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Geostatistical assessment of Pb and the related soil physical and chemical properties in near-surface soil around Sepahanshahr, Isfahan

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Abstract

This paper presents a survey on soil Pb concentration around Sepahanshahr town located in vicinity of Isfahan. Due to the lack of regulation and environmental education and awareness, Sepahanshahr is now a rapid growing residential area suffering from the considerable consequences of poorly regulated mining activities operating in its vicinity. The aim of this study is to explore the spatial structure of Pb distribution and to map Pb pollution using geostatistical techniques. 100 near-surface soil samples were collected and analyzed for Pb and some other related soil physical and chemical variables such as pH, organic matter content, electrical conductivity, clay, silt and sand contents. The variography results show a strong spatial dependency in Pb data due to the dilution effects of natural factors including atmospheric dispersion and precipitation. The almost same range values calculated for both Ln-transformed Pb data and sand content suggest presence of co-regionalization. Kriged Pb map shows a strong gradient of Pb concentration around the three mining sites activating in the area. The results of this study provide insight into identification of the extent and the spatial variability of Pb pollution in the mining sites and surrounding area.

Keywords: Geostatistics; Lead; Pollution mapping; Physical properties; Chemical properties; Sepahanshahr

1. Introduction

Lead pollution and toxicity has been a topic of concern for decades, and its effects on human health have been shown to be harmful at multi-levels (Emory et al., 1999; Ferreira da Silva et al., 2004; Saby et al., 2006; Hooker and Nathanail, 2006). The health hazard due to Pb in soil and dust in now widely accepted (Mielke and Reagan, 1998). Some of the adverse neuro-behavioral effects are irreversible, even when symptoms of Pb poisoning are corrected (Emory et al., 1999). Lead is considered as a general metabolic poison and pre-school children may suffer long-lasting adverse neuro-behavioral effects when blood lead concentration exceeds 10 μg/dl of whole blood. Soil lead therefore is of particular concern with respect to potential chronic risks to human health (Davies et al., 1990; Defra and Environment Agency, 2002 a,b). The major sources of lead pollution in urban environments are exhaust emission from petrol vehicles, mining, smelting and foundry activities, and application of sewage sludge to agricultural soils (Hooker and Nathanail, 2006). Davies (1995) estimated that in remote or recently settled areas, soil-Pb concentrations

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mostly lie in the range of 10-30 mg/kg, but elsewhere, even in rural areas, low-level contamination will have raised Pb concentrations to the 30-100 mg/kg range. Urban soils typically have elevated Pb concentrations reflecting greater vehicle exhaust emission, and perhaps expanding new residential areas on old mining area or in vicinity of operative mining sites (Hursthouse et al., 2004; Hooker and Nathanail, 2006).

Numerous studies have been undertaken on trace element pollution of soils, plants, waters and sediments as a result of industrial and mining activities, in various countries (Merrington and Alloway, 1994; Pestana et al., 1997; Navas and Machin, 2002; Ferreira daSilva et al., 2004; Ungaro et al., 2008). High rate of urbanization occurred in Iran in recent years has been resulted in expanding and growing of new small towns around major cities such as Isfahan located in central parts of Iran. One of these new residential areas is Sepahanshahr located in vicinity of Isfahan. Due to the lack of regulations and environmental education and awareness, Sepahanshahr area expanded close to one old and big mining site called Bama mine and other two new mines were activated in recent years. As a result, this residential area is suffering from the considerable consequences of poorly regulated mining activities. To the knowledge of authors, no scientific published-surveys were carried out in the area to highlight any potential risk of soil pollution by lead. These omissions provided the impetus for the present study.

An unfortunate point that has to be underlined is that there is no national soil and environment guideline values and thresholds for lead and other pollutants in Iran. Global and international published-thresholds might be impracticable since in every country and especially in every mining site, soil types, volume and quality of solid waste, disposal practices and climatic conditions vary considerably. Furthermore, this lack of legislation caused confusion in terms of impact assessment and selection of the appropriate remediation and clean-up scheme.

The first step in the knowledge acquisition and discovery about magnitude and extent of pollution is to collect soil samples, laboratory analyzing for the pollutant concentrations, analysis of spatial distribution of the pollutant (Facchinelli et al., 2001; McGrath et al., 2004; Rodriguez et al., 2008) and investigating statistical relationships with other soil characteristics (Basta et al., 1993). Geostatistics is extensively used to assess the level of soil pollution and calculate the risk in polluted areas, by preserving the spatial distribution and uncertainty of estimates. It facilitates quantification of the spatial pattern and distribution of pollutants and enables spatial interpolation and mapping (Mohammadi, 1997; Komnitsas and Modis, 2006). Some recent geostatistical studied have focused on soil lead at mining sites and in urban soils (Amini et al., 2005; Hooker and Nathanail, 2006; Saby et al., 2006; Rodriguez et al., 2008). They mostly highlight the usefulness of applying geostatistics to address spatial patterns and uncertainties in soil concentration data.

The main focus of this paper was concerned with near-surface total soil lead around an almost new-established town in vicinity of Isfahan. In this study geostatistical methods for spatial interpolation were used to assess the Pb pollution in unsampled area by creating kriging map showing the Pb concentrations in the study area.

2. Material and Methods

Study area

The research focused on the area between mining sites and Sepahanshahr town located about 20 km from Isfahan. This area covers 9000 ha, of which less than 0.5% is cultivated land, the rest is bare lands or under some constructions. This area is affected by mining activities exploiting raw materials containing lead and zinc in southern parts of the study area. Bama mine is one the oldest mining site operating more than several decades. Recently, two other new mining sites were established in the area and operating in vicinity of the Bama mine. Figure 1 shows the area on the false color composite (FCC) of the ETM+ data from 2002 which represents some parts of the Sepahanshahr town. Furthermore, locations of samples and geographical locations of three mining sites were superimposed on the FCC satellite image.

Soil sampling and analyses

In accordance with the extent of the area and collecting soil sampling at different spatial scales, 100 soil samples (as a minimum number of observations required for geostatistical analysis) were collected on a pseudo-grid sampling scheme at an interval range of maximum 8 km and minimum 60 m during summer 2007. At each sampling point, 4 sub-samples were taken, of which 3 were located on a triangle vertices with 30 m apart and the fourth was located in the centre. All soil samples were taken at a depth of 0-10 cm and air-dried. After removing coarse fragments and plant root residuals, sub-samples were thoroughly mixed and ground to pass a 2 mm sieve. Then, the samples were stored in plastic bags prior to the chemical analysis.

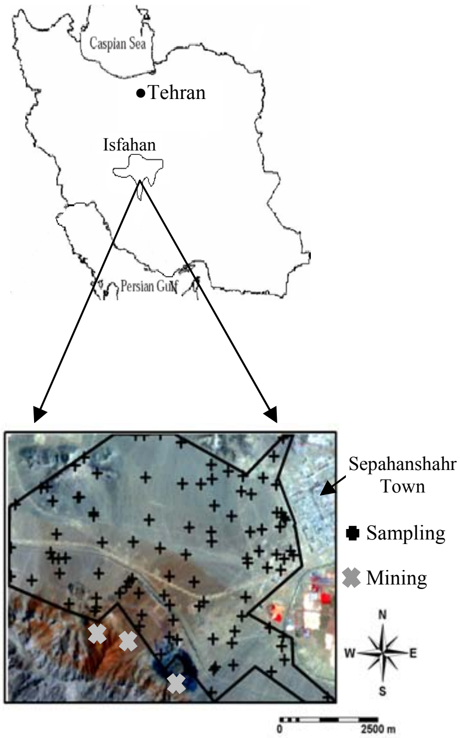


Fig. 1. Study area, sampling locations and mining sites superimposed on FCC satellite image (ETM+, 2001)

Soil pH (soil: H_2O ratio=1:5) was measured using a pH meter with a glass electrode and organic matter (OM) was determined using Walkley-Black method (Nelson and Sommers, 1982). Electrical conductivity (soil:H_2O ratio=1:5) was measured using a EC meter with a cup electrode. Textural fractions (sand, silt, clay) were determined using hydrometer method (Gee and Bauder, 1986). Total contents of lead were analyzed by atomic absorption spectroscopy after soil samples were digested with nitric acid (HNO_3).

Geostatistical methods

Geostatistics is based on the theory of a regionalized variable (Matheron, 1971), which is distributed in space (with spatial coordinates) and shows spatial auto correlation such that samples close together in space are more alike than those that are further apart. Geostatistics uses the technique of variography i.e. calculating variogram or semi-variogram, to measure the spatial variability and dependency of a regionalized variable. Variography provides the input parameters for the spatial interpolation oh kriging (Isaaks and Srivastava, 1989). The variogram function is expressed as:

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

where $Z(x_i)$ is the value of the variable Z at 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 evaluate the possible anisotropic spatial variability, surface variogram was calculated in accordance with the symmetrical property of variogram function for all variables (Pannatier, 1996).

Variogram plots (experimental variograms) were acquired by calculating variogram at different lags. Spherical, exponential and Gaussian models were selected in order to model experimental variograms and acquire information about the spatial structure as well as the input parameters for kriging estimation.
The spherical model is:

$$\gamma(h) = C_0 + C\left[\frac{3}{2}\frac{h}{a} - \frac{1}{2}\left(\frac{h}{a}\right)^3\right] \quad 0 < h \le a$$

$$\gamma(h) = C_0 + C \quad h > a$$

The exponential model is:

$$\gamma(h) = C_0 + C\left[1 - \exp\left(-\frac{h}{a}\right)\right]$$

The Gaussian model is:

$$\gamma(h) = C_0 + C\left[1 - \exp\left(-\frac{h^2}{a^2}\right)\right]$$

Where C_0 is the nugget variance (h=0), represents the experimental error and field variation within the minimum sampling spacing. Typically, the variogram increases with increasing lag distance to attain or approach a maximum value or sill (C_0+C) almost equivalent to the population variance, i.e. priori variance. Cis the structural variance and a is the spatial range across which the data exhibit spatial correlation. For Gaussian model, the practical range is defined as $\sqrt{3}a$.

Information generated through variography step was used to calculate sample weighting factors for spatial interpolation by an ordinary block kriging procedure, using the nearest maximum 12 sample points and a maximum searching distance almost equal to the range distance of the variable. For Pb concentration data, ordinary kriging system was solved for Ln-transformed data. Since we are not interested in an estimate of Ln-transform, but in an estimate for the original scale of the data, a back transformation was carried out according to Webster and Oliver (2001). The general form of this back transformation is:

$$Pb(x_0) = \exp\left[\hat{Pb}_{Ln-transformed}(x_0) + \frac{1}{2}\sigma^2{}_{kriging}(x_0) - \psi\right]$$

where Ψ is the Lagrange multiplier in the ordinary kriging system.

3. Results and Discussions

Descriptive parameters and probability distribution of the data set

The representative statistical summary of the available data set for lead and other soil variables has shown in Table 1. It was noted that the mean of Pb concentration in topsoil was about 4-times higher than median value. The Pb concentrations ranged from 3 to 620 mg/kg with the mean value of 102 mgPb/kg. Mean values reported for background concentrations were 10 mg/kg (Alloway, 1995);15 mg/kg (Page and Chang, 1993); 2 to 44 mg/kg (Kabata-Pendias and Pendias, 1992); 20 mg/kg (Adriano, 1986). The Pb concentrations in our studied area showed that only 12% of sample points contain Pb concentration less than 10 mg/kg. About 50% of sampling points had a Pb concentration between 10 to 50 mg/kg. Considering the guide value of 250 mg/kg for Pb (Saby et al., 2006), 15% of data showed Pb concentration more than this soil guideline value. The above mentioned concentrations of the lead showed that the sampling area was mostly contaminated.

Table 1 and figure 2 represent that topsoil Pb data set was strongly highly skewed. A distribution is considered highly positively skewed when the coefficient of skewness is much higher than 1 (Webster and Oliver, 2001). A Kolmogrove-smirnov test was computed and indicated the Pb concentrations were significantly lognormal ($p<0.0001$). As in conventional statistics, a normal distribution for a variable under study is desireable in linear geostatistics (McGraph et al., 2004). Serious violation of normality, such as too high skewness, can impair the variogram structure and and the kriging results. It is often observed that environmental variables are lognormal or positively skewed, and data transformation is necessary to normalize such data sets. Taking Ln-logarithms of the data removed most of the skeweness (Figure 3).

Meanwhile, the coefficient of variation of Pb was about 72%, less than those reported by Saby et al. (2006) and Hooker and Nathanail (2006). However, this CV value, obtained in our data set, suggests that Pb had a great variation and thus would be possibly influenced by the extrinsic factors such as climatic conditions.

Table 1. Descriptive statistics of 100 sample points for soil variables for the surface soil (0-10 cm)

Variable	Minimum	Mean	Median	Maximum	Standard Deviation	CV (%)
Pb (mg/kg)	3.2	101.9	27.2	620.0	144.0	73.0
OM (%)	0.1	0.2	0.1	0.7	0.1	81.0
EC (dS/m)	0.1	0.5	0.2	3.6	0.6	14.0
pH	7.2	8.1	8.1	8.4	0.2	2.0
Clay (%)	6.0	15.0	15.0	31.0	10.0	31.0
Silt (%)	6.0	27.0	26.0	45.0	8.0	32.0
Sand (%)	39.0	59.0	85.2	83.0	5.0	17.0

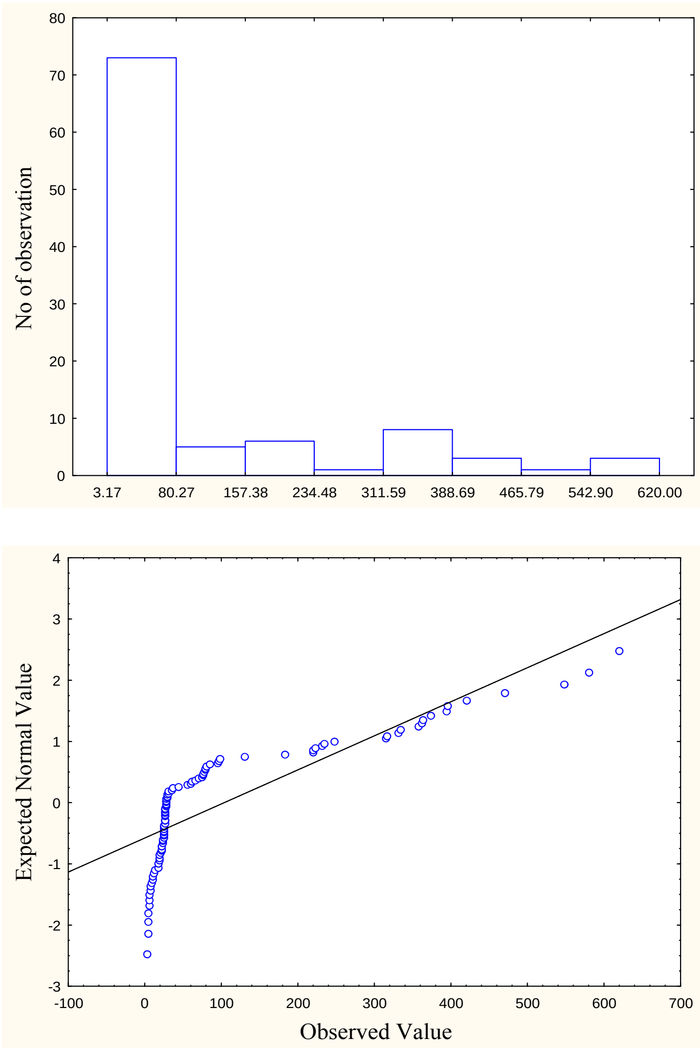
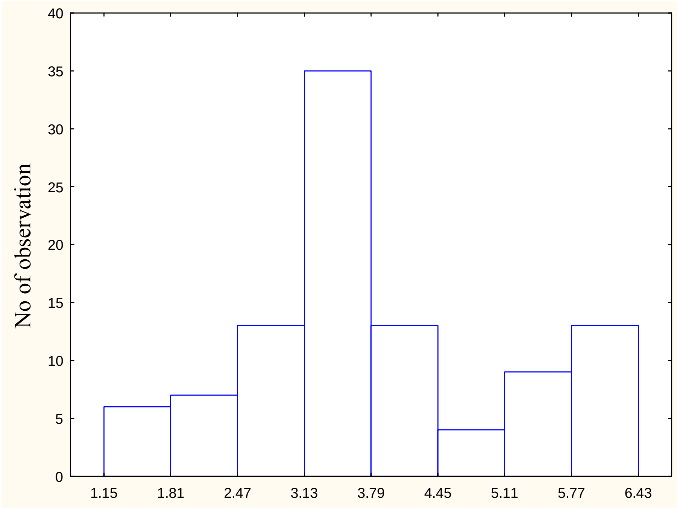


Fig. 2. Histogram and normal probability plot of the original Pb concentration values



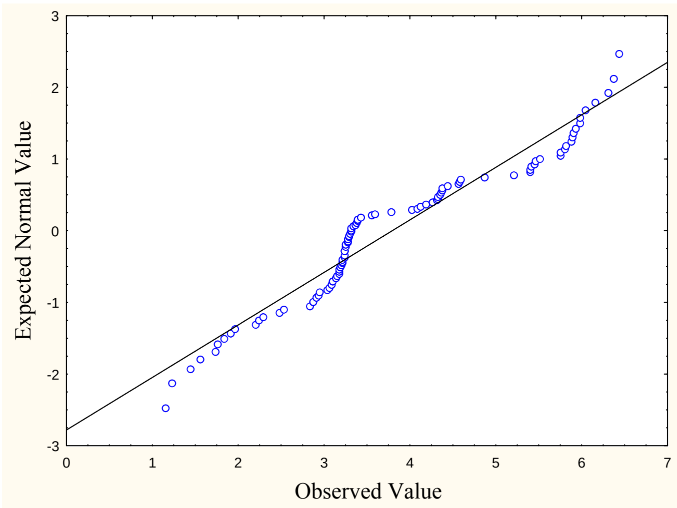


Fig. 3. Histogram and normal probability plot of the Ln-transformed Pb Concentration values

Figure 4 shows the soil-Pb data symbol coded into 5 user-defined classes. The symbols go from a plus "+" symbol for low concentrations to a triangle "Δ" for high concentrations. It can be seen that the extreme north and northeast corner of the study area where is in vicinity of the town tends to have relatively lower concentrations of soil Pb. Higher soil Pb concentrations appear to be more numerous in the south and southwest of the area where mining sites are located.

Calculated summary statistics for other soil properties indicated low organic content of surface soil, low electrical conductivity, almost neutral soil pH, and medium to light-textured class for surface soil in the study area. Furthermore, organic matter and electrical conductivity showed greater variation (coefficient of variation) among the soil samples.

Acquired simple correlation coefficients (Pearson coefficient) between Ln-transformed Pb data and other physical and chemical soil variables indicated that there is only (linear) correlation between Pb concentrations and sand content ($p<0.05$).

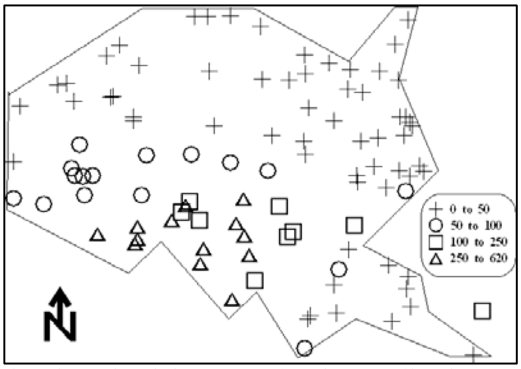


Fig. 4. Classed postplot of Pb Concentrations for 100 soil surface sample points

Geostatistical analysis

Variography analysis

At the first stage of the variography analysis of soil parameters, surface variograms were calculated in order to test for anisotropy. Surface variogram of Pb data set is shown in figure 5. The spatial pattern of isolines seen in this figure suggests a fairly isotropic spatial distribution of the Ln-Pb values, so that the omnidirectional variogram with the fitted Gaussian model was considered an appropriate model (Figure 6). The same testing approach for anisotropy was carried out for other soil variables. According to these results, the omnidirectional variograms with theoretical models of spherical and/or exponential functions were fitted to them. The selection of appropriate model was based on qualitative interpretation of which model best represented the overall behavior of the experimental variogram. The numerical results are given in Table 2.

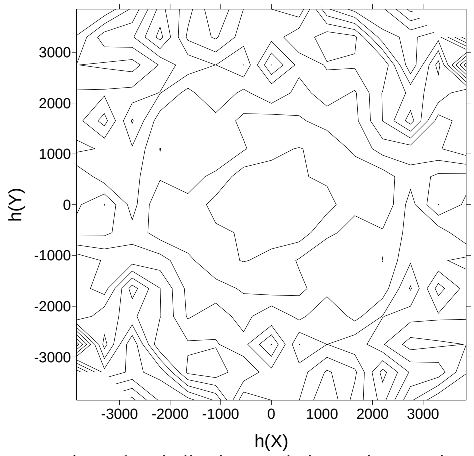


Fig. 5. Surface variogram of Pb concentration values indicating no obvious anisotropy in spatial distribution of soil Pb values

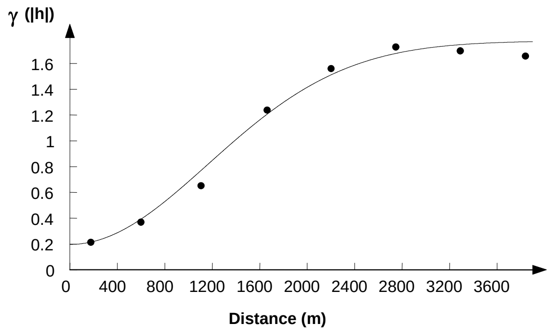


Fig. 6. Omnidirectional experimental variogram (dots) and the fitted Gaussian model (line) for soil-Pb concentrations

Table 2. Parameters of the theoretical models fitted to the experimental variograms for soil variables

Variable	*Model*	*Nugget effect*	*Sill*	*Range* (m)*	R^2	*Spatial dependency*
Ln(Pb)	Gaussian	0.197	1.775	4933	0.98	Strong
OM	Spherical	0.005	0.012	849	0.96	Moderate
EC	Spherical	0.128	0.260	1421	0.97	Moderate
pH	Exponential	0.009	0.020	2558	0.97	Moderate
Clay	Pure Nugget	-	-	-	-	-
Silt	Exponential	19.570	54.870	4045	0.95	Moderate
Sand	Exponential	40.710	81.420	4948	0.97	Moderate

* Practical range for exponential and Gaussian models

To define the degree of spatial dependency, spatial class ratios similar to those presented by Cambardella et al. (1994) were adopted. That is the ratio of nugget variance (noise) to total variance (sill) multiplied by 100. If the ratio of spatial class was less than 25% then the variable is considered to be strongly spatially dependent; if the ratio was between 25% and 75%, the variable was regarded as moderately spatially dependent; and if the ratio was more than 75%, the variable was considered weakly spatially dependent. The resulting variogram of Ln-transformed Pb data indicated the existence of strong spatial dependence. The variograms for other soil variables revealed moderate spatial structure.

Comparing range values calculated for variograms of different soil variables indicated that there is a close similarity between practical range of Ln-transformed Pb data (4933 m) and the range value for sand content (4949 m). This suggests a possible spatial correlation between Pb concentrations and percentage of sand in the study area.

Estimating and mapping soil variables

Mapping pollutant concentrations is often a preliminary step towards decision making, such as delineation of polluted area or identification of zones that are suitable for specific land uses. Ordinary kriging system was used along with isotropic variograms to estimate soil physical and chemical values at about 7400 unobserved locations. For Pb, Ln-transfromed data used for kriging interpolation and then the estimated kriged values were back-transformed. Optimal kriging parameters were found based on the results from the cross validation procedure.

Figure 7 presents the spatial patterns of the Pb concentration values generated from its variogram and corresponding kriging system. The kriged map showes the spatial variation of soil lead estimates. The kriged map of Pb clearly shows high Pb concentrations in vicinity of mining sites with a decreasing gradient from south to northern parts of the study area. It means that the Pb concentrations decrease with increasing distance from mining sites. This gradient might be attributable either to comparable gradients in influencing and controlling factors (e.g. main wind directions and velocities, historical and rate of mining activities) or to an effect of distances of diffuse Pb transportation by air. Although areas close to the town show Pb concentration less than 50 ppm, however existing such a strong gradient and its governing factors indicate a probable risk of Pb concentrations associated with finer soil particles, i.e. particles with size less than 2 mm. Siegel (2002) considers that the true threat to an ecosystem may be obscured by chemical analysis of the total soil sample and, that the chemical analysis of the finer-size of soil matter, undiluted by coarser sizes, can better predict the potential of pollutants to enter an ecosystem and pollute ecosystem components. The results indicate that the concentrations are generally higher in the finer fraction (Ferreira da Silva et al., 2004; Inacio et al., 2002; Navas and Machin, 2002). These finer particles with their associated metals represent particles moved easily through atmosphere. They can enter to human body via respiration or inhalation process. We are now encountering this problem and dealing with chemical analysis of Pb concentrations in finer soil particles.

The calculated error (kriging standard deviation) spatial distribution in the Pb estimation map is shown in Figure 8. The relatively high errors might be due to high dispersion of Pb concentrations in the study area. As it is indicated by the error map, kriging standard deviations drop sharply close to the data points.

Kriged maps of other soil variables are shown in Figure 9. Among them, the spatial distribution pattern of sand shows close similarity with that of Pb concentrations. This can be attributed to the spatial correlation between these two soil properties.

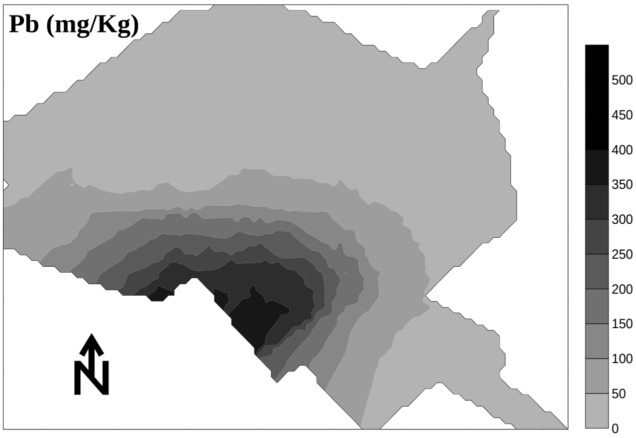


Figure 7. Kriged map of soil-Pb concentration in the study area

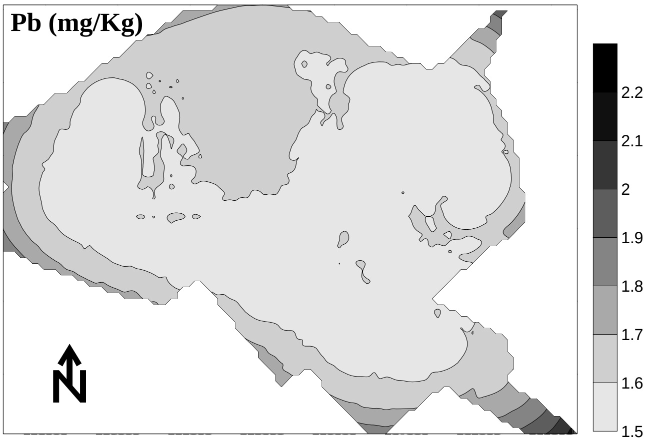


Figure 8. Error map (kriging standard deviation) for the estimated Pb concentration values

4. Conclusion

In this study, a geostatistical approach was adopted to analyzing and interpreting the near-surface soil Pb concentration data and other related soil chemical and physical variables collected around Sepahanshahr town, Isfahan. Descriptive summary statistics of Pb data has shown that there is a significant possibility of an acceptable pollution risk to human health in the studied area. High concentration of Pb was found with maximum value of 620 mg/kg in soil. Variography showed that the soil lead concentrations are spatially correlated and therefore spatial estimation is valid. Among other soil variables only clay content data exhibited lack of spatial structure i.e. pure nugget effect spatial variation. In accordance with significant linear correlation coefficient between Pb concentration data and sand content, both variables revealed nearly the same range values indicating presence of possible co-regionalization.

The kriged map of Pb concentrations revealed that the soils in the vicinity of mining sites, particularly the old Bama mining site, have been severely polluted by mining activities in the past and in the present times. Although there are not any networks of atmospheric deposition measurements in the area, however it seems that the main source of soil pollution related to the diffusion and dispersion pollution. In any case, there is a need using techniques such as relative topsoil enhancement in order to separate Pn content due to diffuse pollution from geochemical background.

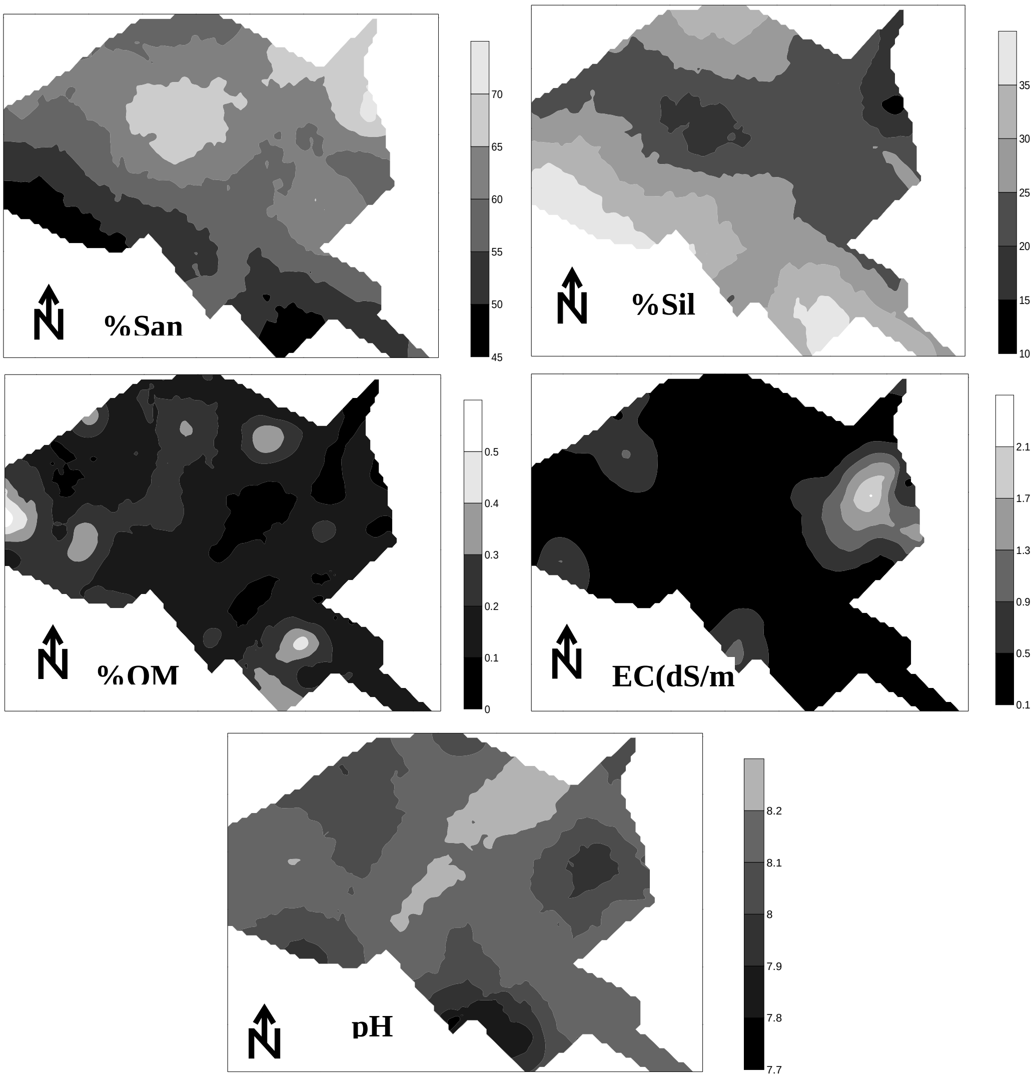


Figure 9. Kriged maps of some chemical and physical soil properties

It is note worthy that the tested soils mainly distributed under the bare areas and only very small parts of the study area are under agricultural cultivation practices. Therefore this does not represent an immediate exposure risk to humans through oral pathways. However, the main exposure pathways for Pb in the study area are considered to be respiration and inhalation pathways. Although, other exposure pathways such as skin absorption and geophagia i.e. eating earth materials, should also taken into consideration.

This work should be considered as a starting point to an orientation survey concerning essentially the identification of the extent and spatial variability of Pb pollution in the mining sites and surrounding area. Whether the Pb in this area pose a human health risk depends on several factors that can only be determined after a proper site specific risk assessment is undertaken.

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