	HCO ₃ ⁻ (MeqL ⁻¹)	Cl ⁻ (MeqL ⁻¹)	SO ₄ ⁻² (MeqL ⁻¹)	As (mg kg ⁻¹)	
Topsoil	Number	83	83	83	83	83	83	83	83	83	83	83	83	
	Mean	7.5	1.2	17.6	34.3	36	29.3	0.93	20.4	12.5	6	5.2	1.03	120
	SD	0.2	0.6	10.1	10.8	7.8	10	0.58	4	9.5	1.33	2.4	0.86	167
	Minimum	7.2	0.2	0.5	12.8	16	13	0.16	10.2	0.32	2.5	1.2	0.25	17
	Maximum	7.9	2.7	43.5	62.5	53	59.2	2	28.7	53.2	10	12.5	5.6	1247
	Skewness	-0.11	0.81	0.56	0.57	0.21	0.46	0.31	-0.006	1.51	0.19	0.33	2.51	4.41
	Kurtosis	-0.41	-0.002	0.02	0.01	0.11	-0.31	-1.31	-0.44	3.21	1.21	-0.04	9.41	25.81
Subsoil	Number	144	144	144	144	144	144	144	144	144	144	144	144	
	Mean	7.6	0.5	22.6	36.2	33.9	30.17	0.98	20.5	6.23	4.9	8.19	2.57	160.4
	SD	0.16	0.37	10.63	11.2	7.3	11	0.6	4.26	7.1	1	6.2	1.6	296.2
	Minimum	7.1	0.02	0.25	12.8	15	12	0.02	7.07	0.39	2.5	1.25	0.4	16
	Maximum	7.9	1.95	46.25	64	52	61.2	2.09	28.9	36.44	8	36.25	8.4	2649
	Skewness	-0.71	1.25	0.41	0.72	-0.11	0.48	0.24	-0.28	2.01	0.62	2.51	1.16	5.45
	Kurtosis	0.76	2.11	-0.29	0.02	0.002	-0.19	-1.51	0.29	4.31	0.45	7.31	1.51	37.91

Topsoil arsenic concentration decreased as the distance from the source parent materials increased showing a range of 17 to 1900 mg kg⁻¹ (Figure 3a). Similarly, subsoil arsenic concentration decreased as the distance from the source parent materials increased showing a range of 19-2650 mg kg⁻¹ (Figure 3b). Moreover, topsoil citrate and dithionate extractable Fe₂O₃ concentrations were found higher nearer to the parent materials source and lower at the distant points, the range being 200 to 14000 mg kg⁻¹ (Figure 4a). Similarly, subsoil citrate and dithionate extractable Fe₂O₃ concentrations were found higher nearer to the parent materials source and lower at the distant points, the range being 700 to 14000 mg kg⁻¹ (Figure 4a). Topsoil and subsoil clay percentages were found higher nearer to the parent materials source and lower at the distant points, the range

being 12 to 60 % (Figure 5a and b). Topsoil and subsoil CEC contents were found higher nearer to the parent materials source and lower at the distant points, the range being 10 to 28 C mol/kg (Figure 5a and b). In the area with high clay, CEC and Fe₂O₃ contents, the concentration arsenic increase.

Table 2 presents the descriptive statistics of the arsenic, CEC, CaCO₃, P, pH, clay, sand, silt, Fe oxides, soluble anions and organic carbon content of the top and subsoils studied. Correlation coefficients of arsenic and CEC, clay, sand, silt, and Fe oxides content of the top soils and sub soils studied are shown in Table 3. As shown, Fe oxides, soil texture and related properties such as CEC showed a meaningful influence on arsenic concentration in soils, and it appeared clearly that the nature of soil parent material controls the

distribution of arsenic concentrations in soils to a relevant extent. Moderately strong and statistically significant correlations are detected between topsoil arsenic concentration and clay content ($r = 0.77$), sand content ($r = -0.45$), CEC ($r = 0.65$), Fe oxides ($r = 0.65$) and silt content ($r = -0.48$). The correlations exhibited with pH, and organic matter, CaCO₃, soluble anions were insignificant. Subsoil arsenic concentrations showed similar correlations with soil properties. Hossain et al. (2008) showed that iron, manganese and phosphorus a clear influence on arsenic concentration in soils. Statistically, significant correlations were detected between soil arsenic concentration and iron, manganese and phosphorus content. The correlations exhibited with pH, and organic matter was insignificant. Ungaro et al. (2008) showed soil texture and related properties such as CEC

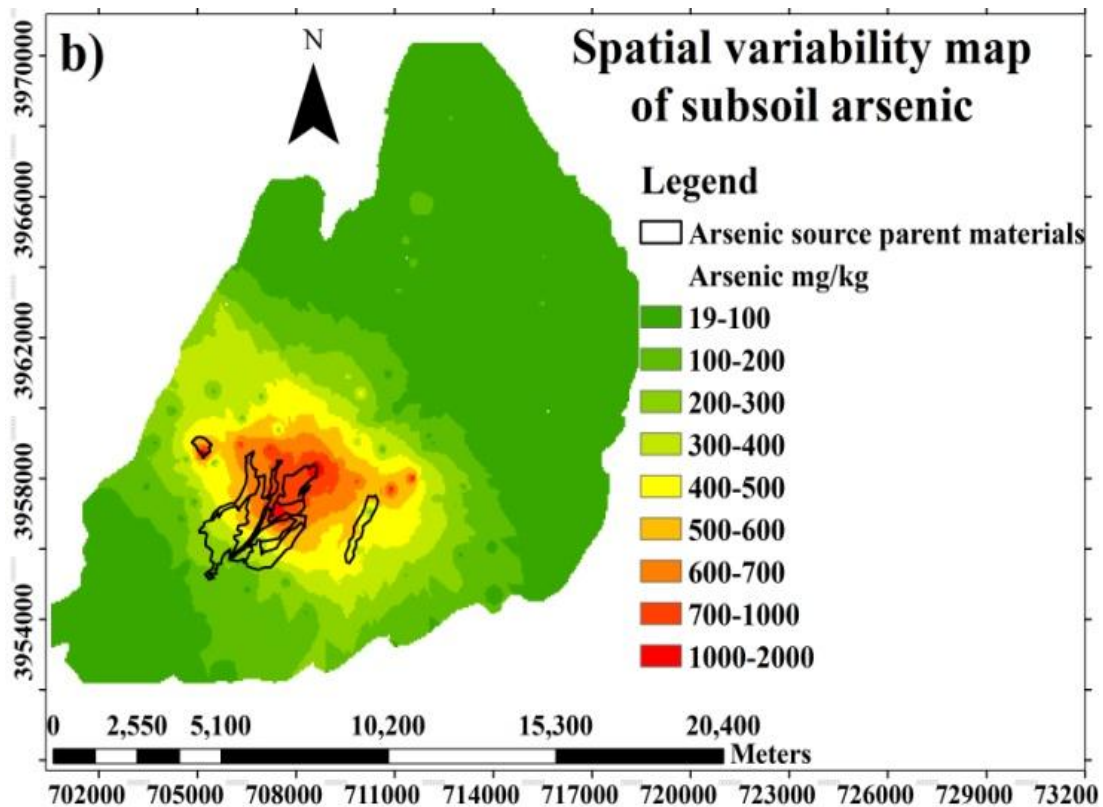
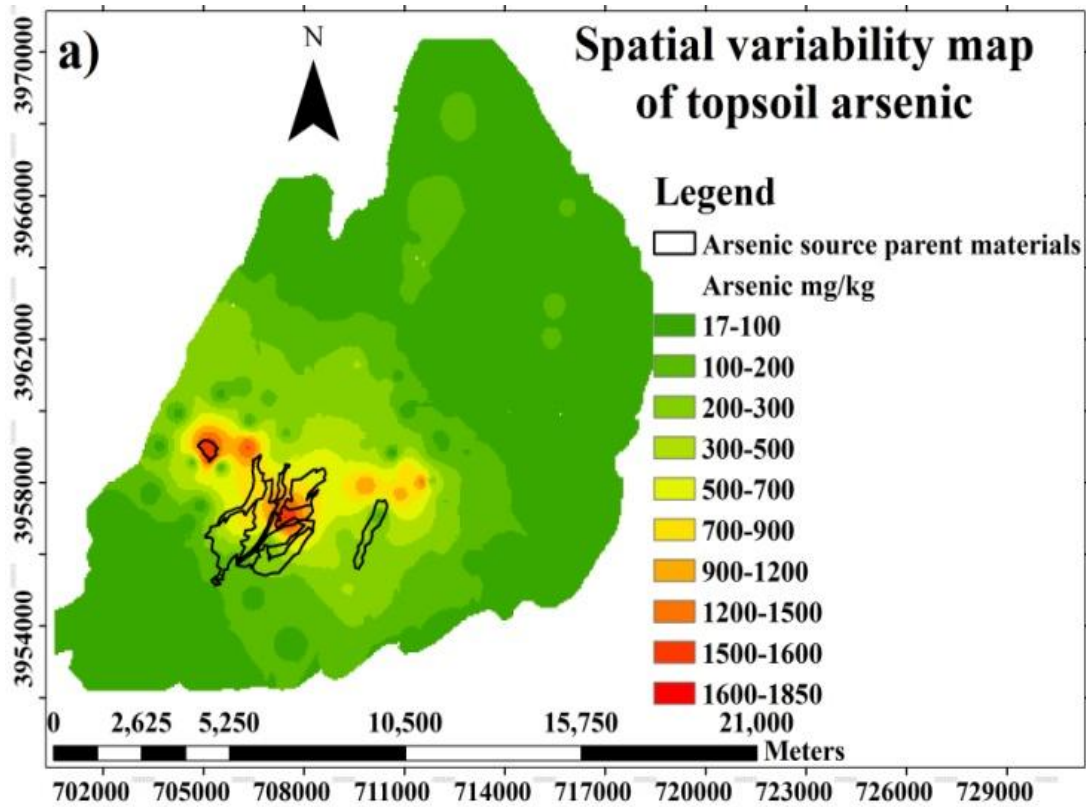


Figure 3. Topsoil (a) and subsoil (b) arsenic spatial variability maps.

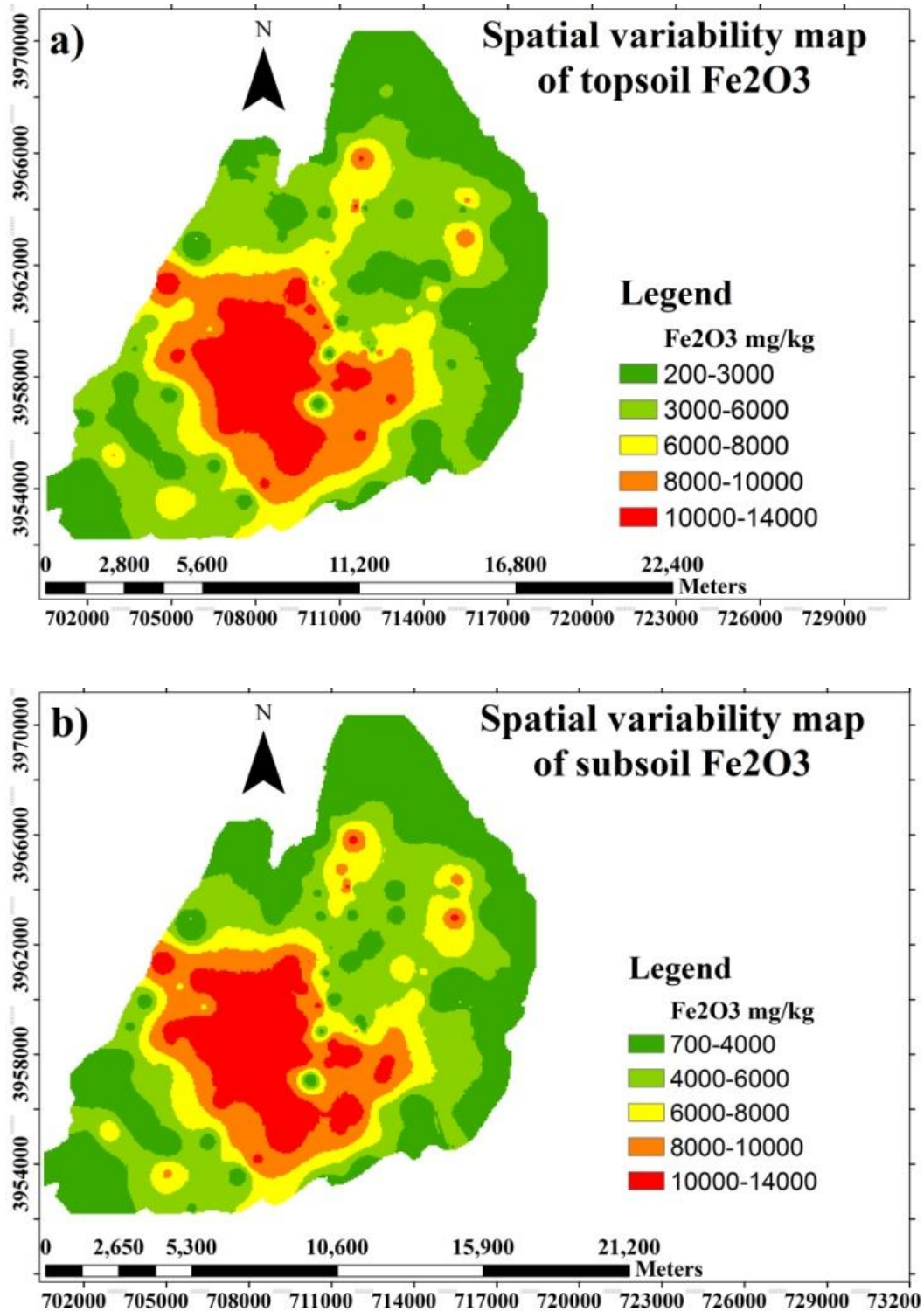


Figure 4. Topsoil (a) and subsoil (b) Fe₂O₃ spatial variability maps.

exert a clear influence on arsenic concentration in soils. Furthermore, moderately strong and statistically significant correlations detected between topsoil and subsoil arsenic concentration and clay content, sand content, CEC,

total carbonates, and silt content. The correlations exhibited with pH, and organic matter was insignificant. The important oxides particularly Fe oxides control the concentration of arsenic in natural environments for a long

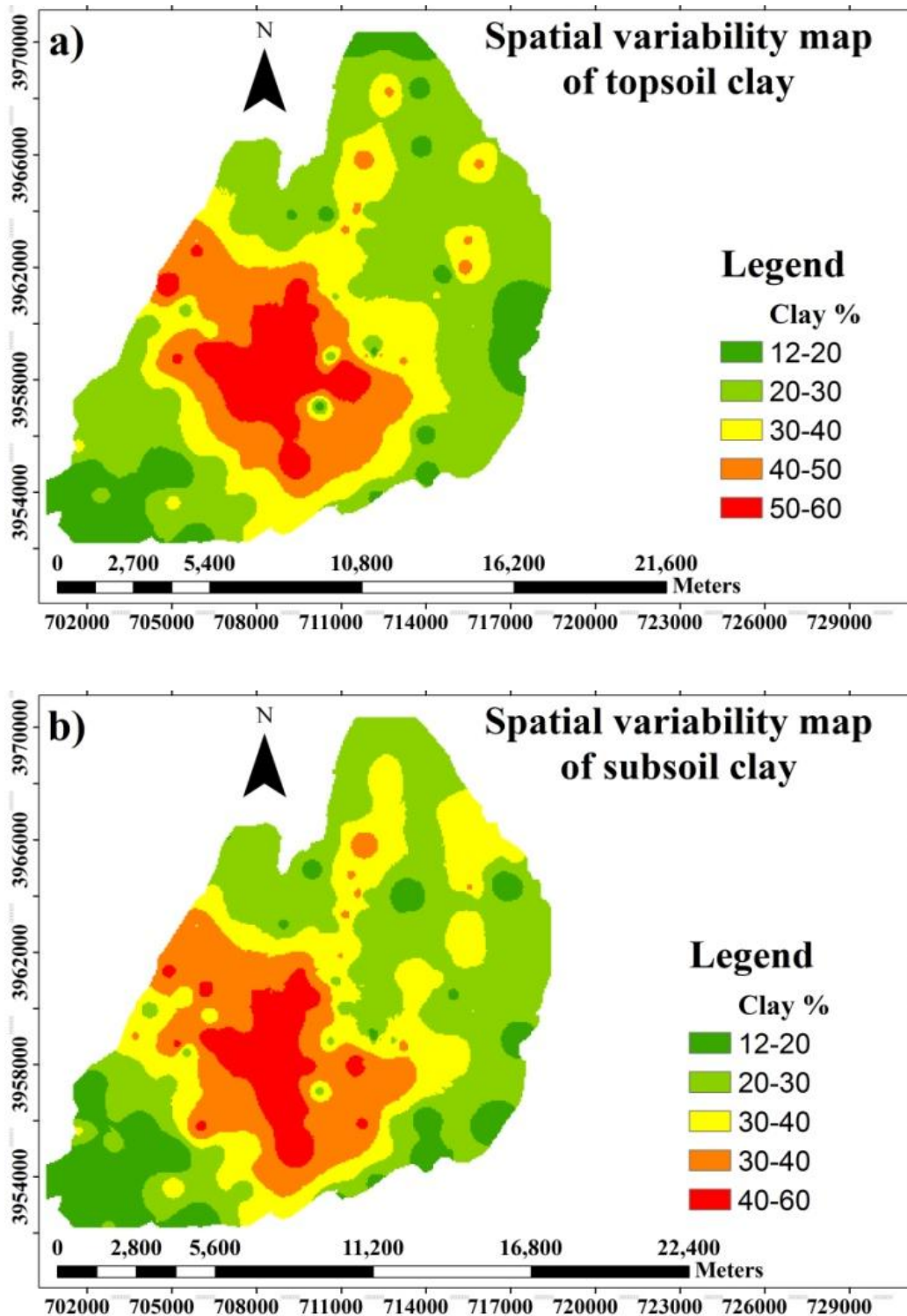


Figure 5. Topsoil (a) and subsoil (b) clay spatial variability maps.

time (Smedley and Kinniburgh, 2002). The positive correlation between arsenic and Fe_2O_3 in the soils suggests that arsenic may be sorbed or co-precipitated on hydrous Ferric oxide. Such adsorption can be explained

by the strong binding affinity of arsenic to hydrous Ferric oxides. This strong affinity of arsenic to Fe oxides is exhibited by a bi-dentate inner-sphere complex that occurs early in mineral nucleation processes (Mitsuo, 2006).

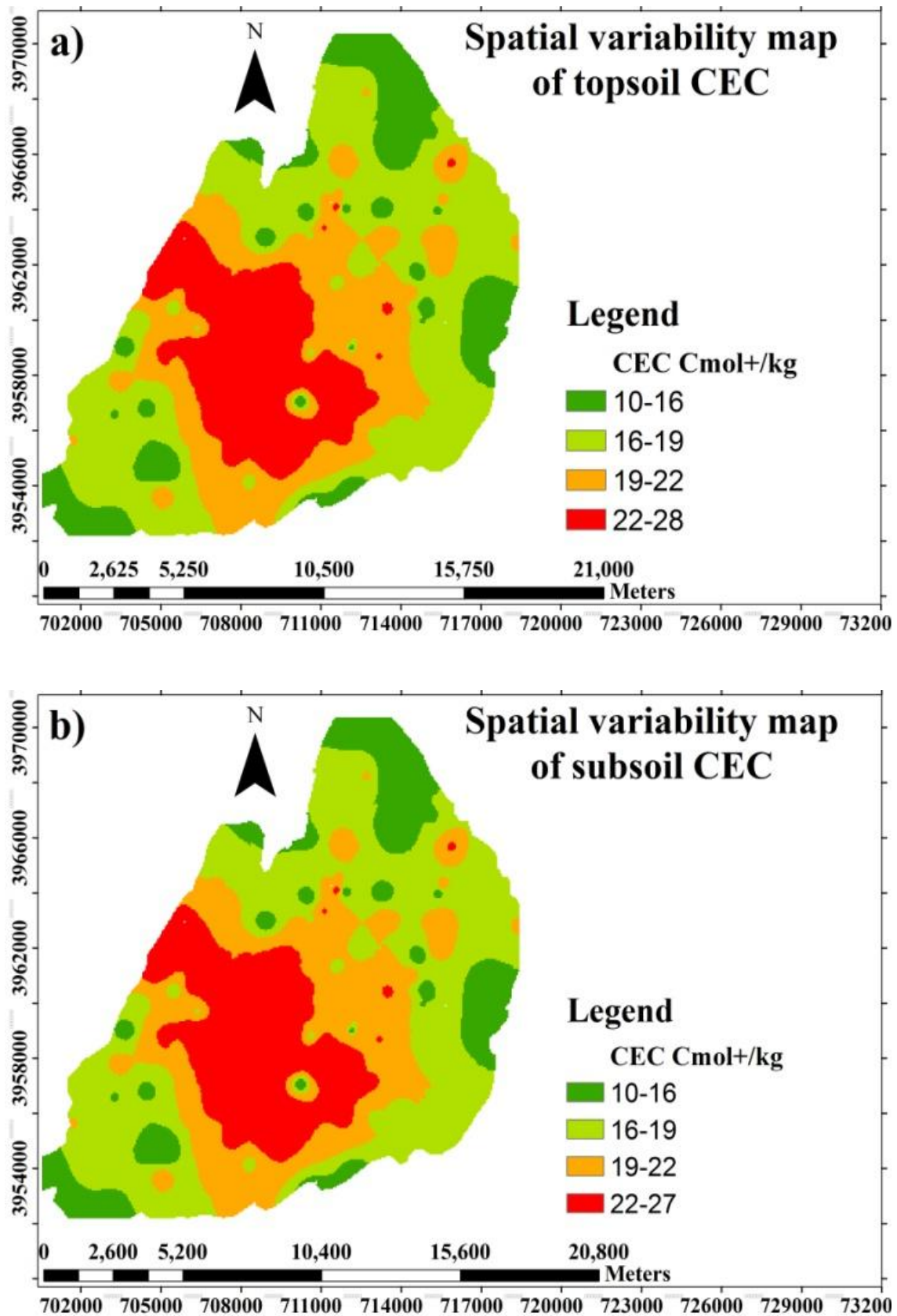


Figure 6. Topsoil (a) and subsoil (b) CEC spatial variability maps.

Table 3. Linear correlation between topsoil and subsoil soil properties and soil arsenic concentration.

Parameter	As topsoil	As subsoil	CEC (cmol ⁺ /kg)	Fe ₂ O ₃ (%)	Clay (%)	Silt (%)	Sand (%)
Topsoil As	1	0.3*	0.65**	0.65**	0.77**	-0.48**	-0.45**
Subsoil As	0.3*	1	0.56**	0.55**	0.7**	-0.48**	-0.39**

**and * indicated that correlation was significant at the level 0.01 and 0.05.

Arsenic adsorption is significantly positively correlated with clay. Soils having higher clay content retain more arsenic than sandy soils with low clay content (Mahimairaja et al., 2005). The effect of pH on arsenic adsorption varies considerably among soils and is dependent on the nature of mineral surface (Mahimairaja et al., 2005). Different results have been shown by researchers on the relationship between arsenic adsorption and CaCO₃. There is a significant positively correlation between CaCO₃ and the arsenic adsorption. Researchers suggest the carbonates being covered by iron oxides and aluminum hydrated oxides (Matera and Hécho, 2001). In contrary, Polemio et al. (1982) showed there was not any correlation between arsenic adsorption and CaCO₃ content. The mobility, bioavailability, and toxicity of arsenic in soils may also be greatly affected by the presence of competitive anions (such as PO₄⁻³, SO₄⁻², HCO₃⁻² and Cl⁻) (Smith et al., 2002).

Averagely, the subsoil arsenic concentrations were, on the average, higher than topsoil concentration; suggesting a prevailing geogenic origin of arsenic in the area studied. The correlation between arsenic concentration at the two depths was significant at $p < 0.05$ ($r = 0.3$), indicating that to some extent topsoil concentration is affected by that of the soil parent material (Table 3). Ungaro et al. (2008) achieved the same results about the topsoil and subsoil arsenic concentrations. These results confirm that the arsenic contents of the soils have geological sources and the

distribution of arsenic in the studied landscapes was controlled by geomorphologic and hydrologic processes.

Risk assessment

Results from spatial variability showed that a considerable area had soils potentially contaminated by arsenic. Soils with such concentrations of arsenic may be hazardous for humans and ecosystems. However, it is still difficult to obtain a meaningful picture of the spatial distribution of heavy metal contamination for the heterogeneity of the soil and the often-accidental nature of contaminating processes; concentrations of pollutants may vary remarkably over very short distances (Carlon et al., 2001). Many studies regarding the spatial variability of heavy metals in contaminated soils indicated that the spatial distribution characteristics of heavy metals were diverse (example, Brooker, 2001; Kim, 2003). In general, the concentrations of pollutants may show complex spatial patterns, with high peak values and large coefficients of variation. Therefore, it is difficult to detect the areas with pollution above a critical level, even if data are collected at a large number of observation points. It has been shown that most soil properties could affect the spatial distribution of heavy metals (Lu et al., 2003; Banat et al., 2005), and these soil properties generally appear to be correlated with each other to a certain

degree (Castrignanò et al., 2000).

In general, based on the spatial variability map, it is clear that area pollution origin are geogenic and Fe-rich rock and mudstone are the most important source of the area pollution. However, such an assessment was made with total concentrations of arsenic in the soil and this is a first approach towards a risk assessment. Future work is needed to determine the needs for remediation. Such work includes the determination of bioavailability of arsenic in the soil, speciation to determine the chemical form of the elements in the soil (As III is more toxic than As V), identification of exposure pathways and probable receptors.

Conclusion

Statistically, moderate to strong significant correlations were observed between topsoil arsenic concentration with clay, sand, CEC, Fe oxides, and silt content of soils. The Fe-rich rock and mudstone parent materials (arsenic source parent materials) are significantly different regarding arsenic content compared to other parent materials. Results from spatial variability showed that a considerable area had soils potentially contaminated by arsenic. The distribution of high arsenic areas rasterized by IDW confirmed that from the arsenic sources (arsenic source parent materials), the arsenic spread to lower positions by erosion-deposition processes. Soils with such

concentrations of arsenic may be hazardous for and ecosystems.

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