World Academy of Science, Engineering and Technology
International Journal of Environmental and Ecological Engineering
Vol:13, No:4, 2019

Open Science Index, Environmental and Ecological Engineering Vol:13, No:4, 2019 publications.waset.org/10010213.pdf

Lead and Cadmium Spatial Pattern and Risk Assessment around Coal Mine in Hyrcanian Forest, North Iran

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***Abstract*—**In this study, the effect of coal mining activities on lead and cadmium concentrations and distribution in soil was investigated in Hyrcanian forest, North Iran. 16 plots (20×20 m^2) were established by systematic-randomly (60×60 m^2) in an area of 4 ha (200×200 m^2-mine entrance placed at center). An area adjacent to the mine was not affected by the mining activity; considered as the controlled area. In order to investigate soil lead and cadmium concentration, one sample was taken from the 0-10 cm 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^2 (the mine as the center) was considered and 80 soil samples were systematic-randomly taken (10 m intervals). Geostatistical analysis was performed via Kriging method and GS^+ software (version 5.1). In order to estimate the impact of coal mining activities on soil quality, pollution index was measured. Lead and cadmium concentrations were significantly higher in mine area (Pb: 10.97±0.30, Cd: 184.47±6.26 $mg.kg^{-1}$) in comparison to control area (Pb: 9.42±0.17, Cd: 131.71±15.77 $mg.kg^{-1}$). The mean values of the PI index indicate that Pb (1.16) and Cd (1.77) presented slightly polluted. Results of the NIPI index showed that Pb (1.44) and Cd (2.52) presented slight pollution and moderate pollution respectively. Results of variography and kriging method showed that it is possible to prepare interpolation maps of lead and cadmium around the mining areas in Hyrcanian forest. According to results of pollution and risk assessments, forest soil was contaminated by heavy metals (lead and cadmium); therefore, using reclamation and remediation techniques in these areas is necessary.

***Keywords*—**Traditional coal mining, heavy metals, pollution indicators, geostatistics, caspian forest.

I. INTRODUCTION

HYRCANIAN forest or Caspian forest (located in the north of Iran and covers the southern coasts of the Caspian Sea). is a proper habitat for some of commercial hardwood species such as beech, oak, maple and alder [1], [2]. However, some areas in the Hyrcanian forest have been degraded by human activities (e.g. traditional coal mining) [3].

Some of the consequences of traditional mining activities threaten forest ecosystems include soil degradation and its effect on of the physical, chemical, and microbiological properties of soils as well as soil contamination with heavy metals such as lead and cadmium [4], which may ultimately replace existing ecosystems with undesirable waste in the form of dumps [5]. Lead (Pb) is one of the main heavy metals and is an important environmental pollutant. Cadmium (Cd) is another important heavy metal and attracted the most attention in soil science and plant nutrition due to its mobility and high toxicity [6].

The first step in the knowledge acquisition about magnitude and extent of coal mining effect on soil is to analysis of spatial distribution of heavy metals concentration [7]. Geostatistics is used to evaluate the heavy metal concentration of soil, by maintaining the spatial distribution and estimating uncertainty. some recent studies have focused on using geostatistics for interpolation of lead concentration around mining areas [8], [9]. They mostly highlight the usefulness of using geostatistics for addressing spatial patterns of lead in polluted soil.

To our knowledge, there has not yet been a study about the effect of traditional coal mining on Hyrcanian forest soil, therefore our main objective was to identify the effect of coal mining on soil lead and cadmium concentration and risk assessment in Hyrcanian forest.

II. MATERIALS AND METHOD

A. Site Description

The study was conducted in Lavij Forest, western Hyrcanian forests, Noor City, Mazandaran Province, Iran (Fig. 1). The natural forest vegetation is temperate deciduous forests containing broad-leaved species 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 about between 950-1270 m. 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 866 mm. Mean annual temperature is 9.8 ˚C. The soil type is brown forest soil and the permeability and stability of bedrock are very weak and has drift conditions [10].

B. Sample Collection

In this study, after determining the location of the mine, 16 plots (20 × 20 m^2) were established systematic-randomly (a 60 × 60 m grid) in an area of 4 ha (200×200 m^2), in the way that mine crater's placed at center [11], [12] (Fig. 1). Then, an area adjacent to the mine, which was not affected by the mining

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activity (approximately 2 km distances), was selected and considered as the control area and plots were established with the mentioned method.

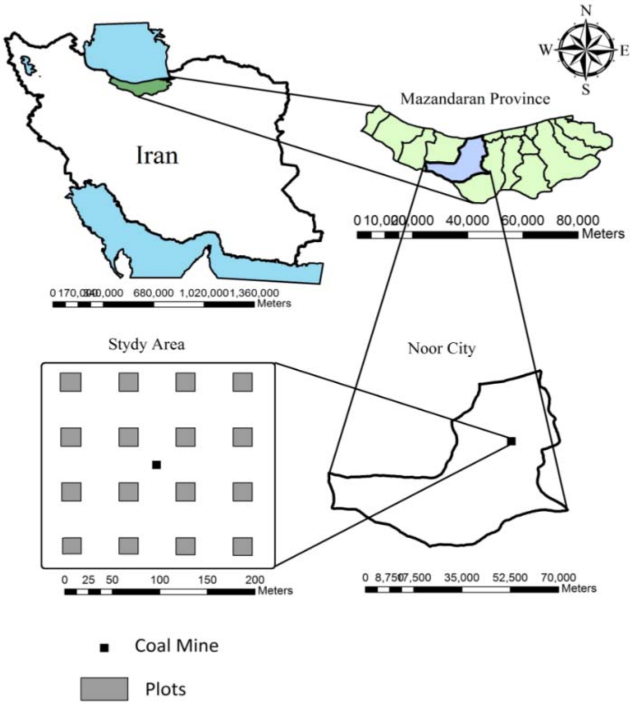


Fig. 1 Location of the study area, western Hyrcanian forests

In order to consider soil lead and cadmium concentration, in the center of each plot, in the control and mining areas, four samples were taken from corners (from the main sides) in plots and the one in the center from the upper layer (0-10 cm) using coring method (cylindrical steel columns; diameter: 8 cm, height: 10 cm). Samples were mixed and finally, one sample was taken and placed in plastic bags.

In order to study the spatial pattern of lead and cadmium concentration in the mining area, an area of 80×80 m^2 with the mine crater's as the center was considered and soil samples were systematic-randomly taken at intervals of 10 m (Fig. 2) from upper layer (0-10 cm) using coring method [13].

C. Laboratory Analysis

The concentration of Pb and Cd were determined by spectrophotometry (AAS, Analitic jena, Contra AA) according to Jackson [14]. For this measuring Pb and Cd concentration, 1 g soil was mixed with 20 ml mixture of HNO_3: HCl: H_2SO_4 (1:2:4) in a flask and digested by heating. 10 ml of HCl: H_2O (1:1) were added and whole digested content was filtered through Whatman filter paper (no. 42).

D. Pollution Indicators

In order to assess the quality of soil and estimation of the effect of mining activities, pollution index was used [15]. According to Lee et al. [16], pollution index (PI) was defined as (1):

$$PI = C_i/C_0 \tag{1}$$

where Ci is the concentration of a given ith metal in soils and C0 is its corresponding reference concentration. The Nemerow integrated pollution index (NIPI) was calculated as (2).

$$NIPI = \sqrt{\frac{PI_{avg}^2+PI_{max}^2}{2}} \tag{2}$$

where PIavg is the average value of all pollution indexes of the metals considered, PImax is the maximum value. According to [17] and [18], the PI is divided into four levels; the NIPI is divided into four six; (Table I).

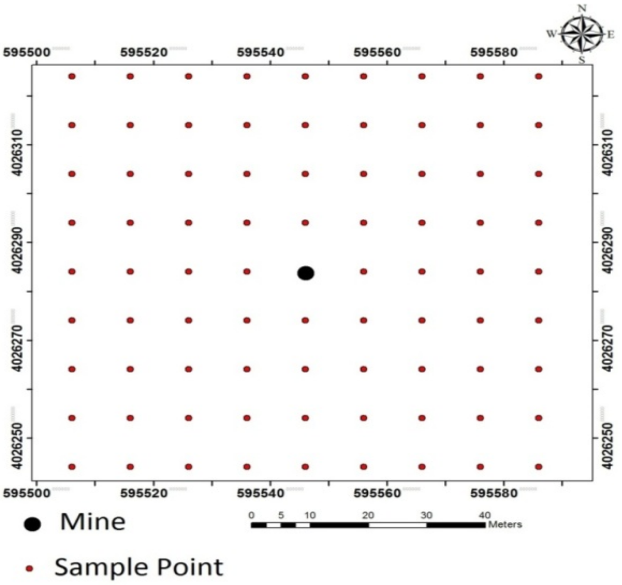


Fig. 2 Sample points in order to consider spatial changes in soil properties

TABLE I
POLLUTION INDEX (PI) AND NEMEROW INTEGRATED POLLUTION INDEX (NIPI)

Index	Value	classification
PI	PI ≤ 1	Unpolluted
	1 < PI ≤2	Slightly polluted
	2 < PI ≤ 3	moderately polluted
	3 < PI	Highly polluted
NIPI	NIPI ≤ 0.7	Safe
	0.7 < NIPI≤ 1.0	Precaution
	1.0 < NIPI ≤ 2.0	Slight pollution
	2.0 < NIPI ≤3.0	Moderate pollution
	3.0 < NIPI	Heavy pollution

E. Statistical Analysis

In this study, exponential model was selected as the input parameters for kriging. In order to estimate the error of the considered models of variogram, two parameters of MBE and MAE were calculated using the following equations (3) and (4):

$$MBE = \frac{\sum_{i=1}^{n}(R_S-R_O)}{n} \tag{4}$$

$$MBE = \frac{\sum_{i=1}^{n} |R_S - R_O|}{n} \tag{4}$$

In these equations, R_S is the estimated value and R_O is the actual value. 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. Geostatistical analysis of soil properties and the map of sampling sites were performed using GS^+ software (version 5.1).

III. RESULTS

A. Heavy Metal Concentration

The results of heavy metals concentration of control and mine areas are presented in Table II. Concentrations of heavy metals (Cd and Pb) were found higher in coal mine soil compared to undisturbed forest soil.

TABLE II
HEAVY METAL CONCENTRATION OF CONTROL AND MINE AREAS

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

B. Risk Assessment of Heavy Metal Contamination in Soil

The calculated pollution index and Nemerow integrated pollution index values of heavy metals in soils were presented in Table III. The mean values of the PI index indicate that Pb and Cd presented slightly polluted. Results of the NIPI index showed that Pb presented the slight pollution, whereas Cd presented the moderate pollution.

TABLE III
POLLUTION INDEX (PI) AND NEMEROW INTEGRATED POLLUTION INDEX (NIPI) OF HEAVY METALS IN SOILS

Heavy metal	PI		NIPI
	Range	Mean	
Pb	0.96-1.67	1.16	1.44
Cd	0.92-3.61	1.77	2.52

C. Geostatistical Analysis

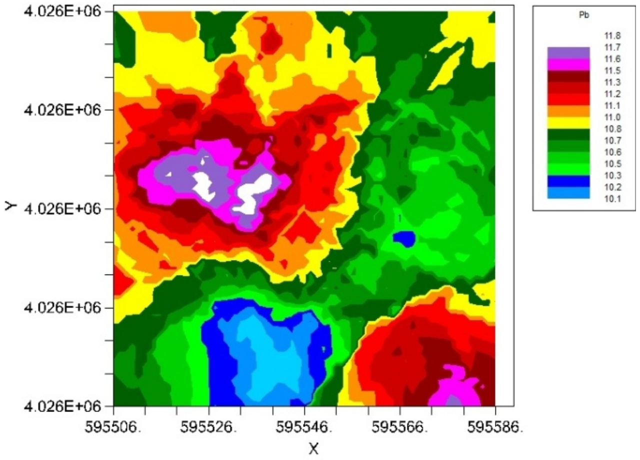
At the first stage of the variography analysis of soil parameters, surface variograms were calculated and the variograms with theoretical model of exponential were fitted to them. The numerical results are given in Table IV. The results of variography indicated that spatial structure of Pb and Cd were relatively moderate.

TABLE IV
PARAMETERS OF THE THEORETICAL MODELS FITTED TO THE EXPERIMENTAL VARIOGRAMS FOR SOIL VARIABLES

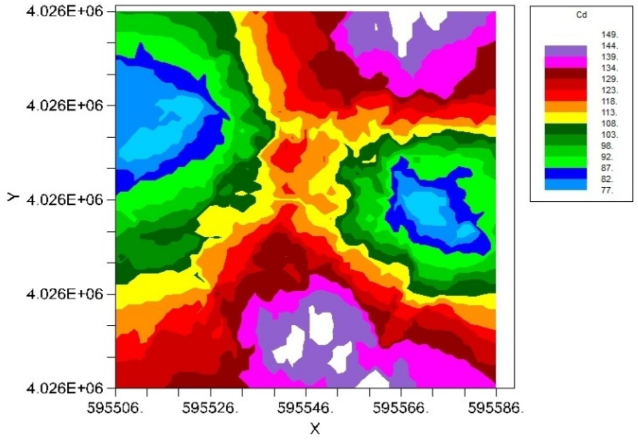
	Model	Nugget effect	Sill	Range	Spatial dependency	MBE	MAE
Cd	Exponential	2.73	5.47	210.90	50	-0.03	1.47
Pb	Exponential	2532	5065	210.90	50	-1/36	41.44

D. Estimating and Mapping

Ordinary kriging system was used along with isotropic variograms to estimate heavy metal concentration around the mine site (Fig. 3). As shown in Fig. 3, the concentration of cadmium and lead near the mine crater is considerably high. It is also observed that in areas where it is more compacted and pH is lower, the amount of cadmium is also higher than adjacent points. It can also be observed that with increasing the distance from the mine crater, concentration of cadmium and lead was increased. It should also be noted that with increasing distance, not only the concentration of cadmium and lead increased, but more surface area was also contaminated. This indicates 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.



Lead



Cadmium

Fig. 3 Kriged maps of the soil lead and cadmium around coal mine

IV. DISCUSSION

Our results 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. Degradation of these wastes into small particles may be due to natural weathering process. Through natural weathering, the large number of fine particles enriched in heavy metals is released into the environment [4], [19].

According to our geostatic results, the spatial distribution of lead and cadmium was important to evaluate the sources of these heavy metals in soils [17], [18], [20]. Our results showed that the concentration of Pb and Cd had exponential model. The exponential model and strong spatial dependency for the concentration of heavy metals around the mine were also reported in previous studies [7], [19]. The degree of spatial dependency was identified by spatial class ratios according to Cambardella et al. [21], that is the ratio of nugget variance (noise) to total variance (sill) multiplied by 100. According to results of spatial dependency, the concentration of Pb and Cd had relatively moderate spatial structure.

In general, the results of spatial pattern showed that the variability of lead and cadmium depend on spatial dimension and distance, so it is possible to prepare a map of lead and cadmium 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 the heavy metals in the soil, was a suitable method for identifying and assessing the range of coal mining activity effects.

In this study, the effect of traditional coal mining activity on forest soil was investigated for the first time in Hyrcanian forest, North of Iran. Concentrations of lead and cadmium were significantly higher in mine area in comparison with control area. According to the results of variography and kriging method, it is possible to prepare interpolation maps of soil properties near the mining areas in Hyrcanian forest. These maps can be very useful in management decisions. In general, knowledge of the forest soil properties in degraded areas can help us to better understand the changes and damages to the forest ecosystem and to provide appropriate solutions for sustainable forest management and reforestation of these areas. Based on the results of pollution and health risk assessments, it was obvious that forest soil was contaminated by heavy metals, for this reason it is necessary to use reclamation and remediation techniques in these areas.

ACKNOWLEDGMENTS

This study was financially supported by *Sari Agricultural Sciences and Natural Resources University*. The authors would like to thank Miss Rasta Rajaei and Dr. Mahya Tafazoli for their kind cooperation.

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