Spatial Variability of Arsenic in Agricultural Area near Sofia

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Introduction

The heavy metals pollution of the soils in the Kremikovtzi region, representing a part of the city-round territory of the Bulgarian capital Sofia, is investigated since beginning of 70ties. The Kremikovtzi region is a productive agricultural area in the Sofia plain supplying the capital in particular with vegetables. All results confirmed contamination of the soils with heavy metals (lead, zinc, cadmium, and copper) and arsenic. The main source of pollution was assumed to be the metal smelter giant with a number of technologic productions "Kremikovtzi". The contamination due to the smelter is a result mainly of atmospheric depositions of dust and aerosols. Other sources of contamination are the tailing dam of the "Kremikovtzi" smelter (120 ha, South-Western of Botunetz village) and the landfills of solid wastes on a sum territory of about 150 ha. There are a lot of data concerning soil pollution in this region as mentioned above. Unfortunately they are fragmentary to some extent and not precisely spatially bound. [1, 2, 4,8,9,10].

The objective of this investigation was to introduce and apply a modern methodology for area assessment and control of sustainable soil management in Bulgaria - advanced geostatistical methods for soil evaluation. This methodology is performed in the case study on the "Kremikovtzi" site which represents an agricultural area with serious environmental problems. The main goal is to obtain a more lucid view on the recent ecological state of the soils by using spatially referenced data which can be further combined with GIS.

Materials and methods

The study area is located around the Kremikovtzi smelter. The plants are situated on an area of 2400 ha in the Northeast part of the Sofia plain, nearly 18 km Northeast from Sofia center. The north part of Kremikovtzi region is situated on the South slopes of the mountain Stara planina and the South part is extended into the Sofia plain. This study is carried out in proximity of Kremikovtsi smelter near Sofia in lands of the village Gorni Bogrov. Surface of studied lands is about 250 ha. Main crop are wheat, barley and oat. Some area with vegetables production exists, too.

Soil sampling is done in 200 m grid and 87 soil samples were collected. GPS technique was used for determination of precise position of sampling points. Every soil sample represent mixture of one sample from the point with precise coordinates and four samples situated at 2.5 m in North, South, East and West from central point. Soil samples are analysed in AAS hydride system. Geostatistical analyses were made with GS+ 5.3 geostatistical software package.

Studied area near the village of Gorni Bogrov has mixt relief. This type of relief is suitable for study of pollutant movement in large scale. Several soil types and different level of soil pollution is good basis to study arsenic spatial variability. Susceptibility of erosion of soils presented in the area could cause arsenic pollution of rivers water.

Kremikovtzi region is situated in the central part of the Sofia Plain which is characterized with a mild continental climate - cold winter and hot summer. The seasonal rainfall distributional differences are ample. Lowest rainfalls occur in winter. In contrast, highest rainfalls occur in late spring, somewhere about May [3]. Total annual precipitation is 566 mm. In summer, air mass circulation entails great fluctuations in the occurrence of wet and dry periods. The hottest months are July and August which provokes the highest deficiency of air moisture (D) and evapotranspiration.

Water balance is positive in winter and in early spring, from October to March. In this period are formed the main quantities of surface water, percolating trough the soil profile. Their value reaches 126 mm (1260 m^3/ha). During the summer insignificant infiltration flow is formed after prolonged or intensive rainfalls.

Main contaminant within the studied area was lead, followed by arsenic. Permissible concentration level (PCL) for As - 25 mg/kg dry soil.

Results and discussion

In Gorni Bogrov area 62 basic sampling points and 25 points from nested sampling points were studied. *Table 1* shows summary statistics for As values. It includes measures of central tendency and variability.

Obtained data were good basis for calculation of semivariograms and kriging. Very high coefficient of determination obtained (*Figure 1*), shows the high credibility of kriging assessment and elaborated maps.

Kriging of obtained data make possible to obtain estimated data for heavy metals content in soils not only in sampling points, but in non sampled soil between them. Good variograms are very important to the reliable and credible results in interpolated area. Block kriging 16x16 was used for all analyses.

Table 1: **Summary statistics for As with data without outliers**

Average	90.80
Stnd. deviation	45.8
Minimum	13.7
Maximum	152.8

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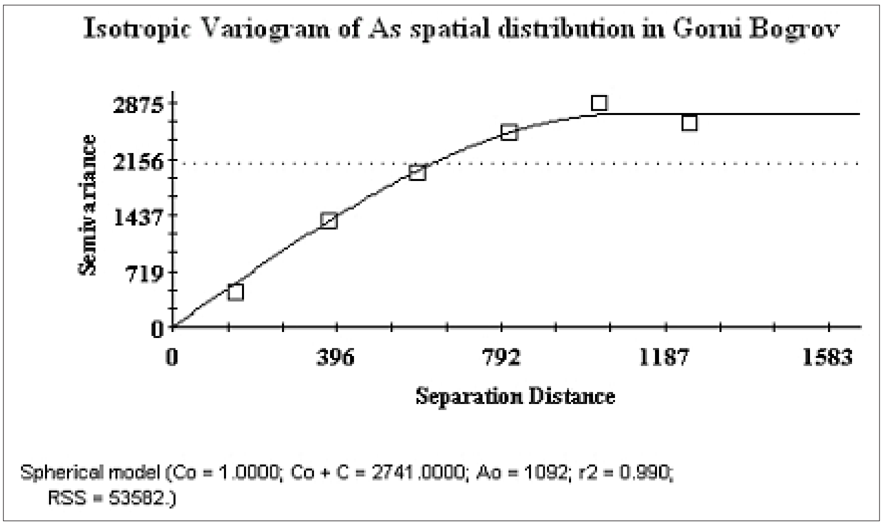

Figure 1: **Variograms of As spatial distribution in soils**

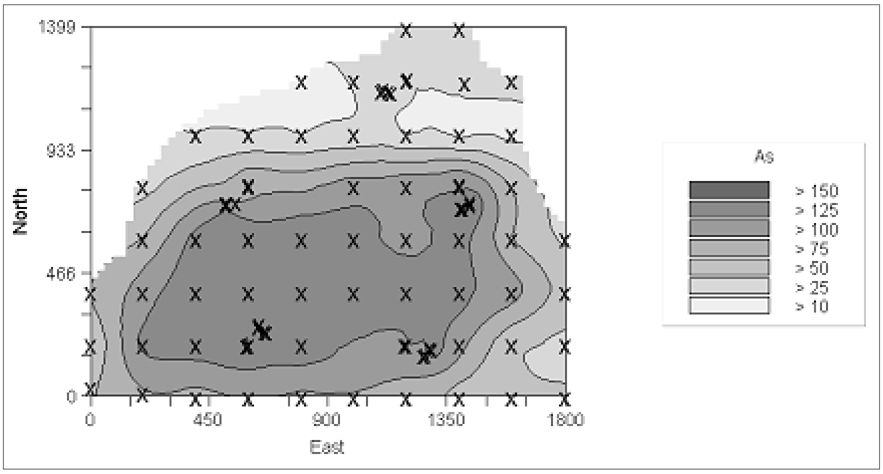

Figure 2: **Kriging maps of As spatial distribution in soils**

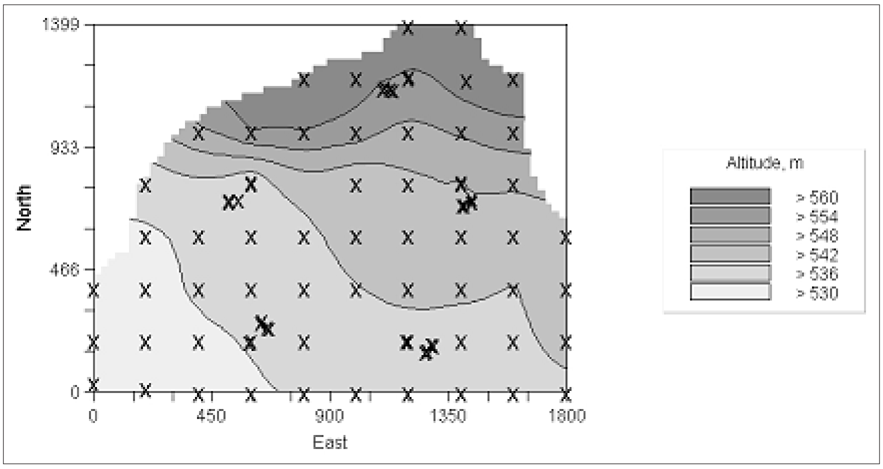

Figure 3: **Kriging maps of altitude of sampling points**

Obtained kriging maps show different distribution of arsenic content in studied area - non polluted part of the North of the area and heavily polluted southern part (*Figure 2*). These two parts coincide with main soil types and the relief (*Figure 3*). Studied area is characterised with very high As content - more than 125 mg.kg^{-1}.

Type of spatial distribution of As in soils and relief of the area make possible some secondary pollution of area by transfer of soil particles by erosion. The lower polluted part of the studied area is covered mainly with Alluvial Fluvisols. Our study shows that these soils have greater filtration coefficient and some leaching of the As down the soil profile in ground waters and after that in the river is possible.

The texture of Chromic Luvisols in Gorni Bogrov subregion was heavy clay. Filtration Coefficient (profile permeability) especially under 20 cm depth (K_f = 0,01 m/24h). The water holding capacity of the plough layer was 830 m³/ha.

The soil texture of Alluvial Fluvisols was light. The soil have a high water permeability (K_f = 0.838 m/24h in Ah and K_f = 0.566 m/24h in the layers). The water retention capacity is low. The water holding capacity of the plough layer was 600 m³/ha.

Filtration coefficient (soil profile water permeability) K_F was determined by Ernst's method (Netherlands), standardized in Bulgaria for investigations due to irrigation and drainage design works [5].

Universal soil loss equation (USLE), adapted for use in Bulgarian conditions was applied for the soil erosion losses calculations [6.7]:

In *Tables 2* and *3* it is possible to see data showing that higher erosion in the Chromic Luvisol is not a hazard for secondary pollution with arsenic - due to the low content of As in soil. In same time on Alluvial Flivisols only 140-170 g As.ha^{-1} is possible to be transferred by erosion.

In same time Alluvial Fluvisols could be a source of secondary pollution for the underground waters and rivers due to their low water retention capacity and relatively high filtration coefficient.

As shown on the *Figure 4* arsenic content increase down soil profile. This is due to leaching of arsenic with water down soil profile. High underground waters could be reached by rain waters during most wet season.

The contamination by arsenic in the case of Gorni Bogrov is mainly by the influence of pedogenic and very low technogenic excess in the soil.

The area of Gorni Bogrov present very well the spatial distribution of As. The pollution with As is detected mainly in the lower part of the field.

Table 2: **Soil erosion losses in Gorni Bogrov - locations with different topographic characteristics**

Location	Soil erodibility factor	Topographic factor	Cover & management factor	Soil erosion loss **(t/ha per year)**
Chromic Luvisols				
A	0.029037	0.3816	0.40 (maize)	6.96548
B	0.029037	4.3947	0.22 (wheat)	44.09764
C	0.029037	1.6371	0.22 (oat)	16.43238
D	0.029037	0.3503	0.22 (oat)	3.515961
E	0.029037	0.8367	0.22 (oat)	8.390073
F	0.029037	1.2095	0.22 (wheat)	12.13870
Alluvial Fluvisols				
A	0.017471	0.2549	0.22 (wheat)	1.535842
B	0.017471	0.3107	0.22 (wheat)	1.873373

Table 3: **Variations of possible losses of arsenic by erosion from different soils in Gorni Bogrov - locations with different topographic characteristics**

Soils	Average	Minimal	Maximal
Chromic Luvisols	320-4000	48-604	537-6740
Alluvial Fluvisols	140-170	21-25	235-286

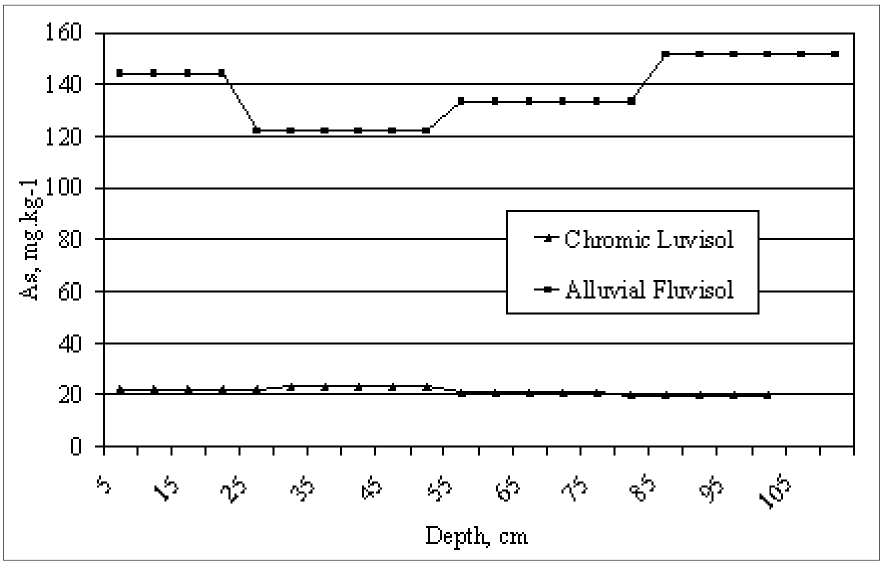

Figure 4: **As content in soils depth profiles**

Higher part of the Gorni Bogrov area is not polluted and As content is low. It could be due to the different soil type and the different parent rocks. Soils in the lower part of G. Bogrov are formed under the influence of rivers originated from the mountain. Other parts of studied area are more recently formed and soil formation process was different (not alluvial). This could be seen very well on the kriging maps.

The present study has some advantages predominantly in the localization of contaminated sites and the opportunity to show the results by computer models.

Conclusion

Obtained results of As patial variability of soils showed that:

❶Maximal contamination of Gorni Bogrov arable land by As is found in the lower part of studied area on Alluvial Fluvisols.

❷The places used for vegetable production of the farmers are located in polluted area.

❸The soil on the northeast part of the field is practically non- polluted.

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