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Geostatistical Interpolation with GIS in Mapping Heavy Metals Concentrations for Preliminary Site Investigation of Old Mining Waste Dump

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Abstract: *Rehabilitation of mining waste facilities, in order to be successful, requires very good knowledge of the contamination levels in the soil. Soil characterization is not an easy process and demands the implementation of a variety of tasks, which include both precise data collection and analysis. Preliminary screening of the area, is often necessary because it leads to valuable conclusions which will determine the range of soil contamination as long as the selection of the right remediation and reclamation methods. Heavy metals can be considered as the key factors of soil contamination. In order to examine the concentrations level of them, data from soil samples have been collected and spatial interpolation methods (Ordinary Kriging and Inverse Distance Weighting) combined with GIS analysis, have been used in order to examine, how they are being distributed among the soil. Maps which display the prediction of the heavy metals concentrations have been produced and the Prediction error was examined.*

Keywords: *Kriging, IDW, Low-cost Site Investigation, Tailings Dump Remediation, GIS, Contamination Mapping, Sustainability*

1. INTRODUCTION

Mining Waste Dumps could be characterized as a necessary evil of mining, due to the environmental concerns they cause. Unfortunately, until a few decades ago, the environmental standards imposed by the legislation were almost non-existent. However, in recent years, the legislation has become too rigid both for the establishment and the operation of these sites. The European Union, in order to promote sustainability, has issued a specific Directive on the environmental specifications to be met by Tailings Management Facilities [1]. This has led to the need to close down old sites and create new ones that are in line with the Environmental Policy. Mining companies, usually are entrusted with the obligation to rehabilitate and restore these old areas (Principle “The polluter pays” Directive 2004/35/EC on environmental liability with regard to the prevention and remedying of environmental damage) [2].

Site Characterization of Old Mining Waste Dumps is not an easy process. A lot of factors have to be considered. Before the process of the Soil Remediation, knowledge of the extent of the contamination is necessary. A broad study should be carried out to confirm the selected methods of rehabilitation and reclamation. Usually, before that a smaller study is being executed for screening and identifying the situation. Soil samples are collected from various depths and chemical analyses are held. Geostatistical tools can be very useful in order to estimate the distribution of the pollution. The combination with the Geographical Information Systems can provide important conclusions of the area.

This paper could be considered as a part of Site Investigation and Soil Characterization of the study area. The main objective is to evaluate and highlight the use of Geostatistical Interpolation Methods in the distribution of the soil elements. The tasks are the production of maps and the analysis of the sample’s data in order produce useful conclusions for the area. Another important issue is to evaluate during the remediation process the depth of the contamination. Knowing this we can determine the amount of soil that has to be removed.

Two spatial methods were chosen in order to compare each other, and to find the optimum. The first was Ordinary Kriging and the second was Inverse Distance Weighting. GIS applications in Geo-statistics can provide excellent low-cost methods for Site Investigation. Collected data is analyzed, and visualized in way that produces quick and accurate conclusions. This is in the range of the Best Available Technique that companies seek to implement in remediation projects.

2. REVIEW OF STUDY AREA

The Old Mining Wasted Dump is located approximately 2.1km North East of Olympias, a village in Chalkidiki, North Greece, and it is part of Mines of Kassandra a large number of Mining facilities. Mining activity has been reported in the wider area since ancient times. Many archeological foundations confirm that during the Hellenistic and Roman period the area was well known for its Gold, Silver, Zinc and Lead. In contemporary times, the mining and processing of mixed sulfur ores began systematically at the end of the

19th century by the Ottoman administration. Today, the Site of Olympias operates as part of Mines of Kassandra. They include, mines of base (Zn, Pb) and precious (Au, Ag) metals and also a sophisticated Flotation Plant. Operations in Mines of Kassandra, include also Site of "Stratoni", Site of "Skouries" and Site "Mantem-Lakos" which exploit mines of mixed sulfur ores, logistic and management facilities, flotation plants, and Tailings Management Facilities with high environmental and legal specifications.

In 1976, the former mining company of Mines of Kassandra, built the Old Mining Wasted Dump, as an area for the disposal of tailings of Olympias mine and Olympias Flotation plant, in the form of pulp, and operated until 1995.The area covers around 30ha, and it is estimated that around 2.4Mt of tailings were aggregated. In 2012, the environmental program was launched for the remediation and reclamation of the area. The program includes the following tasks 1) the removal of the old waste 2) the site investigation and the research for the level of the soil contamination 3) the implementation of the right remediation process 4) the restore of the area back to natural environment by installing natural vegetation 5) the evaluation of the success of the program.

Today, the first task, which requires the removal of the tailings, has occurred for half of the area, except a small part on the north-east area, and the first soil characterizations programs are taking place. The "cleaned" from tailings area is about 15.718ha. The Figure 1 shows the borders of the area. The analysis occurred on the "cleaned" area where the remediation process has begun. This geostatistical method will contribute to determine the so called "depth of remediation", how much soil should additionally be removed.

3. MATERIALS AND METHODS

3.1 Soil Sampling

A drilling program was launched between July and August 2016. The program included 23 drillings (Table 1 shows the coordinates of the drillings) in the cleaned area from which soil-samples were taken. The drill-holes were drilled with mobile drilling equipment COBRA 148/248 and with cores that have diameter 32mm, 52mm and 82mm. In case of drilling in unconsolidated water-saturated material, steel liners were used to prevent caving. The liners were removed after the drilling and sampling was completed. Soil samples have been collected every 0.5 meters until hard-rock soil was found. The samples were kept in special plastic bags, which were clearly identified with the borehole number and depth., and they were delivered to the laboratory for chemical analysis and evaluation.

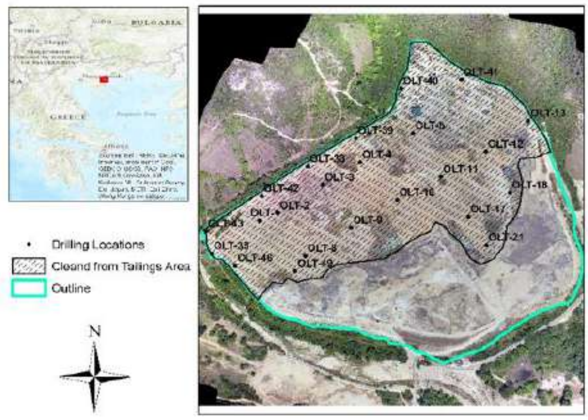

Fig -1: Drilling Locations of Olympias Old Mine Waste Dump

The chemical analysis of the samples involved solubilization with a mixture of HCl /HNO3 in a 1: 3 ratio (royal water) and the determination of 12 elements As, Ca, Cd, Cr, Cu, Fe, Mg, Mn, Ni, Pb, Sb, Zn as well as the Ph and Total Solids. The analysis of the data was performed on the Atomic Absorption Spectrometer PERKIN ELMER 2100 using flame and hydride generator (As). Sulfur analysis was performed on a LECO CS-200 analyzer. In addition, various sample-tests took place like Determination of acid-base balance (Sobek method) and chemical extraction determination test (EN 12457.02, EN 12457.04, CEN/TS 14405, EA NEN 7375:2004).

Table 1: *Drilling Locations in EGSA'87 (Greek Geodetic Reference System)*

DRILLING NAME	*E(m)*	*N(m)*	*Z(m)*
OLT-1	478194.248	4494979.266	39.37
OLT-2	478228.737	4494993.869	38.791
OLT-3	478313.603	4495049.091	38.69
OLT-4	478384.743	4495093.712	38.558
OLT-5	478484.507	4495150.137	38.665
OLT-8	478280.734	4494910.144	35.159
OLT-9	478366.995	4494964.867	34.182
OLT-10	478454.543	4495019.438	36.198
OLT-11	478537.727	4495065.575	35.502
OLT-12	478622.36	4495114.242	36.77
OLT-13	478702.94	4495174.820	44.952
OLT-17	478589.349	4494986.403	33.653
OLT-18	478668.165	4495033.927	34.276
OLT-21	478623.724	4494931.117	32.986
OLT-35	478103.464	4494914.483	40.043
OLT-38	478285.321	4495085.229	44.233
OLT-39	478376.675	4495140.511	45.915
OLT-40	478461.893	4495237.861	46.417
OLT-41	478576.048	4495255.112	50.895
OLT-42	478198.088	4495027.497	44.169
OLT-43	478092.765	4494957.940	41.94
OLT-46	478148.04	4494890.750	44.789
OLT-49	478261.983	4494880.228	34.982

In this research, we deal only with the results from the chemical analysis of As, Pb, and Mn which we considered as the elements with the most crucial impact in the soil contamination. Although many drillings reached even 6-meter depth, the Samples from 4 soil layers were chosen (from surface until 2-meter depth and every 0.5m). This was decided because samples from deeper than 2m showed extremely low concentrations and we assumed that there is no need to do further analysis for them. Table 2 and Table 3 demonstrates the heavy metal concentrations for each soil layer. In total, results from 89 samples were used for the geostatistical analysis.

Table 2: *Results of Chemical Analysis for Soil Layers 0-05m and 0.5-1.0m (values in mg/kg)*

DRILLING NAME	*Depth 0-0.5m*			*Depth 0.5-1.0m*		
	Pb	*Mn*	*As*	*Pb*	*Mn*	*As*
OLT-1	38	1069	76	40	740	10
OLT-2	42	398	10	13	376	40
OLT-3	109	659	81	200	330	60
OLT-4	56	656	10	19	728	10
OLT-5	336	1904	725	49	489	10
OLT-8	55	389	10	10	286	10
OLT-9	10	522	10	10	389	10
OLT-10	137	611	296	42	452	14
OLT-11	284	1790	950	400	2100	950
OLT-12	40	1350	53	55	1306	40
OLT-13	644	3854	1346	50	603	35
OLT-17	100	879	137	36	545	10
OLT-18	2327	10090	4350	401	1918	578
OLT-21	232	1096	278	31	356	10
OLT-35	1397	19450	2452	588	7578	1076
OLT-38	364	3253	927	72	550	63
OLT-39	418	4436	1028	39	288	78
OLT-40	746	8234	2084	384	3979	938
OLT-41	14470	4499	6235	4339	2556	3536
OLT-42	597	5210	1447	179	1524	384
OLT-43	122	766	124	117	492	46
OLT-46	699	7435	3046	219	1539	10
OLT-49	182	432	63	127	380	38

Table 3: *Results of Chemical Analysis for Soil Layers 1.0-1.5m and 1.5-2.0m (values in mg/kg)*

DRILLING NAME	*Depth 1.0-1.5m*			*Depth 1.5-2.0m*		
	Pb	*Mn*	*Pb*	*Mn*	*Pb*	*Mn*
OLT-1	110	840	110	840	110	840
OLT-2	15	698	15	698	15	698
OLT-3	77	385	77	385	77	385
OLT-4	25	735	25	735	25	735
OLT-5	81	860	81	860	81	860
OLT-8	10	215	10	215	10	215
OLT-9	10	468	10	468	10	468
OLT-10	43	710	43	710	43	710
OLT-11	22	381	22	381	22	381
OLT-12	10	737	10	737	10	737
OLT-13	221	1817	221	1817	221	1817
OLT-17	111	932	111	932	111	932
OLT-18	104	744	104	744	104	744
OLT-21	122	704	122	704	122	704
OLT-35	126	597	126	597	126	597
OLT-38	38	414	38	414	38	414
OLT-39	43	622	43	622	43	622
OLT-40	10	324	10	324	10	324
OLT-41	1454	1275	1454	1275	1454	1275
OLT-42	21	424	21	424	21	424
OLT-43	157	1015	157	1015	157	1015
OLT-46	No values	91	No values	91	No values	91
OLT-49	69	311	69	311	69	311

3.2 Geostatistical Analysis

The Geostatistical analysis was implemented in the ArcGIS suite of ESRI and by using the Geostatistical and the Spatial Analyst Extension. The analysis was executed separate for each soil layer and for each heavy metal, and by using two different methods: 1) Kriging and 2) Inverse Distance Weighting (IDW). In total, there were produced 24 Geostatistical rasters (3 Heavy Metals 'As, Pb, Mn' x 4 Soil Layers 'Depths: 0-0.5m, 0.5-1m,1-1.5m, 1.5-2m' x 2 Methods 'IDW, Kriging') for the graphical representation. In addition, each of the Geostatistical raster is combined with a summary statistic table, a histogram and a semi-variogram.

GIS applications provide an all-in-one software suite that it can be used for Geostatistical Interpolation. All steps of Interpolation (including statistical analysis) can be handled with it. Inverse Distance weighing

is based on the assumption that values that are spatial close with each other are more likely than those that are farther apart. Kriging method distinguishes from other interpolation methods due to the fact that it is based on a statistical model that uses autocorrelation (the statistical relationships among measured points) [3].

The steps of Geostatistical Analysis were: 1) Data was under statistical analysis and was examined for outliers, homogeneity, and the distribution of the data. 2) semi-variograms were produced for each case, and the spatial correlation between the points was ensured 3) the IDW and Kriging models were executed and the surfaces were created 4) The variance surfaces were explored in order to evaluate and check the model 5) a summary statistics table was added in each case 6) maps displaying the distribution of the error, for the Kriging method were created.

4. RESULTS

Figures 2,4,6 display the distribution of each Heavy Metal with each method. In addition, Mean Error and the Root Mean Square Error have been also calculated for each case. The values of the drillings are being displayed, as labels in each map. Figures 3,5,7 show the distribution of the Standard Prediction Error from the Kriging method, as long as the Semi-variograms and the Normal Q-Q Plot. Same symbology was chosen in order to be easier to extract conclusion from the analysis.

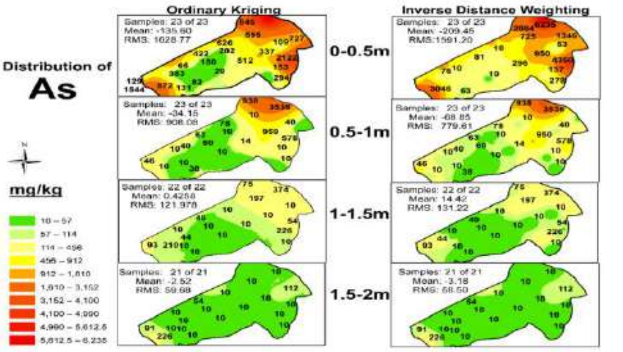

Fig -2: As Concertation Distribution for each Soil Layer

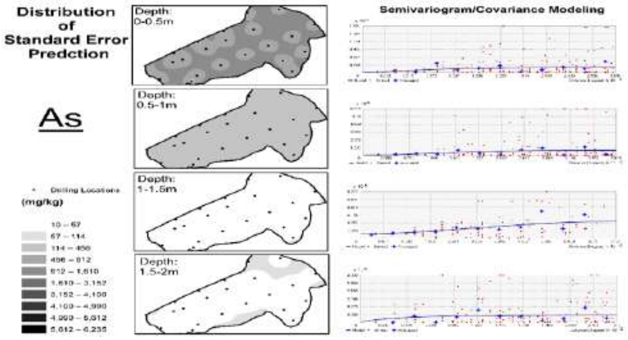

Fig -3: Distribution of As Standard Error for each Soil Layer

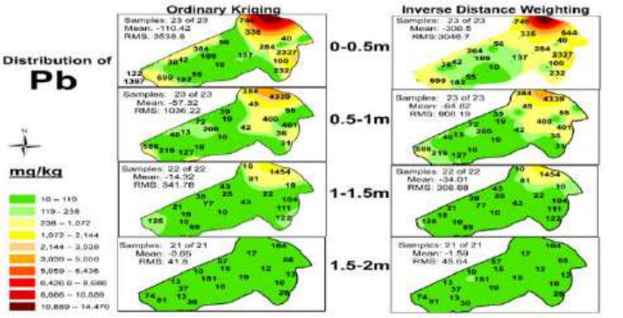

Fig -4: Pb Concertation Distribution for each Soil Layer

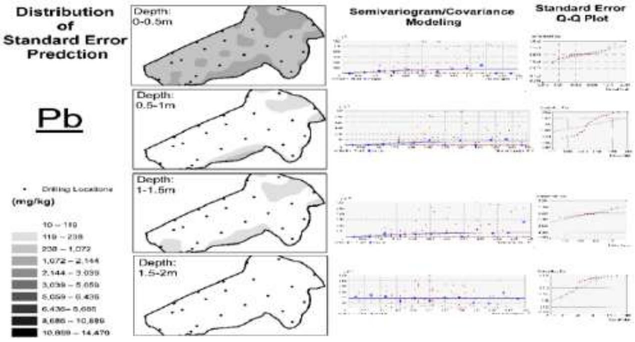

Fig -5: Distribution of Pb Standard Error for each Soil Layer

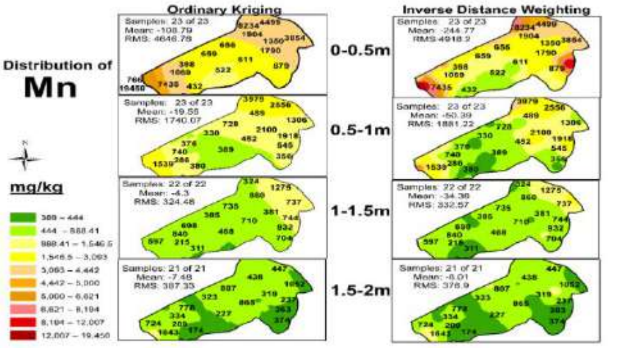

Fig -6: Mn Concertation Distribution for each Soil Layer

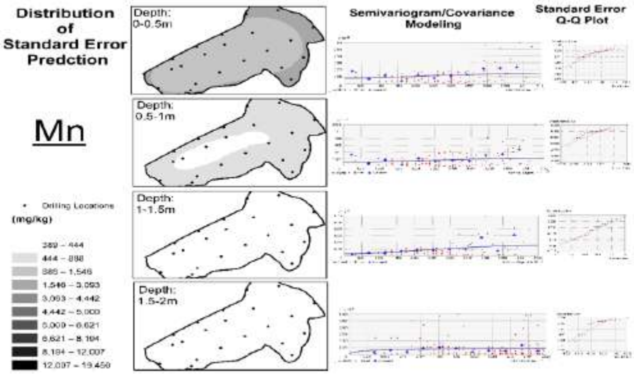

Fig -7: Distribution of Mn Standard Error for each Soil Layer

5. DISCUSSION

The results have been examined in order to determine the contaminated land. As per Flemming, contaminated Land is the land that contains substances that when present in a sufficient quantities or concentrations are likely to cause harm, directly to man, to the environment, or on occasions to other targets [4]. Drilling OLT-41 has the most increased concentrations among the rest. This has led to the deduction that during the removal of the waste in the first phase, some tailings in the north-east area has been left. The high-values of this location, affected the mean error of the geostatistical model.

Values in the drillings of the first and second soil layer have higher variance than the others. This explains the higher and more irregular distribution of the Standard Prediction Error. The same deduction comes out also from the examination of the semi-variograms. In the third and fourth Soil layer, where values are in general terms without major differences, the Standard Error is approximately minimized.

If more drillings were carried out, and therefore more data were exported, the distribution using Interpolation Models would certainly be more accurate. However, the execution of drilling programs is a very expensive procedure. Thus, data has to be acquired and analysed in the most efficient way, which minimizes the cost, and provides the best results. In this case, our scope was to determine how much more soil should be removed. If a program with more drilling was launched, the results would be more accurate but the conclusions for our scope might be the same and more money would have been spent without any particular reason.

One of the most critical issues in examination of Land Contamination is to know the accepted limits of the heavy metals concentrations. Unfortunately, Greek legislation doesn't have specified limits of heavy metals concentrations for soils. Further research must occur in order to determine those limits. Parameters concerning the surrounding area and the soil's physiognomy must be taken into account. Soil concentrations limits can be implemented as indicators in order to examine which area is above contamination limit and which is not.

6. CONCLUSIONS

In general, Interpolation tools have been found really useful in the site examination of an Old-Mining Waste Dump. For an area of 15.178ha, analyzing data from 23 locations has produced very good results. The number of the drilling was enough to get the conclusions we want.

The areas where concentrations are high have been distinguished. The top two soil layers (until 1m depth) , are those that we estimate that need to be removed. Below them concentration levels seem to be fine. If the Concentration limits were specified, then the depth which remediation must reach could easily have been determined. Maps are easy to be used, even from not skilled personnel, and give a very good outlook.

The concept of the paper, was a preliminary assessment of the level of the contamination. In general, for the determination of the remediation process, not only the distribution of the heavy metals but a lot of geological and environmental factors should be considered. However, heavy metals have the most crucial impact in the soil contamination. Thus, for a preliminary assessment heavy metals are the key factors.

In a nutshell, we strongly recommend the usage of Interpolation methods in combination with Geographical Information Systems, for the estimation of Heavy Metal Concentrations in Mining Waste Dumps and as a part of Site Characterization Procedure. It is a low-cost, easy to execute and fast method, which provide accepted results and it can be considered as the Best-Available-Technique.

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