Metals distribution in soil contaminated by gold and copper mining in Georgia

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Abstract

Aim: The study aimed analyzed of heavy metal distribution in soils and their contamination in the metallogenic province of the Lesser Caucasus (Kvemo Kartli region), due to mining of gold and copper ores.

Methodology: Samples were taken from the soil and analyzed using ICP-MC and AAS. Special formula was developed to reflect the relative distribution of the element in the soil profile.

Results: The research showed that intensive mining has been leading to increase scale and indicator of the contamination. Within the long period of time weather (wind, rain, snow) pollution was being spread over the far distances. Formation of molecular complexes in the soil can reduce certain metals.

Interpretation: The research revealed that atomic mass of elements can influence their movement in air, i.e., lighter the atomic mass of the element farthest its

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Introduction

Currently, the influence of industrial wastes on the environment, especially on water and soil, where heavy metals are accumulated, is increasing significantly (Matchavariani *et al.*, 2012; Sherameti *et al.*, 2010).

Water and soil pollution by heavy metals is a global problem throughout the world. Through precipitation and wind, chemical elements emitted in the air can be spread out to long distances, which increase the risk of ingress into the human body. Heavy metals can change the structure and fertility of soils, destroy bio-ecological varieties, including local plant formations (Baycu *et al.*, 2015; Conesa *et al.*, 2006). The possible change in the absorption and accumulation characteristics of heavy metals by plants depends on the concentration of the element, distribution, soil properties, temperature and types of plants (Khatiasvili *et al.*, 2015; Prasad *et al.*, 2008; Dixit *et al.*, 2015). The existence of a small amount of heavy metals in the soil does not harm plants, although over time they accumulate in the soil, hence the ability of their absorption by plants increases (Alkoeta *et al.*, 2004; Khan *et al.*, 2008; Prasad *et al.*, 2003; Prasad *et al.*, 2008).

Soil is the most specific and complex component of ecosystem. (Bargali *et al.*, 2018; Bargali *et al.*, 2019). In the case of water and air, while removing toxic substances, they easily restore their primary state. As for soil, everything is more complicated. If it is contaminated, the centennial equilibrium is disturbed, its normal functioning decreases drastically, and restoration of primary state requires a considerably longer period.

For millennia, the extraction of various polymetallic resources in Georgia has caused number of environmental problems (Felix-Henningsen *et al.*, 2007; Placek *et al.*, 2016). This study is concerned with RMG Gold and Copper Mine which has been functioning since the 70s of last century in the region of Kvemo Kartli (Georgia). The company uses an open-pit mining method that has been polluting the region with a variety of toxic metals (Avkopashvili *et al.*, 2017).

Mining is significant for the economic development of any country, but its harmful impact on the environment must be considered. Open pit mining is dangerous for the ecosystem, when environmental issues are neglected. Because of intensive production, a large number of toxic elements are released in the air, which pose negative effects on the ecosystem. Through precipitation, heavy metals accumulate in the soil and contaminates.

Similar activities are been carried out in the metallogenic province of the Lesser Caucasus in the region of Kvemo Kartli, where Gold and Copper ores are mined and are polluting the region.

Thus, the aim of this study was to analyze the distribution of heavy metals in the soils and their contamination during three years. To illustrate different distribution of metals in the soil profile, a dimensionless parameter was introduced to characterize the relative distribution of a particular chemical element in the soil profile. The numerical value of this parameter depends both on the method of contamination and also on the type of soil (Avkopashvili *et al.*, 2017).

Materials and Methods

The study area represents the metallogenic province of the Lesser Caucasus in the region of Kvemo Kartli – Bolnisi municipality (Fig. 1). This is one of the most important mining regions of Georgia, represented in the valley of the Mashavera River. RMG (Rich Metals Group) Gold and Copper mine is located in Kazreti, approximately 80 km southwest of Tbilisi, on the bank of Mashavera and Kazretula rivers. It is one of the largest enterprises in Georgia and Caucasus region. It has been functioning since 1970. The main part of the region is being used for agricultural purposes. The presence and distribution of some heavy metals in soils in the surrounding area is the result of the influence of open quarries. Wastewater from the mining and processing plant is released into the Kazretula River, which flows into the river Mashavera. Eventually, Mashavera is used for irrigation.

Geologically, the area is represented mainly by quaternary sediments, limestones, sandstones and magmatic formations of the Cretaceous and Jurassic period. The orographic scheme predominantly includes low elevations; tectonic, volcanogenic and erosional forms are quite common. River terraces are distinctly expressed.

The climate of Kazreti, according to general indicators, belongs to steppe type of dry subtropics with moderately cold winters and hot summers. The annual rainfall is on average 500-550 mm, and the wind speed is 2 m sec⁻¹ both western and eastern winds are widespread, which change directions during day. During daytime, east and south-east winds prevail, while at night – western. Obviously, this phenomenon facilitates the active transfer of toxic chemical elements with winds in different directions and their subsidence in soils (Avkopashvili *et al.*, 2017).

The following types of soil occur in the study area and adjacent territories: Cinnamonic Soils, Grey Cinnamonic Soils, Brown Forest Soils, Raw Humus Calcareous Soils, Mountain Chernozems and Alluvial Calcareous Soils (Avkopashvili *et al.*, 2019).

In 2014, soil samples were collected from 10 locations. These are Gantiadi, Dmanisi, Balichi, Kazreti, Sakhdri, Qveshi, Kvemo Bolnisi, Ratevani, Mtskneti, Nakhiduri (Fig. 1), which are located at different distances from the enterprise. Location

Gantiadi was selected as check point, which is quite remote and pollution free. Samples were taken at the depths of 0-10 and 30-35 cm from the surface. The investigated territory was divided into 1000 x 1000 m sections, where the samples were taken from 5 points and mixed as a single sample. The samples were dried at 105°C for 24 hrs, crushed and sieved through a 0.18 mm sieve. To determine the concentration of metals, 5 ml of 65% HNO₃ was added to 1 g of soil sample. The flasks were then put on steam (100°C) for 3 hrs and after 15 min cooled at room temperature, filtered through Whatman paper (0.45 µm) in 50 ml tubes, the volume was made up to 50 ml with distilled water (Avkopashvili et al., 2019). In the prepared solutions, several elements were determined on the ICP-MS (Inductively coupled plasma mass spectrometry) in the laboratory of the University of Georgia, USA. However, in this study distribution of only certain elements such as: Ti, V, Cr, Co, Ni, Cu, Zn, As, Rb, Sr, Mo, Cd, Cs, Ba, Pt, Au, Pb, Th and also B, Na, P, K, Fe were analyzed.

In 2015, soil samples were taken repeatedly from Gantiadi and Kazreti villages in order to compare with the data the data of 2014. The samples were analyzed on the ICP-MS in the laboratory of the University of South Florida, Center for Geochemical Analyzes. The same elements were determined in 2014.

In 2017, soil samples were taken from same 10 villages as in 2014, and Pb was determined on Atomic Absorption Spectrometer in the laboratory of Elevter Andronikashvili Institute of Physics at Ivane Javakhishvili Tbilisi State University.

Results and Discussion

The results of this research showed a peculiar distribution of elements in the soil. Balichi is the nearest village from the Abulbukhi mine, the distance from the mine to the village center is 2.5 km. (Fig.1) Table 1 shows the data taken in 2014. Table 3 shows the concentration of elements in the soil Gantiadi and Kazreti villages soil, where samples were collected in 2015, revealed that the certain elements are decreased in comparison to the data, taken in 2014. In 2015, Ti, V, Co, Cu, Zn, Sr and Th concentrations significantly decreased in both Gantiadi and Kazreti villages compared to 2014. The level of Cr, Ni, Rb and Cs were reduced only in Kazreti whereas in Gantiadi soil their concentrations were increased. The noted reduction in the level of elements can be explained by the fact that some elements penetrating the soil create molecular complexes with those elements the quantity of which is reduced and migrate together to the depth of the soil. For example, they can create molecules of BaTiO₃, VP, etc (Avkopashvili et al., 2019).

In 2015, Ba level significantly increased Gantiadi, as in Kazreti villages in comparison to the 2014 data. This can be explained that in 2014 Sakdrisi mine opened, whose ore was rich in Ba.

Fig. 1 : Location of the settlements near the mining region where the soil samples were collected

Cu, Zn, Cd and Pb are toxic elements, however, their concentrations in Gantiadi and Kazreti did not exceed the maximum permissible limits. Though Cu, Zn, As and Cd were found to increase in Nakhiduri Table 3 as compared to the permissible limits (Table 2). During the study, the highest level of As was found in Mtskneti soil, compared to other villages (Table 2). Soils in Nakhiduri and Mtskneti villages are irrigated with Mashavera river which is contaminated with heavy metals that causes soil pollution in these villages.

It should also be noted that the amount of Cd in the soil of Nakhiduri is 10 times higher than in Balichi. This also can be explained the fact that Cd atomic mass is light and it can be spread over long distances by dust during the wind blowing.

In 2017, soil samples were collected from same villages. The results showed that Pb was increased in Gantiadi, Dmanisi, Balichi, Kazreti, Sakdrisi and Qveshi villages as compared to 2014 data. As mentioned above, in 2014 Sakdrisi mine was opened and has been intensely active since then. The quarry is close to villages where Pb level was high as compared to other villages. In addition, Pb is heavy and when released in the atmosphere gets it accumulated to the nearest places, as reported in Balichi, Sakdrisi and Kazreti villages.

Wide range of data makes it very difficult to visualize distribution of elements on a single graph. Therefore a special formula was developed, where the parameter is introduced that reflects the relative distribution of element in the soil profile:

$$\delta = \frac{m_{10} - m_{30}}{(m_{10} + m_{30})/2}$$

In the formula, m_{10} and m_{30} are the concentrations of these elements, respectively, at a depth of 10 and 30 cm from the surface in mg kg⁻¹. It is noteworthy that the range of deviation of

Table 1 : Concentration (mg kg⁻¹) of elements present in the soil of study area during 2014

	Gantiadi	Dmanisi	Balichi	Kazreti	Sakdrisi	Qveshi	Kvemo Bolnisi	Ratevani	Mtskneti	Nakhiduri
B	8.42	9.31	18.61	16.93	25.88	14.97	18.51	18.34	18.14	9.31
Na	523	585	300	312	353	255	247	380	483	344
P	298	250	430	450	2282	420	272	551	388	288
K	3700	2228	8390	7261	7435	6957	5021	7778	6931	7216
Ti	937.95	921.28	471.98	453.25	364.16	407.44	335.51	403.36	535.90	188.03
V	83.67	104.15	53.68	65.52	34.45	64.37	53.79	65.08	64.53	46.54
Cr	21.42	24.13	38.75	41.10	22.01	31.66	33.26	38.56	38.78	35.41
Fe	9500	11754	8082	9550	6897	10217	9119	10962	9708	10597
Co	15.32	18.53	12.45	13.66	9.80	14.70	14.01	13.99	15.82	17.97
Ni	21.10	22.21	40.48	47.24	28.48	39.72	47.40	46.56	49.67	49.97
Cu	27.52	28.96	101.23	31.58	87.81	41.17	31.95	148.92	186.07	361.01
Zn	83.91	70.98	93.62	65.91	120.27	113.35	54.83	155.30	233.49	348.26
As	3.56	4.59	8.61	9.26	7.94	10.19	8.90	9.89	13.18	7.33
Rb	13.50	17.24	45.64	42.84	28.81	33.51	27.53	36.77	36.15	39.56
Sr	91.94	105.07	98.46	215.80	232.79	144.91	123.08	113.11	159.73	113.56
Cd	0.16	0.14	0.22	0.17	0.28	0.34	0.17	0.63	1.44	2.34
Cs	0.77	1.19	2.60	2.83	1.44	1.86	1.81	2.25	2.20	1.97
Ba	98.0	104.9	220.3	233.7	487.8	201.5	136.0	170.6	168.6	171.4
Au	0.04	0.02	0.01	0.03	0.04	0.01	0.04	0.05	0.07	0.03
Pb	10.44	10.13	18.73	16.23	16.59	21.74	15.81	20.00	17.18	19.59
Th	6.57	7.38	15.10	16.98	9.04	13.41	13.29	13.04	13.76	14.66

Table 2 : Maximum permissible limits for heavy metals (mg kg⁻¹) in agricultural soil (Georgian standards)

Metals	Cu	Zn	Pb	Cd	As
Limits	132	300	32	2	2

Table 3 : Concentrations of chemical elements in soils at the surface of 0-10 and 30-35 cm in soil profiles (Gantiadi and Kazreti villages, 2014-2015 years)

	Gantiadi, 2014		Gantiadi, 2015		Kazreti, 2014		Kazreti, 2015	
	10 cm	30cm	10cm	30 cm	10 cm	30 cm	10 cm	30 cm
Ti	920.53	1016.49	258.022	239.399	530.04	376.47	178.814	216.228
V	79.48	82.09	34.522	35.949	67.59	63.44	35.107	38.257
Cr	24.36	19.3	24.939	27.225	42.33	39.87	25.298	26.837
Co	15.49	15.4	7.435	7.653	14.19	13.14	7.347	7.335
Ni	21.71	17.79	26.38	27.27	49.46	45.01	25.9	25.858
Cu	28.37	24.5	13.423	13.598	32.77	30.39	14.659	13.899
Zn	86.33	76.17	33.84	33.417	66.85	64.97	32.148	31.983
As	3.58	3.36	5.668	6.129	9.75	8.76	7.052	6.792
Rb	13	15.15	29.056	32.718	45.31	40.37	22.923	25.72
Sr	95.85	95.88	69.275	72.794	234	197.6	149.637	161.838
Mo	0.31	0.22	0.322	0.206	0.51	0.41	0.421	0.409
Cd	0.16	0.14	0.195	0.208	0.18	0.17	0.198	0.196
Cs	0.73	0.84	1.501	1.639	2.89	2.76	1.309	1.438
Ba	101	90.8	346.727	364.256	235.6	231.9	391.202	340.804
Au	0.04	0.06	0.028	0.031	0.05	0.02	0.037	0.04
Pb	11.21	9.13	8.108	-	18.08	14.37	-	-
Th	6.49	6.48	3.326	3.499	17.07	16.88	3.033	3.097

this parameter for any metal is the interval $(-2; 2)$; $-2 < \delta < 2$. Fig. 4 shows how this parameter varies for different elements in the villages of Balichi and Nakhiduri.

The change in this parameter over time also depends on the type of soil contamination. Table 4 gives the distribution of Cadmium at different periods in Balichi and Ratevani at a depth of 10 and 30 cm from the soil surface, respectively.

Fig. 4 shows the curves of dependence of parameter of the relative distribution of cadmium in depth (δ) on the time for the soils of Balichi and Ratevani villages. The graph clearly shows that these curves differed significantly from each other.

Soil types were different in the villages of Nakhiduri and Mtskneti and despite the fact that they were quite close to each other, the elements distribution varied significantly. This is clearly

Fig. 2 : Pb concentration (ppm) in villages, 2014 and 2017 data comparison.

Fig. 3 : Numerical values of parameter of relative distribution of chemical elements in soils (On the horizontal axis there are chemical elements, the points correspond to the numerical values of the parameter of the relative distribution of these elements according to depth).

Table 4 : Numerical values of cadmium concentration (mg kg^{-1}) in different periods of 2012 in the villages of Balichi and Ratevani at a depth of 0-10 and 30-35 cm from the soil

Date	Depth (cm)	Settlements	
		Balichi	Ratevani
17.07.2012	10	0.73	2.13
	30	1.32	1.84
23.08.2012	10	2.65	3.2
	30	2.85	3.7
27.10.2012	10	2.82	3.17
	30	2.7	3.32

Fig. 4 : The curves of dependence of parameter of relative cadmium distribution on depth (δ) on time (on the horizontal axis, the time in days is given, on the vertical axis - numerical values of the parameter of relative distribution of chemical elements in soil profile).

seen from Table 1 where the numerical values of the concentration of some metals in soils are presented.

The concentration of lighter metals B, Na, P and Ti in soil of Mtskneti was higher than Nakhiduri whereas the concentration of Cu, Zn, Rb and Cd was more in the soil of Nakhiduri.

Fig. 5 shows the numerical values of parameter of relative distribution of chemical elements in depth in the soils of Nakhiduri and Mtskneti villages (also known as Kiristcekh).

Obviously, in this case, different distribution of metals in the soil profile is determined by the difference in environmental conditions and type of soils.

Observations showed that for last 5 years, the properties of investigated soils in Nakhiduri and Mtskneti villages have deteriorated sharply. In particular, in Mtskneti the soil surface was

sometimes covered with a waterproof whitish-green gypsum film, this caused reduction of porosity, pH change, and soil fertility loss of. As it turned out, limestone is poured into the waste water of the plant, in order to neutralize the acidity, and to drain the water in the collector. In this case, the gypsum formed with river water falls into the irrigation system, by which the agricultural land is irrigated. Over time, gypsum accumulates on the surface of the soil and covers as a thin film, which in turn worsens their aeration and filtration capacity, and also causes a sharp decline in fertility. This process causes compaction of soil, leading to unfavorable hydrophysical conditions for plants. The data of water capacity of soils varied between 19.0-82.6%, and the amount of productive moisture was rather low (6.0-20.6%).

Soil contamination is directly related to their agrophysical conditions. In heavily contaminated soils, the cementation process occurs, as a result of which the bulk density of soils rises

Fig. 5 : Numerical values of the parameter of relative distribution of chemical elements in the soil profile (on the horizontal axis, there are chemical elements, and above them points corresponding to the numerical values of the parameter of relative distribution of these elements in soil profile are given).

sharply, the total porosity decreases, and the water permeability reaches a critical limit.

The logical result of unfavorable parameters of agrophysical properties of the studied soils is a sharp deterioration in their filtration capacity. In brown soils, water percolates only 46 cm per day, creating conditions that are not optimal for plants.

Thus, the research revealed that in Kvemo Kartli region some heavy metals were reported to exceed the permissible limits due to intensive gold and copper mining. The three year ongoing project confirmed that soil pollution by certain heavy metals is gradually increasing from year to year. Contamination reaches to the long distances that is promoted on the one hand by the wind and on the other by using polluted river water for the irrigation purposes. The results of analysis also figured out that some of elements are significantly reduced compared with the previous year, which can be explained by the fact that elements infiltrating the soil create a molecular compound with the elements, which are reduced, and they migrate together in the soil profile. The study showed that amount of certain elements are higher in the villages located to far distances from the mine and have higher altitude, this factor can be described that by the wind the elements with lighter atomic mass are migrating toward these villages and through the rainfall they are being accumulated in the soil.

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