

Beata GOŁUCHOWSKA^{1*} and Radosław WRÓBEL¹

MAGNETIC SUSCEPTIBILITY SPATIAL DISTRIBUTION AS AN INDICATOR OF SOIL POLLUTION IN THE AREA OF OPOLE CITY

ROZKŁAD PRZESTRZENNY PODATNOŚCI MAGNETYCZNEJ JAKO WSKAŹNIKA ZANIECZYSZCZENIA GLEB NA TERENIE MIASTA OPOLA

Abstract: Soil magnetometry, based on topsoil magnetic susceptibility measurement, has been proven in the literature to be very useful and increasingly applicable screening technique of soils affected by anthropogenic pressure. According to the literature data, this method requires further improvement, especially in the field of magnetometric imaging techniques. The aim of the study was assessment of magnetic transformations of soils in the city of Opole (Opolskie Voivodeship) using soil magnetometry and three magnetometric data interpolation techniques (natural neighbour NN, inverse distance weighting IDW and ordinary kriging OK). The data was collected during field measurements of magnetic susceptibility, carried out in an area of 7.1 km², in a network of 124 measurement points, in the year 2015. The location of the points was determined using the Garmin GPS GPSMap 64st device, and the magnetometric measurements were performed in situ using the MS2 meter and the MS2D sensor from Bartington Instruments. The research showed high values of magnetic susceptibility and occurrence of soil magnetic anomalies in the study area. This was accompanied by geochemical transformations of soils, revealed in previous research. The results suggest that it could be caused by the long-term deposition of cement dusts, emitted in increased quantities in former times by the cement plant, which has left its footprint in the environment. When analyzing the usefulness of the magnetometric data interpolation techniques, the IDW technique best reflected the spatial distribution of magnetic susceptibility in the study area, while the technique of OK, due to the so-called smoothing effect, turned out to be less useful.

Keywords: magnetic susceptibility, soil magnetometry, ordinary kriging, cement dust

Introduction

We live in the era of sustainable development which requires special concern for the environment to leave it better for next generations. Environmental pollution, including soil contamination, particularly in urban and industrial areas, which is an effect of

¹ Faculty of Natural Sciences and Technology, University of Opole, Oleska 22, 45-052 Opole, Poland, phone: +48 77 401 60 01, fax: +48 77 401 60 30.

* Corresponding author: beska@uni.opole.pl

human activity, requires fast and simple techniques of monitoring its level and reliable techniques of its imaging to warn people about the dangers of raised level of pollution and enable dealing with emergencies. Such techniques of early warning help reduce the amount of physicochemical analyzes to the “hot spots” i.e. places of the highest environmental hazard. The most effective techniques of soil monitoring are those which do not need sampling in the field, preparation of samples in the laboratory, and then a series of elaborate analyzes with application of chemical reagents, because such analyzes are not only time-consuming and expensive, but sometimes hazardous to the health of people who carry them out e.g. sequential extraction procedures.

The search for the new *in situ* monitoring techniques which would allow for fast and accurate determination of the state of the soil, has begun in the second half of the 20th century. Some of them apply elements of the biosphere i.e. live organisms called bioindicators, inter alia algae and mosses [1, 2], the others elements of the lithosphere, such as iron-bearing minerals, which can be of geo-, pedo- or anthropogenic origin and hence may be called lithoindicators. The monitoring technique called soil magnetometry applies iron oxides, hydroxides, and sulfides, which can be easily detectable in the soil since they are particularly sensitive to the applied, external magnetic field and respond with changes in their magnetic properties. The most important are two iron oxides: magnetite and maghemite and one sulfide – pyrrhotite, since they stand out very high magnetic susceptibility, so even trace amount of these minerals in soil sample will result in increased strength of magnetic signal. Research revealed, that the above mentioned magnetic minerals can be produced by anthropogenic activity, inter alia in high-temperature processing of fossil fuels i.e. hard coal, lignite, and other materials. They are released to the environment as the so called technogenic magnetic particles (TMPs) and can serve as indicators of anthropogenic activity in integrated geophysical and geochemical environmental studies, which include the recognition of strength and range of anthropogenic impact [3–5].

The science about magnetic properties of soils, which uses geophysical research techniques, is called soil magnetism. Magnetic susceptibility is very popular, fast and simple measureable magnetic parameter to provide information on the quantity and quality of magnetic substances present in soils [6–15]. Not only has it been applied in soil pollution studies, but also in the search for renewable energy sources, in climatology and archeology [16–18].

The technique of soil magnetometry in Poland was initiated by late Professor Z. Strzyszcz [6, 7, 19] and developed by Magiera et al. [inter alia: 9–11]. So far it was successfully applied in the monitoring of areas being under the pressure of anthropogenic pollution, despite diversified sources and levels of emissions and different soil cover [6, 7, 9–11, 13, 20–22]. It is possible thanks to magnetic susceptibility data processing techniques to obtain maps of distribution of soil pollution. Maps of soil magnetic susceptibility distribution allow for the determination of spatial extent and the degree of pollution, identify magnetic anomalies, sometimes called the magnetic hot spots, the proxy of which are highest values of magnetic susceptibility. Moreover, magnetic anomalies are usually accompanied by geochemical anomalies, hence magnetic susceptibility distribution images are a useful tool for the determination of

geochemical anomalies evidenced by high content of heavy metals in the soil in relation to the values of geochemical background [12, 20, 21, 23]. High correlations between soil magnetic susceptibility values and the content of trace elements have been proven [12–14, 20, 21, 23], as well as potential health hazard caused by the contact with polluted soil [20, 23].

Soil magnetometry has been proven to be fast and easy soil screening technique. Moreover, it is cost- and time-effective, since it allows for minimize the number of geochemical measurements [9, 12, 20–23]. There is still a high interest not only in improvement of soil magnetometry procedures, but also techniques of magnetometric data imaging in different areas, that could increase the accuracy of evaluation of pollution level and extent. To determine the spatial variability of magnetic susceptibility, often different techniques are applied to interpolate and map magnetometric measurement results.

It is important to choose the most appropriate techniques of analyzing the spatial variability of magnetic susceptibility. Some authors emphasize the need for research in this field, related to the processing of magnetometric data and assessment of usefulness of selected interpolation techniques [21, 22].

To visualize magnetometric data spatial distribution both non-geostatistical and geostatistical interpolation techniques were applied [24]. The first group of interpolators (non-geostatistical) included popular nearest neighbours NN [inter alia: 9, 10, 11, 20], as well as inverse distance weighting IDW [15]. Geostatistical interpolators included various types of kriging K: ordinary kriging OK [13, 17], and more advanced indicator kriging (IK), empirical Bayesian kriging (EBK) and indicator cokriging IC to assess the probability of soil pollution in Upper Silesian Industrial Region on the basis of magnetometric data set [21, 22]. On the other hand, two interpolation techniques had vital disadvantages and proved to be not very useful in magnetometric data visualization: regression kriging (RK), which combines multiple linear regression and ordinary cokriging, and geographically weighted regression, which is a local form of linear regression used to model spatially varying relationships [22].

The aim of this study was comparison of three magnetometric data processing techniques (2 non-geostatistical and 1 geostatistical) to visualize spatial distribution of soil magnetic susceptibility (κ) on maps. The magnetometric data set was collected during field measurements performed on the soil surface in selected urban-industrial-agricultural area located in the city of Opole (Opole Voivodeship, Southern Poland). The central point of this area was old cement plant, which operates more than 100 years. Magnetometric data distribution maps were basis for the determination of soil pollution. The reason for the research was the necessity for monitoring the condition of soil environment in the area affected by the cement plant, due to the long time of its impact, using soil magnetometry – a modern and fast monitoring technique, being still under investigations.

Methods of research

Study area

The study area was located in the city of Opole (geographical coordinates: 17°55'37" E – 50°39'53" N) and included urban, industrial and agricultural land (Fig. 1).

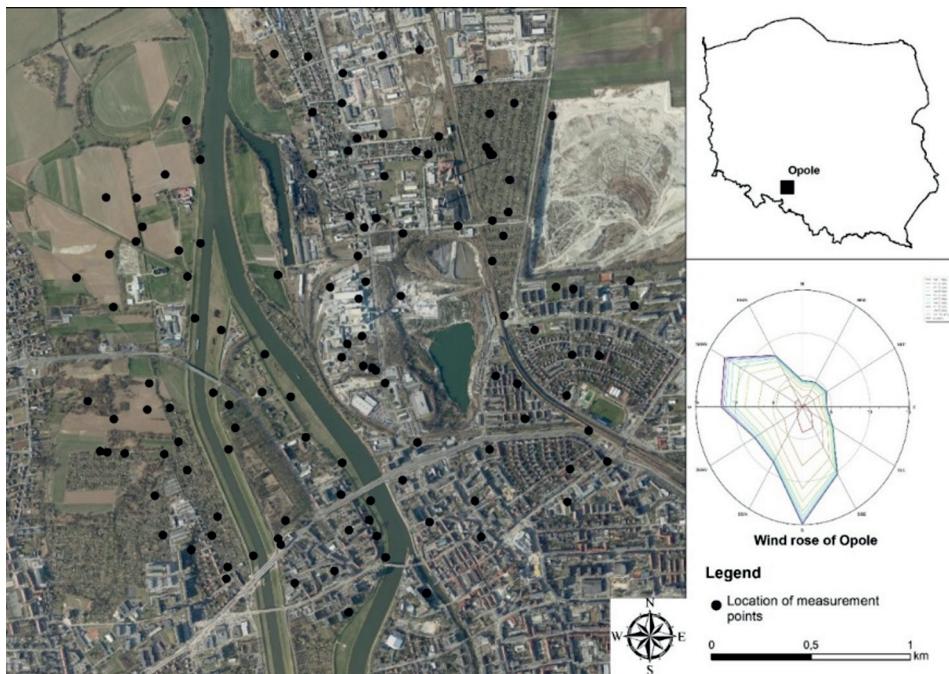


Fig. 1. Location of measurement points

Source: own research.

Opole is one of the oldest cities in Poland with its long history dating back to the 8th century and influences of different cultures: Polish, German and Czech. It is the capital of Opole Voivodeship and the region called Opole Silesia, with the total area of 14 899 km² (since the year 2017), a population of approximately 128 140 and its density 861 people per 1 km² [25]. Opole is inhabited by approximately 12 % of the total population of the voivodeship. According to the physical-geographical regionalization of Poland by Kondracki [26], the city is located in the makroregion Silesian Lowland, and within two mezoregions: the Wroclawska ice-marginal valley and Opolska Plateau, on the both banks of the Odra river, which length in the city is about 20 km. The elevation of the highest point in the city above the sea level is 176 m. One of the main characteristic features of Opole are abundant, high quality deposits of carbonate sedimentary rocks, mainly the Cretaceous marls. Therefore, since the middle of 19th century, opencast mining and cement production has dominated the landscape of Opole. In the past, the city had been considered a cement industry capital and cement contamination hotspot as a result. At the beginning of the 20th century there were 7 cement plants, of which one has remained until today.

The research was carried out in the territory of 706.86 ha (7.069 km²), in the shape of a circle of a 3 km diameter, with the cement plant in the centre of it. This plant has been operating for almost 110 years and uses its own deposits of marl, which has been extracted by opencast mining in the nearby quarries. In addition to this basic raw

material, various additives have been used for the production of cement for many years, mainly industrial waste. The fuel is pulverized hard coal with addition of municipal waste, which has been burnt in clinker kiln since the year 2014. Before the year 2000, very dust-generating wet method of clinker production was applied in 4 rotary kilns. In the years 1998–2000 the cement plant was modernized and began using the dry production method, in accordance with the BAT requirements, thanks to which the emission of dust decreased significantly [27]. Currently the clinker is fired in one rotary kiln, with a 4-stage cyclone exchanger. Dust emission was further reduced after the change of the dust removal system of the clinker kiln, involving the replacement of an electrostatic precipitator with a bag filter in the second decade of this century.

Sampling grid

In the area of the study 124 measurement points of volume magnetic susceptibility (κ) were chosen in such a way that they were distributed evenly in all directions around the cement plant, in a systematic 240×240 m sampling grid. One point fell on the area of 0.057 km^2 . However, due to obstructions present in the field, the locations of selected measurement points sometimes had to be altered. A big impact on this alteration had the availability of terrain, which was examined by walking, to reach the largest possible number of available points and carry out measurements. Magnetometric measurements were made on the soil surface in the immediate vicinity of the cement plant emitters and along with the increasing distance from them.

A portable global positioning system Garmin GPS GPSMap 64st was used to locate and record sampling point coordinates. Measurement points were located in city squares and other recreational areas (playgrounds, tennis courts, meadows nearby the Odra river, situated less than 10 m from the river), as well as in areas and vicinity of other manufacturing enterprises, not only the cement plant, such as heat and power plant, as well as municipal sewage treatment plant. Moreover, some points had to be located in allotment gardens, orchards, arable soils, and in the vicinity of communication routes i.e. car roads and railway tracks, in the distance less than 10 m from the tracks (Fig. 1).

Magnetic measurements

Volume magnetic susceptibility (κ) measurements were performed *in situ* using a Bartington MS2 susceptibility meter by Bartington Instruments, with a MS2D loop probe, which has a diameter of 185 mm and generates alternating magnetic field, which magnetizes the soil underneath. Although the field penetration depth of the MS2D sensor is approximately 10 cm, the majority of the susceptibility signal comes from the upper soil layer (about 90 % of the response from a depth of 6–7 cm, and 95 % from the upper 8 cm). The affected soil volume has a shape of a hemisphere of the same diameter as the loop probe [8]. The κ values are expressed in dimensionless SI magnetic units. In contrast to mass specific magnetic susceptibility (χ), this parameter has advantages, because it can be measured both in the field and in the laboratory and it is considered to

be a non-concentration-dependent, i.e. its values do not depend on soil sample concentration.

At least 20 readings (sub-measurements) were performed in each measured spot (location), which was in the shape of a circle 2 m in diameter with the measuring point, for which GPS coordinates were determined, in the middle. Such a large number of individual measurements resulted from the fact that high variability can occur between field measurements even at close distances [15]. To maintain the accuracy of field measurements, air measurements were taken in between surface measurements, to allow for any change in the measurement sequence to be identified. Air measurements were performed before and after each individual surface measurement and the meter was zeroed if the result fell outside the $\pm 1.0 \cdot 10^{-5}$ SI tolerance level applied. The arithmetic mean of the 20 readings was then used as the representative measurement for each point.

The measured spots were free of grass and litter although detailed research into urban soils [15], revealed that organic (vegetative) coverage does not affects volume magnetic susceptibility, κ , of underlying soil. In-situ magnetic measurements were performed in 2015.

In the case of this study, taking into account diversified land use in the study area, resulting various land availability, a considerable number of measurement spots, and the fact that 20 individual measurements were taken in each of them, field measurements proved to be time-consuming.

Interpolation techniques of spatial analysis of magnetic susceptibility

Magnetometric data set obtained in the field, consisted of arithmetic means of 20 measurements of κ taken in each of 104 measurement spots, were firstly subjected to statistical analysis consisting in the determination of statistical measures of the dispersion of results (the smallest and the largest, arithmetic mean, median, standard deviation) (Table 1). Next, the values of arithmetic means of the κ were processed using three interpolation techniques, to obtain maps of spatial distribution of the analyzed parameter. On the basis of data obtained during fieldwork, point shapefiles were created with the magnetic susceptibility values stored in the attribute table. Files created in this way were then analyzed in terms of the κ distribution in the study area. Spatial interpolation maps (Fig. 2–6) were prepared using the extensions Spatial Analyst and Geostatistical Analyst within ArcGIS – ArcMap v. 10.6 software.

To choose the best technique of the κ distribution imaging, which would highlight the complexity of the magnetic enhancement signature in the study area, the following techniques were applied for spatial interpolation of field magnetometric data:

- nearest neighbours NN,
- inverse distance weighting IDW,
- ordinary kriging OK with 3 semivariogram models: linear, spherical and exponential.

The NN, as a non-geostatistical interpolator, is the simplest approach to the κ spatial distribution, which bases the κ value at the point where it was not measured on the value from the nearest point to its location. This technique is easy to use and transparent,

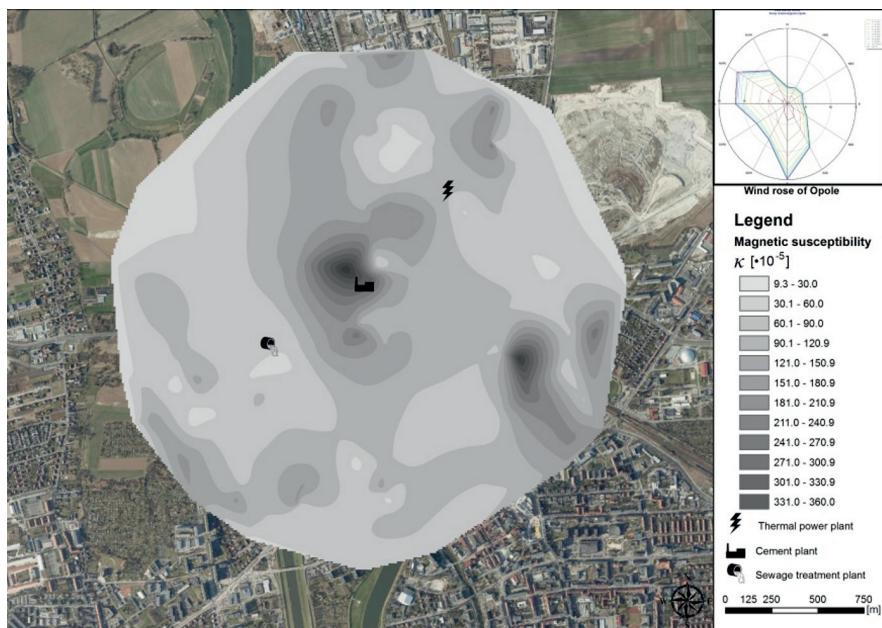


Fig. 2. Magnetic susceptibility spatial distribution with the use of the NN interpolation
Source: own research.

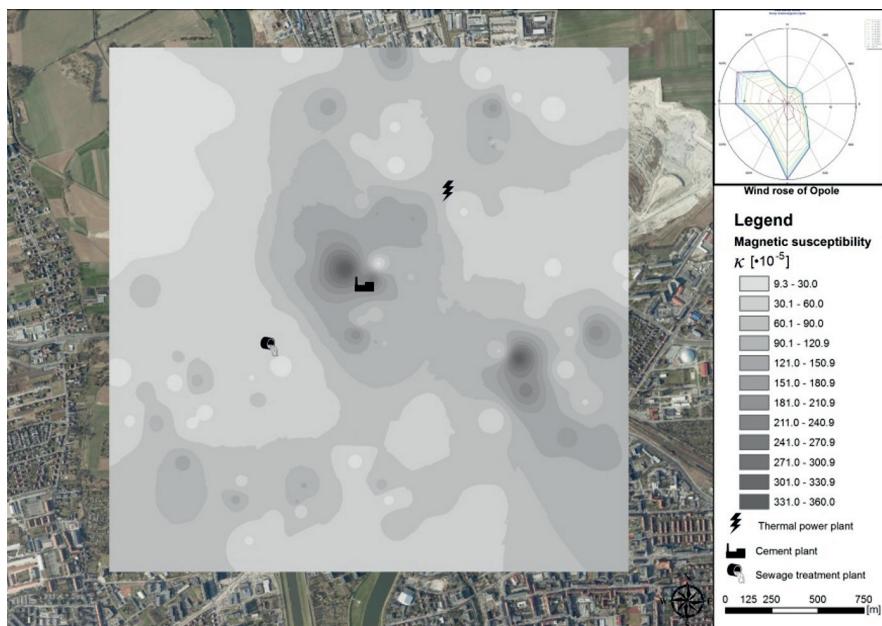


Fig. 3. Magnetic susceptibility spatial distribution with the use of the IDW interpolation
Source: own research.

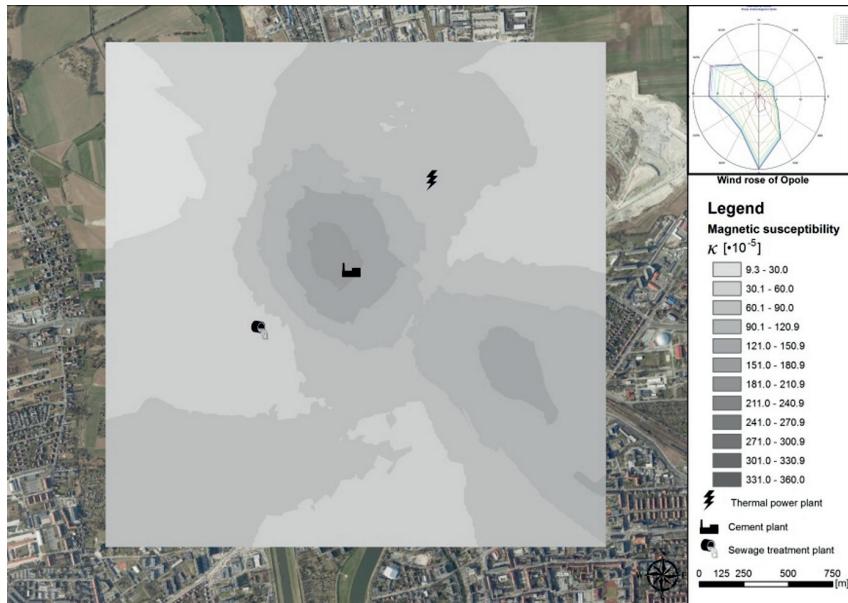


Fig. 4. Magnetic susceptibility spatial distribution with the use of the OK interpolation and the spherical semivariogram model

Source: own research.

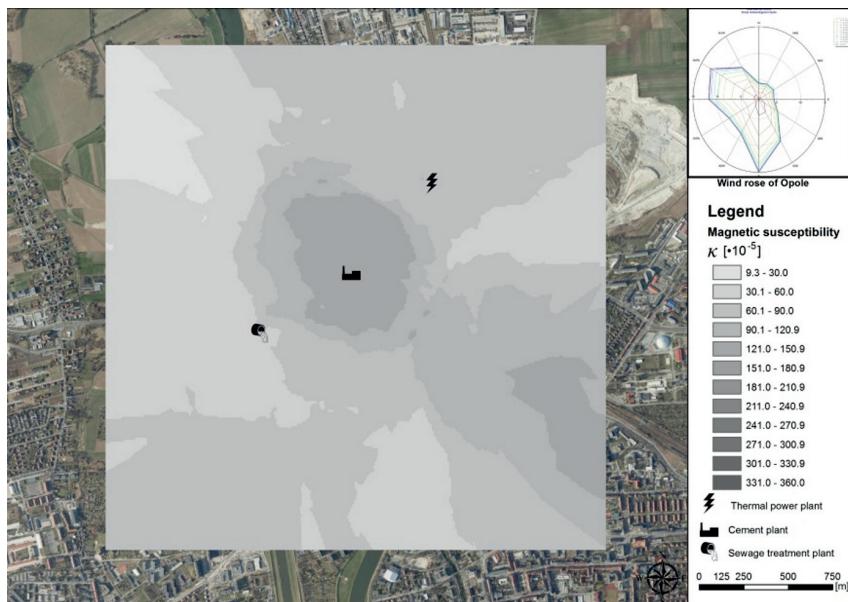


Fig. 5. Magnetic susceptibility spatial distribution with the use of the OK interpolation and exponential semivariogram model

Source: own research.

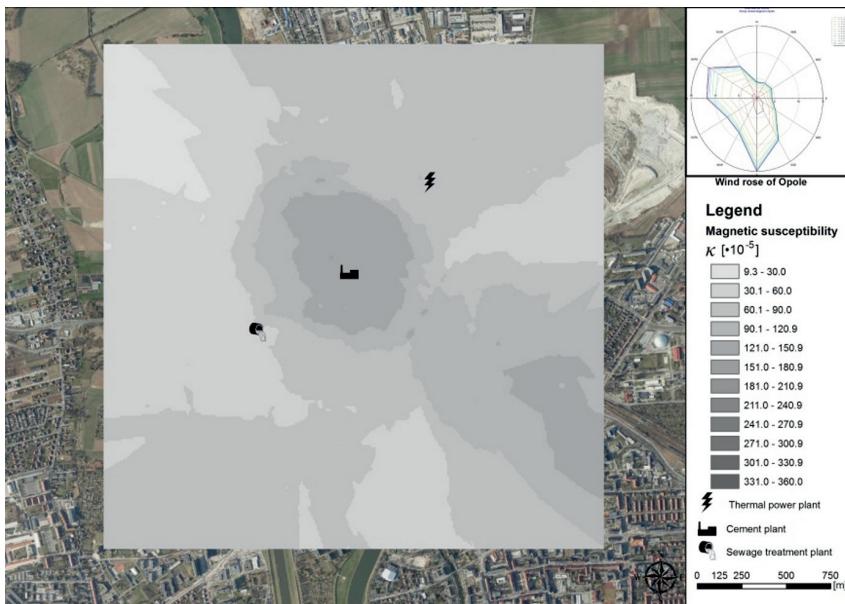


Fig. 6. Magnetic susceptibility spatial distribution with the use of the OK interpolation and linear semivariogram model
Source: own research.

however, it ignores additional available data, and this may lead to some interpolation errors [24, 28, 29]. The NN predicts the value of the κ at an unmeasured point based on the κ value in the nearest point by drawing perpendicular bisectors between measured points (n), forming Thiessen (or Dirichlet/Voronoi) polygons (V_i , where $i = 1, 2, \dots, n$). This produces one polygon per measured point and the measured point is located in the centre of the polygon, such that in each polygon all points are nearer to its enclosed measured point than to any other sample points [24]. The estimations of the κ at unmeasured points within polygon V_i are the measured value at the nearest single measured data point x_i that is $z^*(x_0) = z(x_i)$. All points (or locations) within each polygon are assigned the same value. A number of algorithms exist to generate the polygons [24]. The NN can be expressed by the formula:

$$f(V) = \sum_{i=1}^n p V_{P_i}$$

where: $f(V)$ – interpolation function of the κ at the point P at n ($i = 1, 2, \dots, n$)
natural neighbors
 p – weight assigned to each point.

The weights (p) depend on the coordinates of the P point and take values from 0 to 1. In the case where the point is not a natural neighbour, it receives the value 0.

Second non-geostatistical interpolator – the IDW – is similar to the NN approach but uses multiple measured points instead of only one to make a weighted interpolation of the κ . Weighting is determined based on distance to nearby measured points and by an *a priori* distribution rate that is defaulted to $p = 2$. The predicted κ value at a given location is then a weighted average of the κ values recorded at nearby measured points. In the IDW interpolation technique weights are determined by distance, and all available measured points are used as input unless a maximum search radius is pre-determined [29].

In other words, the IDW is based on the assumption that the value of the κ at a location which has not been measured is the weighted average of known values of the κ within its vicinity. Weights are inversely associated with the distances between the unknown value point location and determined value point locations. The inverse distance weight is dependent on a constant, known as a power parameter. Points closer to the unknown value point can much more influence the determined κ value based on the power parameter [17].

Kriging was initially developed for mining applications by Krige in 1951 [30], but is widely used in environmental studies [24, 28, 29]. It is similar to NN and IDW in that it uses measured point location information, but it does not use other covariates in the model and does not include information regarding spatial autocorrelation structure to improve interpolation estimates [29]. Kriging as a geostatistical interpolator is more reliable interpolation technique. The core of geostatistics is the variogram which expresses the spatial dependence between near measurement values [28, 31]. Kriging is categorized into subdivisions such as simple kriging SK, ordinary kriging OK, universal kriging UK and more advanced kriging techniques, *inter alia* indicator kriging IK, empirical Bayesian kriging EBK and indicator cokriging IC.

In the SK it is assumed that the average value is known and constant in the whole area. In OK the average value is determined locally on the basis of surrounding points. Local fluctuations in the average are taken into account by limiting the area of constancy of the average to the local neighbourhood. The variable has a linear trend. The value of V at point P is calculated using the weighted average p_i of the P_i measurement points:

$$V(P) = \sum p_i \cdot V(P_i)$$

The interpolation result and the mean interpolation error depend on the assumed weights. The best weights are those which give the smallest error of assessment [34].

The OK, in turn, depends on the fact that the closer measurement results are more correlated and similar [32]. It assumes that the spatial autocorrelation function is homogeneous in all directions (assumption of isotropy). The OK predictions are performed in the similar way as the IDW predictions. Unknown values are estimated by a weighted combination of values in measurement (known) points. The basis for calculating weights is semivariogram, which displays the structure of the spatial autocorrelation. Common semivariogram models include linear, spherical, exponential, and Gaussian models [34, 35].

The OK belongs to faithful and continuous interpolation techniques. It can occur in both local and global varieties, which is useful for modelling phenomena which change in space continuously, including many natural phenomena. The values do not change by leaps and bounds but seamlessly.

Results and discussion

The values of volume magnetic susceptibility (κ) in the examined area of Opole varied in very broad range from $9.1 \cdot 10^{-5}$ to $362.1 \cdot 10^{-5}$ SI units. The average κ of all 124 points amounted to $71.7 \cdot 10^{-5}$ SI units, and a median $51.4 \cdot 10^{-5}$ SI units (Table 1).

Table 1

Descriptive statistics of the $\kappa \cdot 10^5$ SI in the study area

Total number of measurement points	Min.	Max.	Median	Arithmetic mean	Standard deviation
124	9.1	362.1	51.4	71.7	61.4

Taking into account the results of the κ measurements, literature data on threshold values of this parameter for different levels of soil hazard, close relationship of the soil κ values with emission and deposition of industrial and urban dusts as well as geochemical status of soils, particularly heavy metal content, attempts were made to assess the strength and extent of anthropogenic transformations of the soil cover in study area. To determine soil pollution levels authors apply various threshold values of the κ . According to Schmidt et al. 2005 as cited after Jordanova et al. [13], the threshold κ value indicative of pollution, corresponds roughly to a value 50 % above the mean of all κ measurements in the area. Taking this condition into account and a mean value of the κ in the study area $71.7 \cdot 10^{-5}$ SI (Table 1), the threshold value to the data set collected in the city of Opole equals $107.6 \cdot 10^{-5}$ SI. Exceedances of the above mentioned threshold value occurred in 25 measurement points, so 20.2 % of the analyzed area can be considered as polluted.

On the other hand, long-term detailed studies carried out in urban and industrial areas of Upper Silesia, strongly affected by industrial and urban emissions, indicate a significantly lower threshold of soil magnetic enhancement in the study area, at which it can be considered polluted: $30 \cdot 10^{-5}$ SI [9–14, 20–23, 36].

Some authors suggest two threshold values of the κ parameter: $30 \cdot 10^{-5}$ SI and $50 \cdot 10^{-5}$ SI and resulting three-level scale of soil hazard: $\kappa \leq 30 \cdot 10^{-5}$ SI – low hazard, $30 < \kappa \leq 50 \cdot 10^{-5}$ SI – average hazard, $\kappa > 50 \cdot 10^{-5}$ SI – high hazard [20]. Meanwhile, other authors suggest the three-level scale at four threshold values: $30 \cdot 10^{-5}$ SI, $50 \cdot 10^{-5}$ SI, $75 \cdot 10^{-5}$ SI and $100 \cdot 10^{-5}$ SI, $30 < \kappa \leq 50 \cdot 10^{-5}$ SI moderate hazard, $50 < \kappa \leq 75 \cdot 10^{-5}$ SI elevated hazard, $75 < \kappa \leq 100 \cdot 10^{-5}$ SI and above $100 \cdot 10^{-5}$ SI strong hazard respectively [22].

Taking into account the above mentioned threshold values of soil hazard, for the purpose of this article five-level scale was accepted (Table 2).

Table 2

Evaluation of soil pollution level in the study area based on $\kappa \cdot 10^5$ SI

Hazard degree	Low $\kappa \leq 30$	Moderate $30 < \kappa \leq 50$	Elevated $50 < \kappa \leq 75$	Strong $75 < \kappa \leq 100$	Very strong (magnetic anomaly) $\kappa > 100$
Number of points	29	31	23	11	30
Percentage	23.4 %	25.0 %	18.5 %	8.9 %	24.2 %

Source: own research.

The research shows that over half (52 %) of the analyzed measurement points were characterized by a significant enhancement of the magnetic signal, from elevated to very strong, including about 1% points with very strong enhancement, which can be considered as a magnetic anomaly. Numerous magnetic and geochemical studies of soils, carried out in urban-industrial regions, involving the integration of magnetic and geochemical techniques have shown that magnetic transformations are usually accompanied by geochemical transformations, expressed in increased content of heavy metals in soils [9–15, 19–23]. It is known from the literature that under conditions of significant magnetic enhancement, at the value of $\kappa > 50 \cdot 10^{-5}$ SI, there is an increased content of easily accessible to the environment forms of heavy metals, especially Pb, which poses a threat to living organisms [20].

The analysis of the spatial distribution maps of field magnetic susceptibility (Fig. 2–6) shows that the highest values indicating the highest degree of soil pollution occur within the premises and in the immediate vicinity of the cement plant, which distinguishes itself unfavorably from other sources of emission of dust pollutants in the study area (heat and power plant, municipal wastewater treatment plant, municipal economy facilities including home furnaces). As can be seen from the archival literature [37], in the years 1951–1975 the described cement plant emitted a huge amount of dust, from 2618 to 29000 Mg per year, with dust emission factor ranging from 33 to 714 kg of dust per 1 Mg of cement, which means that the dust accounted for more than half the amount of cement production. This was reflected in a huge – in comparison to the Polish standard of 200 g/(m² · year) [42] – fall of dust in the area of this plant, which in the years 1966–1968 ranged from 558 to 744 g/(m² · year). According to archival data from later years (1990–1994), dust emissions dropped to 697 Mg/year, which resulted in a decrease in dust fall to values ranging from 151 to 335 Mg, but it still exceeding the protection standard. Another magnetic “hot spot” was revealed in the so-called Mountainous Housing Estate, located about 2 km from the cement plant in the direction of the prevailing winds (SE) in Chabry District of Opole, characterized by compact, 3-storey block of flats. The analysis of the data from Table 2 suggests that the soil magnetic anomalies in this area could have been caused by falling cement dusts. Data from the above-mentioned table indicates the relationship between the amount of falling dusts in the study area and the distance from the cement plant, as well as the occurrence of exceedances of the threshold value of 200 g/(m² · year) at the points closest to the plant. Remarkably, there was cement in the collected dustfall, which – as results from the research – has high magnetic susceptibility, about $250 \cdot 10^{-5}$ SI (Table 4).

Moreover, the so-called. law emission of fly ash from home furnaces fired with coal and waste, could contribute to the increase in soil magnetic susceptibility in the Mountainous Housing Estate. Ashes from various sources of thermal coal processing are mentioned in the literature as the most important source of magnetic substances in soils [3–5, 13, 18], while cement dusts are far less recognized in this respect. The third area, where soil magnetic anomalies were found at some points, were allotment gardens located in the NE direction from the cement plant and the municipal heat and power plant, burning the hard coal, so it may results from the emissions of dusts from both plants.

Table 3
Dustfall in the selected points of studied area in years 1990–2000 [g/(m² · year)]

Point No. and location	Distance and direction from cement plant	Min.	Max.	Average	Mean	SD	Notes
1 St. Anna street	0.7 km N	198	351	279	290	54	In 5 years (1994 1995, 1996, 1997, 1999) cement was identified in falling dust
2 Luboszycka street	1 km SSE	107	266	206	201	54	In 3 years (1995, 1996, 1999) cement was identified in falling dust
3 Kusocinskiego street	2 km SE	68	193	144	155	37	
4 Chabry street	2.5 km E	87	182	127	126	26	

Source: archival monthly data of WSSE in Opole, own elaboration.

Table 4
Volume magnetic susceptibiliy $\kappa \cdot 10^5$ SI of cement produced in the examined cement plant
(Bartington MS2D field measurements performed for 3 cement bags)

Min.	Max.	Median	Mean
245	258	252	252

Source: own research.

When analyzing concentrations of two fractions of suspended dust: PM10 and PM2.5 in the city of Opole, as measured by Voivodship Inspectorate of Environmetal Protection (WIOS) [41], it can be noted that over the last decade there were annual exceedances of permissible, threshold values of these dust fractions. As a result, the city has been still classified in C class areas, for which it is necessary to determine the places of exceedances, achieve threshold values, and update the air protection program.

The long-term impact of dusts emitted from the described cement plant on soils combined with high dust fall in previous years could have contributed to the magnetic transformation of soils around the plant, which is confirmed by maps of the magnetic susceptibility distribution (Fig. 2–6). Research has shown that cement dusts collected in

Opole Voivodship contain technogenic magnetic particles TMPs formed in the high temperature process of clinker burning, which – in terms of morphology and mineralogical composition – occur mainly in the form of sharp-grained crystals about 20 μm in size, and their characteristic component are calcium ferrites [3–5]. The magnetite is the dominant magnetic mineral of TMPs, and apart from it there is maghemite, hematite and goethite.

According to previous research carried out in the area of the described cement plant, geomagnetic soil transformations are accompanied by geochemical transformations, expressed by the increased content of heavy metals, decreasing as the distance from the cement plant increases [38].

Maps of spatial distribution of volume magnetic susceptibility κ in the studied area (Fig. 2–6) created by the two non-geostatistical interpolators: NN and IDW, and one geostatistical interpolator: OK, revealed a slightly different distribution of the same values of κ , which results from different interpolation algorithms. In the case of the NN technique, interpolation was carried out based on the values of κ at neighboring measurement points. The map created with this technique indicates a very clear distribution of magnetic enhancement in the studied area, and also allows to plot isolines of the distribution of the analyzed parameter. Unfortunately this technique, although allows for imagine the actual location of magnetic enhancement in the studied area, is limited to it only.

The other interpolation techniques (OK, IDW), in turn, show different variants of the κ distribution depending on the weights of the measurement data and the distance between the measurement points. Great advantage of these technique is the modeling of the spatial distribution of magnetic susceptibility, not limited only to the studied area. On the OK and IDW maps (Fig. 3–6), it can be seen that the phenomenon of magnetic enhancement has a wider dimension and does not occur only in the studied area.

Kriging as geostatistical interpolator, allows for modeling the propagation of the phenomenon of the occurrence of magnetic enhancement. Comparing to the NN technique, maps created by kriging interpolation technique (Fig. 3–5) show a wider surface differentiation of κ , and on their basis the extent of the occurrence of magnetic enhancement may be predicted. In this technique, it is possible to determine the weight of kriging, which is dependent on the spatial correlation degree, hence the role of semivariograms is important. In calculations carried out for the purpose of this article the weight of Kriging was determined automatically by software, which does not have the option of setting the required weight. The kriging technique allows to show the actual distribution of the magnetic signal intensity, because the κ values are also estimated in the unmeasured locations based on the known values collected from the measurement points, multiplied by the kriging weights.

The OK interpolation sets the trend of the κ distribution, taking into account the dependence of the distance between measurement points with specific values of magnetic susceptibility. In the OK, the mean is treated as an unknown value. This interpolation technique takes into account local fluctuations of the mean value by applying the so-called moving window. The OK assumes that the spatial autocorrelation function is homogeneous in all directions (assumption of isotropy). Unknown values are

estimated by a weighted combination of values in known measurement points. The basis for calculation of weights is semivariogram.

The disadvantage of this interpolation technique is the fact that it can omit measurement points, which are less numerous than others, for example points with the highest κ values, treating them as those which do not indicate the trend. As a result, kriging may not show on the maps actual values obtained for these points in field measurements, so – in the case of study of magnetic susceptibility spatial distribution in a very diverse area – do not show magnetic hot spots i.e. locations of highest values of this parameter, but rather smooths the results of field magnetic measurements. This ‘smoothing effect’ of kriging, which makes it less useful interpolation technique for highly diverse areas such as industrial and urban areas, is confirmed by other authors [39, 40].

When analyzing the usefulness of the applied techniques of visualization of magnetic susceptibility spatial distribution and the interpolation algorithm, the IDW technique should be distinguished as the one that most realistically reflects the distribution of the studied parameter. The spatial distribution of the κ values is the closest to the measured values in individual measurement points. At the same time, the interpolation algorithm allows to determine the spatial distribution of the κ in a wider area than designated by the boundaries of the extreme points.

Another useful technique for assessing the distribution of the studied parameter is the NN interpolation. This technique, in the most faithful way, presents the spatial distribution of the studied parameter, creating isolines between measurement points, but it is restricted only to the area limited by the range of the furthest measurement points.

As results from research, the technique of OK proves to be the least useful. Due to the fact that in the applied software it was impossible to choose kriging’s weight, the automatic selection of this parameter caused, on the one hand, the rejection of extreme values, and – on the other hand – a significant spatial overestimation of the studied phenomenon of magnetic enhancement, not reflected in the measured point values. Moreover, the differentiation of the type of semivariogram within this technique (exponential, linear, spherical) did not show any significant differences in the presentation of the spatial distribution of the studied phenomenon of magnetic enhancement.

The results of the research indicate, that due to elevated values of topsoil magnetic susceptibility and occurrence of magnetic anomalies indicating geochemical transformations of soils (which was proved by previous research), it is vital permanent monitoring of the soil quality in the studied area. Maps created on the basis of the results of soil magnetometric measurements can be a useful tool in this monitoring, as a base (starting material) for analysis and assessment of the dynamics of soil pollution. The results suggest, that soil cover in the area of research has retained the impact of high emission of cement dusts from previous years and – in the first place – was affected by cement plant. It is situated in the center of urban agglomeration of Opole, which requires special attention and constant monitoring of environmental condition. Nowadays the plant produces cement in accordance with the BAT requirements and implements the principles of sustainable development in the cement production process but uses waste-derived substances as substitutes for fuels and natural raw materials.

This imposes special requirements for the monitoring of the environmental condition in the vicinity of this plant and further integrated magnetic and geochemical research.

Conclusions

1. Although topsoil magnetic susceptibility, κ , in the study area varied in a broad range, the average κ value and a median exceeded $50 \cdot 10^{-5}$ SI units indicating high level of soil pollution and necessity for regular environmental monitoring.
2. Taking into account the threshold value of the κ in the study area ($107.6 \cdot 10^{-5}$ SI), calculated on the basis of results of research, 20.2 % of this area can be considered as magnetic anomaly. According to the literature, it is usually accompanied by geochemical anomaly manifested by increased content of heavy metals, easily accessible and hence hazardous to the environment and living organisms. It has been confirmed in previous geochemical research carried out in the study area.
3. The analysis of the spatial distribution of soil magnetic susceptibility in the study area clearly shows the footprint of cement production, consisting in the highest degree of soil pollution within the premises and the vicinity of the cement plant.
4. Although cement dust emissions have decreased nowadays, high and long-lasting emissions since the beginning of the 20th century has been preserved in soils in the form of increased magnetic susceptibility in the area of research.
5. When assessing the applied interpolation techniques of magnetic susceptibility spatial distribution in the study area, non-geostatistical techniques (IDW, NN) proved to be more useful, than geostatistical one – ordinary kriging (OK).
6. The inverse distance weighting (IDW) most realistically reflected the spatial distribution of the κ values in the study area. The nearest neighbors (NN) interpolation proved to be another useful technique, despite some disadvantages of it.
7. The results suggest that ordinary kriging (OK) interpolation, due to its ‘smoothing effect’ is less useful technique for highly diverse – in terms of land use – area of the study. This fact has been confirmed in the literature. Ordinary kriging semivariograms of different types did not show any significant differences in the spatial distribution of the studied parameter.

References

- [1] Rajfur M, Krems P, Kłos A, Kozłowski R, Józwiak MA, Kříž J, Waclawek M. Application of algae in active biomonitoring of the selected holding reservoirs in Świętokrzyskie province. *Ecol Chem Eng S*. 2016;23(2):237-247. DOI: 10.1515/eecs-2016-0016.
- [2] Rajfur M, Świsłowski P, Nowański F, Śmiechowicz B. Mosses as biomonitor of air pollution with analytes originating from tobacco smoke. *CDEM*, 2018;23(1-2):127-136. DOI: 10.1515/cdem-2018-0008.
- [3] Magiera T, Jabłońska M, Strzyszcz Z, Rachwał M. Morphological and mineralogical forms of technogenic magnetic particles in industrial dusts. *Atmos Environ*. 2011;45:281-290. DOI: 10.1016/j.atmosenv.2011.04.076.
- [4] Magiera T, Goluchowska B, Jabłońska M. Technogenic Magnetic Particles in Alkaline Dusts from Power and Cement Plants. *Water Air Soil Pollut*. 2013;224(1):1389. DOI: 10.1007/s11270-012-1389-9.

- [5] Szuszkiewicz M, Magiera T, Kapicka A, Petrovsky E, Grison H, Gołuchowska B. Magnetic characteristics of industrial dust from different sources of emission: A case study of Poland. *J Appl Geophys.* 2015;116:84-92. DOI: 10.1016/j.jappgeo.2015.02.027.
- [6] Strzyszcz Z. Magnetic Susceptibility of Soils in the Areas Influenced by Industrial Emissions. *Soil Monitoring: Early Detection and Surveying of Soil Contamination and Degradation.* In: Schulin R, Desaules A, Webster R, Vonsteiger B, eds. Book Series: Monte Verita. Proc. of the Centro Stefano Franscini Ascona;1993:255-269. ISBN 978-3-0348-7544-8.
- [7] Strzyszcz Z, Magiera T, Heller F. The Influence of Industrial Immissions on the Magnetic Susceptibility of Soils in Upper Silesia. *Stud Geophys Geod.* 1996;40(3):276-286. DOI: 10.1007/BF02300743.
- [8] Lecoanet H, Leveque F, Segura S. Magnetic susceptibility in environmental applications: comparison of field probes. *Phys Earth Planet In.* 1999;115:191-204. DOI: 10.1016/S0031-9201(99)00066-7.
- [9] Magiera T, Strzyszcz Z, Kapicka A, Petrovsky E. Discrimination of lithogenic and anthropogenic influences on topsoil magnetic susceptibility in Central Europe. *Magprox Team Geoderma.* 2005;130(3-4):299-311. DOI: 10.1016/j.geoderma.2005.02.002.
- [10] Magiera T, Strzyszcz Z, Rachwał M. Mapping particulate pollution loads using soil magnetometry in urban forests in the Upper Silesia Industrial Region, Poland. *Forest Ecol Manage.* 2007;248(1-2):36-42. DOI: 10.1016/j.foreco.2007.02.034.
- [11] Magiera T, Parzentyń H, Łukasik A. The influence of the wind direction and plants on the variability of topsoil magnetic susceptibility in industrial and urban areas of southern Poland. *Environ Earth Sci.* 2016;75(3,213). DOI: 10.1007/s12665-015-4846-0.
- [12] D'Emilio M, Chianese, D., Coppola, R. et al. Magnetic susceptibility measurements as proxy method to monitor soil pollution: development of experimental protocols for field surveys *Environ Monit Assess.* 2007;125(1-3):137-146. DOI: 10.1007/s10661-006-9246-1.
- [13] Jordanova N, Jordanova D, Tsacheva T. Application of magnetometry for delineation of anthropogenic pollution in areas covered by various soil types. *Geoderma.* 2008;144(3-4):557-571. DOI: 10.1016/j.geoderma.2008.01.021.
- [14] Kapicka A, Petrovsky E, Fialová H, Podrazsky V, Dvorak I. High resolution mapping of anthropogenic pollution in the Giant Mountains National Park using soil magnetometry. *Stud Geophys Geod.* 2008;52(2): 271-284. DOI: 10.1007/s11200-008-0018-y.
- [15] Golden N, Zhang C, Potito AP, Gibson PJ, Bargary N, Morrison L. Impact of grass cover on the magnetic susceptibility measurements for assessing metal contamination in urban topsoil. *Environ Res.* 2017;155:294-306. DOI: 10.1016/j.envres.2017.02.032.
- [16] Tan Z, Han Y, Cao J, Huang CC, An Z. Holocene wildfire history and human activity from high-resolution charcoal and elemental black carbon records in the Guanzhong Basin of the Loess Plateau, China. *Quaternary Sci Rev.* 2015;109:76-87. DOI: 10.1016/j.quascirev.2014.11.013.
- [17] Saepuloh A, Haeruddin H, Nur Heriawan M, Kubo T, Koike K, Malik D. Application of lineament density extracted from dual orbit of synthetic aperture radar (SAR) images to detecting fluids paths in the Wayang Windu geothermal field (West Java, Indonesia). *Geothermics.* 2018;72:145-155. DOI: 10.1016/j.geothermics.2017.11.010.
- [18] Petrovsky E, Remes J, Kapicka A, Podrazsky V, Grison H, Boruvka L. Magnetic mapping of distribution of wood ash used for fertilization of forest soil. *Sci Total Environ.* 2018;626:228-234. DOI: 10.1016/j.scitotenv.2018.01.095.
- [19] Strzyszcz Z, Magiera T. Record of industrial pollution in Polish ombrotrophic peat bogs. *Phys Chem Earth.* 2001;26(11-12):859-866. DOI: 10.1016/S1464-1895(01)00133-8.
- [20] Strzyszcz Z, Rachwał M. Zastosowanie magnetometrii do monitoringu i oceny ekologicznej gleb na obszarach objętych wpływem emisji przemysłowych (Application of magnetometry for monitoring and ecological assessment of soils in areas affected by industrial emissions). *Zabrze: Inst Environ Eng PAS;* 2010;78. PL ISSN 0208-4112. ISBN 978-83-60877-60-9.
- [21] Fabijanczyk P, Zawadzki J, Magiera T, Szuszkiewicz M. A methodology of integration of magneto-metric and geochemical soil contamination measurements. *Geoderma.* 2016;277:51-60. DOI: 10.1016/j.geoderma.2016.05.009.
- [22] Fabijanczyk P, Zawadzki J, Magiera T. Magnetometric assessment of soil contamination in problematic area using empirical Bayesian and indicator kriging: A case study in Upper Silesia, Poland. *Geoderma.* 2017;308:69-77. DOI: 10.1016/j.geoderma.2017.08.029.

- [23] Rachwal M, Kardel K, Magiera T, Bens O. Application of magnetic susceptibility in assessment of heavy metal contamination of Saxonian soil (Germany) caused by industrial dust deposition. *Geoderma*. 2017;295:10-21. DOI: 10.1016/j.geoderma.2017.02.0070016-7061.
- [24] Li J, Heap AD. A Review of Spatial Interpolation Methods for Environmental Scientists. *Geoscience Australia, Record* 2008;23:137 pages.
- [25] Statistics of Opole). Statistical Office in Opole, Opole 2018. ISSN 2451-1390. <http://opole.stat.gov.pl>.
- [26] Kondracki J. *Geografia regionalna Polski* (Regional geography of Poland). Warszawa: PWN; 2011. ISBN 9788301160227.
- [27] The Odra Cement Plant – 1911-2011, 100 years which cemented our success). OPOLGRAF SA Ed., Opole, 2011, K. Kownacka – text preparation. (in Polish and German).
- [28] Wackernagel H. *Multivariate Geostatistics, An Introduction with Applications*. Third ed. Berlin-Heidelberg: Springer-Verlag; 2003. ISBN: 978-3-642-07911-5. DOI: 10.1007/978-3-662-05294-5
- [29] Roznik M, Porth CB, Porth L, Boyd M, Roznik K. Improving agricultural microinsurance by applying universal kriging and generalised additive models for interpolation of mean daily temperature. *The Geneva Papers on Risk and Insurance – Issues and Practice*, 2019. DOI: 10.1057/s41288-019-00127-9
- [30] Krige DG. A Statistical Approaches to Some Basic Mine Valuation Problems on the Witwatersrand. *J S Afr I MIN METALL*. 1951;52:119-139.
- [31] Isaaks EH, Srivastava RM. *An Introduction to Applied Geostatistics*. New York: Oxford University Press; 1989. <https://pl.scribd.com/document/266290280>.
- [32] Yajie H, Zhen L, Huichun Y, Shiwen Z, Zhiqing Z, An X, Yuanfang H. Mapping Soil Electrical Conductivity Using Ordinary Kriging Combined with Back-propagation Network. *Chinese Geogr Sci*. 2019;29(2):270-282. DOI: 10.1007/s11769-019-1027.
- [33] Abel OE, Rabiu AB, Bolaji OS, Oyeyemi EO. Modeling African equatorial ionosphere using ordinary Kriging interpolation technique for GNSS applications. *Astrophys Space Sci*. 2018; 363:168. DOI: 10.1007/s10509-018-3387-x.
- [34] El-Shejmy N, Valeo C, Habib A. *Digital Terrain Modeling: Acquisition, Manipulation and Applications*. Artech House Remote Sensing Library. Artech House, Inc. Norwood 2005, 270 pages. ISBN 1-58053-921-1.
- [35] Gold C. *Spatial context. An introduction to fundamental computer algorithms for spatial analysis*, 1st Edition. International Society for Photogrammetry and Remote Sensing Book Series, CRC Press; 2016. ISBN 9781138029637.
- [36] Zawadzki J, Fabijanczyk P, Magiera T, Rachwal M. Geostatistical Microscale Study of Magnetic Susceptibility in Soil Profile and Magnetic Indicators of Potential Soil Pollution. *Water Air Soil Pollut*. 2015;226:142. DOI 10.1007/s11270-015-2395.
- [37] Jaworowicz A, Liberska-Szmidt H, Grzybowski J. Udział cementowni opolskich w stanie zanieczyszczenia powietrza na terenie miasta Opola (Share of cement plants of Opole in the state of air pollution in the area of the city of Opole). *CWG* 1970;9-70:272-278.
- [38] Kusza G, Ciesielczuk T, Gołuchowska B. Zawartość wybranych metali ciężkich w glebach obszarów przyległych do zakładów przemysłu cementowego w mieście Opolu (Content of chosen heavy metals in soils of areas adjacent to cement plants in the city of Opole). *Ochr Środ Zasob Nat* 2009;40:70-75.
- [39] Ghiasi Y, Nafisi V. The improvement of strain estimation using universal kriging. *Acta Geod Geophys*. 2015;50:479-490. DOI 10.1007/s40328-015-0103-y.
- [40] Qiao P, Lei M, Yang S, Yang J, Guo G, Zhou X. Comparing ordinary kriging and inverse distance weighting for soil as pollution in Beijing. *Environ Sci Pollut Res*. 2018;25(16):15597-15608. DOI: 10.1007/s11356-018-1552-y
- [41] V Annual reports on the state of the environment for the years 2007-2017. Voivodeship Inspectorate of Environmental Protection (WIOS) in Opole. <http://www.opole.pios.gov.pl>
- [42] Regulation of the Minister for the Environment of 26 January 2010 on reference values for certain airborne substances. Appendix No. 1. *J Laws* 2010, No. 16, Item 87, page 1252).

ROZKŁAD PRZESTRZENNY PODATNOŚCI MAGNETYCZNEJ JAKO WSKAŹNIK ZANIECZYSZCZENIA GLEB NA TERENIE MIASTA OPOLA

Wydział Przyrodniczo-Techniczny, Uniwersytet Opolski, Opole

Abstrakt: Magnetometria glebową, polegającą na pomiarze podatności magnetycznej wierzchniej warstwy gleby, jest bardzo przydatną i coraz powszechniejszą stosowaną techniką monitorowania stanu gleb objętych wpływem antropopresji. Jak wynika z danych literaturowych, metoda ta wymaga dalszego udoskonalania, szczególnie w zakresie technik obrazowania danych magnetometrycznych. Celem badań była ocena przekształceń magnetycznych gleb na terenie miasta Opola (woj. opolskie) z zastosowaniem magnetometrii glebowej oraz trzech technik interpolacji danych magnetometrycznych (naturalnego sąsiedztwa NN, ważonych odwrotnych odległości IDW oraz krigingu zwykłego OK). Dane zostały zgromadzone podczas pomiarów terenowych podatności magnetycznej, wykonanych na powierzchni $7,1 \text{ km}^2$, w sieci 124 punktów pomiarowych, w 2015 r. Lokalizacje punktów określono za pomocą urządzenia Garmin GPS GPSMap 64st, a pomiary magnetometryczne wykonano *in situ* za pomocą miernika MS2 i czujnika MS2D firmy Bartington Instruments. Badania wykazały wysokie wartości podatności magnetycznej oraz występowanie glebowych anomalii magnetycznych na badanym terenie. Towarzyszą temu przekształcenia geochemiczne gleb, wykazane we wcześniejszych badaniach. Wyniki sugerują, że przyczyną tego stanu mogła być długofletna depozycja pyłów cementowych, emitowanych w latach ubiegłych w zwiększonej ilości przez cementownię, która pozostawiła swój ślad w środowisku. Analizując przydatność zastosowanych technik interpolacji danych magnetometrycznych, technika IDW najlepiej odzwierciedlała rozkład przestrzenny podatności magnetycznej na badanym terenie, podczas gdy technika OK, z powodu tzw. efektu wygładzającego, okazała się mniej przydatna.

Słowa kluczowe: podatność magnetyczna, magnetometria glebową, kriging zwykły, pyły cementowe