

EVALUATION OF GEOCHEMICAL PECULIARITIES OF MINING ROCKS FOR ECOLOGICAL RESTORATION OF THE POST-MINING LANDSCAPE

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ABSTRACT

In regions with complex geological history, diversity of climate, soils and parent rocks, it is possible to predict a large number of paths for soils recovery in the areas that have been disturbed by human activity. The purpose of this work was to identify territorial difference of rocks within the Crimean Peninsula basing on the content of the trace elements and macroelements, assessment of potential suitability of disturbed rocks of various age and chemical composition for efficient pedogenesis during implementation of projects for vegetative topsoil recovery in post-mining landscapes. The studied objects have been classified as per the results of cluster analysis, and seven main groups of rocks have been determined based on the content of 16 chemical elements. The most informative geochemical indicators and ratios have been determined; this allowed justifying three general types of rocks based on the difference of their properties related to their mineral composition and the results of weathering and migration processes in the past. In order to determine the initial conditions for soil recovery, geochemical particularities of upper horizons of soils and parent rocks composing them in three regions of the Crimean Peninsula have been compared. The ordered series of chemical elements have been created based on their ability to accumulate in soils compared to rocks. This allowed determining the deficiency of those elements that can restrain the activity of biological and soil processes during soil recovery in post-mining landscapes. The results of the study create a bioecological basis for ecological restoration of disturbed soils represented by sedimentary rocks of different geochemical composition in the development of the mineral deposits of solid useful minerals.

Keywords: parent rocks, geochemical indicators, Crimean Peninsula, post-mining landscapes, ecological restoration.

INTRODUCTION

In the situation when the land is disturbed in the result of mining the biological reclamation of lands is required. The most economic way is dumps overgrowth or ecological restoration of landscape, which requires taking into account the geochemical behaviour trace elements in the soil-rock interface [1 et al.]. Linking parent material to soil properties is an active research topic in the emerging Critical Zone science [2]. Now, when distribution of typomorphic elements and some essential trace elements:

Mn, Cu, Zn, Co, Mo, B, I in the soil is rather well studied, their biological role of the rare earth elements: Ti, Cr, V, Ni, Ga, Ba, Zr, Sr, Be [3] is still less studied.

The results of the study which has been carried out with purpose to identify the source for major and trace elements, this role showed that the association of soil-forming major element (Al, Fe, K, Mg) with some trace elements (Co, Ni, Zn, Pb) denotes their geogenic origin [4].

A significant peculiarity of anthropogenic substrates is heterogeneous in their lithological composition caused by mechanical mixing. Disturbed rocks and their technical mixtures can represent Anthropogenic, Neogene, Palaeogene, Jurassic period and can be a substrate for the formation of groups of autotrophic plants. Often dumping leads to formation of substrates which have no analogues in the surrounding landscapes and have qualitatively new (emergent) properties due to various rocks content in the anthropogenic mixture. At dumps overgrowing the process of trace elements accumulation in recent soils is positively influenced by a substance with increased content of organic matter, silt fraction, clay minerals with expanding lattice.

The content of trace elements in rocks is primarily determined by their mineralogical composition. The more quartz and feldspars are found in the rock, the less heavy accessory minerals (elements carriers of trace elements) will be in it and the poorer the rock will be as a whole [3]. The composition of minerals in sedimentary rocks, if they become parent rocks in post-mining landscapes, creates a structural and functional matrix for recent soils.

Coarse-grained primary minerals mostly determine the agrophysical properties of soils [5], and they are a backup source of secondary minerals and ash elements for plants nutrition. With secondary minerals, accumulated as the result of weathering, for example, clay with montmorillonite [6], the conditions for pedogenesis commencement become more favourable. It should be noted that in assessing the suitability of different types of anthropogenic substrates for biological restoration their edaphic (soil) indicators are usually used: hydrophysical properties, saturation with mineral nutritive elements, and content of toxic substances. In the geochemical cycle the essential elements as well as toxic elements should be in focus soil monitoring [7, 8].

The present study aims to identify territorial difference of parent rocks within the Crimean Peninsula basing on assessment of potential suitability of disturbed rocks of various age and chemical composition for efficient pedogenesis during implementation of recovery projects in post-mining landscapes.

DATA AND METHODS

Description of the study area. The diversity of soils on a relatively small territory of the Crimean Peninsula (25880 km²) is very great (51 soil units in the systematic list of Crimean soils). Anthropogenically transformed soils and soils developed on cultural layers of different ages are widespread in Crimea [9]. On the Crimean Peninsula the proportion of soil on dense rocks and their deluvium is 33.7% of the whole area.

Shelly limestone formed in the place of seas, which in Neogene covered the plain and piedmont areas of Crimea. In the North-Western Crimea Miocene and Pontian limestones crop out, and their eluvium can act as parent rocks of the Rendzic Leptosol.

In the South-Western Crimea Pre-Quaternary sediments are represented by limestones of Miocene and (closed to the mountains) by limestones of Upper Jurassic. Marbly limestone of Upper Jurassic is represented as a discontinuous path running in the mountainous part from the eastern border of Sevastopol to Feodosia.

In the structure of mineral-resources complex of raw minerals of Crimea (123 deposits of 12 groups of useful minerals) the leading position belongs to sawn and building ones – 43 and 15% deposits respectively. Building limestone is developed by way of open-pit mining in more than 100 quarries, the total area of which is 13 000 ha (0.5% territory of the Crimean Peninsula).

The minerals deposits of the Crimean Peninsula are inhomogenously distributed in this area due to differences in spatial distribution of lithological and stratigraphical complexes in the age range from Jurassic to recent sediments.

Data used. The geochemical peculiarities of parent rocks have been studied in the main geographical areas of Crimea (34 most typical objects were selected). The concentration of 22 macroelements and trace elements (CaO, Al₂O₃, SiO₂, P₂O₅, K₂O, Na₂O, MgO, MnO, Fe, TiO₂, V, Cr, Co, Ni, Cu, Zn, Sr, Pb, Rb, Ba, Zr, As) in each rock has been determined. The processes of accumulation and dispersion were studied on the basis of 16 macroelements and trace elements, which showed a considerable variation of the content among the groups. Reference soils in three different climate regions of the Crimean Peninsula: Rendzic Leptosol (Skeletal) in the North-Western Crimea (S1); Calcic Chernozem (secondary carbonates) – virgin soil in the foothill Crimea (S2); S3 – cinnamon red calcareous deep light-clay soil on cluvium-deluvium limestone of the upper Jurassic (Cape Martyan) were selected for comparative analysis of the geochemical particularities of upper horizons of soils (A, 0-17(36) cm) and parent rocks composing them.

Methods. The colour was determined in the dry and moist state, using the Munsell color charts. The chemical properties of soils were analyzed by routine methods: the Corg content, by Tyurin's method; content the CO₂ of carbonates by acidimetry; the available P₂O₅ (mg kg⁻¹) by Machigin's method (spectrophotometer UNICO-1200, USA, 2012). Concentration of macroelements and trace elements within the soils were determined by technique of measuring metals mass fraction and oxides in powder samples using the method of X-ray fluorescence analysis (Spectroscan Max-GV). The conversion coefficients were used for conversion of oxides into elements. The suitability characteristics of rocks for recent pedogenesis were based on integral assessment of nutritive elements content (required macroelements, trace elements and useful elements [10]). A useful index of potential soil fertility (FI) is the ratio of the base cation elements, calcium and magnesium, and total phosphorus to silicon [11]. We have modified the eluviation's coefficient $K_e = \text{SiO}_3/(\text{MnO}+\text{CaO}+\text{K}_2\text{O}+\text{MgO}+\text{Na}_2\text{O})$ [12] and in K_e calculation we used the formula: $K_e = \text{Si}/(\text{Ca}+\text{Mg}+\text{Mn}+\text{K}+\text{Na})$. Basing on the fact, that in the process of formation and growth the soils accumulate and keep the main volume of trace elements, and the plants demand is determined by content of necessary macroelements and trace elements in soil, we can make a list of main typomorphic elements and essential trace elements. Total number of rocks (RQ) and soil (SQ) for separate geographical areas was assessed using the formula: $RQ(SQ) = \sqrt[n]{K_1 \cdot K_2 \dots K_n}$, $K_1, K_2 \dots K_n$ – content of essential elements: Si, Al, K, Mg, Ca, Mn, Fe, Zn, Ni, Cu, Co. The assessment of rocks suitability for restoration was

performed on the basis of indicator EQ, which was calculated as geometric mean value of content of such essential elements: Si, Al, K, Fe, Zn, Ni, Cu, Co. In the cluster analysis of soil data, we used Ward's method, in which clustering procedures are based on the criterion of squared Euclidean distance.

RESULTS AND DISCUSSION

The data on elements content in 34 typical objects was analyzed and basing on the value of variation coefficient ($V > 20\%$) the most informative elements were selected (Table 1).

Table 1. The geochemical characteristics of the main genetic groups of the parent rocks of Crimea

Chemical elements		Number groups ^a							
		1	2	3	4	5	6	7	
Fe	%	2.0	2.4	1.0	1.5	1.6	2.4	6.6	
Ca		9.14	7.10	27.83	17.40	17.99	15.76	0.51	
Na		1.31	1.05	2.60	1.75	1.91	1.95	0.24	
Al		5.2	5.2	2.8	4.4	4.5	4.9	10.4	
Mg		1.16	0.99	2.25	1.58	1.68	2.70	0.83	
P		0.09	0.06	0.12	0.07	0.15	0.07	0.05	
Mn		0.07	0.08	0.04	0.06	0.08	0.05	0.04	
Si		18.9	20.1	5.9	14.2	13.1	15.5	20.8	
K		1.34	1.34	0.73	0.99	1.23	1.07	1.30	
Cu		37.4	49.9	5.1	20.2	23.3	58.0	88.6	
Ni	44.7	56.9	21.5	34.4	37.1	64.6	120.5		
Co	12.0	19.8	4.4	7.7	6.3	10.0	13.1		
Sr	mg kg ⁻¹	207.5	213.4	392.9	217.4	189.4	144.3	70.4	
Pb		17.6	27.8	9.7	10.5	16.4	25.0	25.0	
Zn		74.7	104.8	57.8	65.5	89.6	79.1	163.6	
Cr		86.9	100.9	65.4	77.0	76.2	63.6	138.1	
EQ		-	11.78	14.45	4.67	8.62	9.13	12.43	21.29

^a1 – carbonate pale brown loam (North-Western Crimea); 2 – carbonate brown and greyish brown loam, underlain by limestone (North-Western Crimea); 3 – very pale brown and light grey loamy eluvium of limestone of Western Crimea; 4 – very pale brown eluvium of limestone and clay eluvium (North-Western Crimea); 5 – eluvium of limestone and carbonate brown and greyish brown loam (Piedmont Crimea); 6 – brownish and reddish yellow eluvium of limestone Herakleian Peninsula; 7 – dark red *terra rossa* in cracks limestone of Herakleian Peninsula.

Using the average values of macro- and trace elements content (Table 1) and values of geochemical coefficients, normalized basing on standard deviation, was carried out cluster analysis (Figure A, B).

Geochemical differences of parent rocks are mainly based on the geographical areas of Crimea. In the analysis dendrogram of the cluster analysis (see Figure) it is shown that the rocks of Western Crimea, including Herakleian Peninsula (groups 3, 7, 6), significantly differ from the carbonate rocks Foothill Crimea, but to the greatest extent – from loams and eluvium limestone. In classification terms, eluvium limestone, clay

cluvium and carbonate loam, as in the North-Western (group 4), and in the Foothill Crimea (group 5), have a significant geochemical similarity.

At some differences between carbonate loam and loam, which underlain limestone in North-Western Crimea (groups 1, 2), they together with previous groups form relatively homogeneous common parent rocks mass of the Plain and Foothill Crimea.

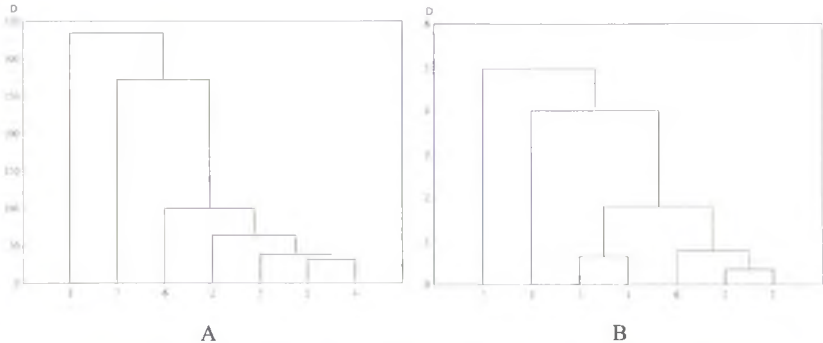


Figure. Dendrogram of clusters analysis of soil-forming rock by the content of 16 macroelements and trace elements (A) and by geochemical coefficients (B). D – Linkage distance; No groups, see note to the table 1

Grouping by geochemical composition allowed defining seven main groups of parent rocks based on the content of 16 chemical elements. Prevailing colour of dry rocks (Munsell charts) is different for selected types: 1 – very pale brown and yellowish brown; 2 – greyish brown and brown; 3 – very pale brown and light grey; 4 – very pale brown and brown; 5 – brown and greyish brown; 6 – brownish yellow; 7 – dark red.

Are fundamentally different geochemically from the abovementioned common parent rocks mass of South-Western Crimea (groups 6, 7 and 3 as the difference is growing). In the most generalized form, this identity of rocks is reflected by the ratios that are represented in Table 2.

Table 2. Geochemical indicators of weathering and pedogenesis for major groups of the parent rocks

Geochemical indicators	Groups						
	1	2	3	4	5	6	7
Na/Al	0.25	0.21	1.06	0.40	0.42	0.42	0.02
(Ca+Mg+K)/Si	0.65	0.48	5.66	1.47	1.72	1.35	0.13
Al/(Ca+Na+K+Mg)	0.41	0.52	0.08	0.20	0.20	0.25	3.64
(Fe+Al)/(Ca+Na+Mg)	0.63	0.87	0.12	0.28	0.29	0.40	10.81
FI=(Ca+Mg + 10·P ₂ O ₅)/Si	0.63	0.45	5.76	1.45	1.75	1.33	0.09
(Fe+Mg+Al)/(Na+Ca+K)	0.72	0.93	0.19	0.37	0.37	0.57	8.71
Ke	1.51	1.97	0.18	0.66	0.59	0.79	7.13
Ka = (Si·Ca·P·K·Co·Cu·Fe) ^{1/7}	3.96	4.26	2.09	3.23	3.64	4.21	3.37

The most informative geochemical indicators and ratios of weathering and pedogenesis for major groups of the parent rocks have been determined; this allowed justifying three

general types of rocks based on the difference of their properties related to their mineral composition and the results of weathering and migration processes in the past.

Special Group basing on geochemical peculiarities is dark red *terra rossa* in the fissure of limestone (group 7). As per the values of the geochemical indicators of weathering and pedogenesis the group 3 in the Western Crimea – very pale brown and light grey loamy eluvium limestone differs from the other group the most. The coefficient of weathering represents the ratio of Al_2O_3 (the clay component) to the main cations removed to the soil solutions showed a high degree of transformation of the dark red *terra rossa* (group 7) compared to eluvium limestones (group 3), although the objects are in the same area – Western Crimea. A comparison of rock suitability for renaturation showed the excesses EQ (Table 1) for rocks of group 7 over group 3 in 4.6 times. Intragroup differences are met in carbonate brown and greyish brown loams, as well as brown and yellow eluvium limestones and clay eluvium in the plains and piedmonts (the other five groups).

The soils and their geochemical composition in particular experience significant transformations as the result of long farming activity on plains [13], as well as in mountainous areas. [14]. So the difference between the upper soils and parent rocks can increase first of all due to elements of high migratory mobility and organogene elements.

Table. 3. Chemical properties of the upper horizons of virgin soils in different regions of the Crimean Peninsula

Soils ^a	Colour (moist)	Colour (dry)	Layer, (cm)	pH (H ₂ O)	Corg (%)	CaCO ₃ (%)	P ₂ O ₅ (mg kg ⁻¹)
S1	10 YR 4/3	10 YR 5/3	0-36	8.37	2.47	36.06	5.23
S2	5 YR 3/4	5 YR 4/4	0-20	7.67	2.60	2.50	0.38
S3	10 YR 3/2	10 YR 5/2	0-19	7.46	1.52	20.01	2.91

^aS1 – soil of North-Western Crimea (groups 1-4); S2 – soil of the Foothill Crimea (group 5); S3 – soil the South Coast of Crimea (groups 6-7).

Estimates soil quality (SQ) for 11 essential elements defined such distribution of soils by ranks: S3 (6.6) > S2 (6.3) > S1 (5.3). The greatest differences between the seven groups of rocks, which are established by the ratio of content of the chemical elements in soil and parent rock, are determined Co, As, Cu, Pb, Ni, Mg, Mn, Si, Ca, Al, Fe, Na (V>33%), and Cr, P, Zn, Sr, K (V<33%). The analysis S/P of the seven groups of parent rocks (Table 4) allowed substantiating the list of seven elements of life, which were included in the structure of the accumulation coefficient (Ka). The rocks the most saturated with these elements parent rocks in group 3 (Table 2).

Table 4. The values of the ratio of the content of chemical elements in the soil in relation to the parent rocks (S/P)

Chemical elements		No groups							$\bar{X} \pm S_x$
		1	2	3	4	5	6	7	
K	%	0.98	1.00	1.82	1.33	0.92	1.44	1.18	1.54±0.3
Si		0.83	0.76	2.75	1.11	1.09	1.18	0.85	1.39±0.3
P		1.16	1.36	0.85	1.15	0.65	0.69	1.01	1.21±0.2
Ca		1.67	2.15	0.53	0.86	0.55	0.10	2.85	1.13±0.2
Mg		1.15	1.35	0.58	0.82	0.79	0.25	0.80	1.10±0.2
Mn		0.78	0.68	1.39	1.02	3.62	1.89	1.87	1.07±0.1
Fe		0.86	0.71	1.83	1.15	1.37	1.30	0.45	1.04±0.1
Na		1.09	1.40	0.53	0.79	0.90	0.26	2.01	0.98±0.1
Al		0.97	0.96	1.97	1.15	1.02	1.58	0.72	0.76±0.1
Co		0.57	0.33	2.63	0.90	2.12	2.40	1.84	1.61±0.4
As	mg kg ⁻¹	0.78	0.84	1.05	0.80	1.48	1.76	1.76	1.24±0.1
Cu		0.82	0.61	2.45	1.62	2.00	1.33	0.86	1.24±0.4
Cr		0.91	0.78	1.21	1.03	1.08	1.71	0.78	1.22±0.3
Pb		0.82	0.51	1.54	1.41	1.87	0.89	0.89	1.19±0.2
Sr		0.85	0.85	0.45	0.77	0.65	0.65	1.07	1.10±0.2
Ni		0.87	0.69	1.81	1.14	1.45	1.14	0.60	1.00±0.2
Zn		1.00	0.71	1.33	1.14	0.97	1.46	0.68	0.82±0.1
Groups	Increasing series of accumulation				Waning scattering series				
1	Zn<(Mg, P)<Ca				Co>Mn>(Cu, Si)>(Fe, Ni)>(Al, K)				
2	K<(Mg, P)<Ca				Co>Cu>Mn>Ni>Fe>Zn>Si>Al				
3	Zn<Mn<(Ni, K, Fe)<Al<Cu<Co<Si				Ca>Mg>P				
4	Mn<Si<(Ni, Zn, Fe, Al, P)<K<Cu				Mg>Ca>Co				
5	Al<Si<Fe<Ni<Cu<Co<Mn				Ca>P>Mg>K>Zn				
6	Ni<Si<Fe<Cu<(K, Zn)<Al<Mn<Co				Ca>Mg>P				
7	P<K<Co<Mn<Ca				Fe>Ni>Zn>Al>Mg>Si>Cu				

Territory of the Crimean Peninsula is a region with complex geological history, diversity of climate, soils, sedimentary and parent rocks. Therefore, here is possible to predict a large number of paths for soils recovery in the lands that have been disturbed by human activity. The change in the rates of weathering and pedogenesis, which both in Holocene [15-17] and in the modern era [18] were due to climatic rhythm, formed the accumulated effect in the geochemical composition of soils and rocks.

Using the obtained series, we calculated the rock quality (RQ) for each of the 7 groups and soil quality (SQ) for the soils of the three areas of Crimea. When comparing the obtained values, it was found that in the 1st and 4th group the contents of elements in parent rocks are equal to the soil. In the 2nd group there is a deficiency of Ca in the rock, in the 3rd group – Si, Cu, Al, K, Ni, in the 5 group – Mn, Cu, Ni, Fe, in the 6 and 7 groups – Co, K, Ca.

Among the essential elements in the synthesis of humus are involved Mn, Ni, Cu, Zn, Co, and in the synthesis of colloids and clay formation – Mn, Fe, Ni, Cu, Zn, Co [3]. A comparative assessment of the quality of rocks (RQ) has shown that the most complete set of essential elements with their high content is group 6 (eluvium limestone of South-Western Crimea, where, it is worth noting, 30 deposits of limestone are located). But the soil of this area has the highest quality (1.4 times more than parent rocks). Group 5 concedes to group 7 (1.5 times in value of RQ), the rocks from groups 4, 1 and 3 have lower quality, however the lowest content of elements of life have the rocks from group 2 and especially 7.

In general, there is a close correlation between the soils and their parent rocks (by values SQ and RQ), which is however most weakened in rocks from group 3, as well as from group 4 and 1. The differences in the quality assessment of rocks (RQ) when compared to the quality of the soils (SQ) that have formed on these rocks are most different in the three groups of seven. The objects in the group 3 (eluvium of Jurassic limestone) have the lowest values RQ when compared to values SQ (deficit Si, Co, Cu, Al, Fe, K, Ni, Mn, Zn).

It can be assumed that the process of recent pedogenesis on such rocks will take place at the lowest rates. The rocks from the following groups 1 and 4 (loam and eluvium Neogene limestone) have another potential for restoration: they are the closest in geochemistry to the soils, so the restoration of soils and vegetation on these rocks will be the most efficient. However, it is necessary to note, that the soils which are formed on carbonate loam (group 1), have accumulation only Ca, P, Mg, Na, and by 9 essential elements concede to the rock. Essential trace elements are most fully (9 in 11) represented in groups 3, 4 and 6, a bit less in the 5th group (7); groups 1, 2 and 7 are the poorest by the content of these trace elements (from 4 to 5).

Such division into groups differs from the general geochemical classification of rocks, where the concentration of 16 chemical elements was taken into account (see *Figure*). A comparison of values SQ and RQ of the seven established groups of parent rock showed that in the restoration of soils achievement in post mining landscapes of soils characteristics in the corresponding bioclimatic region could be prevented by a deficiency in the source rocks the following elements of life: Si, Cu, Al, K, Ni in the 3rd group, Mn, Cu, Ni, Fe in the 5th group, Co, K, Ca in the 6th and 7th groups. Deficient elements may slow down the activity of biological and soil processes during soil recovery when spontaneous succession. However, in managed succession (selection of tolerant and adapted grass mixtures) in post-mining landscapes these problems can be minimized.

CONCLUSIONS

High ecological lability and plasticity of young ecosystems open possibilities for management of the renaturation process (control over succession, built-in subsidy actions). Thus, the renaturation should not be limited only by stimulation of overgrowth of exposed rocks in post-mining landscapes. If in the development of mineral deposit of the solid useful minerals there are sedimentary rocks of different geochemical composition, as is observed in the area of our study, an adapted strategy of the ecological restoration of disturbed lands is needed for the conditions of their natural recovery. Even similar rocks, but with different geological age and palaeogeographical

history, as eluvium limestone, differ significantly by the geochemistry and the potential of the recent pedogenesis. The soil formation capacity of parent rocks allows adjusting the conditions of plants growth to favourable, and accordingly to stimulate effective restoration of soils in post-mining landscapes. Natural processes (spontaneous or managed succession) in post mining landscapes should be integrated into the legal regulations and legal practice [19], as by estimation of suitability of sedimentary rocks it is possible to define the most effective scenarios of renaturation in post-mining landscapes with the help of optimal biolithological combinations.

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