JEB
Journal of Environmental Biology

Journal Home page : www.jeb.co.in
E-mail : editor@jeb.co.in
ISSN: 0254-8704 (Print)
ISSN: 2394-0379 (Online)
CODEN: JEBIDP

Soil quality changes in response to their pollution by heavy metals, Georgia

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Publication Info

Paper received:
25 July 2013

Revised received:
30 October 2013

Re-revised received:
18 June 2014

Accepted:
22 September 2014

Abstract

The present study deals with the composition, migration and accumulation of heavy metals in irrigated soils, plants and partially natural waters; and also, establishing the possible sources of pollution and their impact on environmental situation. The content of toxic elements in the irrigated soils adjacent to ore mining and processing enterprise were studied. Content of toxic elements in the irrigated soils adjacent to ore mining, showed that more than half of territory was seriously polluted by copper and zinc. Some part of the area were considered catastrophically polluted. Expressed technogenesis taking place influenced irrigation. Heavy metals like copper, zinc and manganese negative by effected the properties of soil, thus composition and soil-forming processes taking place in the soil. It was especially well represented in the deterioration of hydro-physical potential of the soil. Irrigation of agricultural land plots by water, polluted with heavy metals changed the pH. Balanced correlation among solid, liquid and gas phases was disrupted. In highly polluted soil, the cementing processes took place that sharply increased the bulk density of the soil, deteriorated the porosity of soil and reduced water permeability critically.

Key words

Georgia, Heavy metals, Soil quality, Technogenesis

Introduction

Soil is the most informative part of the landscape because it is formed as a result of interaction of all other elements of the landscape. Therefore, all anthropogenic and natural processes that are taking place within the concrete ecosystem are reflected in soils (Blume, 2004; Hanauer *et al.*, 2010; Kalandadze and Matchavariani, 2011; Matchavariani and Lagidze, 2012). It is a specific and complicated component of nature. In case of water and air pollution, if the toxic substances are removed they will easily return to their original condition. In case of soil, this process is much more complicated. If soil gets polluted, the centuries old balance gets upset and restoring that balance will take a very long time.

Rocks are the main source of micro and macro-elements in the soil. The concentration of various elements in rocks defines the elemental make-up of the soil. During soil formation process elements are distributed in layers, some of them are lost and some of them are accumulated however, the main characteristics received from rocks are always preserved.

The environment is polluted as a result of human economic activities with industrial waste, wastewater, various radioactive substances, chemicals/pesticides used in agriculture etc. One of the most important problem is pollution of environment due to heavy metals. Some metals are characterized with high toxicity and if a considerable amount of such metals accumulate in living organisms it can have a strong impact on human health, in particular if taken into account the fact that period needed for semi-disintegration of heavy metals can last to hundreds and thousands of years. Heavy metals also decrease the productivity of soil. Therefore, investigating the accumulation and migration of heavy metals in soils is currently an important issue.

The composition of micro and macro-elements in soils is conditioned by soil-formation processes, chemical make-up of rocks and concrete landscape conditions. These are the factors which define the processes of opening, accumulation and migration of various substances. The first “blow” is taken by the upper layer of the soil, where the micro and macro elements and other toxic substances get intensively accumulated. The ability of soil to resist the polluting elements is also diverse.

Using wastewater rich with heavy metals for irrigation purposes is especially alarming. It has a significant impact on the chemical make-up of soil and brings about disastrous results. According to some researchers (Saet *et al.,* 1982), use of such water for irrigation is dangerous only when level of some heavy metals in such water is high.

With the development of ore-dressing industry, the issues connected with environment pollution due to heavy metals are becoming more and more relevant. Similar geo-chemical processes are taking place practically in all copper-sulfide mining sites, situated in the region. Accumulation of large quantities of heavy metals in water and soil has an adverse impact on the region's biosphere. There is a direct impact of hazardous substances on vegetation, besides increasing level of heavy metals in hydro-system and soils which can have a serious impact on the microflora of soil; it can change their make-up and have a negative influence on the self-recovering processes (Ozturk et al., 2008). Many interesting investigations have been undertaken on this topic by several researchers (Shaw, 1990; Prasad, 2003; Prasad, 2004; Lewinsky, 2007; Agarwal, 2009; Ashraf *et al.*, 2010; Dragicevic *et al.,* 2010; Grabicet *et al.*, 2011; Pantelici *et al.*, 2012; Rouina *et. al.*, 2012).

Anthropogenic accumulation of heavy metals in the soils in biogeochemical province increases due to presence of chemical elements. Near ore mines it is conditioned by exogenous mines and emission of industrial wastes into the environment. Intensity of accumulation depends on the ability of soils to absorb these metals. Heavy clay loam absorbs heavy metals more intensively than other types of soil.

Distribution and presence of some heavy metals in soils, plants and natural waters has been investigated in order to determine the impact of mines on the environment existing in study area in Bolnisi-Georgia.

The present study aimed at determining the composition, migration and accumulation of heavy metals in soil, plants and partially natural waters in Bolnisi district; because the soil of this region have developed on the weathered crust of volcanogenic and sedimentary rocks. Their chemical composition is defined by the chemical composition of rocks, soil-formation processes and various anthropogenic factors connected with human economic activities. The study futher aimed to establish the possible sources of pollution and their impact on environmental situation. No such study has earlier been conducted in the study area. Only surveys have been done concerning the geochemical aspects and ore deposits. All data from this study would be used as background materials for further investigation of this region.

Materials and Methods

Study area : The study site was Bolnisi district – part of Lesser Caucasus mountains' metallogenic province. It is one of the most important mining areas of Georgia. Madneuli multimetallic mining combine is situated 80 km south of capital Tbilisi. The main part of the territory is occupied by orchards and vineyards, partly by crops. Presence and distribution of some heavy metals in soil, plants and natural waters was investigated in order to determine the impact of mines existing in the area on the environment. Highly acidic waters containing sulfuric acid move deep underground, with large amount of copper, iron and zinc sulphates. Currently, the mine is open and the ore is mined in the quarry. Wastewater from ore-dressing and processing enterprise is added to them and it goes to Kazretula and Mashavera Rivers; subsequently these waters enter the ground through irrigation. Both hydro-systems and soils are getting polluted. At the same time, the river system becomes the main source of pollution for the district. Eventually the hazardous substances get accumulated in the soils and river sediments.

The study area comprise of brown soil. They have a deep genetic profile, differentiated morphogenetic structure, heavy clay loam and light clay granulometric composition, medium and high amount of humus. Nitrogen and phosphorus are not represented in large quantities, but soil is rich with absorbed bases. The soil reaction is alkaline.

There are various methods of land reclamation of polluted soil: mechanical, physical-mechanical, chemical etc. Use and effectiveness of each land reclamation method depends on the climatic, bio-climatic, geo-ecological and soil-edaphon (granulometric composition and structure of the soil, humus concentration, absorption volumes, oxidation-restoration potential, pH etc.) factors.

The field study carried out in the present study was based on the following three parameters: Agricultural land plot from which the soil samples were taken in order to define the level of heavy metal pollution; amount of soil sample, which was necessary for forming representative, mixed soil sample; Selection of the essential agricultural land plot. Essential agricultural land plot was the smallest geomorphological unit, which accurately reflected the soil's genesis, characteristics, soil-formation rocks, relief, vegetation, hydrological conditions of the site.

Soil samples were taken from ploughed fields, vineyards, orchards as well as crofts of some villagers. Average sampling method was used by taking 10 samples from an approximately 10 ha land plot with a field drill, after mixing these samples together one mixed sample was obtained. The samples were also taken from the vertical profile of the soil. All samples from soil and water were analyzed to identify heavy metals by atomic-adsorptive method (Zeien, 1995).

Concentration of acceptable maximum level of heavy metals in agricultural lands was defined taking into consideration the following factors: initial level of microelements in soils; dynamics of increasing level of concentration of heavy metals as a result of anthropogenic factors; general accumulation of heavy metals in the soils; adequate evaluation of toxic impact of heavy metals on plants; maximum level of concentration of microelements in soil; mutual antagonism and synergy of heavy metals and quantitative and qualitative evaluation of these factors, physico-chemical and agro-physical characteristics of the soil (carbon levels, humus levels, granulometric composition, moisture, moisture balance, aeration, filtration capacity etc.); balance between penetration and extraction of heavy metals in soil; plants tolerance towards heavy metals.

The criteria of soil pollution was made according to Saet *et al.* (1982). Various criteria and norms were adopted for evaluating level of soil pollution. Clarke Concentration Coefficient is one of the notable examples of such norms/criteria. The coefficient was calculated by the following formula: $Kk=Cf/K$; where Kk is Clarke Concentration Coefficient; Cf – is actual concentration of chemical elements in the soil; K – is the clarke of a chemical element. This criterion showed how high or low was the concentration of a concrete chemical element in comparison with that of Clarke's.

The geo-ecologic condition of soil was also evaluated using the Pollution Concentration Coefficient, which was calculated by the following formula: $Hc=Cf/level$; where Hc – is pollution concentration coefficient; Cf – is actual concentration of a chemical element, f – is local or general established levels of a concrete chemical element, which indicated how the concentration increased in comparison with the general levels (Saet *et al.,* 1982).

The pollution Probability Concentration Coefficient was also used for evaluating the level of soil pollution. The formula used was: $C=Cf/max$ permissible level, where C – is coefficient of pollution probability concentration; Cf – is actual concentration of a chemical element. These indicators showed how high the actual concentration of a chemical element was in comparison with the maximum permissible levels of concentration of that element. Higher the coefficient in comparison with 1, higher was the probability of soil pollution and negative impact of chemical elements on living organisms.

The correlation between various elements of the soil and their joint impact on the soil was also considered, in order to establish/evaluate the maximum level of concentration of microelements in the soil. Soils were grouped according to the total actual concentration of toxic chemical elements-pollutants. This grouping was based on the methodology elaborated by Vinogradov (1957); the first group was general level + 1 Clarke; the second group was general level + 2 Clarke. When soils were grouped this way the level of pollution in soil was classified as follows: slightly polluted; moderately polluted; medium pollution (or averagely polluted); higher than medium pollution, strong pollution (or high pollution), very strong pollution (or extreme pollution/extremely polluted).

Territories adjacent to strong pollution sites such as non-ferrous metal processing plants, ore-dressing and processing enterprises etc. were extremely polluted and sometimes their level of pollution exceeded 10 Clarkes. The pollution levels could be used as approximate indicators of adverse impact of chemical elements on environment. For example: at I and II (group) level pollution the soil's biota was strongly deteriorated, biochemical processes were suppressed. At III and IV (group) level of pollution agrochemical characteristics of the soils were worsened, vital functions of plants were disrupted and their chemical composition was changed. At V and VI (group) level of pollution, the plants became sick and died; plant and animal products were not fit for use, due to sanitary-hygienic considerations. Chemical composition of the upper layer of the soil changed and all agrochemical characteristics quickly deteriorated.

Results and Discussion

According to the data obtained during this study, total amount of copper, zinc, cadmium and sulphate-ion in Kazretula River, which was flowing under the above-mentioned tanks; exceeded several times the maximum permissible limit (Matchavariani and Kalandadze, 2012) established for surface waters. River was rife with ore elements. Due to low pH of water, these metals were mainly present in soluble form and could travel long distances. pH of irrigational water fluctuated from 3 (at the source of Kazretula) to 5 (in Mashavera, depending on distance of inflow Kazretula). At the same time, pH and turbidity of water increased, as a result of the metals flowing in the water and continued to migrate in that form. Despite this, the level of sulphate-ion in water remained quite high.

The copper level in soil was very high (more than 200 mg kg^{-1}), which was found in 18.3% of the total number of soil samples (Table 1, 2).

The soil in Bolnisi district was diverse both in their genesis and soil-formation pattern. Character and intensity of agricultural

Table 1 : Levels of heavy metals found in soils of arable land (mg kg^{-1})

Place of sampling & quantity of samples (n)	Cu	Zn	Mn	Pb
Ratevani, n=12	60-3625	75-2250	625-1000	19-36
Abdalo, n=5	40-88	100-625	1125-1375	20-35
Kazreti, n=10	35-200	85-100	750-1000	15-25
Khatisopeli, n=4	60-100	110-212	1000-1250	14-19
Balitchi, n=8	55-85	90-120	750-1125	25-25
Savaneti, n=4	40-115	105-135	875-1000	14-17
Kveshi, n=7	42-125	100-120	1000-1250	15-20

Table 2 : Levels of heavy metals found in soils of vineyard (mg kg^{-1})

Place of sampling and quantity of samples (n)	Cu	Zn	Mn	Pb
Ratevani, n=20	255-3125	165-2125	750-1625	22-41
Pakhralo, n=3	130-625	150-255	1000-1375	22-31
Kianreti, n=2	290-305	100-110	875-1125	32-35
Bolnisi, n=5	100-170	115-175	750-1375	17-22

Table 3 : Average levels of heavy metals (mg kg^{-1}) in the surface layer of the soil, 0-20 cm

Place of sampling and quantity of samples (n)	Cu	Zn	Mn	Pb
Ratevani				
Arable land, n=12	597	501	979	25
Vineyard, n=20	682	480	1150	30
Orchards, n=12	349	250	781	29
Kveshi, Abdalo				
Arable land, n=6	198	256	1208	20
Savaneti				
Arable land, n=3	73	118	958	15
Orchards, n=3	585	335	1208	25
Kianeti				
Arable land, n=11	55	80	897	24
Vineyard, n=2	297	105	1000	33
Khatisopeli				
Arable land, n=4	86	161	1125	16
Vineyard, n=1	140	160	1125	22

activities resulted in dramatic difference in the amount of various elements. High quantities of heavy metals (more than 200 mg kg^{-1}) were found in soils of several villages. It should be noted that sections that especially have high level of metals are often situated between the river basin and railway. Naturally, copper level in vineyard soil was much higher than in the ploughed fields. Copper level was also high in orchard soil. Most of the territory was covered by vineyards and orchards and more than half of the territory was occupied by wheat crops. More than half of this territory was quite highly polluted by copper and zinc; most of the area had 200-700 mg kg^{-1} copper and zinc concentration and about 8-9% of the area was considered to be catastrophically polluted. This area was intensively irrigated with of water Mashavera River. We can actually say that we're dealing with a clear case of anthropogenic impact, which means the impact of the irrigation system using polluted waters from the ore-dressing and processing enterprise. In other areas of the district where copper and zinc concentration (500 mg kg^{-1}) were detected, the pollution was fragmented.

Level of lead in wastewater and pit-run waters was minimal due to low dissolubility of its sulphates. Anthropogenic lead gets in to the environment mainly from vehicles. The maximum concentration of copper was 875.0 mg kg^{-1}. High level of zinc was detected in the same area. Thus, it can be concluded that these territories were under clear anthropogenic influence, which was intensified, even more, due to several other factors.

It's a well-known fact that manganese plays an important physiological role in the life of living beings and also very important as far as geochemical processes are concerned in soil, plant and water. Maximum manganese content in soil was 1,125-1,375 mg kg^{-1}. Pollution Concentration Coefficients was 5.6-6.8. Minimal concentration of manganese was 875 mg kg^{-1}. Manganese is an element of foundation rocks and gets accumulated in these rocks, especially prevalent in clay; smallest quantities of manganese are present in sandy soil (Hanauer et al., 2010). In the investigated area, the maximum level of manganese was found in basalt rock' crusts while minimal level was found in alluvial soils, where sand was prevalent (Table 3).

According to Vazhenin's classification (1987), 61.3 % of all analysed lands were slightly and moderately polluted with copper; pollution of 17.3% soil (30 ha) was more than average and 21.2% (24 ha) of land was either highly or extremely polluted.

As far as zinc concentration was concerned, the soil was slightly or moderately polluted in about 61.3% of the studied land. Pollution in 20 ha of land (18.4% of the analysed lands) was more than average; 24 ha of land (21.2% of all investigated lands) was either highly or extremely polluted. 81.5% of analysed soil had a medium or higher than medium manganese level and 18.4% of the soil had high manganese level.

The total indicator of concentration coefficient showed that only 19 ha of upper layer soil (0-20 cm) had small pollution level; 91.0 ha of the land were strongly polluted.

Chemical elements were represented in the following diminishing order: Mn>Zn>Cu in slightly polluted soil. In the average polluted upper layers of soil (0-20 cm) under minimal values of total quantities of pollution concentration (according to values of concentration coefficients) chemical elements were represented in the following diminishing order: Mn>Zn>Cu, the same diminishing order for Zn (n-1) was completely changed when applying maximum values: Cu>Zn>Mn.

During the last few years the soil characteristics have sharply deteriorated. At some places, the ground was covered with whitish/greenish waterproof coats. Porosity of the soil, as well as, its productivity had diminished. The limestone was added to the wastewaters of Madneuli Enterprise in order to neutralize acid, after that this wastewater was discharged into the sewer. Gypsum formed here, flowed in to the river water and further this water was used for irrigation of agricultural land plots. With the course of time, gypsum got accumulated on the ground surface and coated the soil, which worsened the aeration and filtration capabilities of soil, subsequently leading to sharp decrease in the productivity.

Characteristics of average polluted soil in the investigated areas according to total values of pollution concentration coefficient revealed that the soil was heavy clay loam, solid-phase density of soil (specific weight d) in the cross-section was unnaturally differentiated, it was higher in the ploughed layer (3.53 g cm^{-1}), and lower at greater depth (2.49-2.33 g cm^{-1}). Optimal bulk density (dv) of soil was 0.90 cm in the ploughed layers, and naturally increased up to 1.18 g cm^{-1}. According to the analysed data, soil was not consolidated in the lower layers of ploughed field, which was a positive factor as it conditioned positive indicators of the rest of the parameters. General porosity of the soils in ploughed field and below was good and the whole cross-section of soil showed that the growth conditions for plants were satisfactory (45.3%-64.4%). The favourable aeration conditions for plants were created in soil, favourable hydrological characteristics were formed and thick and capillary pores were able to maintain moisture. This was evidenced by large quantities of productive moisture and high level of maximum moisture volume in the fields. The amount of productive moisture and level of maximum moisture volume in the soils was 30.5-51.0% and 19.3-32.7%, that was considered to be the best values for irrigated soils. Table 4 shows the average content of heavy metals in soils in vertical profile.

Due to low bulk density (dv) and high general porosity (P) soil had good filtration capability – water traveled 1.5 m during the twenty-four hour period.

Soil grouped under medium pollution category (soils polluted with Zn (n-1) had satisfactory hydro-physical characteristics. The specific weight of soil (d) was normal (2.51-2.60 g cm^{-1}), and was naturally distributed in the cross-section of soil. Volume weight (dv) in the cross-section was not high (from 1.18 g cm^{-1} to 1.28 g cm^{-1}), which was quite acceptable. The level of maximum moisture volume in soil was average. The important hydro-physical component in the ploughed part of soil was about 3.5%-20%. The range of productive layer was relatively low (16.9%-25%). As far as filtration ability was concerned, water traveled 0.93 m during the twenty-four hour period, which was not a bad result.

Table 4 : Average level of heavy metals (mg kg^{-1}) in soil profile

Deep of soil horizons	Cu	Zn	Mn
0-20	155	116	967
20-40	71	104	960
40-60	55	95	95
60-80	47	90	1050
80-100	48	86	930

As far as highly polluted soil and their hydro-physical characteristics were concerned, the whole cross-section of soil indicated that there were no favourable conditions for normal development of plants. According to granulometric analysis results, the soil was light clay loam, which indicated the presence of heavy metals in it. The specific weight of soil along the whole cross-section was not optimal (from 2.32 g cm^{-1} to 2.47 g cm^{-1}). Volume weight of soil starting from the upper layer of the ploughed soil was pretty high (1.20 g cm^{-1} in the ploughed portion up to 1.35 g cm^{-1} in the lower portions). This indicated that soil was significantly solidified, which created unfavourable hydro-physical conditions for plants. The level of maximum moisture volume was not acceptable (19.0%-82.6%), and the amount of productive moisture in soil was very low (6.0%-20.6%).

Vegetation is a defining factor of geo-chemical processes taking place in soil. Plants have selective absorption ability. They are able to receive various chemical elements from soil disproportionately from their composition; in other words, they are able to select the chemical elements which are necessary for their growth. In numbers this phenomenon is reflected in the biological coefficient of absorption. The level of heavy metals in different plants (corn, pumpkin, grapes) was as follows: Grapes: Cu – 597 mg kg^{-1}; Zn – 501 mg kg^{-1}; Mn – 979 mg kg^{-1}; Pb – 25 mg kg^{-1}; Pumpkin: Cu – 682 mg kg^{-1}; Zn – 480 mg kg^{-1}; Mn – 1150 mg kg^{-1}; Pb – 30 mg kg^{-1}; Orchards: Cu – 349 mg kg^{-1}; Zn – 250 mg kg^{-1}; Mn – 781 mg kg^{-1}; Pb – 29 mg kg^{-1}, respectively.

Productive moisture was the most important component of moisture categories existing in soil. Productive moisture assist the plants to absorb dissolved substances. Its deficit was acutely felt by crops, which resulted in deterioration of plants, decreased yield and in many cases led to death of plants.

Polluting heavy metals – copper, zinc and manganese especially have negative impact on soil characteristics, its composition and soil-formation processes, which results in deterioration of hydro-physical potential of the soil. Balanced correlation between solid, liquid and air phases in the soil was disturbed. Summation of the agro-physical parameters of less, average and highly polluted soil provided a clear evidence of soil-formation process. Important anthropogenic factors causing soil degradation were irrigation with polluted water and using various chemical pesticides. Agro-physical characteristics of soil were directly connected with pollution of soil with heavy metals. The cementing processes taking place in highly polluted soils, sharply increases the bulk density of soil, the porosity of soil deteriorates and water permeability is critically lowered.

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