Review

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Geostatistical modelling as an assessment tool of soil pollution based on deposition from atmospheric air

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ABSTRACT: Research in the scope of geostatistics is applied in many fields of study, including soils and atmospheric air. Geostatistics can constitute a tool for interpretation of results of research on the natural environment. For example, the semivariogram permits the estimation and analysis of the variability structure of selected phenomena. Stochastic interpolation techniques allow for obtaining the value of the studied variable with no necessity of field studies with consideration of a dense network of measurements owing to information obtained from other research. Research on the quality of atmospheric air conducted by the European Environmental Agency (EEA) presents the state and forecasts of atmospheric air in particular European countries based on a low number of measurement points throughout Europe. In Poland, only four measurement stations function in the scope of the European Monitoring and Evaluation Programme (EMEP). An important aspect in geostatistical modelling is later assessment of uncertainty as to the estimated value of the analysed variable. Results of such an assessment are usually presented in the form of a map of probability of exceeding critical values. The last stage of geostatistical modelling usually involves stochastic simulations performed by means of an increasingly broad range of available algorithms. The assessment of generated effects combined with expert knowledge permits e.g., the identification of polluted areas. The quality of atmospheric air affects the degree of soil pollution (primarily as a result of the phenomenon of dry and/or wet deposition). Due to this, it is necessary to analyse such impact with consideration of all environmental and geochemical conditions. The application of the generally available data permits the estimation of the degree of soil pollution with no necessity of sampling in a given place, or performing costly laboratory analyses. The aim of the study was the presentation of the commonly used geostatistical methods and good practices in geostatistical modelling for the assessment of soil contamination by heavy metals based on deposition data from atmospheric air. The work was divided into two parts: (i) geostatistical modelling, presenting individual stages of the use of various tools and techniques, as well as (ii) kriging and cokriging interpolation methods used as a tool to integrate spatial data from different sets. The workflow in geostatistical modelling in environmental sciences using existing data sets was proposed.

Key words: geostatistics, geostatistical modelling, soil, atmospheric air, deposition of pollutants

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1. INTRODUCTION

Geostatistics and Geographic Information Systems (GIS) are basic tools used for the georeferential analysis of spatial information, and investigation of spatial variability at various scales (Chang et al., 1999). Geostatistics introduce new tools to many scientific disciplines, permitting interpretation of data in space, and obtaining information necessary in the decision making process. Geostatistical modelling involves the application of numerical methods for particular features of spatial attributes for the purpose of generating the probability model (Olea, 2009).

Geostatistical methods are optimal estimation methods, if two conditions are met, i.e., data show normal distribution, and are stationary. Considerable deviations from normality or stationarity may cause problems with interpretation of results. Due to this, the first stage in the analysis of spatial data should be the preparation of a data histogram (Bohling, 2005).

The histogram is the most popular way of presentation of the empirical distribution of a parameter (Fig. 1). It is primarily applied when analysing high amounts of data differing to a low degree. If a given value is a total or average value of many

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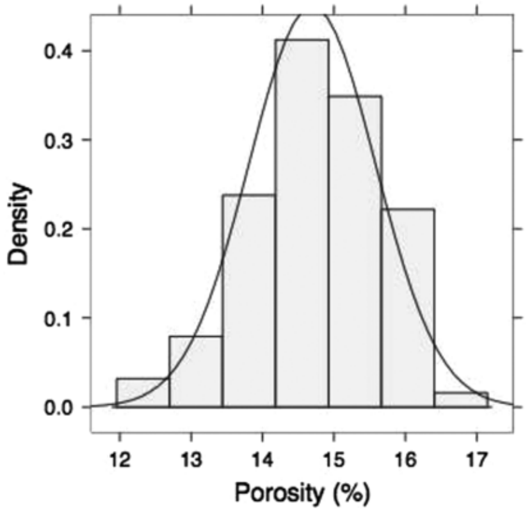

Fig. 1. Spatial data histogram – normal distribution (Bohling, 2005).

inconsiderable random factors, irrespective of the distribution of each of the factors, its distribution will be approximate to normal.

The term of stationarity of a variable means that its values do not change with time, and the mean and variance of a given variable should not differ considerably in space. The assumption of stationarity of variables in a model is necessary in the case of introduction of distributions of typical test statistics applied in testing hypotheses. Results of many studies show that when a model involves non-stationary variables, the asymptotic distributions of test statistics are non-standard. This may lead to inaccurate results of statistical inference. During research on air quality and degree of soil pollution, the variability of natural conditions and anthropogenic activity may cause disturbances in measurements, leading to weak stationarity. This should be considered at the further stages of geostatistical modelling among others in the preparation of a semivariogram and interpolation of spatial data by means of the kriging or cokriging method (Brenning, 2001). The determination of the spatial distribution of regionalised variable Z(x) or its moments such as among others semivariance requires obtaining many realisations of $z_1(x)$, $z_2(x)$, … $z_n(x)$. In research on the natural environment, however, only a limited number of realisations of a phenomenon with location x is often available. Such observed distributions are frequently unique, e.g., distribution of soil pollution. In order to obtain the relevant number of realisations of the analysed regionalised variable, it is assumed that the studied phenomenon in a certain area is repeated in space (Zawadzki, 2011). Results of research on stationarity are best

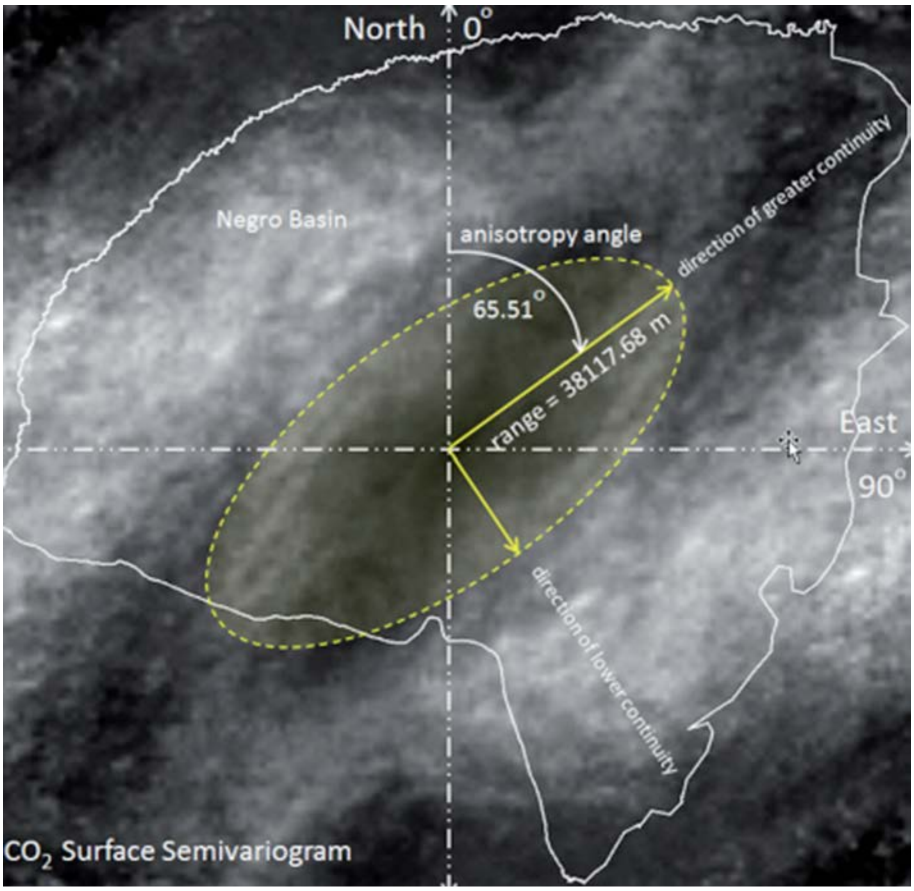

Fig. 2. Phenomenon of anisotropy during CO_2 emission (Felgueiras et al., 2014).

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verified by means of one of popular tests such as Kwiatkowski Phillips, Schmidt, and Shin (KPSS), or Dickey-Fuller test (ADF) (Xiao, Lima, 2007).

During geostatistical estimation by means of interpolation methods, the dependency of the phenomenon on direction is interpreted – so-called anisotropy (Fig. 2), or its lack – so-called isotropy. The emission of pollutants to atmospheric air, as well as their deposition in the ground, is determined by many factors which usually cause anisotropy of the analysed phenomena (Felgueiras et al., 2014).

Before the commencement of the geostatistical analysis of content of heavy metals in the soil, so-called natural contents of the analysed trace elements in the soil should be determined, and the degree of pollution from anthropogenic sources should be estimated (Kabata-Pendias and Pendias, 1993). Several methods of determination of the "background" for trace elements in soils exist, permitting obtaining values approximate to their natural occurrence (Kabata-Pendias, 1991). Adopting the value of the geometric mean and calculated range of natural contents permits high level of approximation of determination of so-called geochemical background, necessary for further analyses and further interpretation of results. Geochemical maps of selected trace elements are of fundamental importance for the assessment of changes in the natural environment caused by anthropogenic activity (Sprovieri et al., 2007).

2. GEOSTATISTICAL MODELLING

Geostatistics are a group of statistical tools considering the spatial and temporal location of data in their analysis. They

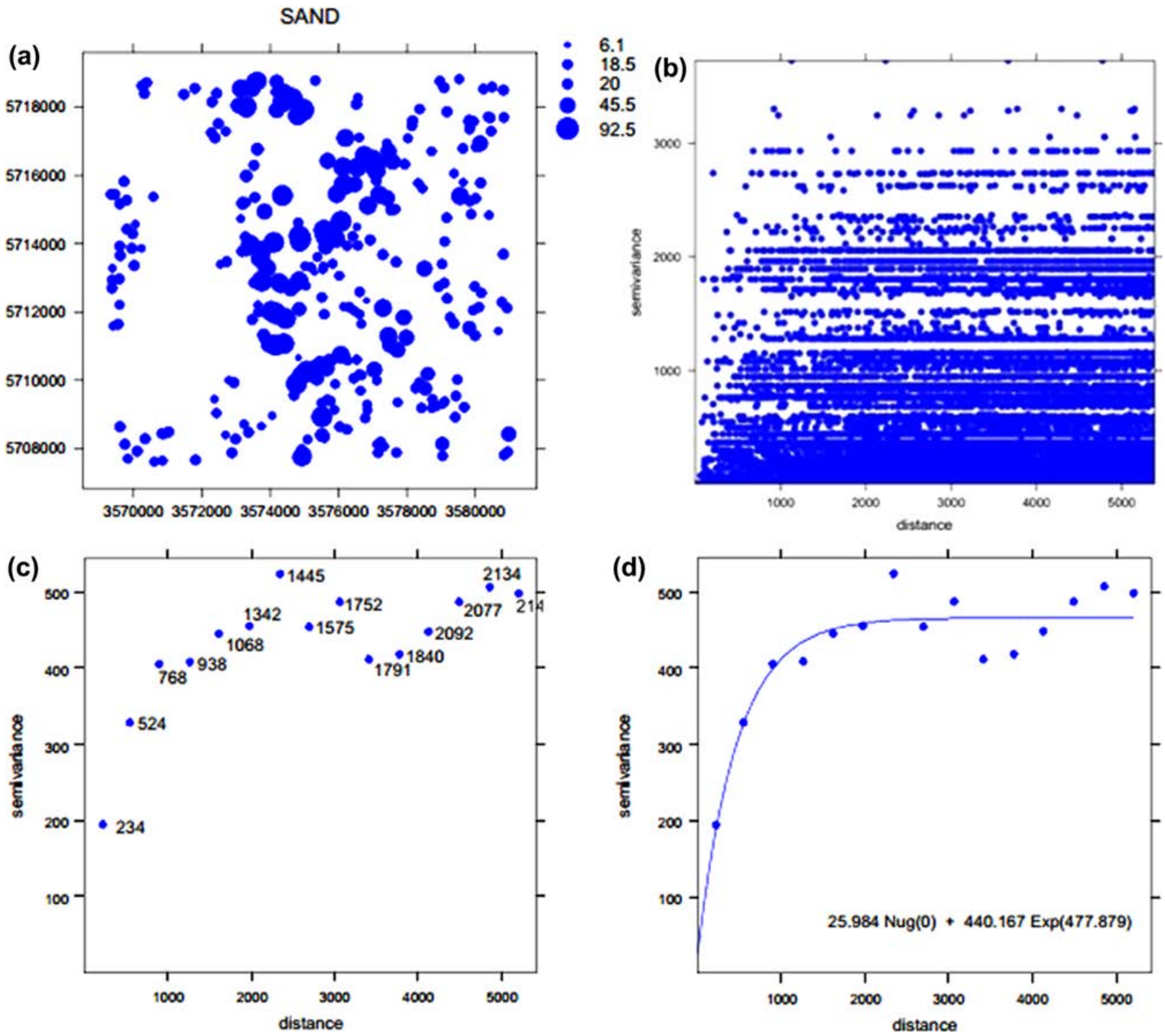

Fig. 3. Stages of variogram modelling: (a) location of points, (b) cloud of 44850 pairs of points, (c) semivariance with lag h = 300 m, and (d) final variogram graph (Hengl, 2007).

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assume that elements of the analysed group located near one another in space or time show more similarities than those located at a certain distance. Geostatistical analysis permits first of all the determination of the spatial variability of the studied elements, i.e., how the probability of occurrence of a phenomenon decreases or increases with an increase in distance between them. The semivariance function is the most frequently applied in analyses of atmospheric air and soils (Hengl, 2007). The semivariance function is used for analysing temporal and spatial correlations, and is expressed as half of squared differences between paired data (1):

$$\gamma = \frac{1}{2N(h)}\sum_{\alpha=1}^{N(h)}[z(u_\alpha - z(u_\alpha + h)]^2, \quad (1)$$

where, N(h) – number of pairs of points, z(u) – regionalised variable, h – lag vector.

The determination of a moment such as semivariance requires the presentation of the location of particular measurements, and then development of a spread plot presenting all pairs of equally distant points along the specified direction (the first value is Z(x), second Z(x + h)). The next steps involve the development of the semivariance graph with determined lag h (so-called lag vector) and final variogram graph (Fig. 3).

The standard variogram model can be characterised by means of three basic parameters: (i) range of impact, (ii) nugget effect, (iii) sill. The range of impact determines the range of the variogram above which no spatial correlations exist. The nugget effect specifies the internal credibility of data, comprising among others measurement errors. The sill is the value of variances of all measurements (Fig. 4).

The range of the variogram is frequently approximate to the mean value of physical spatial variabilities of regionalised variable Z(x). In order for the variogram modelling to be considered credible, the sill should meet the following three conditions (Barnes, 2008):

1. The data are evenly distributed in a given study area.

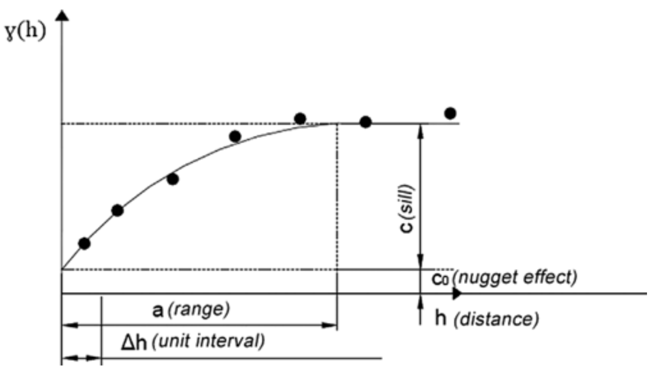

Fig. 4. Graph of dependence of semivariance on distance between measurement points h.

2. No significant tendency occurs which affects values throughout the study area.

3. The dimension of the studied space (max-min) is at least three times larger than the value of the range of the variogram.

The nugget effect occurring on axis γ (h) usually results from two factors:

1. Variability caused by measurement error.

2. Variability of regionalised variable Z(x) occurring at a scale smaller than the distance selected during variogram modelling.

In addition to the semivariogram, geostatistics also employ the following measures such as (2,3,4) (Cressie, 1993):

• semirodogram, expressed as:

$$\gamma_R(h) = \frac{1}{2N}\sum_{i=1}^{N}\sqrt{|Z(x_i) - Z(x_i + h)|}, \quad (2)$$

where N – number of pairs of points, Z(x) – regionalised variable, h – lag vector,

• semimadogram, expressed as:

$$\gamma_M(h) = \frac{1}{2N}\sum_{i=1}^{N}|Z(x_i) - Z(x_i + h)|, \quad (3)$$

where N – number of pairs of points, Z(x) – regionalised variable, h – lag vector,

• semivariance of logarithm values, expressed as:

$$\gamma_R(h) = \frac{1}{2N}\sum_{i=1}^{N}[\ln Z(x_i) - \ln Z(x_i + h)]^2, \quad (4)$$

where N – number of pairs of points, Z(x) – regionalised variable, ln – natural logarithm, h – lag vector. The measures are more resistant to outlier observations, but are rarely used in kriging interpolations.

Variogram modelling is a very important stage of geostatistical analysis. The knowledge of semivariable values for any distances (vector h) is necessary in further interpolation by means of the kriging method. In variogram modelling, the variance of regionalised variable Var{Z(x)} cannot be negative, and the semivariance function necessary for the development of the variogram model should be in accordance with the following Formula (5):

$$\gamma(|h|) = \sum_{i=1}^{N} w_i \gamma_i(|h|), \quad (5)$$

where N – number of pairs of points, w_i – combination weights (must be non-negative), γ_i – semivariance values. The most popular variogram models are presented in Figure 5.

The choice of the model depends on the spatial variability of the analysed natural phenomenon. This is particularly important in the analysis of the quality of atmospheric air and soils,

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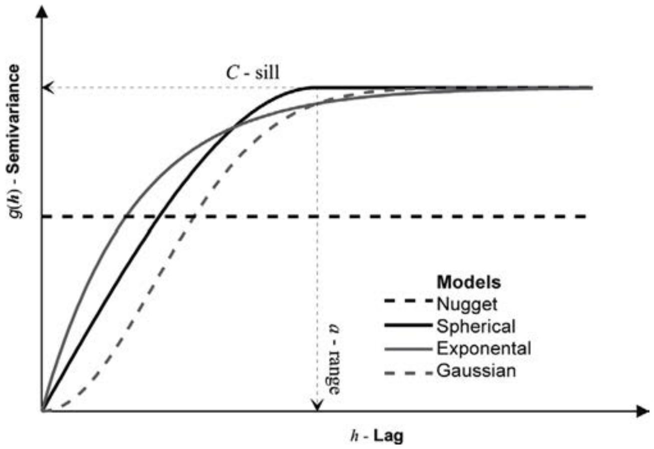

Fig. 5. The most popular variogram models (Stach, 2007).

because they are strongly variable in space (Biswes and Si, 2013). The selection of the appropriate model of data analysis permits the assessment of the simulation, and determination of uncertainty of results (Papritz and Dubois, 1999). The semivariogram should be possibly regular, and points belonging to it distributed possibly close to one another. Large distances between points suggest high spread of data (high variance value), which is frequently observed in studies on the quality of atmospheric air, when measurement points are relatively scarce over a large area. Moreover, the semivariogram should be analysed for vector $h \approx 0$, and for possibly large vectors h.

The shape of semivariance frequently changes with the change in direction of certain impact. This is related to the anisotropy of a given phenomenon. In studies concerning the degree of atmospheric air pollution in smaller areas (local scale), the anisotropy phenomenon is observed, among others related to the wind rose, e.g., in Poland westerly winds are dominant, determining transport of pollutants from west to east. In that case, variogram modelling should employ anisotropic semivariance. In this case, both axes of anisotropy should be determined, i.e., axis with the greatest continuity of the phenomenon, and axis with the lowest continuity of the phenomenon. Both of the axes develop a directional ellipse permitting finding a transformation reducing all directional semivariances to one model (Webster, Oliver, 2007).

A phase of analysis of the data, often generated over 50% of errors in geostatistical modelling (Zawadzki, 2011) and for a limited number of data such as is in the deposition of atmospheric air even more. The results of numerous studies indicate that the deposition of pollutants can provide 40–70% of the total accumulation of contaminants in the soil, depending on factors such as geographic location and quantity of emissions in an area (Nielsen, 1984; Harrison and Laxen, 1981; Fowler et al., 2004; Brunner and Rechberger, 2016). Therefore the stage of data analysis is extremely important for the further procedure interpolation and interpretation of research results.

3. INTERPOLATION BY MEANS OF THE KRIGING AND COKRIGING METHOD

One of the best methods of point estimation is kriging, permitting obtaining the best linear unbiased estimators (BLUE). BLUE can be characterised as follows: (i) it is linear because its estimates are a linear combination of available data, (ii) it is unbiased because its objective is to obtain the average of errors equalling zero (iii) it is the best due to the minimisation of error variance (Srinivasan et al., 2010).

The standard version of kriging is called ordinary kriging (OK). It is expressed as the following Formula (6):

$$Z(s) = \mu + \varepsilon'(s)\,, \tag{6}$$

where Z – variable of interest with deterministic distribution, μ – trend (s), ε – error autocorrelation, s – specifies location as specific coordinates x (length) and y (width).

All deterministic interpolation algorithms (inverse distance squared, splines, radial basis functions, triangulation, natural neighbour) estimate values of the studied phenomenon considering location, and decrease weight with an increase in distance between measurement points (Kwiatkowska-Malina and Borkowski, 2015). Kriging ascribes values in accordance with a model based on the theory of random functions in which bias and error variance are calculated, and then weights are selected for values obtained during the measurement, minimising error variance (Vargas-Guzman and Jim Yeh, 1999). This is particularly important in research on the quality of atmospheric air and soils, where distances between measurement points can amount to several tens, and sometimes several hundred kilometres at the regional or national scale.

In the case of occurrence of a trend in data (e.g., wind rose affecting spread of pollutants), the best estimation method is the universal kriging method (UK), also called kriging with a trend (Bohling, 2005). The trend model is considered in interpolation, and expressed with the following Formula (7):

$$m(u) = m(x, y) = a_0 + a_1 x + a_2 y\,. \tag{7}$$

All known kriging methods are popular and commonly applied among others in research on soils, although due to the lack of data, the majority of them may make estimation of soil properties difficult. Due to this, the kriging method is combined with regression analysis (regressing kriging, RK). Results of studies conducted so far suggest that RK has potential for considerable improvement of the accuracy of spatial forecasting,

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even in the case of application of a weakly correlated auxiliary variable (Meng et al., 2013). RK permits obtaining considerably better results of spatial interpolation than any other interpolation method. The basic condition for the RK method is the application of an auxiliary variable, significantly correlated with the variable of interest. Remote sensing can be useful in this case, providing abundant information on various phenomena. In addition to RK, further combinations are developed, e.g., using scripts written in application R, where in addition to basic soil variables such as granulometric composition, grain size, or content of elements, also mean annual precipitation totals are applied, as well as digital elevation model (DEM), land use, land relief, and geology of the study area (Omuto and Vargas, 2015). In such an approach, it is very important to validate the final model. For example, in the estimation of depth of a soil profile in a given area with the application of seven variables, additional model validation was performed using 148 soil depth measurement points, and the efficiency of a given method was confirmed at a level of 67% (Sarkar et al., 2013). A multithreaded approach to modelling soil properties permits a more detailed and certain interpretation of the analysed phenomena.

In environmental research, values of a given phenomenon are usually estimated based on one variable. The set of input data, however, can include more than only one main variable of interest. Additional variables correlated with the main variable can include useful information helpful in the further interpolation process. Additional variables usually contribute to a further decrease of error variance (Yalçin, 2005). The selection of auxiliary variables should be based on the rule in which they should have higher spatial resolution than the variable of interest (Robeson, 1997). An example can be the digital terrain model (DTM), used in the estimation of the spatial distribution of salinity of soils (Shahabi et al., 2016). Another example can be the atmospheric precipitation variable directly related to the deposition of pollutants from atmospheric air (Gunawardena et al., 2013), permitting obtaining a higher degree of detail of results of research on the content of pollutants in atmospheric air owing to a considerably broader set of data (more measurement points and higher data regularity). The estimation method employing auxiliary variables is the so-called cokriging method (Zare Chahouki et al., 2014). The cokriging method requires generating variograms and cross variograms between the analysed variables, e.g., content of pollutants in air and soil, atmospheric precipitation, wind rose, distance of measurements from roads, etc. (Fig. 6). To confirm the correlation between the data can create a correlation matrix (Ko et al., 2009).

The verification of results of interpolation and selection of relevant parameters requires generating a map of errors, and calculating mean relative interpolation errors (Wasilewska and Mucha, 2005) (Fig. 7).

Additionally, cross validation is worth performing, omitting the location of some samples from the analysis (points), and then estimating values using the remaining sampling sites (points) (Lyon et al., 2010). The most frequently applied method of cross validation for small data sets is the "leave one out" method, where an N-element sample is divided into N subsets including one element each (Witten and Frank, 2005).

Interpolation with a selected method (kriging, kriging with trend, cokriging) permits obtaining credible maps of spatial distribution of the phenomenon, and correct interpretation of results (Yang et al., 2004). The selection of the interpolation method should involve a comparison of the selected methods by choosing relevant criteria with the application of the cross validation procedure (Hamzehpour et al., 2013). Following the interpolation of data on the deposition of pollutants from the atmosphere the sensitivity of the applied model should be

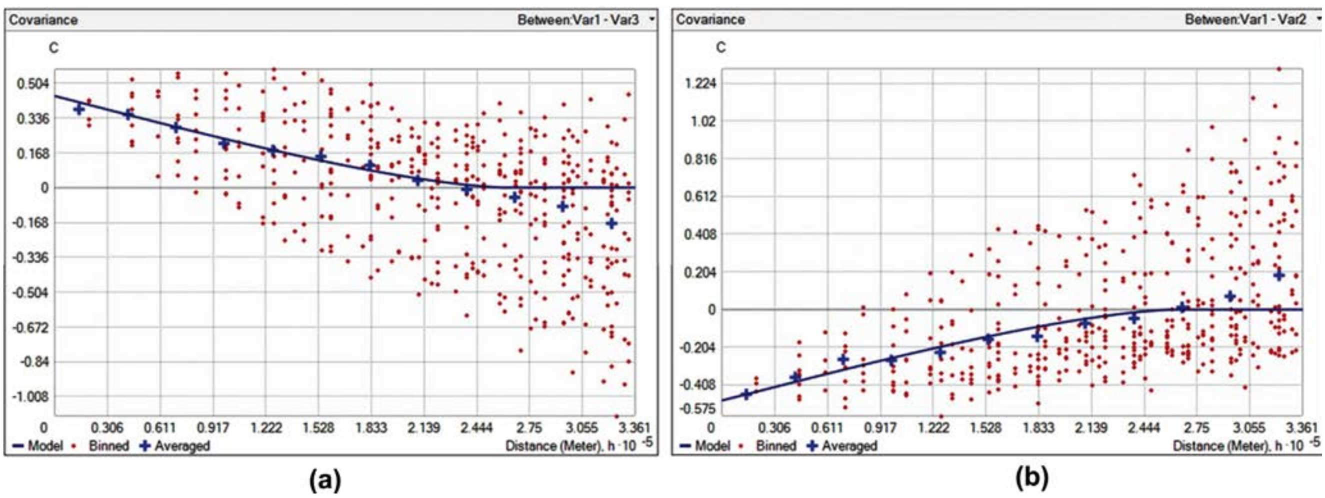

Fig. 6. Cross covariance between two variables (a) positive correlation between two variables, (b) negative correlation (Krivoruchko, 2014).

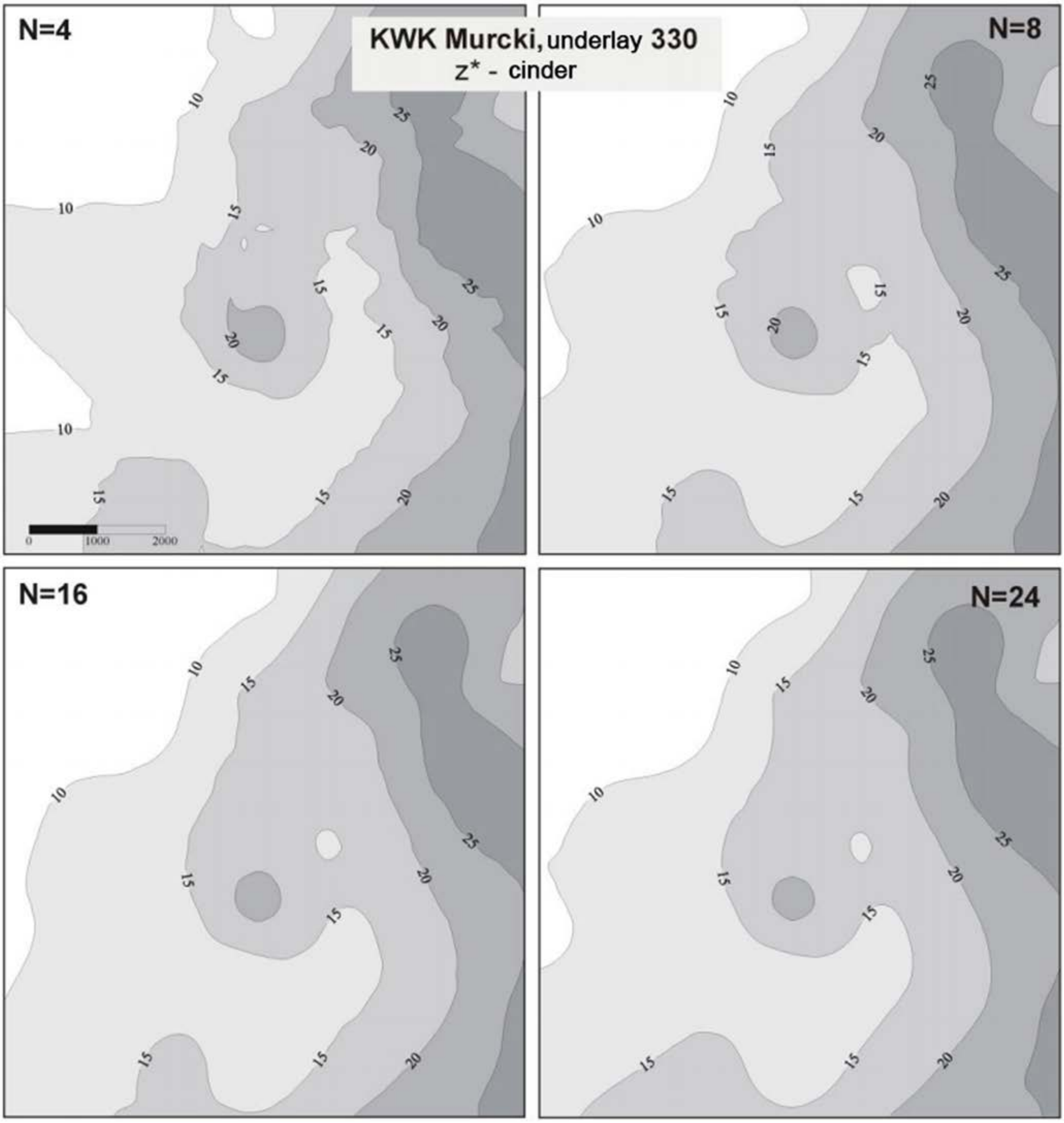

Fig. 7. Isoline maps of interpolation errors indicating uncertainty of results (Wasilewska, Mucha, 2005).

analysed (Cohen et al., 2000). Obtaining credible results requires the selection of the appropriate pattern of analysis of the studied phenomenon (Fig. 8).

4. SUMMARY

The course and selection of parameters in geostatistical modelling undoubtedly affects the quality of obtained results. This is confirmed by results of numerous studies concerning both atmospheric air and soils. A detailed analysis of available data is important. The result of the geostatistical analysis in the form of a variogram model is the most important part of the modelling. It ensures that the obtained results reflect the reality to the highest possible degree, and are subject to minimum error.

In the case of research on the quality of atmospheric air and

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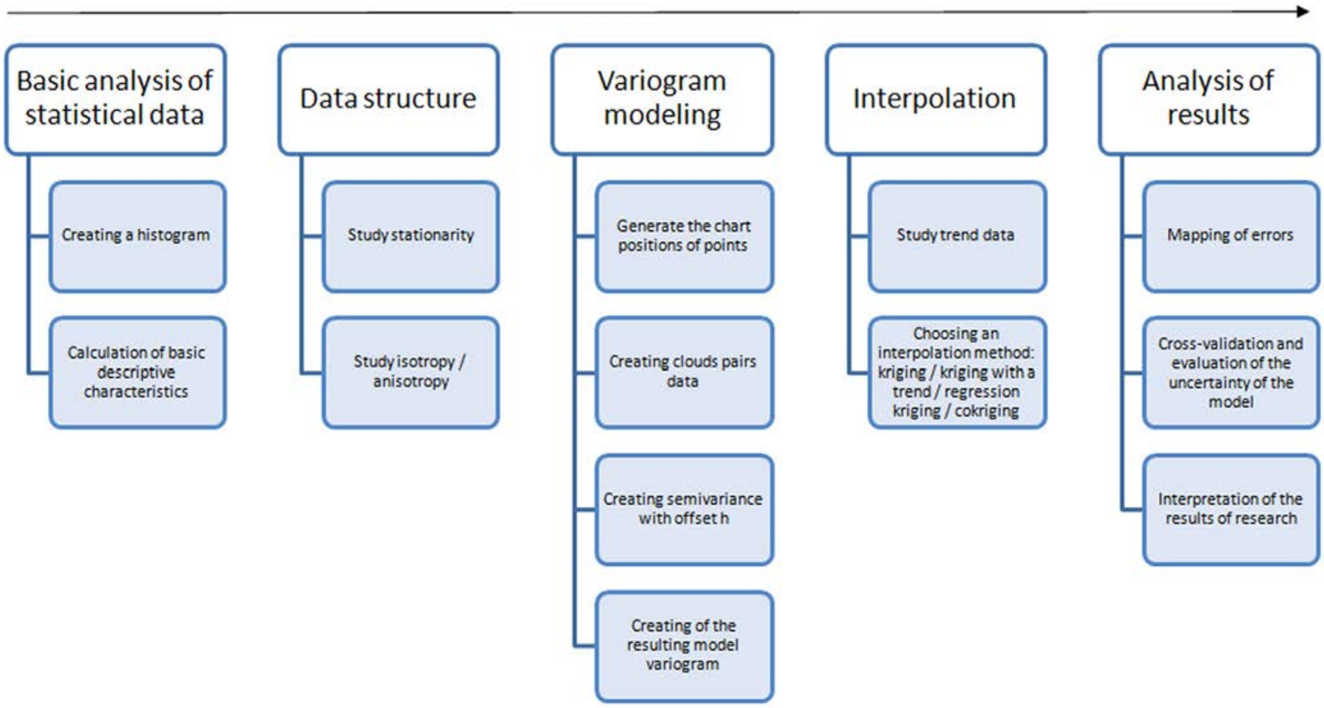

Fig. 8. Diagram of geostatistical modelling in research on the degree of pollution of atmospheric air and soils.

soils, all factors possibly affecting the character of data and further generated information should be taken into consideration. The proposed new approach to the geostatistical modelling in accordance with a scheme of procedure allows its use in the study of environmental sciences. Owing to such an approach, the drawn conclusions and interpreted phenomenon will be credible.

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