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ORIGINAL PAPER

Spatial distribution of heavy metals in soils and plants in Zinc Town, northwest Iran

M. A. Delavar[1] · Y. Safari[1]

Abstract Identification of heavy metals spatial variability in soil and plants may provide useful information on how to manage the polluted sites. The main objective of this study was to determine the spatial distribution of selected heavy metals in soils and natural plants of Zanjan city, northwest Iran. A total of 184 composite topsoil samples (0–10 cm) and 98 natural plant samples were systematically taken from an area of about 4000 ha located around an industrial complex, covering rangeland and agricultural and industrial land uses. All samples were analyzed for their total concentration of Zn, Pb and Cd. The results showed that the average concentrations of Zn, Pb and Cd in the soil samples were up to 294.2, 152.8 and 5.6 mg kg^{-1}, respectively, whereas in the plant samples, these values decreased to 131.4, 113.2 and 2.5 mg kg^{-1}, respectively. These contents are much higher than the normal range in soil and plant communities, leading to classify the studied area as a polluted site. Variography analyses revealed a similar spatial structure for the studied heavy metals in the soil and plant samples. Based on interpolated maps, the highest concentrations of the selected heavy metals in the soil and plant samples were found in the vicinity of industrial complex. These findings clearly highlight the role of industrial activities in simplifying the entrance of dangerous trace elements to the human food chain. Application of ordinary kriging technique to predict the heavy metals spatial variability in the plant community resulted in logical estimations with acceptable error values.

Introduction

Soil as the most important human food resource serves as both a sink and a source for heavy metal (HM) pollutants (Sun et al. 2010). These pollutants are generally discharged into soil by industrial activities (Alloway 1995; Möller et al. 2005), agricultural additives (Pang et al. 2011) and various urban activities (Jing et al. 2007). Concentrated HMs in soils may present a serious risk of human health because they are non-degradable pollutants with a large spectrum of negative effects (e.g., nervous or digestive system disturbances and carcinogenic effects) (Maas et al. 2010). Contaminated urban soils directly expose humans to HMs due to their close proximity to human activities (Sun et al. 2010), whereas agricultural soils rich in HMs play this role indirectly due to possible transfer of metals through food webs (Burger 2008). Hence, determining the spatial distribution pattern of metal pollutants in soils for human and ecological risk assessment is necessary.

Such spatial data help environmental scientists in defining the areas where risks are high and decision makers in identifying locations where remedial efforts should be made (Li et al. 2009). Because of the complexity of the soil body and various resources of HMs, these pollutants vary greatly over the land surface, and their distribution patterns generally have high spatial heterogeneity, paralleling variations in pedological parameters (Maas et al. 2010). Thus, it is very difficult to acquire an accurate spatial distribution pattern of HMs (Xie et al. 2011). Considering the insufficiency of classical statistics to reveal the spatial autocorrelation of soil pollutants (Hu et al. 2006), many

✉ M. A. Delavar
amir-delavar@znu.ac.ir

[1] Soil Science Department, College of Agriculture, University of Zanjan, P.O. Box 45371-38791, Zanjan, Iran

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researchers have focused on and referred to new techniques like geographical information systems (GIS) and geostatistical methods as more useful approaches (Wu et al. 2008; Maas et al. 2010; Karanlik et al. 2011; Pang et al. 2011; Dankoub et al. 2012; Guo et al. 2012; Li et al. 2013). The main advantage of geostatistical method is its unbiased estimation of variable values for spatial objects at not-sampled locations, i.e., interpolation (Goovaerts 1999; Webster and Oliver 2001).

Geostatistics is a powerful interpolation tool that can quantify and reduce sampling uncertainties, minimize investigation costs (Wu et al. 2008) and as well provide a set of statistical tools popularly applied in surveys of soil contamination (Zhang et al. 2008). Using geostatistical estimators, e.g., different kriging methods, spatial variability of soil contaminants can be displayed as a continuous map. This detailed information about HMs distribution pattern in the soils with different land uses assists in developing strategies to protect urban environments and human health against long-term accumulation of HMs (Guo et al. 2012). Although geostatistics is used commonly for risk assessment in contaminated soils, few studies have been carried out to describe the spatial distribution of HMs in natural or cultivated plants on polluted sites.

The process of HMs uptake by various plants is influenced by such different factors as plant species (Alloway et al. 1990), physicochemical soil properties (Qian et al. 1996) and elements concentrations (Thornton 1999). The presence of high levels of HMs in soils, however, exerts a selection pressure on plant populations leading to extra uptake of HMs (Ghaderian and Ghotbi Ravandi 2012). Hence, it is predictable that HMs distribution pattern in the soil affects the spatial distribution of these elements in natural plants grown in the polluted soil without regard to complexity of the plant communities. Therefore, we tested this hypothesis (accurately described by geostatistics) that the spatial distribution of selected metal elements has a similar trend in the soil and plant communities.

Zinc Specialized Industrial Town (briefly Zinc Town), located in the south of Zanjan city, northwest Iran, is one of the most productive metallurgical plants in the country. This industrial complex was established in 1996 with a current consumption of about one million tons of raw ore and a production of 0.19 million tons of Zinc (Zn) per year. It is located 6 km southwest of the city and produces high volumes of metallic wastes undesirably collected around the town; thus, contamination of the adjacent environment consisting of air, soil and plant communities with HMs is almost unavoidable. The main objectives of this study were first to determine the concentration and distribution of Zn, lead (Pb) and cadmium (Cd) in the soils and natural plants of the studied area and then to assess the efficiency of geostatistics in revealing HMs spatial variability in plant community.

Materials and methods

Study area

The area under investigation has a size of approximately 4000 ha. It is located between 36°35′ and 36°40′ north and 48°24′ and 48°28′ east in Zinc Town and the surrounding lands, Zanjan Province, northwest Iran (Fig. 1). This area has a semiarid climate with an average annual temperature of 11.5 °C and rainfall of more than 300 mm. Old alluvial fans are the main geomorphologic unit of the study area, and the soils under investigation have generally been formed on Quaternary alluvial and colluvial deposits. Rangeland and agricultural and industrial areas are major land uses in this area (Fig. 1). *Hulthemia persica*, *Alhagi maurorum*, *Ephorbia macroclada* and *Cardaria draba* are the main endemic plant species in the area. Among these natural plant species, the herbaceous annual plant of *H. persica* was recognized as the dominant species in the studied area and was used for assessing the HMs spatial distribution pattern in the natural plant community. Plant samples were collected from all locations that *H. persica* existed (Fig. 1).

Sample collection and analyses

The sampling points were systematically distributed in the lands surrounding Zinc Town, based on a regular grid sampling method 500×500 m, which is the most usual sampling approach for geostatistical purposes (Webster and Oliver 2001). Thus, 184 grid cells were sampled (Fig. 1). Within each cell, five subsamples (to obtain a representative sample) were collected from the topsoil (0–10 cm). Besides, plant samples from the areal parts of *H. Persica* as the dominant endemic plant species were collected at all sampling points (98 points) located in the rangeland sites; areas consist of a combination of rangeland and agricultural land uses. All the soil and plant samples were collected in August and September 2012.

Soil samples were air-dried and sieved through a 2-mm plastic sieve for removal of large debris, plant roots, gravel-sized and other waste materials. The samples were digested with a 5:2:3 mixture of HNO_3–$HCLO_4$/HF (Li et al. 2012). For the analysis of plant dry matter, leaf materials were washed well with double-distilled water and dried at 70 °C for 48 h. About 1 g of dry leaf sample was added to a 25-ml beaker and burned to ash in a muffle furnace for 14 h at 480 °C. The ash was taken up in 5 ml 10 % HNO_3, and the digest was finally made up to 20 ml in

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Fig. 1 Map of the study area along with sampling points under different land uses
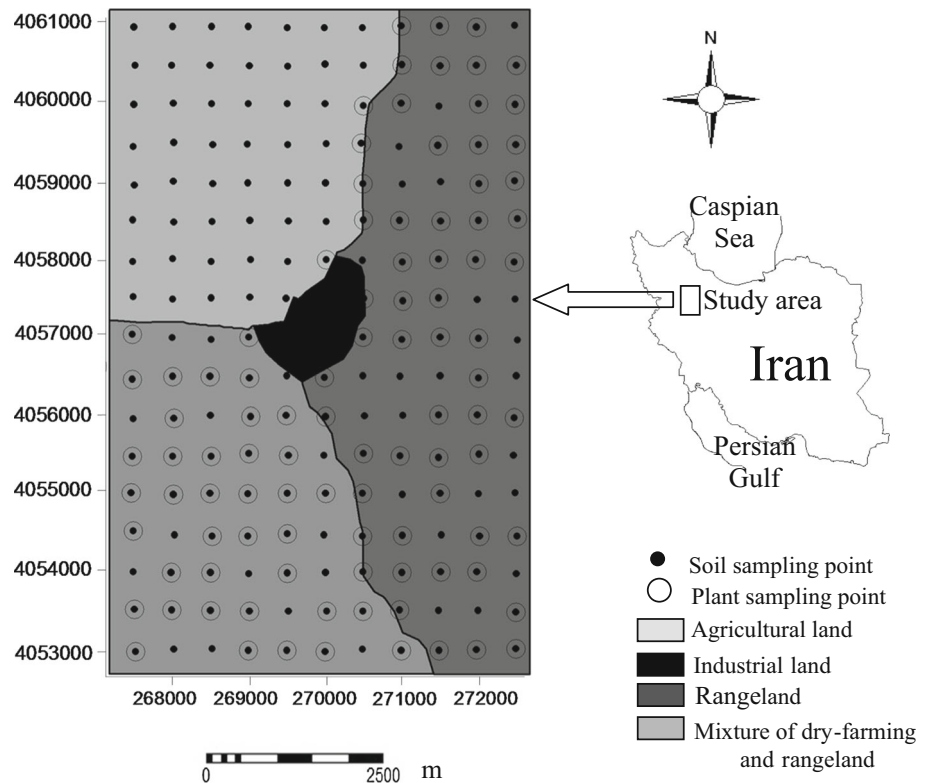
10 % HNO₃. All of the plant and soil digested solutions were analyzed via atomic absorption spectroscopy for their total concentrations of Pb, Zn and Cd.

Data processing

Statistical analysis

Descriptive statistics including minimum, maximum, mean, standard deviation, skewness and kurtosis were determined using the SPSS software version 17.0 for Windows. The normality of data was checked by the Kolmogorov–Smirnov (K–S) test. Since serious violation of normality can impair the quality of geostatistical results (Webster and Oliver 2001), the abnormal data series were transformed to normal distribution.

Geostatistical studies

Geostatistics provides a set of statistical tools for incorporating the spatial and temporal coordinates of observations in data processing, and its increasing use in environmental applications testifies to its utility and success (Saito and Goovaerts 2000). Semivariogram, functioning as the basic tool of geostatistics, is the mathematical expression of the square of regional variables $Z(x_i)$ and $Z(x_i + h)$, namely the variance of regional

variables. Semivariogram $\gamma(h)$ is computed as half of the average squared difference between the components of data pairs (Goovaerts 1999):

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

where $N(h)$ is the number of sample value pairs within the distance interval h; $Z(x_i)$ and $Z(x_i + h)$ are sample values at two points separated by the distance interval h.

The semivariogram function is used by geostatistical interpolation methods to quantify the spatial variability of regionalized variables. Ordinary kriging (OK), as the most common geostatistical estimator, provides an estimate for the whole area around a measured point. The OK estimator is expressed as (Webster and Oliver 2001):

$$Z^*(x_0) = \sum_{i=1}^{n} \lambda_i Z(x_i) \qquad (2)$$

where $Z^*(x_0)$ and λ_i are the estimated values of variable Z at location x_0 and the associated weight with the measured value of Z at location x_i, respectively. The weights are assigned to the observed values so that the estimated error variance is minimized. Interpolated maps of HMs spatial distribution were generated using Surfer software version 9.

In order to check the validity of performed estimations, the jack-knifing technique, the common technique of assessing the predictive capability of different regression models (Lesch and Corwin 2008), was used. Consequently, interpolated and actual (observed) values were compared using the error measurement of standardized root-mean-square error (RMSE %), which is calculated as follows (Hengl et al. 2004):

$$RMSE\% = 100 \times \frac{\sqrt{\frac{\sum (x'-x)^2}{n}}}{\bar{X}} \quad (3)$$

where n is the number of observations, $\bar{X}$ is average of the observed values, and x' and x are estimated and observed values, respectively.

Results and discussion

Statistical analyses of HMs contents in the soil and plant samples

Table 1 presents the descriptive statistics of the studied HMs concentrations in the soil and plant samples.

There was a remarkable change in the content of HMs among the sampled soils: Total concentrations of Zn, Pb and Cd varied between 1.8 and 5400, 0.7 and 3440, and 0.2 and 75 mg kg^{-1}, respectively. Mean concentrations of the studied elements in the soils decreased in the order of Zn > Pb > Cd. Average global values for total Zn, Pb and Cd in uncontaminated soils are about 80–120, 20–100 and 1–5 mg kg^{-1}, respectively (Adriano 2003; Kabata-Pendias 2010). The mean values of the studied HMs contents in the studied soils were significantly higher than those for non-contaminated soils (Table 1). Thus, the studied area can be considered as a polluted site, requiring specific management strategies for preventing the spread of HMs pollution.

For all metals, the total concentrations showed a great degree of variability, indicated by large coefficients of variation (CV) from 234.2 % of Cd to 354.8 % of Pb. The elevated CVs reflected the non-homogeneous distribution of concentrations of anthropogenically emitted HMs. It has been reported that CV values of HMs dominated by natural resources are relatively low, while CV values of HMs affected by anthropogenic resources are quite high (Han et al. 2006). Accordingly, Zn, Pb and Cd contents in soils tend to be affected by anthropogenic activities in the Zinc Town of Zanjan. In other words, the observed high CV values of the measured HMs may show a primary evidence for the effect of Zinc Town HMs on the pollution status in the surrounding lands.

Large standard deviations were found in the studied HMs except Cd. This also indicated a wide range of concentrations in the studied soils. The distribution of concentrations was skewed by a small number of large values (contamination hot spots). These outliers lead to high positive skewness and consequently serious violation from the normal distribution (Table 1). The results of the K–S test ($P < 0.05$) confirmed that the concentrations of the measured HMs were not normally distributed (Table 1). Therefore, the row data of the HMs concentrations were transformed by natural logarithm to a normal distribution prior to geostatistical analyses.

The Zn, Pb and Cd contents of plants in this study ranged from 4.5 to 960.2, 2.5 to 380.5 and 0.4 to 8.4 mg kg^{-1}, respectively. These contents are also higher than their typical contents in the plants (Adriano 2003), but according to Baker and Brooks (1989), the studied plant species does not match the requirements of the hyperaccumulator plants. Notably, Zn and Pb hyperaccumulation does not occur in most plants, say Pb- and Zn-contaminated soils (Wenzel and Jockwer 1999; Reeves et al. 2001). The majority of Zn and Pb hyperaccumulators have been reported from Europe (Reeves and Baker 2000). Analyzing

Table 1 Summary statistics of total concentrations (mg kg^{-1}) of studied HMs in the soil and plant samples

Variable	Mean	Minimum	Maximum	Coefficient of variation (%)	Standard deviation	Skewness	Kurtosis	K–Sp[a]
Soil								
Zn	294.2	1.8	5400	297.6	875.44	3.54	12.51	0
Pb	152.8	0.7	3440	354.8	542.16	4.39	20.23	0
Cd	5.6	0.2	75	234.2	3.26	3.18	9.88	0
Plant								
Zn	131.4	4.5	960.2	161.7	212.41	2.43	5.41	0.001
Pb	113.2	2.5	380.5	90.3	102.2	0.8	0.54	0.11
Cd	2.5	0.4	8.4	77.4	1.95	1.08	0.23	0.035

[a] K–Sp: statistics of Kolmogorov–Smirnov test at significance level 95 %

HMs concentration in natural plants collected from a contaminated site in the Central Iran, Ghaderian and Ghotbi Ravandi (2012) reported that just one of the different studied plant species may be considered as the hyperaccumulator for HMs. Whether or not the plants species grown in the contaminated soils at the present study are physiological races of metal-tolerant species requires further investigation and experimentation.

Similar to soil samples, mean concentrations of studied HMs in the plant samples decreased in the order of Zn > Pb > Cd, with the values of 131.4, 113.2 and 2.5, respectively. This similarity may be considered as an evidence for the impressibility of the HMs distribution pattern in plant communities from their variability pattern in the basement soil. Mean concentrations of Zn and Cd in the plant samples are about half of their total contents in the soil (Table 1). Although simple relationships are seldom found in natural soil systems between plant metal levels and total metal concentrations in soils (Qian et al. 1996), regarding the relatively low values for the ratio of metal content in plant to soil, it is logical and probable that all the plant Zn and Cd uptakes are ensured by soil. However, for Pb, the ratio of metal content in plant to soil increases to about ¾. Considering that the high proportion of the total Pb is an insoluble fraction not immediately bioavailable for plants (Kopittke et al. 2008), founding another source of Pb in the studied area seems to be inevitable. Totally, metal pollutants mainly enter the plant system through the soil or via the atmosphere (Uzu et al. 2010). Because of its strong binding with organic and/ or colloidal materials, it is believed that only small amounts of lead are available in the soil for plant uptake (Punamiya et al. 2010). On the other hand, airborne contamination of plants by lead was evidenced in previous studies (Tjell et al. 1979; Kozlov et al. 2000; Uzu et al. 2010). Hence, it seems that contaminated dusts arising from metal smelting activities in Zinc Town may somewhat have added to the elevated Pb concentration in the natural plants grown in the surrounding lands. However, accurate portioning of HMs of different resources requires further investigation.

Spatial structure analysis of HMs

The spatial behaviors of the selected HMs in the soil and plant samples were evaluated through their variogram surfaces and semivariograms analyses. Since the variogram surfaces did not show any spatial anisotropy (not shown), omnidirectional variograms for the studied HMs in the soil and plant samples were calculated and fitted to a spherical model (Fig. 2). This fitted model was chosen due to the lowest RMSE in comparison with other possible models. The parameters and cross-validation results for fitted spherical models are presented in Table 2.

As presented in Table 2, the range of the optimal model for all the soil and plant HMs concentration data varied between 2100 and 2850 m. This key parameter, which is a function of scale, the distance between sample points and position of landscape (Cambardella et al. 1994), establishes the outer limit at which points in space still interact spatially (Webster and Oliver 2001). Knowledge of the range of influence for various properties allows construction of independent datasets for classical statistical analysis. However, the range of all variables was exceeded 500 m, indicating the presence of spatial structure beyond the original sampling distance. This finding can be a good indicator of decrease in the sampling density for pollution monitoring in the future.

The spatial dependency levels for the selected variables were evaluated by the ratio of the random variance (nugget) to total variance (sill) multiplied by 100, namely relative nugget effect (Cambardella et al. 1994). For the ratio of <25 %, the variable was considered to be strongly dependent on space, or strongly distributed in patches; for the ratio between 26 and 75 %, the variable was considered to be moderately dependent on space; and for the ratio >75 %, the variable was considered to have a weak spatial dependency. The results of this study showed that all the soil and plant HMs contents have moderate spatial dependency level (Table 2), although some differences exist between soil and plant data. RNE values in soil data were quite close to threshold value 25, but these values increased in plant data except for Zn. This observation might indicate that OK had slightly better performance in soil community compared to plant community. Other researchers have reported moderate spatial correlation class for different HMs in the soil (Wu et al. 2008; Karanlik et al. 2011; Dankoub et al. 2012).

As shown in Table 2, HMs distribution pattern in plant samples was similar to the spatial variability pattern of HMs in the soil too. Having the same spatial dependency level and spatial variability model and negligible differences in the effective range in the studied soil and plant, the selected HMs seem to have the same impressibility from natural and anthropogenic resources of pollutants through the space in the studied area. Regarding the low correlation between plant metal levels and total metal concentrations in soils (Qian et al. 1996), it can be concluded that whereas HMs pollutants are just derived from natural resources, sameness of the HMs spatial distribution in the plants and soil is less probable. Hence, a high similarity between soil and plant HMs distribution patterns at the present study may indicate that anthropogenic activities have likely brought a large amount of industrial pollutants and as a result affected the spatial variability of selected HMs concentration.
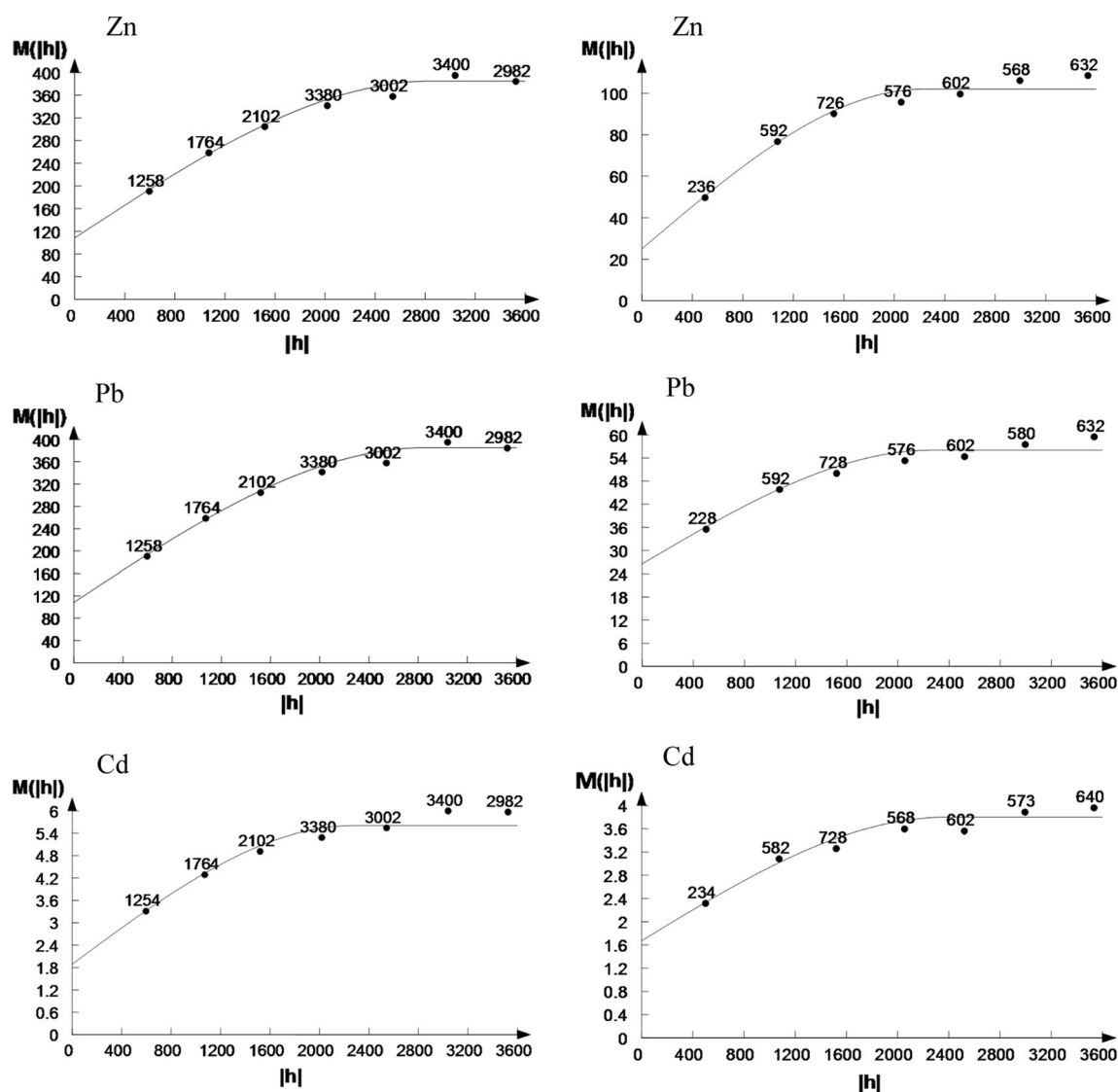
Fig. 2 Omnidirectional variograms for studied HMs in the soil (*left*) and plants (*right*)

Table 2 Authorized models, interpolation parameters and cross-validation statistics of soil and plant HMs contents

Variable	Model	Range (m)	Nugget	Sill	RNE (%)	Spatial correlation class	RMSE %
Soil							
Zn	Sph	2850	108.3	385.2	28.1	Moderate	38.51
Pb	Sph	2520	58.8	197.4	29.8	Moderate	35.23
Cd	Sph	2268	1.9	5.6	33.6	Moderate	34.2
Plant							
Zn	Sph	2100	25.7	102.2	25.1	Moderate	32.25
Pb	Sph	2268	26.5	56	47.3	Moderate	28.54
Cd	Sph	2376	1.67	3.8	43.9	Moderate	30.23

Sph spherical, *RNE* relative nugget effect, *RMSE %* standardized root-mean-square error

Effectiveness of soil and plant pollution assessment depends on an accurate and efficient mapping of the pollutants distribution. Based on the spatial structure of the studied variables, predictive capability of applied interpolation method can vary greatly. The cross-validation statistics given in Table 2 shows how well selected

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variables can be estimated by OK method. All RMSE % values for the studied HMs in the soil and plants are lower than 40 %. Hengl et al. (2004) argued that a value of RMSE % below 40 % means a fairly satisfactory accuracy of prediction. Therefore, the kriging model performed best for all of the selected variables in the studied area. The capability of the OK interpolator to provide relatively accurate estimations of various soil properties, especially total contents of HMs in the soil, has already been proved (Xie et al. 2011). Our findings support the efficiency of OK in estimating selected HMs contents in the natural plant community.

Spatial distribution of HMs

Figure 3 shows the spatial distribution of selected HMs for the soil and plant samples created by OK. As shown in Fig. 3, the HMs values significantly differed in different locations. It appeared that the higher HMs values were concentrated mostly in the central parts of the study area—the industrial zones, whereas the lowest soil HMs contents were found in the south and north of the study area. These observations clearly demonstrated that Zinc Town plays a key role in the pollutants distribution in this area. For all studied HMs, total contents in the soil decreased as the distance from Zinc Town increased. Previous researches have justified that HMs distribution in the soil is strongly influenced by various human activities (Facchinelli et al. 2001; Franco-Uria et al. 2009; Cai et al. 2012; Guo et al. 2012; Qu et al. 2013).

The similarity of spatial distribution pattern of different soil HMs (Fig. 3) indicated that these pollutants were strongly controlled by industrial activities. In comparison with Zn and Pb, some small difference in Cd distribution pattern was observed (Fig. 3). This observation may indicate lower impressibility of Cd total contents in the soil from the industrial activities. In other words, it seems that alternative factors like natural resources affect Cd distribution in the area. However, more studies are required to clarify the relatively accurate portion of all factors controlling the distribution pattern of HMs in this area.

Likewise, the measured Zn, Pb and Cd in plant samples showed high values in the vicinity of Zinc Town, whereas a downward trend was observed when the distance from the pollution source increased (Fig. 3), i.e., duplication of this HMs distribution pattern in the soil. Hence, it can be concluded that Zinc Town controls the spatial distribution of Zn, Pb and Cd not only in the soil but also in other ecosystem components in the study area. These findings clearly highlight the role of industrial activities in simplifying the entrance of dangerous trace elements to the human food chain. Unfortunately, lack of scientific management strategies for controlling the ecosystem pollution by industrial pollutants deteriorates the HMs risks in the area. In accordance with these findings, other researchers have reported some similarities between the contents of the HMs in the soil and plants grown in the vicinity of metal smelter and production factories (Barcan et al. 1998; Ghaderian and Ghotbi Ravandi 2012).

Comparing the trends of studied HMs spatial variability between soil and plant revealed some small differences. Distribution trends of studied HMs in soil were more similar compared to their trends in plant (Fig. 3). This might be due to the complexities of plant uptake process and movement of elements between different parts of plants (Adriano 2003), and this fact that the spatial distribution of elements in plant is more difficult to explain in comparison with soil community.

However, some differences may exist in the distribution pattern of each special HM in the studied soil and plant (Fig. 3). Such differences may reflect a low correlation between total and bioavailable contents of elements in the soil (Kabata-Pendias 2010) as well as existence complexities in plants uptake processes (Adriano 2003). In other words, Pb distribution in the plant samples is to some extent different from Zn and Cd as its maximum levels spread from central parts to the north and northeast of the study area (Fig. 3). Regarding low mobility and consequently low bioavailability of Pb in soil (Kopittke et al. 2008), these relatively high Pb levels in plants may result from the presence of Pb in the industrial emissions, absorbed by aerial plant organs. It has been reported that industrial emissions and dusts contain high concentrations of various HMs, particularly Pb (Han et al. 2006; Dankoub et al. 2012). Based on other related reports, atmospheric deposition of Pb (in the emissions from metal processing factories) could make large amounts of Pb attach to the aboveground parts of the crops (McLaughlin et al. 1999).

The two main purposes of pollution mapping are the analysis of the pollutants spatial structure and identification of the contaminated areas (Xie et al. 2011). Based on the smallness of the calculated estimation error index (Table 2) and logical levels of estimations compatible with actual values of each specific variable (Fig. 3), it can be stated that applied geostatistical method performed well in recognizing the spatial structure of HMs in the soil and plants. In addition, based on the interpolated maps, a maximum of HMs concentrations in plant samples was observed in the areas surrounding Zinc Town, as it was logically expected. Therefore, the OK estimator was able to reveal the spatial distribution pattern of HMs contents in the studied plant samples.
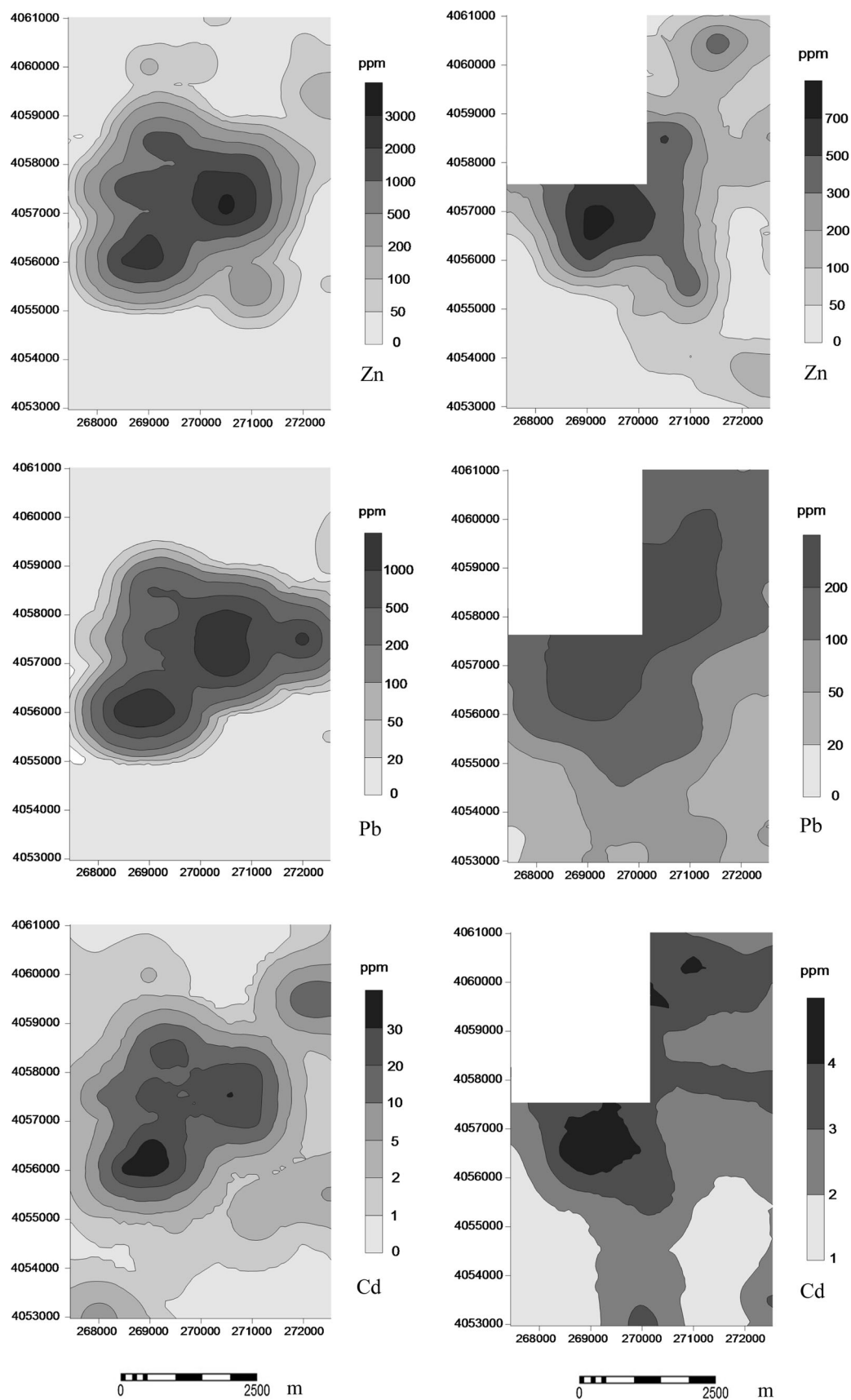
◄**Fig. 3** Spatial distribution maps of the selected HMs in the soil (*left*) and plant (*right*) samples in the study area

Conclusion

Mean total contents of the soil Zn, Pb and Cd in the studied area were higher than those in non-contaminated soils. Thus, the studied area can be considered as a polluted site, requiring specific management strategies to prohibit the spread of HMs pollution. The highest concentrations of Zn, Pb and Cd in the soil and plant samples under study were found in the central parts of the area, around Zinc Specialized Industrial Town. A downward trend in HMs concentration in the soil and plants was observed as a function of the distance from the pollution source. Hence, it can be claimed that the distribution of selected HMs in the ecosystem components is mainly affected by industrial activities in the area. Considering the slightly different distribution pattern of the studied HMs, especially Pb, in the soil and plant samples, it seems that polluted emissions arising from Zinc Town are responsible for elevated HMs contents in natural plants in the study area. However, analyzing HMs contents in the air may provide some useful information about different sources of plant community pollution. Applied geostatistical method has managed to reveal the HMs distribution pattern in the endemic plants grown on polluted soils.

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