

Soil Contamination Pattern Affected by Coal Mining Activities in a Deciduous Temperate Forest

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ABSTRACT

Aims The present investigation aimed to study the effect of traditional-coal-mining on the spatial distribution pattern of soil properties in the Lavij-Forest located in the western part of the Hyrcanian area-North Iran.

Materials & Methods After selecting a mine area, 16 plots (20×20m) were established by systematic-randomly (60×60m) in an area of 4 ha (200×200m-mine entrance placed at center) which was affected by coal-mining. An area adjacent to the mine (same site conditions) that was not affected by the mining activity considered as the control-area. In order to investigate soil Physio-chemical properties one sample was taken from the 0-10cm depth in each plot. To study the spatial-pattern of soil properties and lead and cadmium concentrations in the mining area, an area of 80×80m (the mine at the center) was considered and 80 soil samples were systematic-randomly taken (10m intervals). Geostatistical analysis was performed via Kriging method and GS+ software.

Findings Results showed that mining had led to a significant decrease in soil moisture, pH, EC, nitrogen, and potassium level. Lead and cadmium concentrations were significantly higher in mine area (Pb: 10.97±0.30, Cd: 184.47±6.26mg.Kg⁻¹) in comparison to control-area (Pb: 9.42±0.17, Cd: 131.71±15.77mg.Kg⁻¹). The range value calculated for variograms of cadmium and lead was 210m. The kriged maps showed that the concentration of cadmium and lead near the mine crater was considerably higher in comparison with adjacent points.

Conclusion The findings showed that coal mining activity had negative effects on the forest soil and it is necessary to consider reclamation of contaminated soil in these areas.

Keywords Cadmium; Geostatistic; Hyrcanian Forest; Kriging; Lead

CITATION LINKS

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Introduction

The Hyrcanian forest located in the north of Iran (covering the southern coasts of the Caspian Sea and is expanding over the northern slopes of the Alborz Mountains) is a suitable habitat for several commercial hardwood species such as beech, oak, maple, alder [1-3]. The Hyrcanian forests are one of the last remnants of natural deciduous forests in the world. The Hyrcanian forests appear to be very similar to the broad-leaf forests typical of central Europe, northern Turkey, and the Caucasus [4]. These forests with an area of 1.84 million ha are vital sources of biodiversity, genetic variation as well as various environmental services such as preservation of the groundwater, wildlife habitat, and erosion control. However, some areas in the Hyrcanian forest have been degraded by human activities such as traditional coal mining [5, 6].

According to Bahrami *et al.* [7], about 20% of deforestations in developing countries are attributed to mining activities. Some consequences of mining activities that threaten the forest ecosystem include destruction of the forest, reduction in biodiversity, degrading of the soil surface and its effect on the physical, chemical and microbiological properties of soil as well as soil contamination with heavy metals such as Lead (Pb) and Cadmium (Cd) [8-10], which might finally replace the existing ecosystem with some mine waste materials [11, 12]. Lead is one of the main heavy metals which have gained considerable importance as an environmental pollutant. Cadmium is another important heavy metal and has attracted the most attention in soil science and plant nutrition because of its mobility in the soil-plant systems and high toxicity to human health [13]. High concentrations of Cd in soil cause long-term risks to ecosystems and humans [14].

Mine waste has an anthropological nature that creates a range of problems for the establishment and maintenance the vegetation. The mine waste are deficient in plant nutrient [15, 16], consequently causing problem for revegetation, restoration and inhibit the plant growth [8, 17, 18]. Loss of vegetation and plants in mining areas cause soil erosion, compaction and temperature fluctuations [8]. In addition, it leads to drainage of mineral acid and heavy metal contamination [19, 20] along with increase in heavy metal concentration (e.g. Pb and Cd) there would be a decrease in some soil parameters such as organic Carbon (C) content and available

nutrients [21]. Previous studies on this subject have also reported different mining effects on soil [8, 9, 18, 22-25].

The first step for knowledge acquisition about the magnitude and extent of coal mining effect on soil is to analyze the spatial distribution of soil properties and heavy metal concentration [26, 27] and study of their relationships [28]. Therefore, geostatistics is being used to evaluate the spatial distribution of soil heavy metals and estimating uncertainty and finally it provides spatial pattern of pollutants [29]. Some previous studies have focused on using geostatistics for interpolation of soil heavy metal concentration, especially lead concentration at mining sites [27, 30, 31].

Iranian Hyrcanian forests have been registered in UNESCO world heritage list. Conservation of world heritage sites is an important strategy [32]; unfortunately studies about the effect of traditional coal mining on valuable Hyrcanian forest ecosystem are rare, especially on the forest soil; therefore main objective of this study is to consider the effect of traditional open-cast coal mining on soil properties in the Hyrcanian forest for the first time. The geostatistical technique was used to assess soil physio-chemical properties in the unsampled area by creating kriging map. The hypotheses of present study were that traditional coal mining can adversely affect forest soil and can increase Pb and Cd concentration in soil; and soil contamination pattern is affected by mining activities. The results of this study can be used for appropriate management of forest ecosystems related to soil reclamation.

Materials and Methods

Study site

The study was conducted in Lavij Forest, western Hyrcanian forests, Nur City, Mazandaran Province, Iran (Figure 1). The natural forest vegetation is temperate deciduous forests containing broad-leaved trees such as Beech (*Fagus orientalis* Lipsky), Oak (*Quercus castaneifolia* C. A. M), Hornbeam (*Carpinus betulus* L.), Maple (*Acer velutinum* Boiss., *Acer cappadocium* Gled.). The elevation range is between 950-1270m. The average slope is 30% and the main aspects are West and South-west. The climate is temperate humid and meteorological data provided by synoptic meteorological station indicated that annual average rainfall is 866mm. Mean annual

temperature is 9.8°C. According to the United States Department of Agriculture (USDA) Soil Taxonomy, soils can be classified as Alfisols; the permeability and stability of bedrock are very weak and has drift conditions [33].

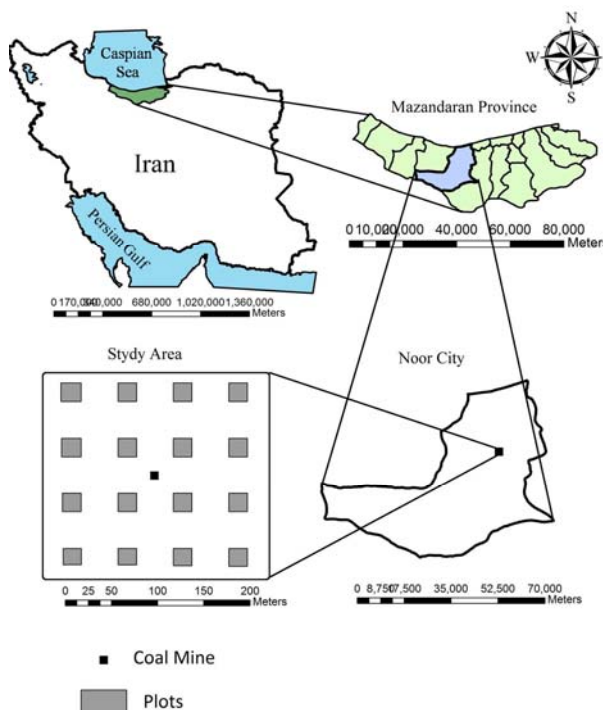


Figure 1) Location of the study area, western Hyrcanian forests

Experimental design and sample collection

In this study, after the field visiting, a mine was selected that located at least 1km far from the road, and mining activities had been carried out for several years and in addition it was located within the approximately flat forest area with minimum change in slope percentage. In general, it was considered that other anthropogenic activities (e.g. forest logging, animal husbandry) have not taken place in that area. After determining the location of the mine, 16 plots (20×20m) were established systematic-randomly (a 60×60m grid) in an area of 4ha (200×200m) [34, 35], in the way that mine crater's placed at the center (Figure 1). Then an area adjacent to the mine, which was not affected by the mining activity (approximately 2 Km distance), was selected and considered as the control area and plots were established by the mentioned method. According to available information, the mine was used for 4 years (2012-2016), and the mine was abandoned for six months before the sampling date.

In order to consider soil physical properties, in the control and mining areas, one sample

was taken from 0-10cm depth in the center of each plot. In order to consider chemical properties, 4 samples were taken from corners (from the main sides) in plots and the one in the center of the top soil (0-10cm depth) using the coring method (diameter: 8cm, height: 10cm). Samples were mixed and finally, one sample was taken and put in plastic bags.

In order to study the spatial pattern of soil physical and chemical properties as well as the concentration of lead (Pb) and cadmium (Cd) in the mining area, an area of 80×80m² with the mine crater's as the center was considered and 80 soil samples [11] were systematic-randomly taken at intervals of 10m (Figure 2) from upper layer (0-10cm depth) using the coring method [36].

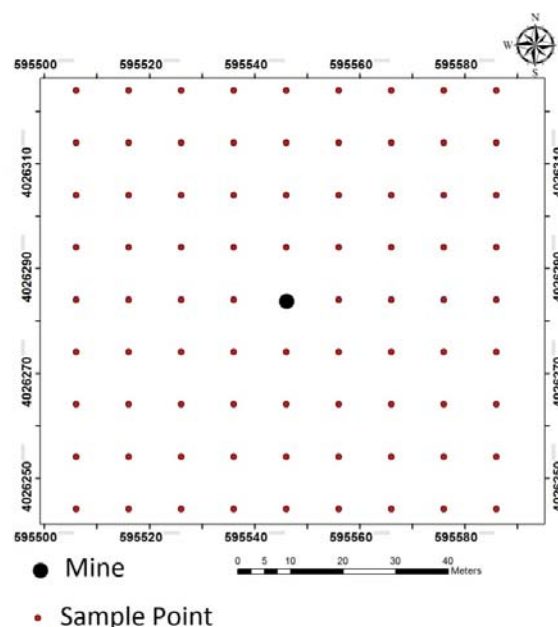


Figure 2) Sample points in order to consider spatial changes in soil properties

Laboratory Analysis

In the laboratory, the samples were passed through a sieve (2mm mesh). Soil moisture content (%) of samples was determined by drying the given amount of fresh soil in a hot air oven at (105±5°C, 24h). Clod method was used to measure soil bulk density [37]. Hydrometer method was used to measure fine earth fragments (<2mm) in order to determine the soil texture. Soil pH (in a 1:2.5 soil/water suspension), EC (EC meter, in water-saturated soil extract), total nitrogen (N) (Kjeltec System Instrument), available phosphorus (P) [38], available potassium (K) (extracted by ammonium acetate), lime content, as well as

organic carbon (C) (Walkley-Black technique) were measured [39].

The concentration of Pb and Cd were measured by spectrophotometry (AAS, Analiticjena, Contra AA) according to Jackson [40]. For measuring Pb and Cd concentration, 1 g soil was mixed with 20ml mixture of HNO₃: HCl: H₂SO₄ (1:2:4) in a flask and digested by heating. 10 ml of HCl: H₂O (1:1) was added and the whole digested content was filtered through Whatman filter paper (no. 42). The total volume of the filtrate adjusted to 50ml with distilled water.

Geostatistical Methods

In geostatistics, the variography technique is used to calculate variogram or semi-variogram in order to measure the spatial variability and dependency of a regionalized variable [41]. Variography provides the input parameters for the spatial interpolation of kriging [42]. Equation 1 expresses the variogram function:

$$\gamma(h) = \frac{1}{2N(h)} \sum_{i=1}^{N(h)} [Z(x_i) - Z(x_i) - Z(x_i + h)]^2 \quad (1)$$

Where $Z(x_i)$ is the value of the variable Z at the location of x_i and $N(h)$ is the number of pairs of sample points separated by the lag distance of h . In order to model experimental variograms, acquiring data about the spatial structure and the input parameters for kriging estimation, spherical, exponential and Gaussian models were chosen [43, 44].

Statistical Analysis

In order to estimate the error of considered models of variogram, two parameters of Mean Bias Error (MBE) and Mean Absolute Error (MAE) were calculated using the Equation 2 and 3:

$$MBE = \frac{\sum_{i=1}^n (R_s - R_o)}{n} \quad (2)$$

$$MAE = \frac{\sum_{i=1}^n |R_s - R_o|}{n} \quad (3)$$

Which in these equations R_s is the estimated value and R_o is the actual value. The normality of the variables was checked by the Kolmogorov-Smirnov test and the homogeneity of variances by the Levene's test. Independent sample t-test was used to compare soil physical and chemical properties between control and mine areas. For all statistical analyses, SPSS v.20 software was used. In addition, relationships between soil properties were analyzed by Principle Component Analysis (PCA) using PC-ORD version 5.0. Geostatistical analysis of soil

properties and the map of sampling sites were performed using GS+ software (version 5.1).

Findings

Soil physical and chemical properties

The soil type in both control and mine areas was Silt Loam. The results of our study showed that open cast coal mining has altered the forest soil characteristics: pH, EC, N, and OC were significantly higher in the control area than the mine area, but the amount of P and K was significantly lower in the mine area (Table 1).

Table 1) Physical and chemical properties of coal mine soil and undisturbed forest soil

Soil properties	Site	Mean±SE	T
Bulk density (gr.cm ⁻³)	Control	1.92 ±0.11	2.32*
	Mine	1.61 ± 0.63	
Moisture (%)	Control	52.34 ± 1.87	7.24**
	Mine	32.53 ± 1.99	
Clay (%)	Control	6.63 ± 0.91	-3.28**
	Mine	12.47 ± 1.52	
Sand (%)	Control	20.92 ± 1.43	0.195 ^{ns}
	Mine	20.49 ± 6.93	
Silt (%)	Control	72.44 ± 1.59	1.98 ^{ns}
	Mine	67.03 ± 2.21	
pH	Control	6.52 ± 0.16	2.28*
	Mine	6.09 ±0.08	
EC (ds.cm⁻¹)	Control	0.88 ± 0.02	3.81**
	Mine	0.71 ± 0.03	
N (%)	Control	0.38 ± 0.01	2.10*
	Mine	0.31 ± 0.03	
P (mg kg⁻¹)	Control	9.18 ± 0.92	-2.31*
	Mine	12.94 ± 1.33	
K (mg.kg⁻¹)	Control	308.98 ± 17.81	-3.76**
	Mine	414 ± 21.58	
OC (%)	Control	7.30 ± 0.35	2.68*
	Mine	5.97 ± 0.34	
Lime (%)	Control	3.30±0.21	2.19**
	Mine	4.11±0.30	

*: Significant difference at 5% level; **: Significant difference at 1% level

Heavy metal concentration

The concentrations of heavy metals (i.e. Cd and Pb) were found higher in the coal mine soil compared to the undisturbed forest soil (Table 2).

Different soil properties represented different locations in the PCA ranking output. Results of the PCA showed the first and second axes accounted for 30.85% and 16.39% of the total variance, respectively (Table 3). The first axis is related to soil pH, EC, BD, N, moisture, Cd, Pb, clay and sand and the second one is related to N, P, C, lime, and silt. Soil pH, EC, N, C and moisture had a negative correlation with soil Pb and Cd (Figure 3).

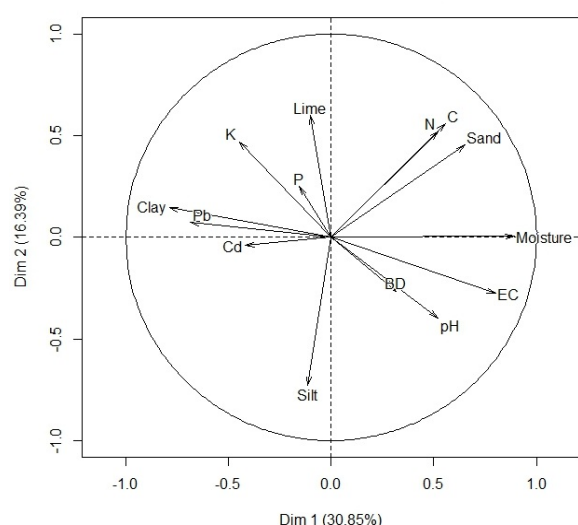
Table 2) Heavy metal concentration of control and mine areas

Heavy Metal (mg.kg ⁻¹)	Site	Mean±SE	T
Cd	Control	131.71 ± 15.77	3.11**
	Mine	184.47 ± 6.26	
Pb	Control	9.42 ± 0.17	4.35**
	Mine	10.97 ± 0.30	

** : significant difference at 1% level

Table 3) Initial Eigenvalues of PCA of soil properties in control and mine areas

Component	Initial Eigenvalues		
	Total	Variance (%)	Cumulative (%)
1	4.319	30.847	30.847
2	2.294	16.387	47.234

**Figure 3)** Principle components analysis (PCA) of soil properties in the study area

Geostatistical Analysis

Surface variograms were calculated in the first stage of the variography analysis, and then theoretical models of spherical and exponential functions were fitted to them. The results of variography indicated that bulk density, moisture, EC, OC had no spatial dependency. However, other soil properties including sand, silt, clay, lime, P, N, pH as well as Pb and Cd had relatively moderate spatial structure (Table 4).

Estimating and mapping soil properties

The bulk density as it approaches the Northeast-Southwest increases significantly with the distance from the mine span, indicating a high compaction in these areas (Figure 4). As shown in Figure 4 it is observed that in areas where traffic is higher and the compaction is higher, the pH value is also lower. In addition, in areas where the moisture content was high, the amount of K was low. There was less N in the area where the pH was higher. The concentration of Cd and lead near the mine crater is considerably high. It is also observed that in areas where it is more compact and the pH is lower, the amount of cadmium is also higher compared to the adjacent points. It should also be noted that with increasing distance, not only the concentration of cadmium and lead increased, but also more surface area was contaminated. This shows the spread of these heavy metals in the area. It can also be observed that in the area with high clay content, the lead content was higher.

Table 4) Parameters of the theoretical models fitted to the experimental variograms for soil variables

Parameters	Model	Nugget effect	Sill	Range	Spatial dependency	MBE	MAE
Bulk density	linear	0.38	0.38	83.36	0	-0.04	0.54
Moisture	linear	119.12	119.12	83.36	0	0.38	8.03
Clay	Spherical	149.30	298.70	210.90	50/01	0.33	9.59
Sand	Exponential	306.60	613.30	210.90	50	0.10	8.42
Silt	Spherical	81.90	163.90	210.90	50	0.58	14.71
pH	Exponential	1.99	3.99	198.90	50.12	0.00	1.04
EC	linear	0.087	0.087	83.36	0	-4.30	241.97
N	Exponential	0.01	0.021	210.90	52.38	-0.00	0.07
P	Spherical	46.50	107.62	210.90	56.79	-0.05	5.49
K	linear	31640.61	31640.61	83.36	0	-0.05	130.49
OC	linear	9.03	9.03	83.36	0	-0.04	1.54
Lime	Exponential	6.30	12.60	210.90	50	0.93	147.5
Cd	Exponential	2.73	5.47	210.90	50	-0.03	1.47
Pb	Exponential	2532	5065	210.90	50	-1/36	41.44

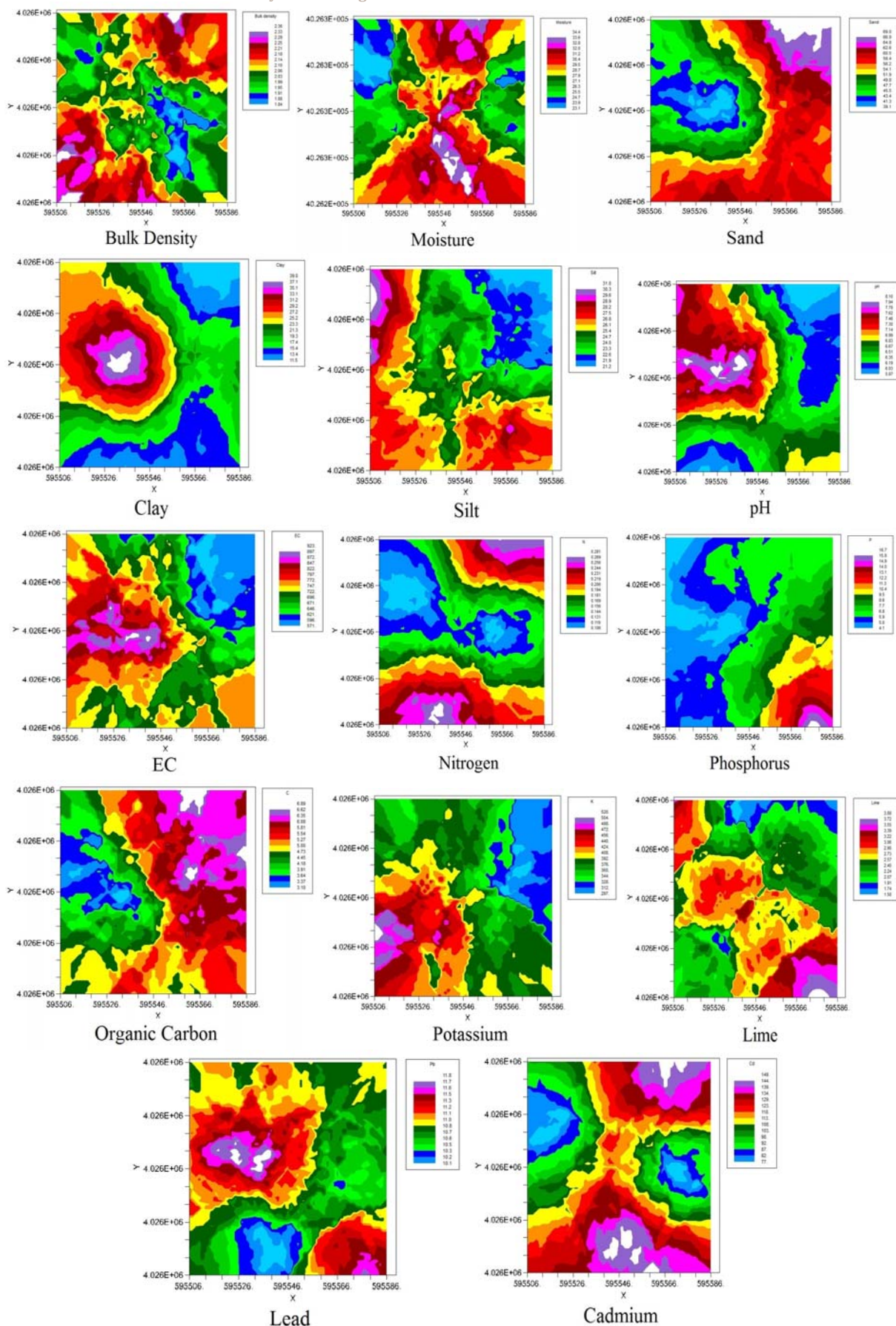


Figure 4) Krige maps of the soil properties

Discussion

Results of the present study showed that clay percentage was higher in the mine area regarding the forest soil and no significant difference was observed in sand and silt content between the mine soil and forest soil. Weathering and erosion processes might have occurred on mine soil and a more or less homogenization was achieved between the mine spoils and surrounding soil by a dynamic equilibrium [23].

The percentage of soil moisture in the mine soil was lower than the forest soil. The difference of water content between these two soils depends on the time of sampling, the organic carbon content and soil texture [21]. However, low moisture content in mine soil might be related to higher stone content, sandy texture and lack of organic matter [23].

We found that bulk density of mine spoils was lower compared to the forest soil. This finding was contrary to previous studies such as Sadhu *et al.* [23], Ghose [45], Maiti [21], and Pandey *et al.* [9] which reported that dumping sites in the mining areas are evidently influenced by human activities since the Bulk density is higher in the mine area.

According to our results, the pH of forest was significantly higher than mine soil; which was reported by Singh and Agrawal [46] and Pandey *et al.* [9]. Coal mining typically exposes sulfur containing pyrites that oxidize to sulfuric acid when exposed to oxygen, water and certain aerobic bacteria, leading to low soil pH [17,47]. The electrical conductivity of the mine soil was significantly lower than forest soil. This finding was similar to Sadhu *et al.* [23] and Pandey *et al.* [9] which reported that mine soil had lower EC than the forest soil. Generally, leaching losses of base cations from soil increase with the decrease in pH, consequently the EC will decrease. The reduction in soil pH (increase in H^+ due to acid release by mining activity) leads to replacement of cations (e.g. Ca^{2+} , Mg^{2+} , K^+) on exchange sites by H^+ . Consequently, cations in soil solution will leach out from the soil surface [48-50].

The mine soil had lower total soil organic carbon than forest soil. The soil organic carbon plays a pivotal role in biological activities and soil fertility of the soil and its main sources in forest soil include plant debris, dead roots, rhizomes and the litter [9]. The soil organic matter (SOM) increases the soil porosity, which supports microorganism growth in the soil. Since the

trees were harvested in the mine area therefore the content would decrease [23]. Pandey *et al.* [9]; de Quadros *et al.* [8]; Pandey *et al.* [18] reported the lower soil organic carbon content in mine soil in comparison with forest soil.

The N concentration of the mine soil was lower compared to the forest soil sites. The concentration of N is more due to litterfall and microbial activity causing transformation of inorganic forms around the roots of plant species; therefore removing vegetation in mine soil leads to the loss of large amounts of nitrogen from the top soil [9, 21]. Available P was significantly lower in forest soil than mine soil. In the mining area, soil clay particle content was higher than forest soil and high clay will fix phosphorus. The K content of the mine soil was lower compared to the forest soil, which can be related to the low amount of organic matter and lower litter decomposition in mine soil. This result was similar to results reported by Sadhu *et al.* [23] and Talukdar *et al.* [51].

Our data showed that the concentration of lead and Cadmium were significantly higher in the mine area than control area. In the process of coal mining, huge quantities of mine waste and dust are produced along with coal. The natural weathering process may degrade these wastes into small clay-sized particles. Through this process, the large number of fine particles enriched in heavy metals is released into the environment [9, 21, 22,52].

According to PCA results, heavy metals showed significant negative correlations with the soil pH. In line with the report of Nan *et al.* [53] and Zang *et al.* [25], results of this study showed a significant positive correlation among the heavy metals. The strongest correlations were observed between pH, organic carbon and heavy metals [25, 54, 55]. However, weak correlations were also reported by Manta *et al.* [56]. A negative correlation between heavy metal concentrations and pH was found by Gjoka *et al.* [54].

The spatial distribution of Pb and Cd was important to evaluate the sources of these heavy metals in soil [57,58]. Results showed that some of the soil properties including bulk density, moisture content, electrical conductivity, potassium, and organic carbon had linear models, and other properties had a spherical or exponential model. So the bulk density, moisture, EC, OC had no spatial dependency. However, the percentage of sand, silt, clay, lime, P, N, pH and the concentration of Pb and Cd had

relatively moderate spatial structure. Kooch *et al.* [34] reported that the exponential model is the best model for estimating soil pH in the Hyrcanian forests. The spherical model and strong spatial dependency for the concentration of heavy metals around the mine were also reported in previous studies [22, 26, 59].

In general, results of spatial pattern showed that the variation of the soil properties depends on the spatial dimension and distance, so it is possible to prepare a map of soil properties interpolation using geostatistical methods. The MBE and MAE analysis also showed that Kriging method was able to make a fairly good estimate; therefore, the estimation and interpolation of soil properties, especially the concentration of heavy metals in the soil, was a suitable method for identifying and assessing the range of coal mining activity effects.

Variograms of Pb and Cd showed similar range by comparing calculated range values; there was a close similarity range value of clay, sand and silt contents with heavy metal concentrations; therefore this showed a possible spatial correlation between Pb and Cd concentrations and the percentage of sand, silt and clay over the study area. The kriged maps of Pb and Cd indicated high heavy metal concentrations in the surrounding of the mine entrance which a decreasing gradient was observed from south to northern parts of the study area (Figure 4). These results may be due to some factors such as: main wind directions, historical background and rate of mining activities.

The results of the present study show that the Pb content was higher in the area with high clay content. Recent studies showed that concentration of heavy metals in soil increases with decreasing particle size [60, 61]. Fine particles retain high amounts of metals because of the highest specific area [62].

Conclusion

In this study, the effect of traditional coal mining activity on the forest soil was investigated for the first time in the Hyrcanian forests, North Iran. The findings showed that the coal mining activity had negative effects on the forest soil. Mining activities had led to a significant decrease in soil moisture content, pH, EC, N and K concentrations. Concentrations of Pb and Cd were significantly higher in mine area compared with the control area. In general, knowledge of the forest soil properties and their spatial

pattern in degraded and polluted areas can help us to better understand the changes and damages of the forest ecosystem and to provide appropriate solutions for sustainable forest management, soil reclamation and reforestation of these areas. According to obtained results (negative effects of mining activities), it is necessary to consider reclamation of contaminated soil by using amendments (e.g. biochar and organic amendments) in these areas. Due to the lack of information about the impact of traditional coal mining on soil properties in the Hyrcanian forest, more experiments are needed to obtain comprehensive data about forest soil nutrients' dynamic and biological properties like enzyme activities and microbial biomass.

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