GEOSTATISTICAL METHODS AND TURNING-BANDS SIMULATION ASSESSMENT OF HEAVY METALS POLLUTION OF SOILS IN DĄBROWA GÓRNICZA AREA

Introduction

An assessment, by means of geostatistical methods, of surface soil pollution with six heavy metals (lead Pb, cadmium Cd, zinc Cd, copper Cu, nickel Ni and chromium Cr) was carried out using data from the monitoring of soils in selected areas of the Silesian province in the period 1982-1991. The pollution of the surface soil layers with some heavy metals (Cd, Pb, Zn) in different areas of the Upper Silesia has been the subject of various publications e.g. [4-5]. The extent and degree of pollution with these metals and the probability that the Cd, Pb and Zn content values exceeded the soil pollution limits (fixed by IUNG – Institute of Soil Cultivation, Fertilising and Pedology in Puławy [1]) in the areas of Bytom, Będzin [4], Dąbrowa Górnicza [6], Tarnowskie Góry and Piekar Śląskie [5] have been investigated.

The use of linear geostatistics methods combined with turning-bends simulation made it possible to identify fully the extent and degree of pollution in the studied Dąbrowa Górnicza area. The heavy metals variation assessment results – estimated averages $Z^*$, standard estimation deviations $\sigma_2$ and the simulated values – were used to determine the proper sample size and distribution for the farmland area where environmental monitoring is to be reinstituted.

Subject and range of survey

The subject of the statistical analysis was the variation of the six heavy metals content in the surface layer of soils in the Dąbrowa Górnicza area. These are exclusively farmlands. Only the fertile layer down to depth of 20 cm was investigated. The soil layer in the area was sampled by the Institute of Environmental Research and Control in Katowice in 1984. Different kinds of soils occur in the considered area: lightly loamy and loamy sands; loose sands; light, medium and heavy rendzina soils; lowmoor peats (sparsely).

The deposits which make up the soil have mostly clayey character. Metal determinations made on 152 samples provided a basis for the analysis of the variation of the Pb, Cd, Zn, Cu, Ni and Cr content. The pH of the soil layer ranged from 4.70 to 7.50 with the average of 6.90 (pH$_{H_2O}$) and from 4 to 7.5 with the average of 6.06 (pH$_{KCl}$).
Estimation of basic heavy metals statistics

The average Pb content reaches 104.14 mg/kg, exceeding slightly the admissible value in heavy soils (Table 1). This suggests that the soils are lightly polluted with this metal. The degree of soil pollution according to the IUNG guidelines [1]. But in some places of the analysed area maximum contents, as high as 900 mg/kg, were found indicating local zones of higher pollution corresponding to the 3rd degree of soil pollution [1,7]. Here and there, quite high values of standard deviation S, and thus high values of variation coefficient V, occur.

| Chemical Element | Average \( \bar{X} \) [mg/kg] | Variance \( S^2 \) [mg/kg]² | Standard deviation S [mg/kg] | Variation coefficient V [%] | \( X_{\min} \) [mg/kg] | \( X_{\max} \) [mg/kg] | Threshold values in soils light soils [mg/kg] | heavy soils [mg/kg] |
|------------------|---------------------------|--------------------------|---------------------------|-------------------------|----------------|----------------|--------------------------------|----------------|----------------|
| Lead Pb          | 104.14                    | 6679.36                  | 81.73                     | 78.48                   | 10.00          | 890.00         | 50                            | 100             |
| Cadmium Cd       | 4.11                      | 5.05                     | 2.25                      | 54.63                   | 1.00           | 17.00          | 3                             | 3               |
| Zinc Zn          | 420.58                    | 239437.43                | 489.32                    | 116.34                  | 37.00          | 3820.0         | 200                           | 300             |
| Chromium Cr      | 8.00                      | 94.33                    | 9.71                      | 121.40                  | 0.00           | 49.00          | 20                            | 60              |
| Nickel Ni        | 9.71                      | 32.28                    | 5.68                      | 58.51                   | 1.00           | 47.00          | 30                            | 100             |
| Copper Cu        | 11.16                     | 24.41                    | 4.94                      | 44.28                   | 3.00           | 39.00          | 50                            | 100             |

Similar observations concerning the average content were made for cadmium Cd (Table 1). The average Cd content (4.11 mg/kg) exceeds slightly the allowable standard but the S values for Cd are substantially lower than for Pb and so is coefficient. V. The soils are polluted to the 3rd degree and locally to the 5th degree (the highest Cd content is 17 mg/kg [1,7]).

The average Zn content (420.58 mg/kg) exceeds the threshold limit values for soils (Table 1). The very high values of the deviation S and coefficient V statistics indicate that the Zn content is highly diversified over a considerable part of the area. Although on average the 2nd degree predominates, soils heavily polluted with zinc – the 5th degree (3820.0 mg/kg [1,2]) – occur locally.

The averages for the other metals: 11.16 mg/kg for Cu, 9.71 mg/kg for Ni and 8.00 mg/kg for Cr are much below the current standards (Table 1) [1,7].

One notices immediately the very high values of coefficient V and deviation S for chromium Cr, which are indicative of great variability in the Cr content, and the much lower values for nickel Ni and copper Cu (Table 1).
The average Ni and Cu values indicate unpolluted soils with natural heavy metals contents and the maximum values signal slightly (2\textsuperscript{nd} degree) polluted soils occurring locally (Table 1) [1,7].

**Research methods**

For the geostatistical survey of the pollution of soils with heavy metals methods of linear, stationary geostatistics (variogram functions and ordinary point kriging [2,8]) and turning-bands simulation [3] were used.

Kriging was applied to determine the “best” – from the point of view of the minimisation of estimation variance (kriging standard deviation) – local heavy metal content estimates (estimated averages $Z^*$) in the unsampled places, i.e. in the kriging net’s nodes. The analysed variable (a heavy metal content) is interpolated as a moving average calculated on the basis of sampling data with attached weights. Then the closeness of estimation on the local scale is determined.

By applying a kriging procedure based on the variogram function we obtain a single numerical pollution model consisting of estimated kriging areas (averages $Z^*$) for the particular heavy metals and local estimation closeness areas (standard kriging deviation $\sigma_k$).

The turning-bands simulation method is described in detail in [3]. This method is used for the very accurate mapping of the spatial variation of pollution, making it possible to generate multiple stochastic images (multiple realisations of Gauss random function $Z_0(x)$). As a result many alternative numerical models of the spatial variation of pollution are obtained.

The geostatistical analysis began with the calculation of the empirical isotropic and directional variograms of the six heavy metals content values. Then their traces were approximated by theoretical functions.

The determined variogram model parameters (range of influence $a$, sill variance $C$, nugget effect $C_0$) were used for kriging in the second stage of the analysis. The analysed Dąbrowa Górnicza area, within which the environmental data were distributed, was covered with a grid of 250 m (axis x) x 500 m (axis y) elementary fields [6]. The size of elementary fields was 48 along axis x and 63 along axis y.

Raster maps of estimated heavy metal content averages $Z^*$ with the associated maps of standard kriging deviation values $\sigma_k$ and spatial pollution block diagrams for the particular heavy metals were produced as a result of the kriging calculations. The block diagrams are presented in perspective view (longitude – 44°, latitude – 24°).

Then the turning-bands simulation method was tested for the Pb content. The original Pb pollution data were converted into a set of data, representing a Gaussian variable, by applying the Gaussian anamorphosis function. An empirical isotropic variogram of the converted data was calculated and its trace was approximated by a model. 100 bands were used for the turning-bands calculations. The results of 20 simulations were reconverted from the Gaussian scale to the original scale. A result map, representing the average of the 20 simulations, a simulation variance map and maps of the lowest and highest values simulated for each net node were plotted. Furthermore maps of the probability of exceeding the Pb content pollution thresholds (250 mg/kg, 300 mg/kg, 400 mg/kg and 800 mg/kg),
based on the simulation set, and maps of averages above the tested thresholds, simulated for each net node, were produced.

The geostatistical survey was carried out by means of the geostatistical software contained in the SATIS package (version SATIS 3.1 November 1997) by GEOVARIANCES (Avon Cedex, France) – a firm associated with the Centre of Geostatistics in Fontainebleau [9]. The results of the structural analysis of the six heavy metals are discussed in detail in [6]. General remarks concerning them can be found in the present paper’s Recapitulation. Below, because of the paper’s narrow confines, only the results for lead are presented.

**Variogram function results**

In general, the empirical isotropic Pb content variogram shows moderate variation in the value of function \( y(h) \), except for the graph’s first point which is statistically insignificant (\( n=16 \))(Fig.1). The graph’s shape was approximated by a spherical model and the values of the model’s parameters: sill variance (\( C \)) and range of influence (\( a \)) are specified on the figure’s right side.

The traces of the empirical variograms, analysed in four direction (\( 0^\circ, 45^\circ, 90^\circ, 135^\circ \)), show that there is no significant variation in the Pb content along the particular test lines [6]. Only in the case of two variograms calculated along lines \( 45^\circ \) and \( 90^\circ \) one can notice a sharp increase in \( y(h) \) values over a very short distance.

![Fig. 1. Empirical isotropic variogram of lead content (Pb) [mg/kg]^2 with fitted theoretical model](image-url)

**Kriging estimation results**

The Pb content raster map shows four subareas of heavily Pb-polluted soils (maxima: 890 mg/kg, 670 mg/kg, 450 mg/kg, 230 mg/kg), each with one pollution centre.
(890 mg/kg) (Fig. 2). The forecasted Pb content values are shown around the centres, indicating the extent of the danger zones. Estimated averages $Z^*$ within the zones’ boundaries range from 105 to 167 mgPb/kg and from 123 to 136 mgPb/kg. Also a few much less polluted subareas occur with a particularly extensive 80-90 mg/kg zone running around the pollution maximum. The lowest estimation standard deviations $\delta_k$ from averages $Z^*$ (~88 mg/kg) became pronounced in two places where the sampling was relatively dense (Fig 3).

![Fig. 2. Raster map of estimated averages $Z^*$ of lead contents (Pb) in soils](image)

![Fig. 3. Raster map of standard deviation of estimation $\sigma_k$ for estimated averages $Z^*$ of lead contents](image)

**Turning-bands simulation results**

An empirical isotropic variogram was calculated using the Pb data converted to Gaussian variables [9]. The variogram’s trace was approximated by a model consisting of three structures: the nugget effect, a linear model and a spherical model (Fig. 4). Taking this model’s parameters into account, calculations were done using the turning-bands method and 20 different (conditional) simulation images of the soils’ lead pollution were
obtained (Fig 5).

Fig. 4 Empirical isotropic variogramic for transformed lead Pb contents (on Gaussian variables) with fitted theoretical model.

On the map one can discern four centres of heavier pollution (which is heaviest in the central part of the Dąbrowa Górnicza area). The simulated Pb content ranges from 150 to 250 mg/kg. The boundaries of the centres are even more distinct on the map of the highest simulated values [6]. The range of simulated values is wider in this case: 50-90 mg/kg. But on the map of the lowest simulated values [6] the highest Pb contents are associated with an interval of much lower values: 50-90 mg/kg. The map of simulation variance [6] shows interesting data. The highest simulation variance values are found within zones of higher simulated Pb content values but the outlines of such zones indicate that they are much smaller in comparison with the larger zones visible on the map showing the average of the 20 simulations (Fig. 5).
Fig. 5. Simulation map of soils pollution by lead Pb (averaged on basis of 20 simulations)

Generally, the simulation Pb pollution zones are more extensive in comparison with the Pb content area in the case of estimation by ordinary point kriging (Fig. 2).

Fig. 6. Map of averaged simulated values (for each net node), which are above accepted threshold (250 mgPb/kg) of lead contents
Fig. 7. Map of averaged simulated values (for each net node), which are above accepted threshold (400 mg Pb/kg) of lead contents

Figs 6-7 show maps of average simulated values for the particular grid nodes for two Pb content thresholds. Pb pollution areas are largest for the 250 mg/kg and 300 mg/kg thresholds [6]. They become severely reduced, to small fields or single blocks, for the threshold of 800 mg/kg [6]. The highest probability values (20-30 mg/kg) occur for the threshold of 250 mg/kg (Fig. 8).

Fig. 8. Map of probability (for each net node) of exceeding of threshold values (250 mg Pb/kg) of lead contents

Pollution centres (four distinct centres), decreasing progressively to the size of single blocks or small concentrations of blocks, can be seen on the probability maps for the three consecutive thresholds (Fig. 9), [6]. If 400 mg/kg and 800 mg/kg thresholds are as-
sumed, the probability values are low, ranging from 9 to 19. For the latter threshold the blocks with higher probability values are much smaller than those for the threshold of 400 mg (Fig. 9). For lower pollution thresholds (250 mg/kg, 300 mg/kg), the probability that they will be exceeded is higher, ranging from 15 to 35 (Fig. 8).

![Map of probability](image)

**Fig. 9.** Map of probability (for each net node) of exceeding of threshold values (400 mgPb/kg) of lead contents

**Recapitulation of results and conclusions**

The following observations can be made on the basis of the results of the estimation done by means of geostatistical methods for the six heavy metals and by the turning-bands simulation method for one metal.

The anisotropy of the variation in the six heavy metals content was strongest in the case of copper, less pronounced for chromium and zinc and least pronounced for nickel [6]. The strongest anisotropy associated with the upward trend in the value of variogram function $\gamma(h)$ occurred along directions $0^\circ$ and $45^\circ$ for Cu, along directions $135^\circ$ and $0^\circ$ for Cr, along directions $135^\circ$ and $90^\circ$ for Cd, along direction $45^\circ$ for Zn and along direction $135^\circ$ for Ni. No anisotropy in the variation was found in the case of Pb. The observed character of the variation of $\gamma(h)$ values in the particular roses of the variograms may be linked to the susceptibility of some metals to displacement and their mobility in the soil environment (mainly Cd, Zn and Cu). Certain trends in the heavy metals content variation may also reflect wind activity: the raising of metalliferous dust on dumps and the directions in which it is transported.

The pictures of the forecasted Pb, Cd and Zn content for the Dąbrowa Górnicza area clearly coincide as regards the boundaries of the surface area polluted with these metals. Two larger concentration intensification areas and one smaller centre are similar in their spatial distribution for three elements: Pb, Cd and Zn. The rest of the increased heavy metal content zone shows conformity in the case of Pb and Zn. One should notice a subarea of a higher Cd content there.

As regards the other three metals: Cu, Ni and Cr, the regularity in the spatial dis-
tribution of increased metal content zones is not so distinct as for Pb, Cd and Zn. A comparison of Ni and Cr areas shows that there is one intense pollution centre and one centre of less intense pollution. A subarea of a slightly increased copper content coincides with the spatial distribution of only some of the Pb, Cd and Zn anomalies. The second subarea of a higher Cu content coincides roughly with the estimation pictures for Ni and Cr and partly for Cd.

The Zn content block diagrams stands out from the six block diagrams of soil pollution with the heavy metals because of the highest peak value. A considerable jump in the Cd value can be noticed in the estimated Cd content area. The smallest jump is recorded for the Pb content. The multilayer block diagram shows clearly a similar spatial arrangement of the peaks of the three metals. The pollution maxima are situated close to each other.

The block diagrams for Cu, Ni and Cr show that the soils are much less polluted with these metals [6]. The highest is the Ni peak, the Cu peaks are lower and the Cr peaks are still lower. Such moderate pollution centres for Cu, Ni and Cr are visible on the multilayer block diagram. The spatial distribution of these centres, apart from a slight shift, is on the whole similar to that of the Cd, Pb and Zn peaks.

All the six estimation pictures, including the maps of estimated averages, the maps of standard kriging deviation and the estimation and simulation block diagrams, were taken into account [6] in order to determine the boundaries of the area in which environmental monitoring should be reconstituted. The dimensions of the area, the uniform sampling and the sample sizes (N= 182 and N =266) were adopted on the basis of the forecasted Pb and Cd content values (averages Z'), indicating places where danger of pollution with these elements could occur, supported by maps and block diagrams of the conditional simulation of Pb pollution, statistical simulation maps and maps of the probability of exceeding the pollution threshold.

The high simulated Pb content values and the large simulation variance, the picture obtained from the largest realisations and the map of the probability of exceeding the pollution threshold of 250 Pb/kg justify further the proposed uniform sampling of the soils in the Dąbrowa Górnicza area.

The pictures of the conditional Pb content simulation show much more accurately the variation in the Pb pollution than the estimation maps obtained exclusively by kriging.

In the further studies of the character of the heavy metals pollution the heavy metals concentrations of anthropogenic origin should be identified precisely and separated from the natural concentrations of these metals.

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