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Assessing Spatial Distributions of Total Trace Elements Content in Bottom Sediments of Dzierżno Duże Water Reservoir – Geostatistics-Based Studies

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ABSTRACT

The aim of this study was to assess the spatial distributions of total trace elements content in the bottom sediments of Dzierżno Duże water reservoir, along with the comparison of the accuracy and characteristics of Kriging and IDW interpolations. On the basis of regular measurement grid consisting of 53 points, bottom sediments samples were collected. Mean values of total trace elements content in bottom sediments of Dzierżno Duże were as follows: Zn – 410 mg/kg, Pb – 57 mg/kg, Cr – 36 mg/kg, Cu – 40 mg/kg, Cd – 5 mg/kg, Ni – 16 mg/kg and Ba – 267 mg/kg. According to the geochemical quality classification, the concentrations of Cd in 32% of samples were assigned to class IV (heavily contaminated), 45% to class III (contaminated), Zn in 42% samples to class III with 1 sample in class IV and 26% to class II (slightly contaminated), Pb in 9% to class III and 58% to class II, Cu in 4% to class III and 68% to class II, Cr in 17% to class II, Ni in 55% to class II, Ba in 8% to class III and 61% in class II. Coefficient of determination was determined between each case of trace elements content. The highest correlation (R^2 in range from 0.81 to 0.96) was observed between Zn and Pb, Zn and Cu, Zn and Cr, Zn and Ni, Pb and Cu, Pb and Cr, Cu and Cr, Cr and Ni. Significant correlation (R^2 in range from 0.70 to 0.80) occurred between: Zn and Cd, Pb and Ni, Cu and Ni, Cd and Ni. The lowest correlations (R^2 in range from 0.25 to 0.70) were observed between concentration of Ba and the rest of trace elements. Two different interpolation methods were chosen for the purpose of generating spatial distributions – Inverse Distance Weighted and Ordinary Kriging. These methods were chosen for purpose of obtaining optimal accuracy result of spatial distributions. The distributions of trace elements content were classified by means of geochemical criteria. In the case of accuracy comparison between IDW and Ordinary Kriging, the former had slightly better results in terms of mean value and root mean square. The generated spatial distributions allowed to determine the most contaminated areas, which were mainly northern-central and southern-central parts of water Dzierżno Duże reservoir.

Keywords: bottom sediments, spatial distribution, trace elements, geostatistics

INTRODUCTION

The stagnant water reservoirs accumulate a part of the material flowing from the basin in the form of bottom sediments [Ulrich et al. 2000, Wiatkowski et al. 2008]. The accumulated sediment in the reservoir basins contributes to the reduction of their capacity and, as a consequence, restricts the functions fulfilled by these objects [Bąk & Dąbkowski 2013, Bąk et al. 2014]. The chemical composition of sludge is a derivative of many factors, both natural and anthropogenic. It depends primarily on the geological structure of the basin, its management, use, as well as climatic and hydrological conditions, which determine the course and intensity of erosion processes in the basin as well as migration and accumulation of suspended matter as well as trace elements [Jancewicz et al. 2012, Bąk et al. 2013, Bąk et al. 2014]. Trace elements flowing in with river waters are accumulated in bottom sediments as a result of sorption processes and sedimentation of mineral and organic matter [Gałka & Wiatkowski 2010]. The knowledge on the chemical composition of sediments allows for inference about sources of velocity and distribution routes of metals in the reservoir is a better indicator than the more variable chemical composition of surface waters. Trace elements, including lead, mercury, chromium, zinc and cadmium, are one of the main pollutants in bottom sediments. These metals are considered the most toxic [Sidoruk & Potasznik 2013, Kazimierowicz & Kazimierowicz 2014, Szydłowski & Podlańska 2016].

An important feature that distinguishes trace elements from other undesirable or toxic substances is that they are not biodegradable but only biotransformed [Wojtkowski et al. 2008]. Metals and other substances can be immobilised in sediments even for a long time, counted in tens or hundreds of years [Dobicki 2004, Gałka & Wiatkowski 2010].

The aim of this study was to assess the spatial distributions of total trace elements content in bottom sediments of Dzierżno Duże water reservoir, along with comparison of the accuracy and characteristics of Kriging and IDW interpolations. Kriging and IDW are most commonly used interpolation methods. The analysis of their accuracy can indicate which method is more appropriate in terms of generating spatial distribution for bottom sediments of Dzierżno Duże water reservoir.

MATERIALS AND METHODS

The basin of the analysed Dzierżno Duże reservoir constitutes a part of the Kłodnica basin (38.1 km and 32.2 km of Kłodnica [Rzętała 2005]) with an area of almost 530 km^2, which is almost entirely located in the Katowice Upland area [Rzętała 2005, Kondracki 1978]. In administrative terms, the Dzierżno Duże reservoir is located in the western part of the Silesian Voivodeship and is part of the following administrative units: the city of Gliwice, the city of Pyskowice, the commune of Rudziniec and the commune of Zbrosławice. The area of the reservoir is nearly 615 ha, making it one of the largest reservoirs in Poland. From the south and east side, the reservoir basin is natural, the earth barrier limits it from the west, and a dam – from the north. The reservoir is supplied mainly with heavily polluted Kłodnica waters [Rzętała 2005].

The specificity of water management in the industrialised and urbanised area (regulation of watercourses, water discharges and use of foreign water from outside the basin, discharges of municipal and industrial sewage, introduction of underground mine waters to the rivers, construction and formation of numerous water reservoirs) and, to a lesser extent, the climate change caused changes in the natural regime of outflow of the Kłodnica basin [Rzętała 2005, Czaja & Jankowski 1990, Czaja & Jankowski 1991]. Moreover, the process of anthropogenisation of water relations in the Kłodnica basin illustrates the qualitative changes in water [Rzętała 1996, Rzętała 2005, Absalon et al. 1996], which are primarily expressed in the transformation of the chemical structure of water and the related change in its utility values [Żmuda 1973]. As a consequence of large-scale emission of pollutants to surface waters, the watercourses of the discussed area are classified as not meeting the standards because they are characterised by insufficient oxygenation, high content of biogenic substances, presence of significant amounts of trace elements and bacterial contamination [Rzętała 2005].

The sampling of bottom sediments of the Dzierżno Duże reservoir was carried out in September 2016. Using the ArcGIS software, a network of measurement points was developed in the form of a regular grid of squares consisting of 57 measuring points. Next, using the GPS system, the planned network was replicated and samples of bottom sediments were collected. Due to the terrain conditions, material collection in points 19, 36, 55 and 57 was not possible. Bottom sediment test samples were obtained at 53 measuring points at a depth of 0.3 m to 16.5 m below the water surface. The material for the research was collected using a specialist bottom-sediment trap of the Van Veen type from KC Denmark.

In the laboratory, the samples were firstly dried under dry-air conditions and then sieved through a 2 mm screen. They were then dried in an oven at 105°C to constant weight and ground in a vibrating mill until the grain size was lower than 0.2 mm. A sample prepared in this way was used to determine the total trace elements content. *Aqua regia* (a mixture of concentrated hydrochloric acid and nitric acid in a volumetric ratio of 3:1) was used for metal extraction. Mineralisation was carried out at 180°C, for 30 minutes in a high pressure microwave mineraliser manufactured by a German company Berghof GMBH. Three samples were prepared from each sediment sample for analysis. A plasma spectrometer (IRIS ICP-OES Thermo) was used to determine the trace elements content in accor-

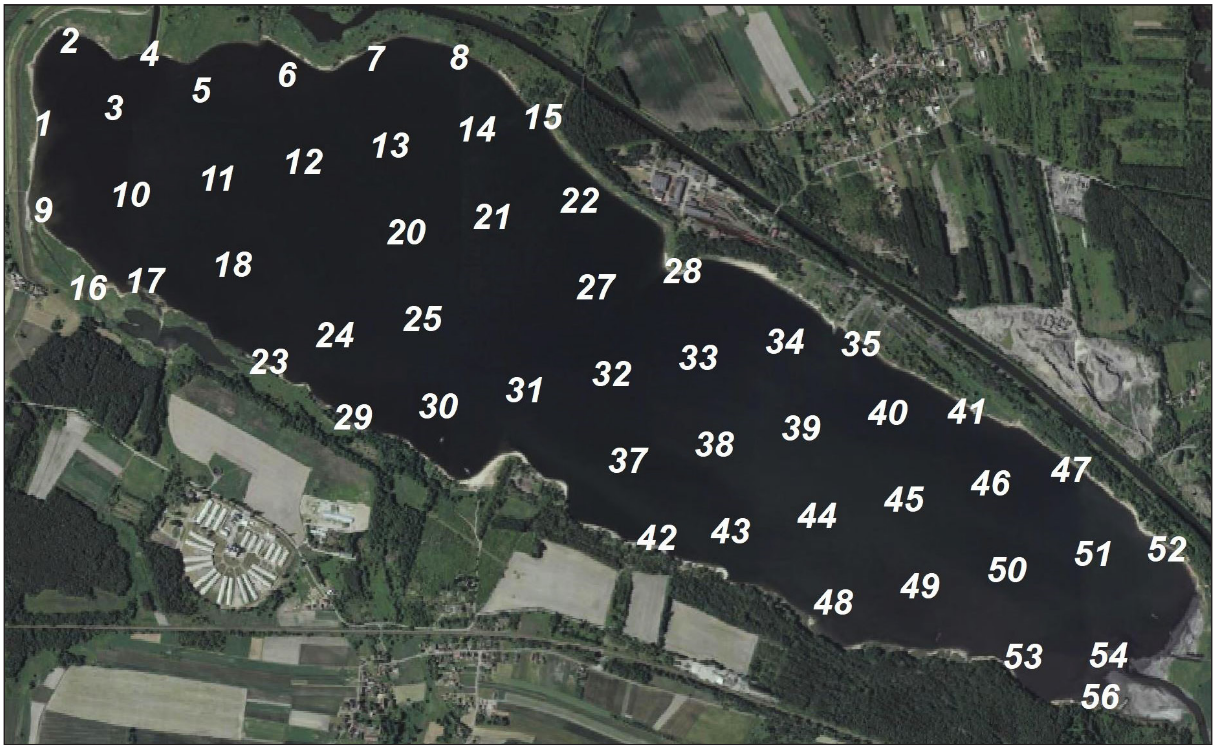

Figure 1. Dzierżno Duże water reservoir and measurement network

dance to Polish Standard PN-ISO 10390:1997 and 11466:2002 [Rozpondek and Wancisiewicz 2016, Rozpondek and Rozpondek 2017].

A statistical analysis has been conducted using the Statistica software. It included the basic statistical characteristics (Table 1) – the arithmetical mean, minimum and maximum, kurtosis and skewness. Using ArcGIS software, the Coefficient of determination (R^2) has also been calculated between the determined content of the individual elements in the bottom Two different interpolation methods were chosen for the purpose of generating spatial distributions, i.e. Inverse Distance Weighted (IDW, which is one of the most common deterministic methods [Gong et al. 2014]) and Ordinary Kriging [Reza et al. 2010]. In terms of accuracy, two main criteria were chosen: Mean and Root Mean Square error (RMS). Additionally, Root Mean Square Standardized (RMSS) and Average Standard Error (ASE) were determined for Kriging. These criteria were calculated with cross validation method in geostatistical analyst, which is part of ArcGIS software. In the case of Ordinary Kriging, along with spatial distribution, a prediction error map was created [Gong et al. 2014]. Descriptive statistics were performed for each trace element. Spatial distributions of trace elements content were classified by geochemical criteria (Table 1) [Bojakowska 2001].

RESULTS AND DISCUSSION

Depending on the localisation of the sampling of bottom sediments, the contamination values were varied. It is indicated by a large range of obtained data (difference between the minimum and maximum value).

The results of statistical analysis (Table 1) indicated high values of standard deviation. Kurtosis and skewness oscillated near values 0.1–0.8 (except for Cd). On the basis of the statistics, there was no need of performing data transformations for purpose of Ordinary Kriging. Mean values of total trace elements content in bottom sediments of Dzierżno Duże were as follows: Zn 410 mg/kg, Pb 57 mg/kg, Cr 36 mg/kg, Cu 40 mg/kg, Cd 5 mg/kg, Ni 16 mg/kg and Ba 267 mg/kg. According to the geochemical quality classification by Bojakowska [2001], the concentrations of Cd in 32% of samples were assigned to class IV (heavily contaminated), 45% to class III (contaminated), Zn in 42% samples to class III with 1 sample in class IV and 26% to class II (slightly contaminated), Pb in 9% to class III and 58% to class II, Cu in 4% to class III and 68% to class II, Cr in 17% to class II, Ni in 55% to class II, Ba in 8% to class III and 61% in class II.

The coefficient of determination was determined between each case of trace element con-

Table 1. Bojakowska's geochemical quality classes of bottom sediments [Bojakowska 2001].

Trace element	Class I	Class II	Class III	Class IV
As	<10	30	50	>50
Ba	<100	500	1000	>1000
Cr	<50	100	400	>400
Zn	<200	500	1000	>1000
Cd	<1	3,5	6	>6
Co	<10	20	50	>50
Cu	<40	100	200	>200
Ni	<16	40	50	>50
Pb	<30	100	200	>200
Hg	<0.1	0.5	1	>1.0
Ag	<1	2	5	>5

tent (Table 3). The highest correlation (R^2 in range from 0.81 to 0.96) was observed between Zn and Pb, Zn and Cu, Zn and Cr, Zn and Ni, Pb and Cu, Pb and Cr, Cu and Cr, Cr and Ni. Significant correlation (R^2 in range from 0.70 to 0.80) occurred between: Zn and Cd, Pb and Ni, Cu and Ni, Cd and Ni. High correlation (above 0.7) indicates a strong spatial relation between studied elements – probably, the contamination source is similar. The lowest correlations (R^2 in range from 0.25 to 0.70) were observed between concentration of Ba and the rest of trace elements – the source of Ba is probably different than the source of other trace elements.

Bottom sediments of the tested water reservoir are mainly contaminated by zinc and cadmium. Significant concentrations of these elements are present in the most of the researched area (Figure 2 – 8). The highest concentration was observed in the northern part – dominant class IV for cadmium and III for zinc, according to geochemical criteria. The distribution of higher pollution values in the northern part coincides with the values of other metals tested. The highest concentration of zinc was observed at point 20. Only the spatial distribution of barium (Figure 8) was characterised by higher values in the southern part and lower in the northern part. On the basis of the generated maps, significantly lower values of pollution in the coastal zone of the reservoir were observed. High values obtained as a result of the interpolation process in the area around points 30, 31, 37 in the coastal zone are caused by the lack of measurement in point 36 (dense vegetation and shallowing). This fact is confirmed by the distribution of prediction errors (Figure 8) – significant error values occur in this area. The distribution of the examined elements can largely be caused by the dominant current in the reservoir (inflow of the Gliwicki Canal near points 4 and 15, outflow at point 54). This likely indicates the occurrence of significant pollution in the waters of the Gliwicki Canal. The increased content of trace elements in the bottom sediments of the Dzierżno Duże water reservoir may be caused by the fact

Table 2. Statistical analysis for total trace elements contents

Parameter	Ba	Cd	Cr	Cu	Ni	Pb	Zn
N	53						
Mean	266.97	5.13	36.25	40.07	16.18	56.58	409.99
SD	175.59	4.47	20.60	28.449	9.61	36.03	264.00
Min	12.84	0.21	2.32	0.37	0.32	0.92	13.36
Max	678.49	22.75	88.23	100.98	36.56	134.42	1056.36
Median	318.45	4.94	41.75	48.3	16.8	67.25	473.25
Curtosis	-0.58	3.84	-0.21	-0.81	-0.64	-0.73	-0.84
Skewness	0.08	1.59	0.31	0.12	0.17	0.00	0.04

Table 3. Coefficient of determination (R^2) between total trace elements contents

Element	Zn	Pb	Cu	Cr	Cd	Ni
Pb	0.96	-	-	-	-	-
Cu	0.80	0.85	-	-	-	-
Cr	0.89	0.86	0.84	-	-	-
Cd	0.78	0.68	0.61	0.81	-	-
Ni	0.86	0.79	0.78	0.92	0.79	-
Ba	0.59	0.72	0.67	0.52	0.25	0.46

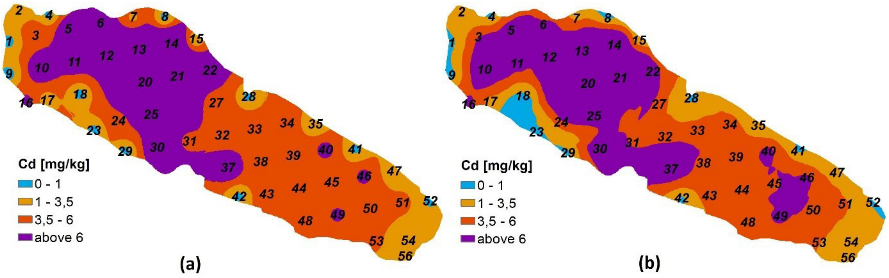

Figure 2. Spatial distribution of Cd interpolated by Inverse Distance Weighted method (a) and Ordinary Kriging (b), classified by geochemical criteria [Bojakowska 2001]

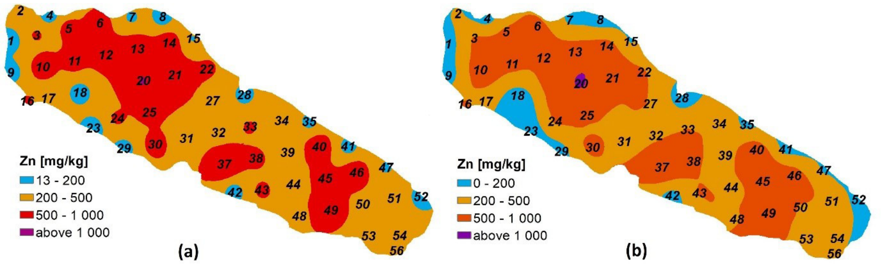

Figure 3. Spatial distribution of Zn interpolated by Inverse Distance Weighted method (a) and Ordinary Kriging (b), classified by geochemical criteria [Bojakowska 2001]

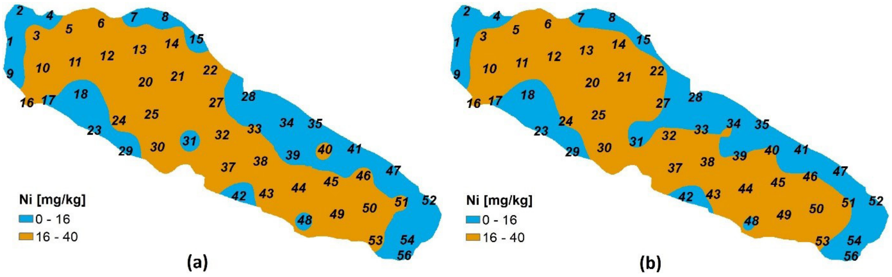

Figure 4. Spatial distribution of Ni interpolated by Inverse Distance Weighted method (a) and Ordinary Kriging (b), classified by geochemical criteria [Bojakowska 2001]

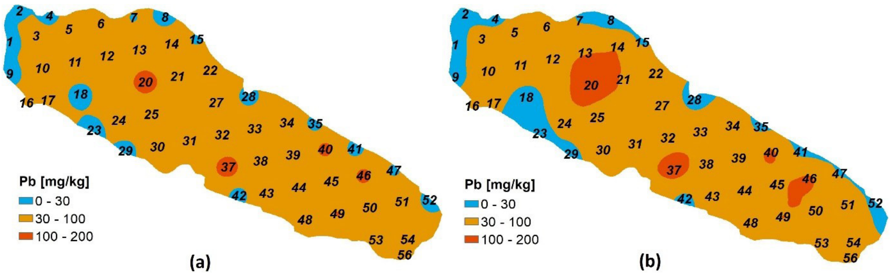

Figure 5. Spatial distribution of Pb interpolated by Inverse Distance Weighted method (a) and Ordinary Kriging (b), classified by geochemical criteria [Bojakowska 2001]

that the tested reservoir and its basin are exposed to the harmful effect of anthropogenic activities. Most likely, this state is related to the supply to the basin of the reservoir, adjacent to urban agglomerations, of municipal and industrial sewage (mainly the chemical industry).

In the case of comparison of accuracy between IDW and Ordinary Kriging, IDW had slightly, but consistently better results in terms of mean value and RMS (Table 4). The results confirm the studies of other authors [Zhou & Michalak 2009, Gong et al. 2014]. It should be noted that Kriging

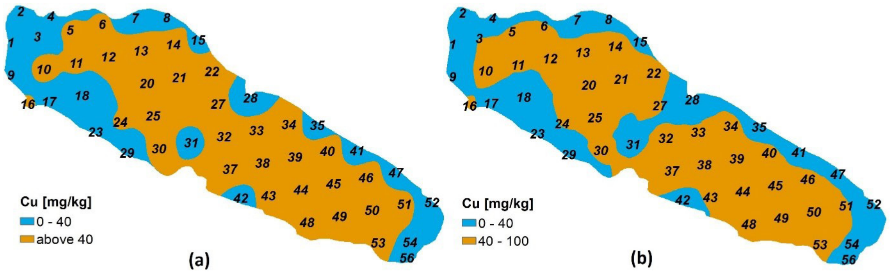

Figure 6. Spatial distribution of Cu interpolated by Inverse Distance Weighted method (a) and Ordinary Kriging (b), classified by geochemical criteria [Bojakowska 2001]

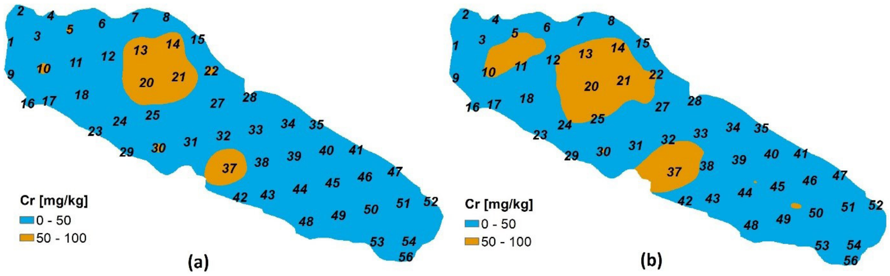

Figure 7. Spatial distribution of Cr interpolated by Inverse Distance Weighted method (a) and Ordinary Kriging (b), classified by geochemical criteria [Bojakowska 2001]

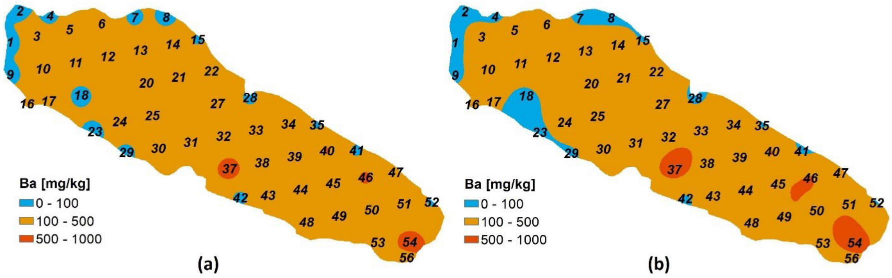

Figure 8. Spatial distribution of Ba interpolated by Inverse Distance Weighted method (a) and Ordinary Kriging (b), classified by geochemical criteria [Bojakowska 2001]

methods are definitely more complex and proper application requires performing preliminary analysis. It concerns mainly the type of statistic distribution, trend analysis, data transformation, nugget effect and size of lag. All of the criteria were determined in geostatistical software. Transformation of the data was not required. Another advantage of Kriging methods involves additional factors that can be determined (smooth, number of points etc.). All of them have an impact on final spatial distributions. Additionally, Kriging provides information about prediction error which can be crucial in planning of future studies (Figure 9) [Mabit et al. 2007, Hu et al. 2005]. Because of its complexity (mainly the possibility of calculating semivariogram and numerous criteria), Ordinary Kriging provides two additional criteria for cross validation process – RMSS and ASE. On their basis it is possible to indicate if the data is underestimated or overestimated [Robinson et al., Mabit et al. 2007]. If RMSS is higher than 1 and RMS higher then ASE, the spatial distribution is underestimated (estimated values are lower than the real values). In the opposite situation, the spatial distribution would be overestimated. All of the obtained Ordinary Kriging results are characterized by much higher RMS then ASE, and RMSS greater than one (Table 4).

In addition to accuracy comparison, the obtained spatial distributions were analysed in terms of the visual quality (Figure 2–8). The spatial distributions generated by Ordinary Kriging methods are smoother. Additionally, the Kriging results minimalize the effects of small circles

Table 4. Results of accuracy analysis for Inverse Distance Weighted and Ordinary Kriging interpolation methods

Trace element	Interpolation Method	Mean	RMS	RMSS	ASE
Zn	Inverse distance weighted	25.79	280.56		
	Ordinary kriging	27.84	309.22	1.38	224.12
Pb	Inverse distance weighted	3.51	38.42		
	Ordinary kriging	3.98	44.33	1.46	30.54
Cu	Inverse distance weighted	2.70	29.65		
	Ordinary kriging	3.35	31.96	1.34	23.99
Cr	Inverse distance weighted	1.96	21.21		
	Ordinary kriging	2.24	22.39	1.27	17.65
Cd	Inverse distance weighted	0.36	4.33		
	Ordinary kriging	0.39	4.34	1.16	3.78
Ni	Inverse distance weighted	0.88	9.91		
	Ordinary kriging	1.01	10.00	1.21	8.19
Ba	Inverse distance weighted	17.95	182.05		
	Ordinary kriging	21.17	210.75	1.49	142.96

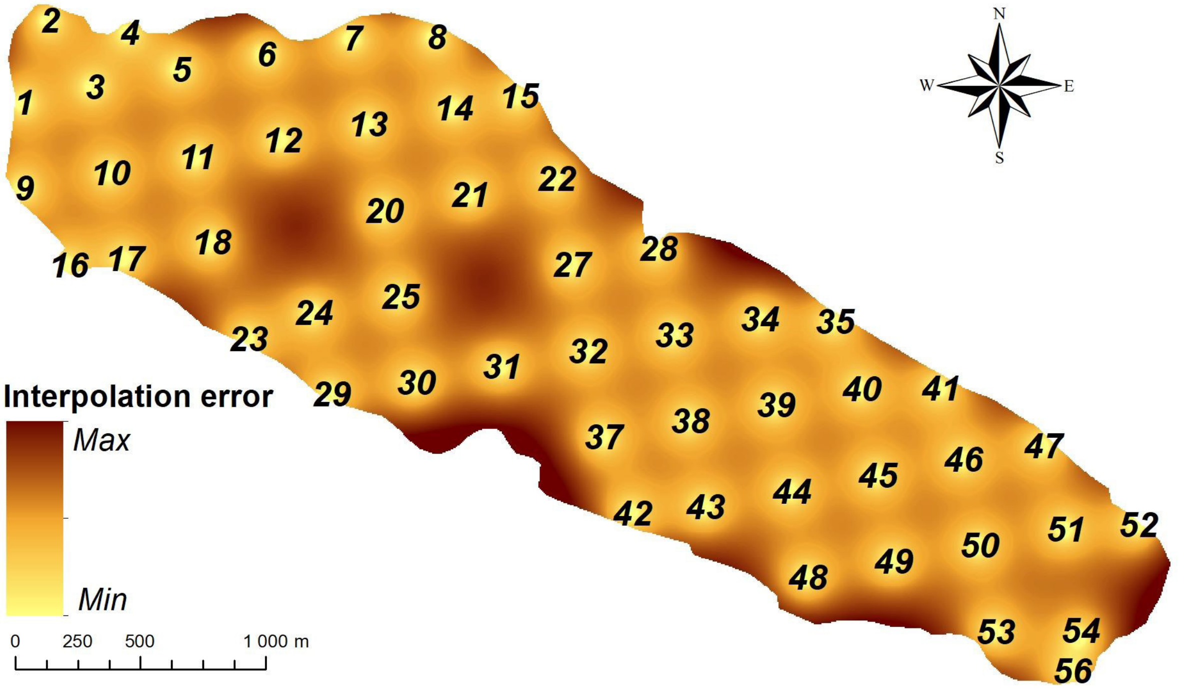

Figure 9. Map of interpolation errors for Ordinary Kriging method

around points with higher/lower contamination values then the close neighbourhood (mostly Figure 2, 3, 4, 5 and 6). Lower concentration of total trace elements content in coastal area has better representation in Kriging methods. The overall spatial distributions generated by Kriging seem slightly better in the case of the visual representation of the studied Dzierżno Duże water reservoir. However, the differences between the methods are not significant enough to determine the most appropriate method for generating spatial distributions of total trace elements content in bottom sediments.

Ordinary Kriging, besides RMSS and ASE, additionally allows generating prediction error maps. Prediction error maps were generated for all studied trace elements. As the output was similar for each of the trace elements, a schematic map of prediction error was created for purpose of this study (Figure 9). Noticeably, the values decrease together with the distance to the entry point. The highest errors were observed between points 18 and 20, 25 and 27. Significant errors also occurred in the southern-eastern, central (near points 30, 31, 37) coastal area. The prediction error map can be a solid base for future studies. The afore-mentioned areas should be additionally examined in terms of total trace elements content. These points should greatly increase the accuracy of spatial distributions.

CONCLUSIONS

The epresented studies allow for drawing the following conclusions:

1. Bottom sediments of water reservoir Dzierżno Duże are heavily contaminated by trace elements (mostly Cd and Zn).
2. The most contaminated area is located mainly in the north and south. Contamination can be related to water flow. Lower concentrations of trace elements were observed in the coastal area.
3. Coefficients of determination were high between trace elements (except for Ba).
4. High contamination values are probably caused by "Kanał Gliwicki", which is contaminated mainly from anthropogenic sources (chemical industry).
5. Th eaccuracy of Inverse Distance Weighted interpolation method is better than in the case of Ordinary Kriging.
6. The visual quality of the obtained spatial distributions was better for Ordinary Kriging than Inverse Distance Weighted.
7. The differences in both accuracy and visual quality of studies interpolation methods were not significant enough to clearly determine the most appropriate method for the purpose of determining total trace elements content in bottom sediments of Dzierżno Duże water reservoir – they can be used interchangeably.
8. Prediction error map can be a solid base for planning of future studies.
9. Spatial distributions can be crucial in proper processing and interpretation of the data obtained in environmental studies. The acquired spatial distributions allow for better understanding of the studied Dzierżno Duże water reservoir.

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