Soil Contamination by Heavy Metals in the Botevgrad Valley Region, Bulgaria

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Abstract

Heavy metal pollution in Bulgaria is a complex issue linked to the country's industrial and mining history. While the industrial progress has been made, much work remains to protect human health and environment. Soils in the Botevgrad Valley show that heavy metal pollution occurs in only in limited places, with concentrations exceeding permissible levels mainly near urban areas affected by anthropogenic activity. Soil contamination by copper and zinc is also observed in areas with intensive agriculture, due to the use of pesticides and mineral fertilizers. Increased levels of heavy metals such as lead and cadmium are found near several roads, especially along unregulated landfills, industrial zones, and scattered household waste.

Keywords: soil pollution, spatial variation, lead, zinc, cadmium, copper

Introduction

Heavy metal pollution in Bulgaria remains a serious environmental concern, primarily resulting from industrial activities, transportation, agriculture, and military exercises. The main pollutants include copper, lead, zinc, cadmium, and arsenic. These contaminants tend to be concentrated around industrial zones, major highways, and intensively cultivated areas. Approximately 43,600 hectares of soil in Bulgaria are contaminated with various chemicals, including heavy metals (Todorova, 2002).

Soils contaminated with heavy metals have been relatively well studied and mapped. Atanasov et al. (2015) pointed out that no statistically proven cases of heavy metal contamination after 1994 were established in Bulgaria

Soil contamination by heavy metals can adversely affect ecosystems, human health, and agricultural productivity (Rashid et al., 2023). In forest ecosystems affected by urbanization, the primary barrier to heavy metal penetration into deeper soil layers is forest litter, where metals tend to accumulate (Doychinova et al., 2005). Naturally, heavy metals exist as mineral ores in rocks, in various chemical forms from which they are recovered as minerals. The risks of deterioration in soil productivity and ecological quality are caused by natural degradation processes under modern conditions, as well as anthropogenic activities.

Industrial activity impacts agriculture mainly through waste deposition on soils (Dinev et al., 2015; Tsolova et al., 2016; Yotova et al., 2018; Malinova et al., 2022).

There are no significant industrial pollutants in the territory of the Botevgrad Valley. According to the Botevgrad Municipality report, some soils are contaminated due to uncontrolled disposal of household, construction, and garden waste, as well as unregulated landfills (Botevgrad Municipality, 2005). The highest pollution levels are near the industrial zone of Botevgrad, the main road E79, and waste landfills at the municipal landfill in the area "Temosh," a plot in the village of Skravena, 3 km northwest of Botevgrad. Additionally, pollution is observed near the non-hazardous waste landfill in “Sadinata”, north of Botevgrad.

Part of the municipal soils is affected by uncontrolled landfills of household, construction, garden waste, and animal husbandry waste. The ecological status of the soil cover on the "Razhana" ridge suggests possible heavy metal contamination due to activities at the "Eliseyna" flotation plant.

Table 1. *Standards for trigger values (maximum allowable concentrations) and intervention concentrations for some heavy metals in Bulgaria, (defined as total content in mg/kg dry soil when extracted with aqua regia)*

Heavy metals	pH	Trigger values (Maximum allowable concentrations)			Intervention concentrations
		Arable land mg/kg	Permanent lawns mg/kg	Correction coefficients	mg/kg
Cd	<6.0 6.0–7.4 >7.4	1.5 2.0 3.0	2.0 2.5 3.5	1.3	12
Cu	<6.0 6.0–7.4 >7.4	80 150 300	80 140 200	1.2	500
Pb	<6.0 6.0–7.4 >7.4	60 100 120	90 130 150	1.3	500
Zn	<6.0 6.0–7.4 >7.4	200 320 400	220 390 450	1.3	900

Soils vary in their resistance to chemical pollution, which is reflected in the different concentration thresholds: "quality values," "trigger values," and "intervention concentrations." These thresholds are determined based on various soil indicators, such as pH, soil texture, depth, and land use (arable land, permanent grassland, settlements, parks, sports grounds, and industrial sites) (Regulation Law 3, August 12, 2008, Council of Ministers of Bulgaria).

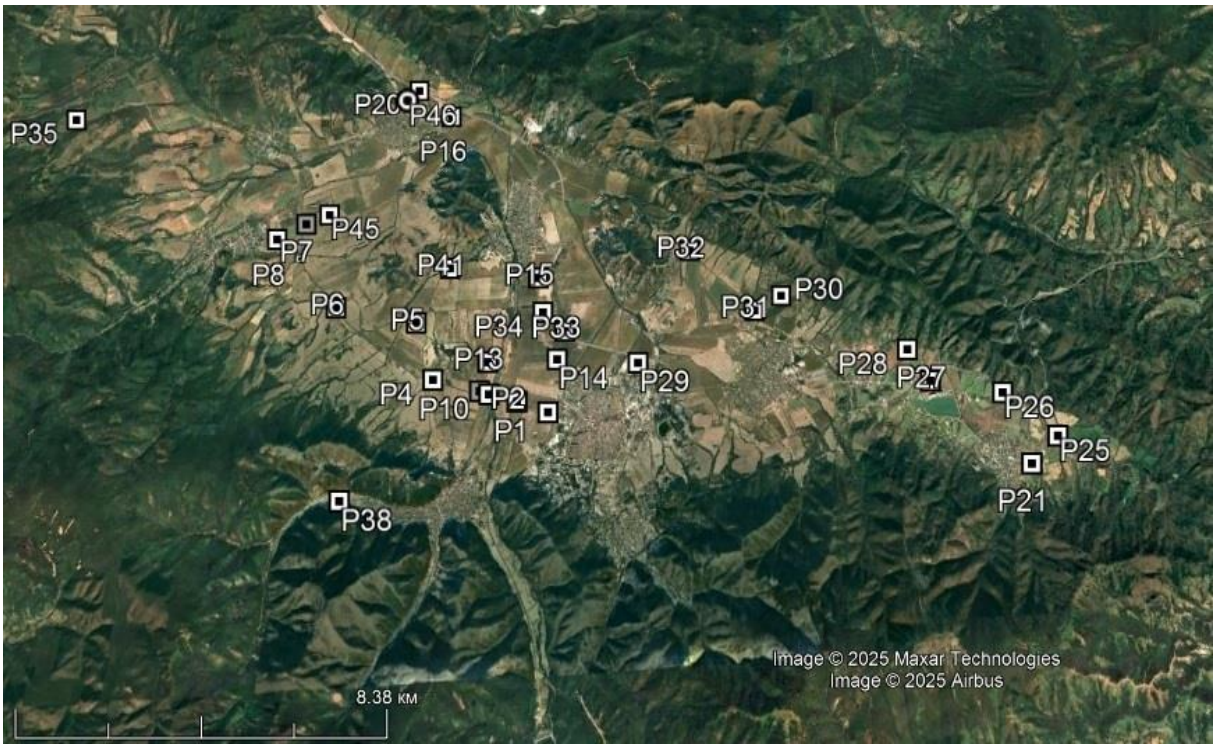


Fig. 1. *Sample plots in Botevgrad Valley (Bulgaria)– Google earth.*

Materials and Methods

The total survey area covers approximately 150 km², as shown in Fig. 1 including the main sample plots in the valley. In spring 2022, twenty-nine soil samples were collected from the surface A horizon and six samples from deeper horizons (B and C). The samples were mineralized using aqua regia (a mixture of HCl and HNO_3 in a 3:1 ratio). The concentrations of Mn, Zn, Pb, Cu, and Cd were determined using atomic absorption spectrophotometry (Perkin-Elmer Model Analyst 5000).

The soil pH was measured in water according to ISO-10390. For mapping purposes, ArcGIS 10.5 was used, employing the Inverse Distance Weighted (IDW) interpolation method within the Spatial Analyst extension. IDW assumes that the value at a location is more similar to nearby sample points than to distant ones. Only surface soil samples are used for interpolation maps. Statistical analysis was performed using MS Excel 2019.

Results and Discussion

The primary heavy metal pollutants affecting Bulgarian soils include lead (Pb), zinc (Zn), copper (Cu), and cadmium (Cd). These pollutants mainly originate from industrial activities particularly ferrous and non-ferrous metallurgy, chemical industries, and heavy traffic. In the Botevgrad area and surrounding territories, increased concentrations of heavy metals have could be found in the surface soil layers due to anthropogenic influences. Similar findings are noted in urbanized areas and regions under urban pressure (Doychinova et al., 2013). Studies also indicate a direct correlation between heavy metal concentrations in topsoil and proximity to settlements and roads (Pin et al., 2000).

Table 2. *Heavy metal content of soils in the Botevgrad Valley.*

Sample code	Profile/ Spot	Horizon	pH (H_2O)	Pb mg/kg	Cu mg/kg	Mn mg/kg	Zn mg/kg	Cd mg/kg
1	1 Spot*	A	6.0	32.8	143.4	1380	118.6	1.07
2	2 Spot	A	6.2	35.7	47.7	1495	130.1	0.97
3	3 Spot	A	6.2	35.7	41.2	2601	119.3	1.01
4	4 Spot	A	6.0	30.0	36.1	1268	116.9	0.86
5	5 Spot	A	7.0	25.0	33.3	1193	71.4	0.74
6	6 Spot	A	6.3	18.9	22.9	878	56.0	0.68
7	7 Spot	A	6.4	21.4	29.8	876	57.0	0.62
8	8 Spot	A	8.2	33.0	35.2	806	100.9	0.83
9	9 Spot	A	6.1	28.1	31.8	628	94.5	0.73
10	1 Profile**	A	6.0	71.9	75.4	3167	208.0	1.29
11	1 Profile	C1	6.7	49.8	59.5	3192	159.2	1.14
12	1 Profile	C2	6.8	35.1	44.8	2655	124.0	1.05
13	13 Spot	A	5.5	30.5	41.5	2030	114.0	0.97
14	14 Spot	A	5.5	38.9	51.1	1347	101.0	1.05
15	15 Spot	A	6.0	35.6	46.2	994	107.2	0.76
16	2 Profile	A	6.0	27.0	30.5	898	74.9	0.65
17	2 Profile	Bt	6.6	22.5	29.3	887	71.1	0.65
18	2 Profile	Bt	6.2	18.4	30.0	801	67.6	0.61
19	2 Profile	C	6.0	15.8	31.0	750	69.9	0.66
20	20 Spot	A	6.0	27.7	40.8	1235	83.9	0.75
21	3 Profile	A	5.2	27.3	41.6	2473	79.9	0.68
23	3 Profile	C1	5.5	16.0	35.3	1384	94.3	0.72
24	3 Profile	C2	5.5	16.4	33.0	1310	84.4	0.76
25	25 Spot	A	6.5	17.3	28.4	914	52.3	0.50
26	26 Spot	A	6.0	21.0	46.6	1276	82.5	0.70
27	27 Spot	A	6.0	34.0	73.1	1234	73.1	0.73
28	28 Spot	A	6.5	32.8	72.5	1348	74.7	1.07
29	29. Spot	A	6.5	30.0	47.1	5998	100.8	3.32
30	30 Spot	A	6.5	268.8	95.3	3045	288.6	1.75
31	31 Spot	A	6.0	98.4	113.5	1927	141.9	0.93
32	32 Spot	A	5.5	28.9	28.6	1242	81.2	0.57
33	33 Spot	A	5.5	29.4	44.5	1327	123.6	0.92
34	34 Spot	A	5.0	30.8	48.5	1342	119.6	0.90
37	37 Spot	A	5.8	31.8	35.8	1748	106.0	0.78
45	38 Spot	A	6.3	22.5	25.6	481	88.7	0.67
46	46. Spot	A	5.9	65.3	27.2	825	93.5	0.54

*- Spot is a place where only one soil sample were taken from the surface

**- Profile is a place where more soil samples were taken from different depth

In different soil types, heavy metals tend to precipitate at around pH 6.0 (Ganev, 1990). This suggests that the release of heavy metals from their inorganic compounds, making them accessible to plants, depends on soil pH. In soils with pH below 6.0, heavy metals become ionic and they are adsorbed onto soil colloids, making them more available for plant uptake. As pH values increase above 6.0, the accessibility of heavy metals to plants decreases. They form insoluble salts or precipitate as insoluble compounds – oxides, hydroxides, carbonates, phosphates, silicates, humates, etc. (Malinova, 2010). In the studied soils, pH values range from 5.0 to 8.2, with most soils being slightly acidic around pH 6, averaging 6.11 (Table 3).

Another key characteristic of heavy metals in soils is their persistence; they are tightly bound as non-degradable substances and are rarely released back into the environment. Heavy metals tend to accumulate mainly in the upper soil layer, which has the highest sorption capacity, hence their concentrations are primarily measured in the surface A horizon.

Table 3. *Descriptive statistics of the studied soils with heavy metal concentration and soil reaction.*

	pH (H2O)	*Pb* mg/kg	*Cu* mg/kg	*Mn* mg/kg	*Zn* mg/kg	*CD* mg/kg
Mean	6.11	39.01	47.17	1582	103.63	0.91
Standard Error	0.10	7.11	4.29	174.82	7.41	0.08
Median	6.00	30.00	41.00	1293	94.40	0.76
Mode	6.00	32.80	#N/A	#N/A	#N/A	1.07
Range	3.20	253.00	120.50	5517.00	236.30	2.82
Minimum	5.00	15.80	22.90	481	52.30	0.50
Maximum	8.20	268.80	143.40	5998	288.60	3.32

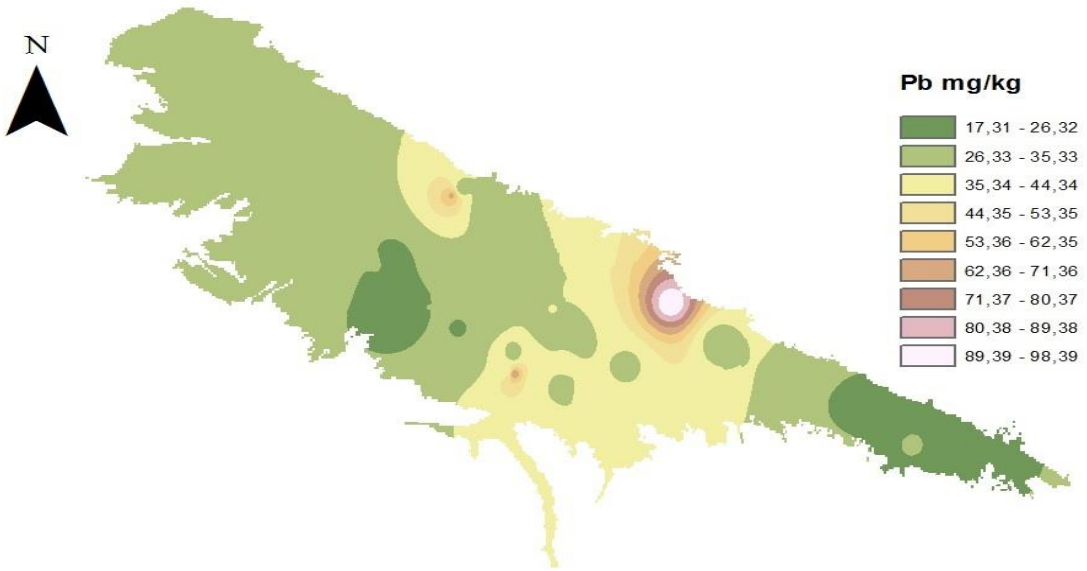


Fig. 2. *Interpolation map (IDW) of lead (Pb) content in Botevgrad valley - mg/kg.* (*Only surface soil samples are used for interpolation maps)*

The studied heavy metals include toxic elements such as lead (Pb), zinc (Zn), copper (Cu), and cadmium (Cd). Manganese (Mn) does not have established trigger values since its toxicity depends on various factors, and its total concentration cannot directly indicate contamination risk. Organic soil horizons tend to have higher manganese levels than mineral ones, with concentrations ranging from 100 to 2000 mg/kg, and sometimes exceeding 10,000 mg/kg (Vanmechelen, 1997). In Botevgrad soils, average manganese levels are approximately 1582 mg/kg, with maximum values reaching 5998 mg/kg.

The average lead (Pb) content in uncontaminated soils worldwide ranges from 10 to 67 mg/kg, with the Bulgarian average around 25 ± 15 mg/kg (Chuldzhiyan, 1989). A value of 26 mg/kg is considered natural. Elevated levels above the trigger value (30 mg/kg) are observed in the area of Trudovets village, where lead concentrations reach about 268 mg/kg, twice the permissible limit. The average lead level for the valley is 39 mg/kg (see Table 3). Potential sources of lead contamination include highways, roads, mines, and industrial enterprises, which can release lead into the environment. Car exhaust gases from vehicles using leaded gasoline also contribute to soil contamination (Fig. 2). Nearby, the *Eliseyna* station, involved in copper mining, has been associated with lead, zinc, and arsenic pollution, which can be transported via fine particles. Improper disposal of waste containing lead, such as batteries and paints, can also contribute to soil pollution. The highest lead concentrations are recorded around Trudovets village and town of Botevgrad.

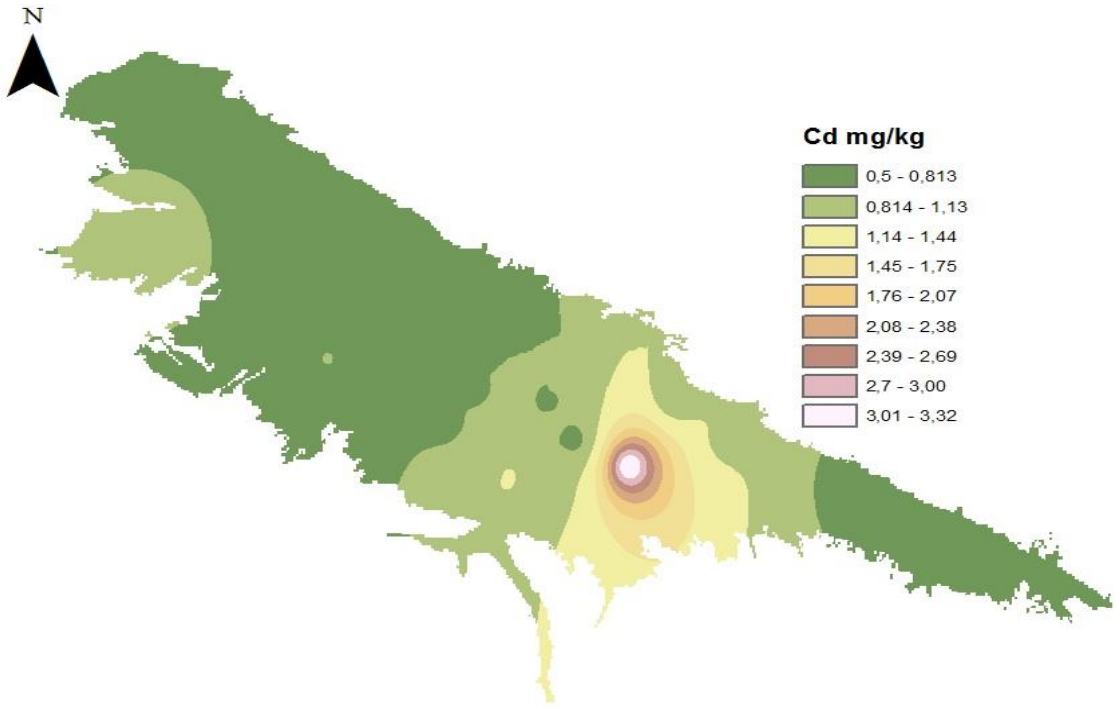


Fig. 3. *Interpolation map (IDW) of cadmium (Cd) content in Botevgrad valley - mg/kg.* (*Only surface soil samples are used for interpolation maps)*

Cadmium (Cd) naturally occurs in the Earth's crust, but human activities significantly increase its soil concentrations. In the study area, only point 29 (3.32 mg/kg) exceeds trigger value norms, located near an industrial zone of the town of Botevgrad (Fig. 3). The average cadmium level in uncontaminated soils in Bulgaria is 0.30 mg/kg (Chuldzhiyan, 1989), with a

natural concentration adopted as 0.4 mg/kg. In industrial areas, cadmium levels can reach 3–30 mg/kg (Petrov, 1984). The value of 0.4 mg/kg was adopted as the natural concentration. In the industrial areas of Bulgaria, the values reach 3–30 mg/kg (Petrov, 1984). The main sources of cadmium pollution being industrial activities—factories, battery production, paints, and plastics. Phosphate fertilizers also contain cadmium as an impurity, which accumulates in soils. Mining activities - specifically the extraction and processing of zinc, lead, and copper - contribute to cadmium release through atmospheric deposits. Waste management issues, such as improper handling of industrial and municipal waste, also lead to contamination. The average cadmium level in the area is 0.91 mg/kg, which is below the maximum permissible concentration.

Soil contamination with copper (Cu) is a significant problem in Bulgaria. In agriculture, copper pollution occurs through the use of copper-containing fungicides and herbicides, most often Bordeaux mixture, Cuprocin, Perocin, etc., also used in the Botevgrad region. Although the average copper levels are below the norm (47 mg/kg), contamination has been identified in specific locations. As mentioned, the nearby Eliseyna station is a source of copper contamination. The average copper content in uncontaminated soils in Bulgaria is higher, around 30 mg/kg (Chuldzhiyan, 1989). The value of 34 mg/kg is accepted as the natural soil concentration. Pollution exceeding trigger values is present in two locations - sample 1 and sample 31 - near the town of Botevgrad and the village of Trudovets, but these levels are only slightly above the norm (Figure 4). Additionally, according to the municipal plan, there are enterprises in the city involved in the production of copper electrical components and electronics (Botevgrad Municipality, 2005).

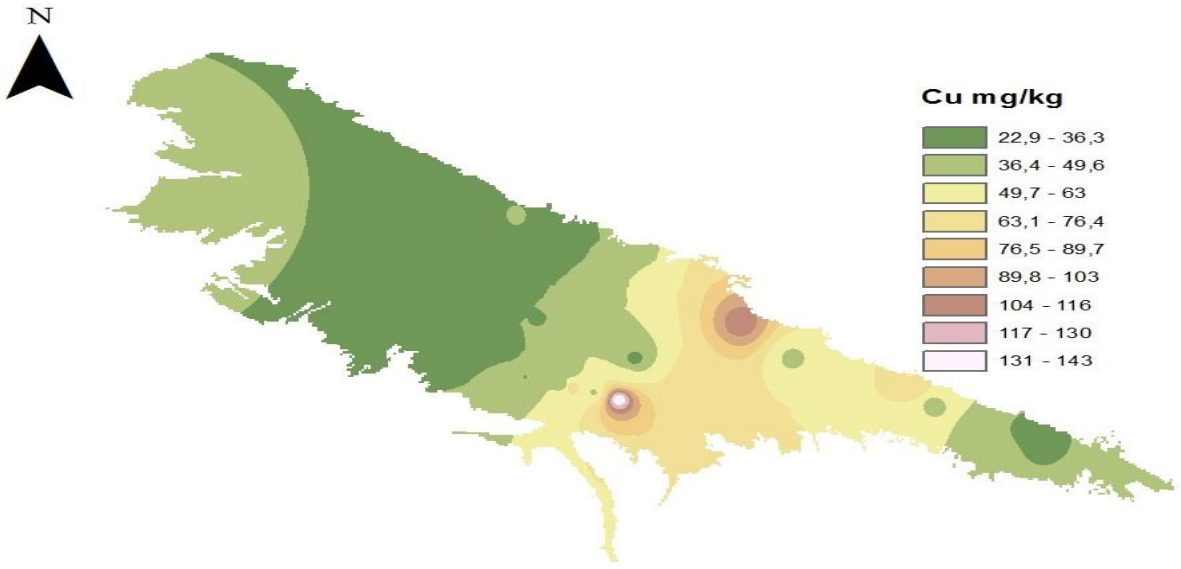


Fig. 4. *Interpolation map (IDW) of copper (Cu) content in Botevgrad valley - mg/kg. (Only surface soil samples are used for interpolation maps)*

Zinc (Zn) is a naturally occurring element in soil; however, excessive accumulation is usually the result of anthropogenic activities. Major sources include metallurgy, battery, tire, paint, and electronics manufacturing, as well as mining and ore processing. Zinc is often found in wastewater and tailings.

Sources of zinc pollution include waste and landfills improper disposal of metal and electronic waste along with the use of zinc-containing fertilizers and pesticides in agriculture. Atmospheric emissions from the combustion of coal and petroleum products containing zinc also contribute to soil contamination.

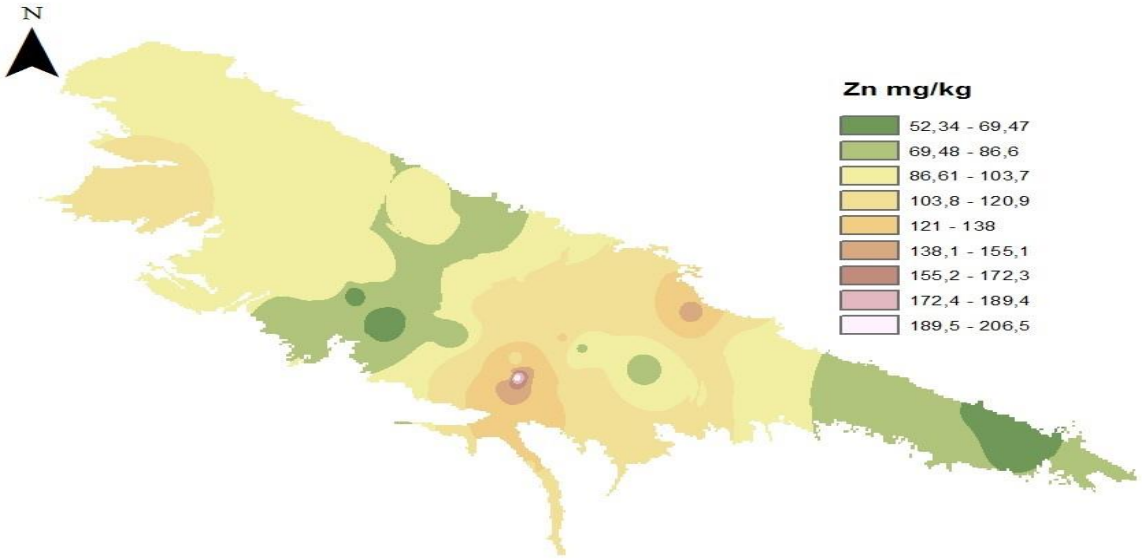


Fig. 5. *Interpolation map (IDW) of zinc (Zn) content in Botevgrad valley - mg/kg. (Only surface soil samples are used for interpolation maps).*

The average zinc content in unpolluted soils across Bulgaria is approximately 75 ± 20 mg/kg, with a natural concentration assumed to be around 88 mg/kg (Malinova, 2010). In the A horizon of profile 1, zinc exceeds the trigger value by about 208 mg/kg, indicating minimal pollution. The average zinc level in the studied soils is approximately 103 mg/kg, which remains within the acceptable range.

Conclusion

The soils studied in the Botevgrad Valley, show that heavy metal concentrations above permissible levels are limited to isolated locations, primarily near areas affected by human activity. Overall, the average values of heavy metals are within the norm. The trigger values for heavy metals are observed mainly in the central part of the Botevgrad Valley, where the industrial zone is located. High levels of copper and zinc are also found in areas with intensive agriculture, due to the use of pesticides and mineral fertilizers. An increase in heavy metals such as lead and cadmium can be observed near some roads, along which unregulated landfills, industrial zones, and scattered household waste are present. Based on the data obtained, it can be concluded that there is no significant risk of soil contamination in the Botevgrad Valley with heavy metals.

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