Caspian J. Env. Sci. 2008, Vol. 6 No. 1 pp. 11~17

CJES
Caspian Journal of Environmental Sciences

[Research]

Quantifying the effect of traffic on lead accumulation in soil: a case study in Iran

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ABSTRACT

Road transport is a ubiquitous source of lead contamination in the soil near highways with direct and indirect impacts on human health. Accumulation of traffic-induced lead in the soils depends on gasoline lead content, traffic volume, as well as meteorological conditions. To evaluate the effect of traffic on soil lead concentration, 113 samples from the topsoil (0-15 cm) were collected in a regularly spaced grid (70 x 70 m) in the vicinity of a busy highway (Tehran-Karaj) ;the west of Tehran. Total lead (Pb) concentration in soil samples was extracted by 4 M HNO_3. Three different kriging techniques (ordinary, universal and indicator kriging) were applied to investigate the spatial distribution of Pb. According to the measured values, the average concentration of Pb (48 mg kg $^{-1}$) was very close to the upper limit of the Swiss guide value of 50 mg kg^{-1} with the maximum value (75 mg kg^{-1}) exceeding this limit. The result also showed an accumulation of 5 to 25 mg kg^{-1} Pb in the soil based on the distance from the highway. The average concentration of Pb decreased exponentially with distance from the highway up to 200 m becoming relatively constant thereafter. The trend of spatial continuity of Pb concentration was in agreement with major wind direction. Comparing different kriging techniques indicated that UK was superior due to smaller error of estimation. The probability map of Pd concentration exceeding the Swiss guide value clearly shows the traffic to be the main cause of soil Pb contamination.

Keywords: Lead accumulation, Traffic, Universal kriging, Highway pollution, Iran.

INTODUCTION

Human activities have significantly increased the global emissions of heavy metals, particularly lead (Pb), in the surface environment (Nriagu and Pacyna, 1988). Lead is among the most hazardous heavy metals that has damaging impacts on human and environmental health (Wong 2004). Traffic induced Pb, as a result of using leaded gasoline, is one of the major sources of lead in the populated and highly industrialized areas (Fakayode and Olu-Owolabi, 2003). Due to growing concerns about the problems associated with Pb, the use of leaded gasoline has been decreasing globally at an annual rate of about 7% (Faiz *et al.*, 1996). The maximum level of Pb in leaded gasoline has been set to be less than 0.15 g L^{-1} since July 1989 (Nriagu, 1990), but there are still many countries that use leaded gasoline with Pb content of about 0.4 g L^{-1} (Faiz *et al.*, 1996; Kaysi *et al.*, 2000). In developing countries such as Iran leaded gasoline with Pb content of more than 0.4 g l^{-1} was used in the past decade (Rahmani *et al.*, 2001). Although the use of leaded gasoline decreased during this time period, but the increasing number of automobiles compensated its effect on the vehicles based on lead emission. In addition, wearing down of vehicle tires can also introduce Pb (Giannouli *et al.*, 2007) to the roadsides soil. Consequently, road transport is still polluting the atmosphere, soil and water near the highways (Caselles, 1998; Fakayode and Olu-Owolabi, 2003; Li, 2006).

Soil represents a major sink for heavy metals in the terrestrial environment. Due to the non-biodegradability and cumulative tendency of Pb, emitted lead from the vehicles accumulates in the surface soils in the long run (Sutherland *et al*., 2000). The concentration of Pb in the roadside soils is influenced by gasoline quality, traffic intensity, as well as meteorological conditions like the velocity and direction of wind and to some degree by precipitation (Viard *et al*., 2004). Several studies indicated Pb pollution in the surface soil due to the use of leaded gasoline in developed countries (Mellor and Bevan, 1999; Al-Chalabi and Hawker, 2000; Sutherland *et al*., 2000; Li, 2006), but the extent of the pollution has not been quantified in developing countries such as Iran. The environmental problems associated with leaded gasoline are exacerbated where highways are close to agricultural lands. This is because accumulated Pb can be transferred to food chain by either direct atmospheric fallout on the plants or plant uptake from the soil.

Geostatistical techniques are powerful means of evaluating spatial trends (Goovaerts *et al*., 1997). These techniques can be used to estimate either concentration of Pb at a particular location, or the probability of exceeding in a given threshold using the measured data in the neighborhood. Such information is essential to map potential risks to environment or human health (Cattle *et al*., 2002; Amini *et al*., 2005).

The objective of this study is to use geostatistical approaches to quantify the effect of traffic on Pb accumulation in the agricultural soil at in he vicinity of Tehran-Karaj highway.

MATERIALS AND METHODS

Study area

The study area is in the research station of the agricultural faculty of Tehran University, which is located at a distance of 10 m north of Tehran-Karaj highway. This area is located in a river alluvial plain and the soil of this region belongs to xeric haplocambids. The soil texture is loamy with an average pH of 7.7, EC of 1.4 dSm^{-1}, organic matter (OM) content of 0.9%, and $CaCO_3$ content of 15.0%. Wind rose indicating the frequency of yearly wind direction (in percent) in Karaj weather station is shown in Fig. 1. In about 45% of days in a year there is no wind (calm) in the region but winds are more frequent in north-west direction. This highway has a high traffic density of more than 70,000 cars per day. Silage maize is the major feeder crop followed by alfalfa and wheat are the main cereal crop in this region. Due to the lack of animal manure, crop production is based on mineral NPK fertilizers. Furthermore, as there is no addition of compost and sewage sludge, the observed lead concentration is mainly assumed to be from traffic.

Sampling and chemical analysis

The study area was discretised into a grid system of 490 m in East-West direction and 420 m in North-South direction, with cell sizes of 70 x 70 m. A finer grid was created within the coarser grid and a total of 113 topsoil samples (0-15 cm depth) were collected on cell nodes (Fig. 2). The samples were air-dried, passed through a 2 mm sieve and extracted for total Pb concentration by 4 M HNO_3. (Cao *et al*., 1984). Lead concentration was measured by an atomic absorption spectrophotometer (Shimadzue 750).

Spatial analysis

The principles of geostatistics have been presented in detail by Goovaerts (1997), Deutsch and Journel (1998) among others. A brief summary of the techniques used in this paper are presented below.

Ordinary kriging

The most frequently used form of kriging is (OK). Ordinary kriging estimates are linear weighted averages of the *n* available observations:

$$\hat{Z}(x_o)=\sum_{\alpha=1}^{n}\lambda_\alpha Z(x_\alpha) \qquad (1)$$

Where Z is the concentration of lead in this study; λ_α's are the weights assigned to the *n* measured points; $Z(x_\alpha)$, around the estimation points; $\hat{Z}(x_o)$, and should sum to one.

Universal Kriging

Ordinary kriging may be used in a generalised form which is designed to deal with a non-stationary mean. This is termed kriging with a trend model, which is also known as universal kriging (UK). In using UK, a trend component (deterministic variation) $m(x)$ is added to a residual component (stochastic variation), $R(x)$ as follows:

$$Z(x) = m(x) + R(x) \tag{2}$$

In UK, the trend is modelled as a deterministic function $f_k\ (x)$ of the co-ordinates (Goovaerts, 1997; Deutsch and Journel, 1998):

$$m(x) = \sum_{k=0}^{K} \beta_k f_k(x) \tag{3}$$

The unknown parameters β_k are then fitted to the data.

Indicator kriging

The utility of OK (as well as UK) is limited in particular by properties such as normality of observed data and the independence of its standard error with the data values. The indicator approach is an alternative method to overcome these limitations and can be used to estimate the probability of exceeding of a specific threshold value (Goovaerts, 1997). In indicator kriging, the variable $Z(x)$ is transformed into indicator variables using k threshold (Z_k) with a binary distribution as follows:

$$I(x; Z_k) = \begin{cases} 1 & \text{if } Z(x) \le Z_k \\ 0 & \text{otherwise} \end{cases} \qquad k = 1,\ldots,K \quad \alpha = 1,\ldots,n \tag{4}$$

Indicator variograms are then required for k thresholds to estimate the conditional cumulative distribution function (ccdf) at each threshold. The ccdf is then constructed by putting together the IK estimates for each threshold. Having the ccdf, the probability of exceeding of a given critical concentration can be computed.

We used the GSLIB library programs KT3D to perform OK, UK kriging, and the program IK3D for IK (Deutsch and Journel, 1998). In IK3D nine deciles of total concentration of Pb were used to estimate the ccdf of the Pb at unsampled locations. In all methods the search radius of 100 m with a minimum of two and the maximum of 20 observations were used. The performance of the three kriging techniques were compared using the mean error (ME) and the mean square error (MSE) statistics.

As there are yet no official reference values for soil pollutant concentrations in Iran, we used the Swiss guide value of 50 mg kg $^{-1}$ (VB Bo, 1998) as a critical concentration of Pb in order to produce the probability maps (fig 3).

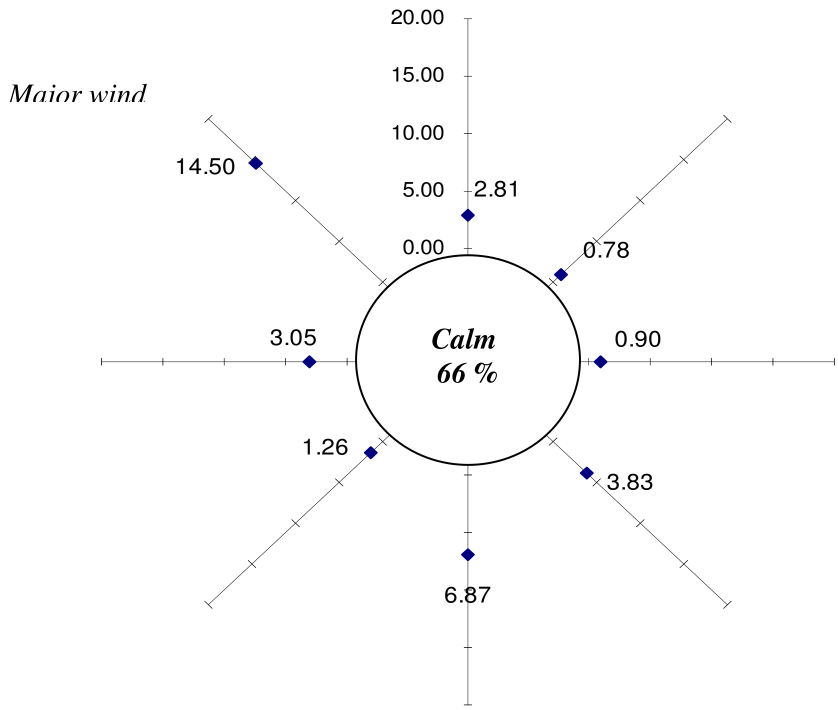

Fig 1. Wind rose indicating the frequency of yearly wind direction (in percent) at karaj weather station. All axes have the same division as in the northern one from 0 to 20. Major wind direction is in the NW at 14.5%. "Calm" indicates 66% of days had no wind.

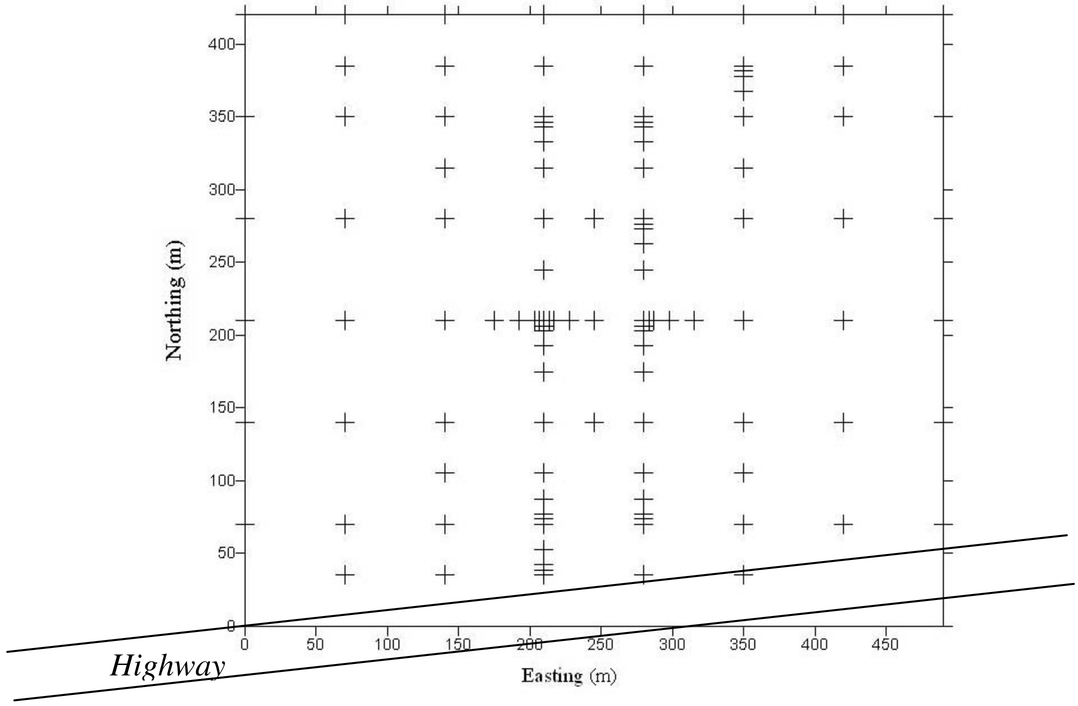

Fig 2. Scheme of the study area and sampling locations.

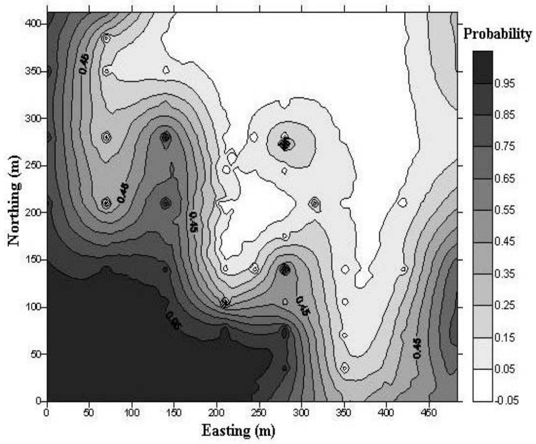

Fig 3. Map of probability of exceeding the concentration of Pb from its respective Swiss guide value of 50 mg kg^{-1}.

RESULTS AND DISCUSSION

Statistical summary

Mean concentration of Pb in the soil samples was 48 mg kg^{-1} with a standard deviation of 7.5 (Fig. 4). The maximum total concentration of Pb was about 75 (mg kg^{-1}), which exceeded its respective Swiss guide value of 50 mg kg^{-1} (VBBo 1998). Distribution of Pb (Fig. 1) was skewed similar to other studies such as Cattle *et al.*, (2002), Chang *et al.*, (1998) and McGrath *et al.*, (2004). Considering that the geology of the study area is uniform, the presence of relatively high concentrations of Pb (higher than 50 mg kg^{-1}) in some locations and alluvial sediment of quaternary age is more likely to be due to anthropogenic sources such as traffic.

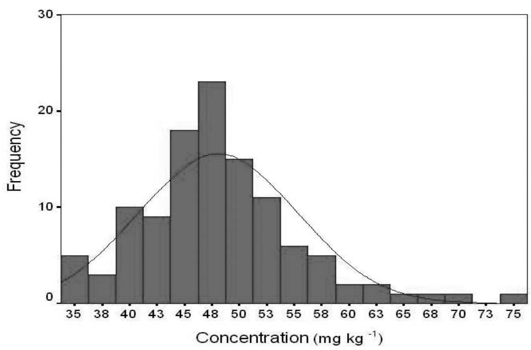

Fig 4. Distribution of total concentration of Pb in the soil sampled from the study area.

Spatial description

To demonstrate the variation of Pb concentration resulted from traffic, observed data were first categorized according to their perpendicular distance from the highway, and then the mean concentration of each category was calculated and plotted against the distance. The results show that the mean concentration of Pb decreases as the power function of distance from the highway up to about 200 m and then it becomes relatively constant (Fig. 5). Although traffic induced Pb can be transported to more than several kilometers by wind, we focused only on a 200-m corridor perpendicular to the highway and assumed that mean concentration of Pb at distances larger than 200 m were equal to the background concentration. In the 200-m corridor, mean Pb accumulation varied from 25 mg kg^{-1} near the highway to 5 mg kg^{-1} at the end of the corridor.

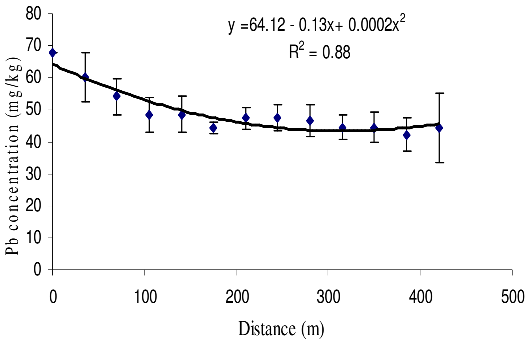

Fig 5. Variation of Pb concentration according to distance from the highway.

Computed surface variogram of total Pb concentration indicated a large spatial continuity in NW-SE direction (Fig. 6), which is in agreement with major wind direction (Fig. 1). This demonstrates the effect of wind direction and traffic on Pb concentration in the soil near the highway.

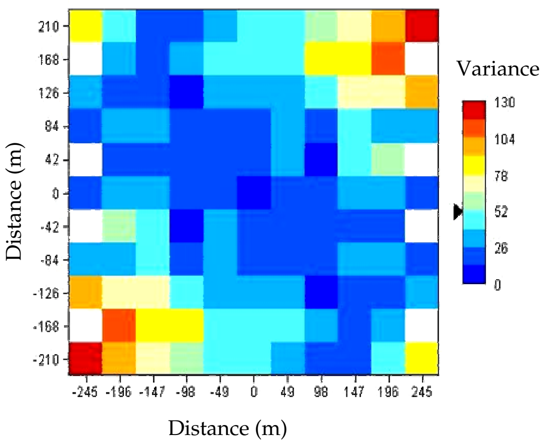

Fig 6. Surface variogram of total Pb concentration.

The omnidirectional variogram of total Pb increases no limit by increasing the distance, which is an indication of a low-frequency trend (Fig. 7a). Similar results have also been reported by Lioyd and Atkinson (2001). Due to the anisotropic feature illustrated in the surface variogram of total Pb and the existing trend, directional-variograms were computed and used for OK. For UK, the trend was first removed using a quadratic polynomial model and then the residuals were used to compute omnidirectional variogram (Fig. 7b). By removing the trend, a linear model of raw data was transformed to a bounded spherical model of variation with the range of 100 meter.

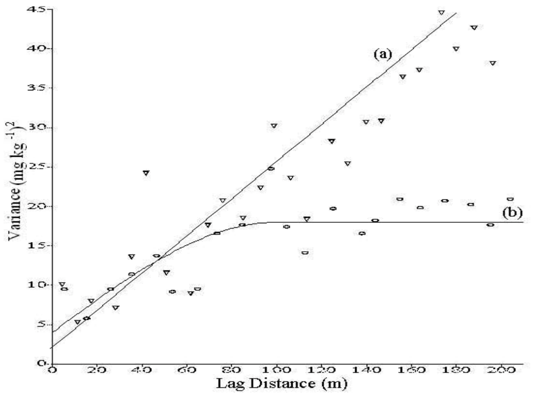

Fig 7. Variograms of original total Pb concentration data and data based on removed trend in universal kriging analysis.

The parameters of fitted indicator variograms are listed in Table 1. The best fits were obtained by spherical or exponential models as well as a nugget effect. The ratio of nugget to sill which is a measure of the spatial continuity is computed for different variograms. This ratio can be regarded as a criterion for classifying the spatial continuity of the variables (Chang *et al.*, 1998). If this ratio is less than 25% the spatial continuity is strong, between 25% and 75% the variable has a moderate spatial continuity, and for larger than 75% weak spatial continuity is suspected. The ratio of nugget to sill in the indicator variograms varies from 11% to 66% which indicates a strong moderate spatial continuity for different thresholds. The first two thresholds show a strong continuity but the remaining thresholds indicate a relatively moderate continuity. This is probably due to the presence of the trend in the data set ensuring that the small values of Pb occur far from the highway.

In OK, UK and IK, the ultimate goal is to estimate the optimal value of Pb concentration at unsampled locations. In order to compare the performance of these techniques, mean error (ME) and mean square error (MSE) of OK, UK and IK were computed using cross validation procedure. The results are summarized in Table 2.

Table 1. The thresholds, (Z_k), and parameters of indicator variograms for Pb.

Z_k mg kg^{-1}	Nugget	Sill-Nugget	Range m	Nugget/Sill %
	(mg kg^{-1})^2			
39.8	0.012	0.09	374	12
41.9	0.026	0.15	222	14
44.5	0.13	0.066	160	66
46.1	0.127	0.14	261	47
47.3	0.12	0.16	290	42
48.8	0.074	0.15	191	33
50.8	0.045	0.13	182	26
52.7	0.037	0.101	186	27
57.1	0.028	0.037	178	43

Table 2. Mean error and mean square error for ordinary kriging (OK), indicator kriging (IK), and universal kriging (UK)

Kriging Method	Maximum Error	Minimum Error	Mean Error	Mean Square Error
OK	234.03	0.004	0.17	25.3
IK	120.08	0.002	0.13	16.2
UK	94.80	0.000	0.06	13.0

The overestimation of OK and IK are about twice that of UK according to the calculated ME. The calculated MSE which is a measure of estimation accuracy indicated a significant difference between UK and IK as compared to OK. The predictions of UK were more accurate than that of OK and IK. However, using a polynomial trend in UK improved the performance of predictions by about 48% compared to OK. The UK outperforming OK has also been reported for point source pollution in Dallas metropolitan area of Michigan by Saito and Goovaerts (2001). However, other researchers found no significant difference between kriging methods (e.g., Papritz and Moyeed, 1999).

We used IK to estimate the ccdf for Pb and then computed the probability of exceeding Pb concentration according to the Swiss guide value of 50 mg kg^{-1} as illustrated in Figure 6. As it was expected, a hot zone of elevated concentrations of Pb can be identified in the soils close to the highway up to about 200m in the southwestern part of the study area.

SUMMARY AND CONCLUSION

Soil Pb pollution of the study area was evaluated using a set of 113 soil samples collected in a regular grid. Three different kriging methods, i.e. ordinary, universal and indicator kriging were used to estimate Pb concentration in unsampled locations. According to the computed mean square error UK outperformed OK and IK. Conditional cumulative distribution functions (ccdf) were estimated using IK for total Pb. Based on the estimated ccdf, the probabilities of exceeding Pb concentrations from Swiss guide value was computed. A hot zone of elevated concentrations of Pb was identified in the soils close to the highway up to about 200 m in the southwestern part of the study area. Recently using leaded gasoline in Iran has substantially decreased. However, the results of this study show a clear accumulation of Pb in road soils which is a footprint of vehicle based lead emission.

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