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SPATIAL DISTRIBUTION OF HEAVY METALS IN SOILS AND WATERS AS EXEMPLIFIED BY KAKHETI REGION (EASTERN GEORGIA)

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Abstract: The subject of research is the chemical analysis of heavy metals and assessment of their impact on soil and water, which is caused by solid waste. The closed landfill of Gurjaani in Eastern Georgia, was selected for the study, where various wastes are accumulated, including agricultural pesticides, herbicides and other wastes and their containers. In order to identify the contaminated areas in the field, soil samples were taken. The samples were processed accordingly in the research laboratory and various elements (pH, Pb, Cu, Zn, Cd) and organic substances were determined in them. As a result of the laboratory research, the facts of metal concentration were clearly identified. Zinc turned out to be especially dominant. It is a fact that the waste of various composition unsystematically located at the landfill is the cause of these metals entering the environment. However, the type and quality of pollution depend on several factors: the amount of waste, morphological composition, climatic conditions, physical-chemical properties of the soil, etc. It was determined that Soil type, in this case brown soils, site reaction and climate contribute to the accumulation and subsequent solubility of heavy metals. Through surface and ground water, even remote areas are put at ecological risk.

Keywords: solid waste, heavy metals, soil, water, environmental sustainability.

1. INTRODUCTION

As it is known, heavy metals have a particularly negative impact on soil properties, its composition and formation processes, resulting in a deterioration of the physical and chemical potential of the soil. Consequently, the balanced ratio of solid, liquid, and air phases in the soil is disturbed; the properties of the soil components change dramatically. The result is degraded soils, disturbed vital activity of crops and decreased bioproductivity (Matchavariani, & Kalandadze, 2012), while the main goal of the society is to get enough food with as little impact on the environment as possible.

Microelements play an important physiological and biochemical role in plant, animal and human life. They are part of vitamins, enzymes, and hormones. Their excessive or insufficient content in food causes metabolic disorders in the body and may lead to severe diseases (Urushadze, et al., 2011).

Relevance of the topic Unsystematic disposal of waste creates an ecologically dangerous zone, because harmful substances released as a result of their decomposition lead to soil, water, and air pollution. It is known that excessive amounts of heavy metals in the environment accumulate in the soil, migrate to groundwater or surface waters, are absorbed by plants and enter the food chain. Lead, zinc, arsenic, and cadmium are heavy toxic metals whose common and mobile forms are distributed differently depending on soil composition and properties **(**Urushadze, et al., 2015).

A closed landfill, virtually without signs of surface contamination, was chosen as a study site. However, the results of the study showed that improperly burying solid waste in the ground is not a solution to the problem, and causes negative environmental effects on the nearby agricultural fields. Rainwater constantly seeps into the waste “hidden” under the soil or other inert material, which dissolves and contaminates the area with harmful substances. Since the vast majority of landfills in

Georgia do not have seeping water collection systems, contaminated rainwater eventually mixes with groundwater and severely pollutes it. What is most important, these processes take place right near the agricultural fields or populated areas.

The purpose of the research is to determine the ecological, social and economic threats to the environment, which will help us to properly plan measures aimed at reducing the impact of waste on the environment and human health.

Therefore, the study of the accumulation and migration of heavy metals in soils is a very important and relevant issue. Through our research, we determine the spatial distribution of heavy metals in soil and water, which will allow us to determine the ways of their entry and movement into the environment.

2. MATERIALS AND METHODS

The study was conducted in Eastern Georgia, in Gurjaani Municipality, which administratively belongs to Kakheti, with the city of Gurjaani as its center, (Figure 1).

Figure 1.Map of Georgia. Gurjaani Municipality.

The Municipality has a low-mountainous terrain, with altitudes ranging from 300-450 meters to 850-1000 meters. Gurjaani Municipality has a moderately humid subtropical climate, with moderate winters and hot summers. Annual temperature is 12.4°C; average temperature in January is 0.9°C and it is 23.6°C in August. The amount of precipitation is up to 800 mm. As the main focus of the study was the impact of solid waste on soil and water, the determinants in this case were soil types and properties, as the nature of distribution of substances in the soil profile is affected by a set of soil factors: humus in the soil, the granulometric composition of the soil, soil reaction and its organic matter content, cation exchange capacity, presence of geochemical barriers, drainability, permeability, etc.

Brown soils (Cambisols - WRB), According to the World Reference Base for Soil Resources, the given soil type is identified as Cambisols (WRB). formed under dry subtropical weather conditions, are widely distributed in the study area, with warm and dry winters, and hot and dry summers. The moisture coefficient is 0.5-0.8. The soil moisture regime is impermacide, i.e. evaporation exceeds precipitations. They include Cambic B horizons, the diagnostic criteria of which are: the coarse grain size, higher clay and sludge content compared to the upper and lower horizons, less dense structure, organic carbon content > 0.6%, high saturation with bases, and slight alkaline or neutral reactions.

The study was performed directly on the closed Gurjaani landfill and in its surrounding area. The following works were done in the field: reconnaissance of the study area, marking the working sites, mapping, identifying humus content in the soil samples, as well as soil reaction (pH), heavy metals, including lead (Pb), copper (Cu), zinc (Zn) and cadmium (Cd), and the water samples were studied for water pH, HCO_3, heavy metals zinc (Zn), copper (Cu), lead (Pb) and arsenic (As). The distribution of heavy metals in soils and waters, their quantification, and comparative analysis will clearly show us the impact of waste on the environment and its environmental resistance.

Figure 2. Gurjaani Municipality. Closed landfill.

The said landfill was closed in 2014, but it is not known since when it operated, or what amount and type of waste was placed in it (Figure 2). However, it is known that all kinds of waste (organic, solid, pesticides, packaging, etc.) were systematically gathered and periodically burnt here (Waste Management Technologies in the Regions, September 1, 2015). Today, the landfill is closed and fenced along its entire perimeter, the waste is compacted and not seen on the surface. During the fieldwork, it was noticed that the surface of the landfill is covered with a clay layer of about 40-50 cm. From the environmental point of view, this can be viewed as positive, as it is a watertight barrier due to its low infiltration capacity, but it can also have a negative effect as it will retain surface water and form an artificial water body. About 20 meters north of the landfill, there is a natural water reservoir used for fishing and irrigation. (Figure 3) As we observed, it is in contact with a portion of the waste that had been dumped in the above basin for years (Waste Management Technologies in the Regions, September 1, 2015). At 30-40 meters from the south, the landfill is bordered by a natural ravine with constant runoff, where the waste from the nearby livestock farm is also accumulated (Figure 4). Therefore, the transparency of the water in the ravine is quite low, and the color and strong smell suggest that it is contaminated with various organic substances. Thus, the landfill is between two water bodies. The water from the natural water body in the north leaks to the landfill by means of underground filtration as confirmed by the existing drainage channel in the area (Kavelidze, & Kalandadze, 2021). The seepage water following the slope of the terrain, flows into the natural depression in the south, the ravine. Therefore, the agricultural lands bordering the landfill from the east, west and north face a serious ecological threat. As a result, an extensive area can be assumed to be contaminated.

Table 1. Soil sample data

Sample	Altitude, asl	Geographical coordinates	Soil sampling depth
P1	266 m	N. 41.766534 E. 45.872232	0-50 cm
P3	485 m	N. 41.716473 E. 45.821673	0-25 cm
P4	286 m	N. 41.671702 E. 45.754346	0-25 cm
Plot P5	270 m	N 41.460491 E. 45.522762	0-60 cm
Reference plot P6	258 m	N 41.455501 E. 45.522054	0-50 cm
P2	273 m	N. 41.756338 E. 45.838953	Sediments from the ravine bed in autumn
P7	264 m	N 41.455744 E. 45.521622	Sediments from the ravine bed in spring

Figure 3. Pond adjacent to Gurjaani landfill.

Figure 4. Ravine adjacent to Gurjaani landfill

Table 2. Water samples from the pond and ravine adjacent to Gurjaani landfill.

Sample	Altitude, asl	Coordinates
N 1 (pond), autumn	265 m	N. 41.767178 E. 45.8714424
N 2 (ravine), autumn	273 m	N. 41.756338 E. 45.838953
N 3 (pond), spring	262 m	N. 41.767445 E. 45.871288
N 4 (ravine), spring	273 m	N. 41.455744 E. 45.521711

The field works on the study sites were carried out for two years. A field survey of soils in the landfill area and mixed sampling were accomplished by the Auger method, (Figure 5). Soil samples were taken at 0-5 cm from the surface and 50-60 cm from the depth. Mixed soil samples were collected both directly from the landfill and from the agricultural fields. (Table 1) (Figure 7) soil sample was placed into a polyethylene bag, labeled, and transported into the laboratory. (Figure 6) Mobile forms of heavy metals in soils are made by inductively coupled plasma emission spectrophotometer (OES) spectrometer. This method refers to the determination of mobile forms of heavy metals in soil extracts by the atomic emission method. Motile forms are extracted from soil with pH 4.8 acetate-ammonium buffer. (Gost ISO22036-2014)

Water and sediment samples were taken from the ravine, the depression located near the landfill. (Table 2) Before sampling from the water bodies, the sampling vessels were rinsed with distilled water. Soil and water samples were taken to scientific laboratory for chemical analysis, where various parameters were measured and the heavy metals were detected by means of atomic absorption spectrophotometry.(AAS) Heavy metals in water are determined by the ISO-11885-2007 standard.

An important stage of the research is the interpolation of the received information using the Inverse Distance Weighted (IDW) method.

3. RESULTS AND DISCUSSION

The solid waste management problem is a global and urgent one today. This issue is a particularly important cause of soil degradation for a developing country such as Georgia. Burying solid waste in the ground is not a solution and it is still an important source of heavy metals (Duruibe, et al., 2007). An increased concentration of heavy metals in soils can seriously affect the biological properties of soil, change its composition, and negatively affect the soil self-recovery processes (Matchavariani, & Kalandadze, 2012).

Pollution of soils is specific, as it is both an acceptor of a substance and its donor to other natural environments. The entry and distribution of heavy metals in soil is influenced by a number of factors: granulometric composition of soil, content of organic matter, soil reaction, cation exchange rate, presence of geochemical barriers, anionic composition of soil solution, water regime, absorption capacity, etc. (Turkadze, 2016).

After entering the soil, heavy metals are absorbed first by fast reactions, followed by slow absorption reactions and their breakdown into different chemical forms with different bio-penetration, mobility and toxicity. It is the upper humus layer of the soil “hit” the first where micro- and macroelements or other toxic substances start to accumulate intensely. The ability of different soils to resist different substances varies (Matchavariani, et al., 2015). The distribution of substances in the soil depends on the reactions of heavy metals in soils, such as mineral precipitation and decomposition, ion exchange, absorption and desorption, water complexation, biological immobilization and mobilization, and uptake by plants (translocation effect).

Soil reaction is variable and depends on the chemical composition of soil as well as the content of salts, organic and mineral acids in soil **(**Urushadze, et al., 2011). The laboratory studies of the soil samples revealed a neutral or slightly alkaline reaction; acidic reaction was observed only in the sediments sampled in spring. As it is known, the brown soils area reaction (pH) is neutral or slightly alkaline, with a characteristic increase in alkalinity with depth, caused by the washing regime.

High copper concentration exceeding the MAC (Maximum permissible concentration) according to the national standard, was observed on the waste dumpsite (Figure 11). No excess rate was detected in

Figure 5. Sampling with Augher method.

Figure 6. Water and Soils samples

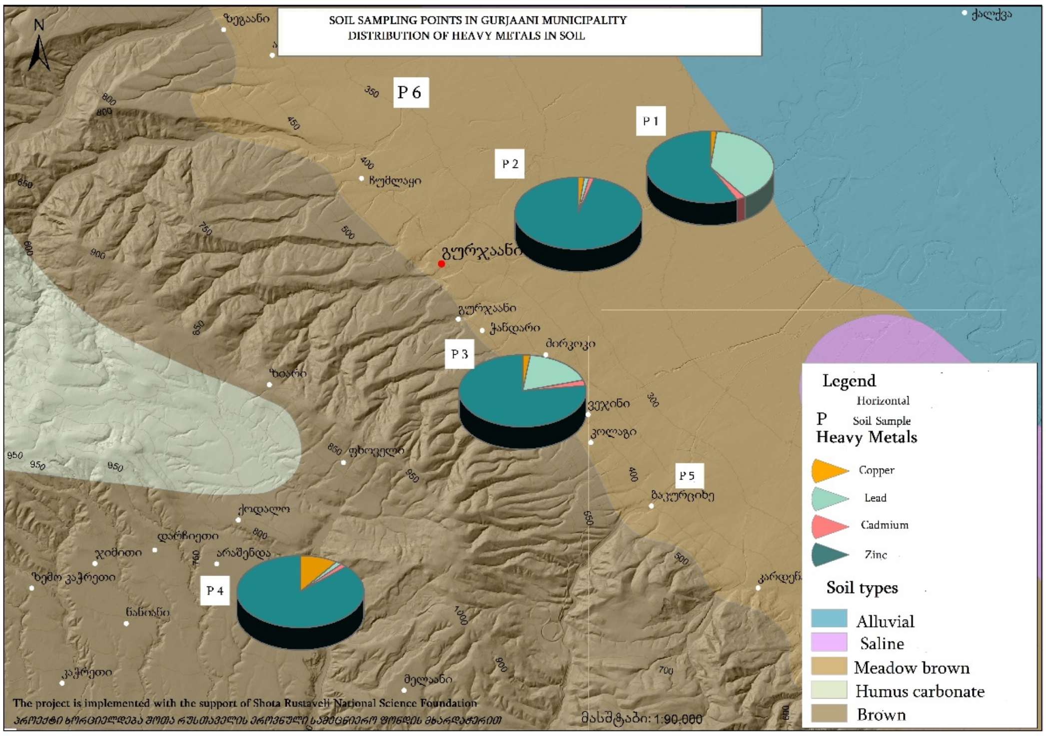

Figure 7. Soil sampling points in Gurjaani municipality. Distribution of some heavy metals in soil.

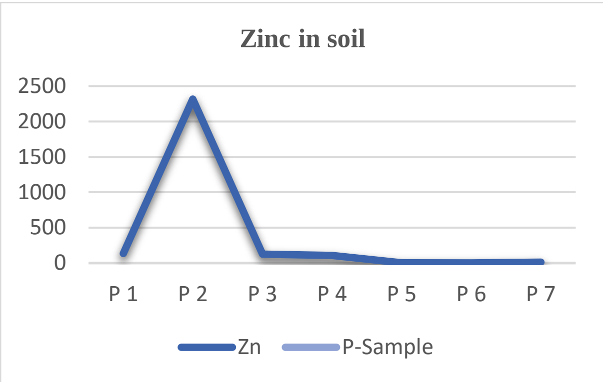

Figure. 8. Absorbable zinc forms.

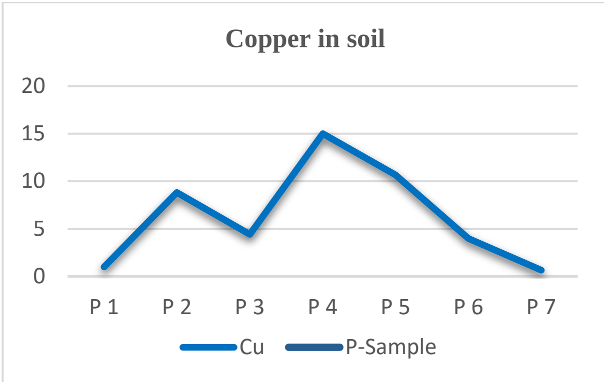

Figure. 9. Absorbable copper forms.

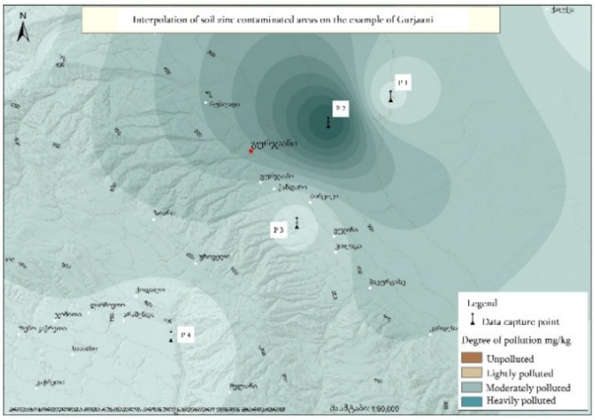

Figure 10. Soil contamination with zinc.

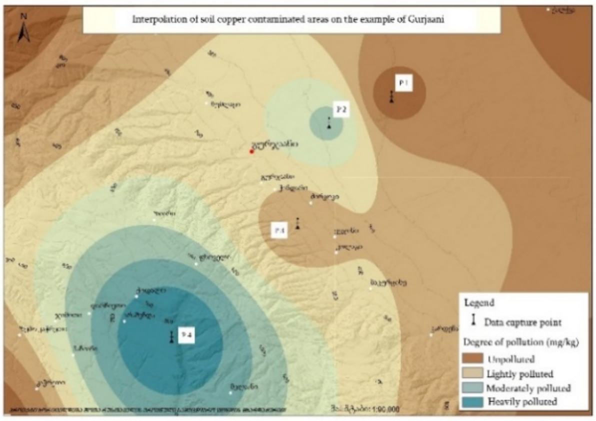

Figure 11. Soil contamination with copper

P1 sample (0-50 cm). However, in its vicinity and in arable plot P5 (10.68 mg/kg) it exceeds the admissible concentration (3.0 mg/kg) (Table 3), and only slightly exceeds the permissible level in the reference sample (3.97 mg/kg in P6), (Table 3). It should be noted that there is a marked difference between the sedimentary precipitations sampled in the upper water segment (P7) on the one hand, which has an alkaline reaction and copper concentration of 0.65 mg/kg that is lower than the maximum permissible concentration, (Table 3) and the lower water segment near the landfill (P2) on the other hand, which has an acidic reaction and copper concentration of 8.8 mg/kg, (Figure 9). These copper concentrations can be explained by the acidic reaction that supports the solubility of heavy metals. This is the evidence of the connection of the ravine water with the buried waste through groundwater. As for the excessive copper content in agricultural soils, it is probably related to the use of the ravine water to irrigate the agricultural plots.

Zinc (Zn) is an important element for plants and for the normal functioning of the human body, but its excessive amount is toxic. (Matchavariani & Kalandadze, 2012) The studied samples contain alarming levels of zinc (Figure 10). It exceeds the maximum permissible level in by ten times, and its concentration in the sediments is a hundred times higher the admissible level, (Table 3)

As the study shows, zinc in soils reaches toxic levels: it exceeds the maximum permissible level in almost all samples. This is particularly true with the soil sampled in autumn, which contain the absorbable forms of zinc from 105.7 mg/kg to 133.2 mg/kg, while the difference with sedimentary deposits is great, (Figure 8). Zinc content is critically high (2316.7 mg/kg) in the lower part of the ravine (P-2) where water acidification takes place, and it is 11.73 mg/kg in the upper section of the ravine (P-7) with neutral water reaction, (Table 3). Zinc is known as the best soluble chemical element. According Kalandadze, (2003), clay and organic substances found in soil are linked with zinc. The ability of zinc to dissolve in the soil is much lower than in $Zn(OH)_2$, $ZnCO_3$, and $Zn(PO_4)_2$ compounds. Zinc is the most mobile element and is easily assimilated by living organisms from light soils (Kalandadze, 2003). Brown soils spread in Gurjaani Municipality have a heavy mechanical composition, in which the amount of organic substances in the samples varies from 3.55% to 54.81%. Low levels of zinc were fixed in the reference plot (1.56 mg/kg in P6) and in the arable field (2.63 mg/kg in P5), (Table 3). Samples taken in the rainy spring season confirmed that the concentration of heavy metals in the soil is higher during dry seasons than during rainy seasons. This is because the solubility of metals in the soil increases due to runoff and infiltration during atmospheric precipitations (Amadi, 2014). The presence of a small amount of zinc can also be explained by the reduced organic substances in soil (3.55% - 6.48% (P5 -P7) (Table 3). As it is known, both common and mobile forms of microelements are distributed differently in the soil through organic substances. It is the organic substances that determine the availability of heavy metals in the soil: the concentrations of heavy metals are reduced in soils with heavy granulometric composition and low concentrations of organic substances.

Lead (Pb) is the most toxic metal. It is persistent and has low mobility and bio-penetration**.** Therefore, its content in sediments is lower than maximum permissible concentration. According to scientific studies, lead does not seep into a soil profile, but Bowen (1979) states that though very slowly, but it still moves through soils. Kabata-Pendias & Pendias (1999), on the other hand, points out that the lead movement velocity in a soil profile may change due to the decreased absorption capacity of the soil or increased soil acidity. This means that this element may migrate to groundwater, representing a serious toxicological hazard

Table 3. Results of the soil study on Gurjaani landfill (mobile forms of elements are given)

Sample	pH	Cu mg/kg	Zn mg/kg	Pb mg/kg	Cd mg/kg	Organic substances, %
P1 Landfill site (in autumn)	7.77	<1.0	133.2	81.5	1.7	9.49
P2 Sediment (in autumn)	5.10	8.8	2316.7	<1.0	<0.1	54.81
P3 Landfill site (in autumn)	7.50	4.4	125.6	30.7	0.82	13.86
P4 Landfill site (in autumn)	7.52	15.0	105.7	<1.0	1.37	12.98
P5 Plot (in spring)	7.79	10.68	2.63	0.96	0.04	6.48
P6 Reference (in spring)	7.53	3.97	1.56	1.17	0.03	4.21
P7 Sediment (in spring)	7.02	0.65	11.73	2.16	0.04	3.55
Maximum permissible concentration according to the national standard (mg/kg)		3.0	23.0	6.0	0.17	

Minister of Labor, Health and Social Protection of Georgia Order No. 297/N August 16, 2001 St. Tbilisi. "On the approval of norms of the qualitative state of the environment" 24. p.154

(Pikula, & Stepien, 2021). The content of lead in the soil of the closed Gurjaani landfill where they used to dispose various types of waste exceeds the baseline value: it is 81.5 mg/kg (P1) and 30.7 mg/kg (P3), (Figure 15). In the soils with alkaline reaction lead mobility is less. Or when this heavy metal enters the soil, it is absorbed by clay minerals, and the carbonate system of the soil acts as a barrier for it. This explains the surface accumulation of metals generally (Matchavariani, & Kalandadze, 2012), whereas in other cases the accumulation rate is low: 1.17 mg/kg in the reference sample (P6), and less than 0.96 mg/kg in land plot P5 (Table 3). Lead content in the sedimentary deposits is within the normal limits, once again confirming the absence of lead solubility and mobility (Figure 12).

Cadmium (Cd) is considered an eco-pollutant because of its high toxicity and accumulation in the environment. The study showed that cadmium exceeds the maximum permissible level only on the landfill and in its immediate vicinity (Table 3). As already discussed, one of the important variables determining the distribution and enrichment of heavy metals in soil is the soil reaction (pH) (Amadi, & Nwankwoala, 2013). The top layer of the landfill is covered with clay soil and the area reaction is alkaline what increases the deposition and bioaccumulation of Cd in the soil because soil reaction is considered to be the main factor controlling the availability of Cd and particularly of Zn (Black, 2010). Bioaccumulation of Cadmium may occur at all levels of the aquatic and terrestrial food chain (Nartey, et al., 2012). However, research revealed that when the level of cadmium in the soil varies from 0.5 mg/kg to 1 mg/kg the uptake of cadmium by the plant is higher. (Avkopashvili, et al., 2022) The results of our research show that cadmium ranges from 0.1 to 1.7 mg/kg.

According to Vinogradov's classification (Vinogradov, 1957), based on the clarks of heavy metals, soils with Cd value higher than the clark norm of 0.17 mg/kg should be considered polluted (Figure 14). The source of high concentration of metals in the area is the buried waste. The intensity of the accumulation of metals in soil depends on soil properties, including absorption ability. Brown soils are characterized by argillization, carbonization, and significant absorption ability. Heavy loams absorb

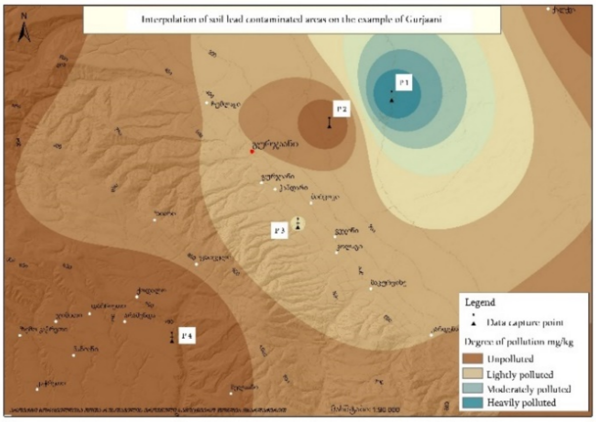

Figure 12. Soil pollution with lead.

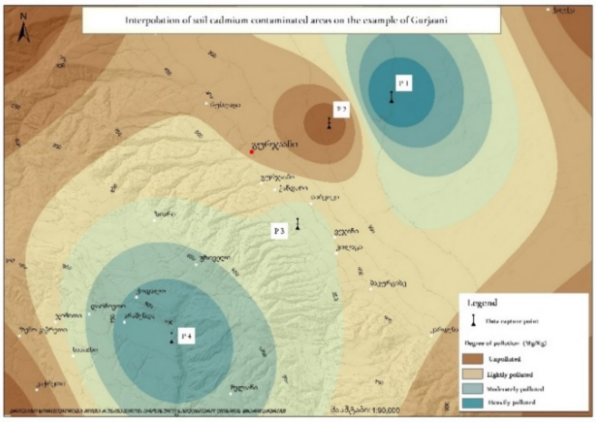

Figure 13. Soil pollution with cadmium..

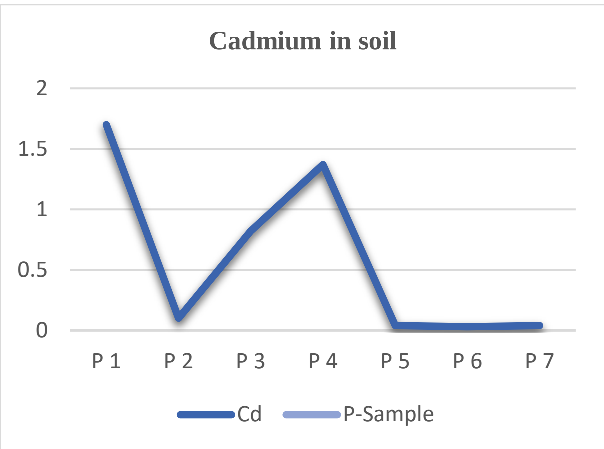

Figure.14. Absorbing forms of Cadmium.

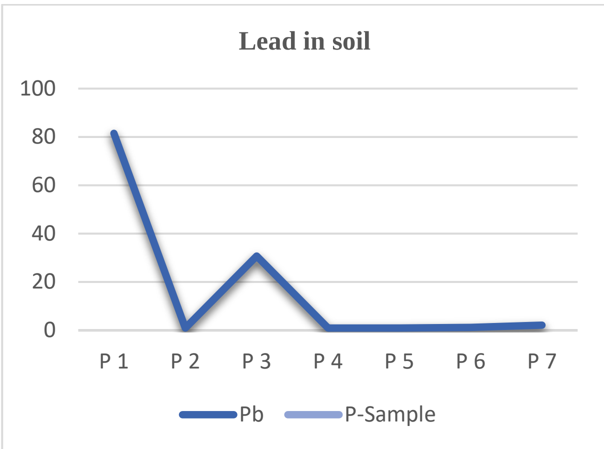

Figure.15. Absorbing forms of Lead.

Table 4. Results of water study in the pond and ravine adjacent to Gurjaani landfill

Sample	pH	Cu mg/l	Zn2+ mg/l	Pb mg/l	As mg/l	Hydrocarbonate (HCO_3)
N 1 (pond), autumn	7.35	0.10	0.017	0.0023	<0.01	183.0
N 2 (ravine), autumn	5.03	0.09	0.033	0.0012	0.04	292.8
N 3 (pond), spring	7.12	0.01	<0.01	0.01	<0.01	192.2
N 4 (ravine), spring	7.60	0.01	<0.01	0.01	<0.01	253.2
Maximum permissible concentration according to the national standard (mg/kg)		1.0	1.0	0.03	0.05	

Minister of Labor, Health and Social Protection of Georgia Order No. 297/N August 16, 2001 St. Tbilisi. "On the approval of norms of the qualitative state of the environment" (XXIII. Appendix 8).

better than other soils (Hanauer, et al., 2011). It is a fact that cadmium is not migrated through groundwater, as its concentrations in sediments, reference plot and land plot is low (Figure 13).

Table 4 shows the results of the study of water samples and maximum permissible concentration according to the Georgian legislation (Order of the Minister of Environment and Natural Resources Protection of Georgia No. 130 of September 17, 1996), evidencing that pollution is not hazardous. Although the visual appearance of the water in the ravine was unfavorable, a strong odor was indicative of the decomposition of organic waste. As a result, water transparency was quite low.Reaction is an important factor in evaluating soil and water quality because it affects many biological and chemical processes. High water acidity has been found to increase the solubility and mobility of metals and other toxic substances in water. Changes in water characteristics can affect the aesthetic and economic properties of water, such as transparency, odor and taste. This can increase its toxicity and compromise water safety (Nartey, et al., 2012). Among the water bodies we studied, acidic reaction was observed only in the water sample taken from the ravine (5.3 mg/l in P2). However, against the effective standards of the country, the concentrations of heavy metals in the water are not high and does not exceed the maximum admissible concentrations, including arsenic, which is a carcinogenic element, can bioaccumulate and causes severe pollution (Table 4). It slowly dissolves in water. Its concentration was <0.01 mg/L in pond water and 0.04 mg/L in ravine water. Arsenic and its compounds are used mainly as a melting agent in lead batteries. Arsenic has been widely used as a pesticide and herbicide, although its use for this purpose is gradually decreasing. Thus, no dangerous concentrations of arsenic were observed in the water bodies we studied.

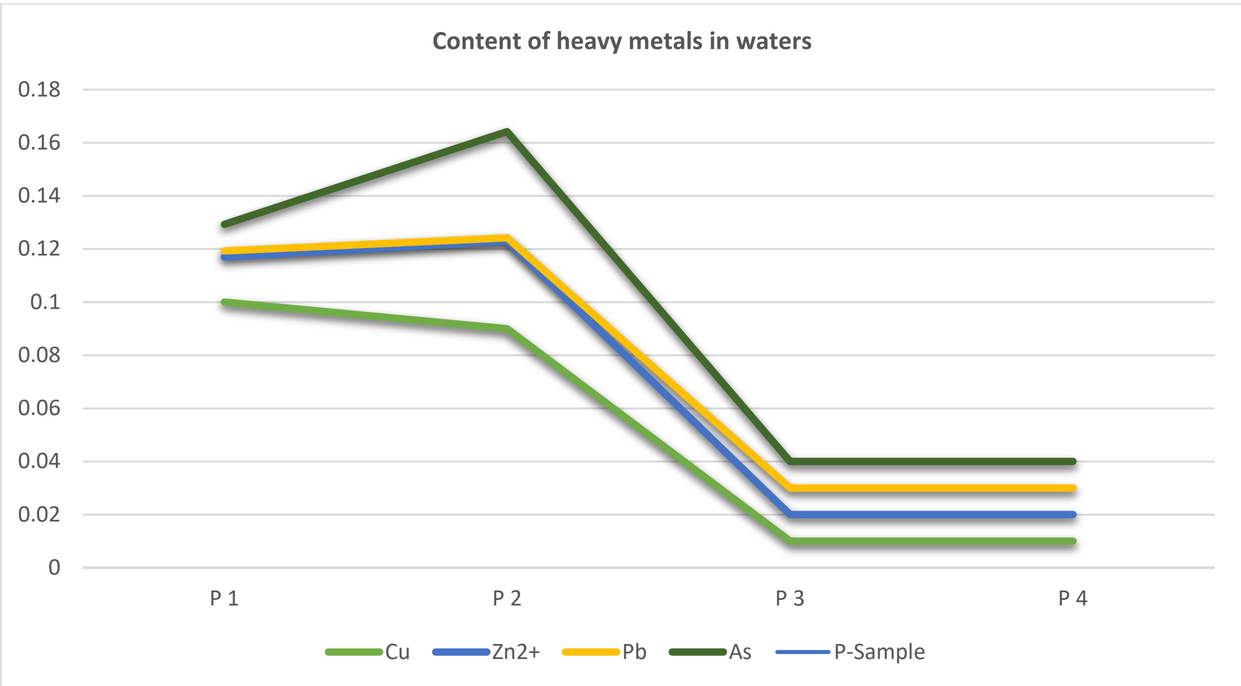

Figure 16. *Content of heavy metals in ravine and pond in spring and autumn*

Copper in water samples does not exceed the maximum permissible concentration, but its content is higher in the current of the lower ravine section (0.09 mg/l in P2) than in the upper section (0.01 mg/l in P4), (Figure 16). At the same time, water reaction in the lower ravine section is acidic and the ability to absorb heavy metals is high, while in the upper segment the reaction is alkaline, thus reducing the copper solubility in water. The total solubility of cationic and anionic forms decreases when water pH value varies between 7 and 8 (Kalandadze, 2003).

Thus, despite the excess concentration of heavy metals in soils, in the widespread heavy clay soils, whose porosity is less and the colloidal part is compacted, deep migration is slow. Inorganic colloidal part of soil is most responsible for absorption of mineral particles (Dube, et al., 2001).

The conclusion is that presently, heavy metals are passively leaking from the landfill into the groundwater. However, we can assume that this is temporal and they may reach even the water reservoir after some time. If the soil acidity changes, the solubility of the metals will be activated. It should be noted that the presence of alkaline salts in the water prevents the solubility of heavy metals. It is known that bicarbonates and carbonates in water can drive away toxic metals such as lead, cadmium, and others. As the laboratory results show, the content of hydrocarbons is not high. It varies only slightly across the seasons, ranging from 183.0 mg/L to 292.8 mg/L what may be related to the nearby livestock farm and salt added to the livestock feed.

4. CONCLUSION

Nowadays, studying the accumulation and migration of heavy metals in soils is very important and topical, because in addition to their impact on human health, heavy metals also reduce the productivity of soils. Therefore, the results of the study carried out within the scope of the project were found interesting, because we thought that solid waste management is only one side of the problem, but another very important fact is to assess their impact on the environment. Studies have shown that solid waste negatively affects the physical and chemical properties of soil and water. Soils are affected by any anthropogenic intervention in the ecosystem. It is a fact that wastes release various substances, including heavy metals, with zinc predominating but also high levels of copper and cadmium. The main problem is caused by the chemical nature of heavy metals, as they are not biodegradable and accumulate in the environment. Depending on the area reaction of soil and water, heavy metals may reach toxic levels at the expense of their persistence and enter the food chain. It is a fact that wastes of different compositions, haphazardly disposed at the landfill, cause these metals to enter the environment.

As for water, the data show that heavy metals do not exceed the maximum allowable concentrations permitted by the effective standards of the country. However, this does not give us an excuse to ignore the issue, as the threat to the sustainability of the ecosystem and the environment is evident.

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