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## Rare and trace elements of the Verkhniidniprovske deposit of the Dnipro brown coal basin as an indicator of Ukrainian Shield metallogeny

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Рідкісні та розсіяні елементи Верхньодніпровського родовища Дніпровського буровугільного басейну як показник металогенії Українського щита

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**Ключові слова:** Дніпровський буровугільний басейн, Український щит, рідкісні та розсіяні елементи, мікрокомпоненти вугілля, кореляційний аналіз.

The geochemical features of the coal from the Verkhniidniprovske deposit of the Dnipro brown coal basin, located within the Dnipro megablock of the Ukrainian Shield, have been studied. The position of the deposit within the Shield was defined according to metallogenic, geochemical, and landscape-geochemical zoning. It is shown that the crystalline rocks of the Ukrainian Shield are the source of rare and trace elements in the coal. Correlation analysis was used to evaluate the associations between the considered elements. Two stable associations have been found: Yb-Y-Be and Ti-Zr-Sc-V-Cr. The migration pathways of the elements into the peat bed and the possible forms of fixation of the elements in the organic matter of the coal are demonstrated. For the first time, an analysis of the correlations of the considered elements with the microcomponents of the coal organic matter of the lignite, huminite-helite, fusinite and liptinite groups is presented. The most coalphilic elements are Be, Yb, Y, Ge, Mo, Sc. Elements such as Ti, Zr, V, Cr, Pb, Ga, Cu, La, Mn, Ni are more often associated with terrigenous material, which does not exclude their presence in coal in sorption and organically bound form. A highly correlated dependence of Ge on the resin content in the benzene extract was found, which makes it possible to recover Ge in the technological process of bitumen extraction from coal.

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## Introduction

The study of rare and trace elements in coal, which are considered as associated minerals and are used in almost all areas of science and technology, is of great scientific and applied importance.

The study of the metal content of brown coal, especially of the Dnipro brown coal basin (Dniprobass), began in the 1960s. The conditions of migration and concentration of elements in coal, the role of tectonic processes in the metal content of coal, etc. were studied (Mitskevych, 1962; Saprykin, Bogdanov, 1967; Kler, 1979; Lepigov et al., 1986; Radzivil et al., 1987 et al.). However, many issues of coal geochemistry and the role of its individual microcomponents in this process remain to be solved. Previous studies on the metallogeny of the Ukrainian Shield (USh), relating to its metallogenic, geochemical and landscape-geochemical zonation, are mentioned below with appropriate references.

The aim of this study was to identify the pathways of elements entering the peat beds from the source areas, the form of their accumulation, and to determine the role of coal as a concentrator of rare and trace elements.

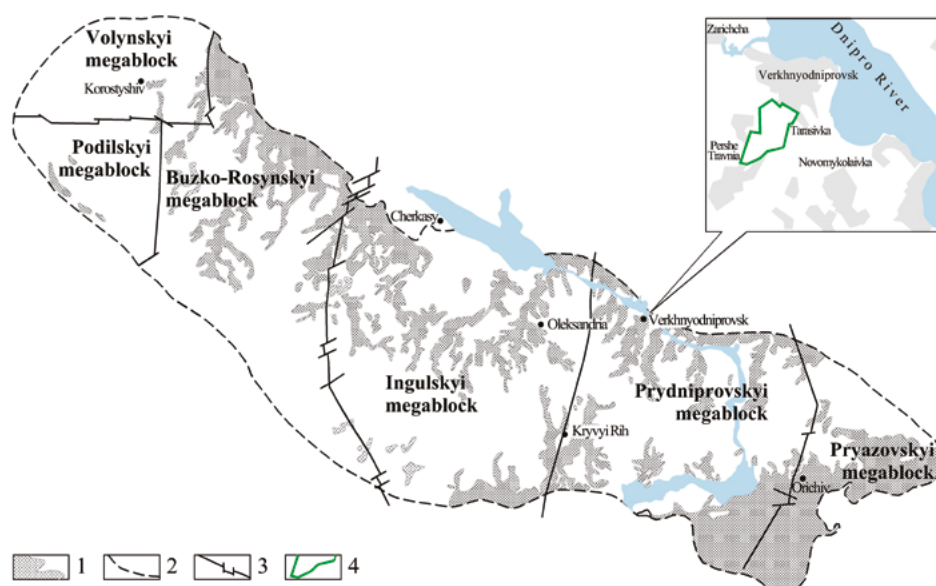
150 petrographic sections were analyzed in transmitted polarized light, the results of 146 determinations of chemical and technological indicators of coal quality, 144 determinations of the chemical composition of ash and spectral analysis were used. Coal petrographic, geochemical, chemical, and statistical research methods were applied.

The materials of this article were presented at the All-Ukrainian scientific conference "Geological Structure and History of Geological Development of the Ukrainian Shield" (Ivanova et al., 2024).

## Object of study

The geochemical characteristics of coal from the Verkhniodniprovske deposit of the Dniprobass, which is located within the USh, were studied (Fig. 1).

The main source of rare and trace elements in the peat beds were the rocks of the crystalline basement of the Dnipro granite-greenstone catastable megablock of the USh (Kyryliuk, Shevchenko, 2023). According to the metallogenic zoning (Bochay et al., 1999; Goshovskiy, 2003), the deposit belongs to the Middle Dnipro subprovince with a wide development of greenstone rocks, the metallogenic specificity of which is determined by the presence of Ni, Co, Cu, Cr, Mo, Au ore occurrences and titanium-zirconium placers. According to the metallogenic zoning by individual stages of the development of the USh, this area belongs to the Middle Dnipro metallogenic region, the Paleogene deposits of which are characterized by the presence of Ti, Zr, and partially Sn (Shevchenko, Goyzhevskiy, 1984). According to the map of rare metal mineralization of the USh, the deposit belongs to the Dnipro metallogenic subprovince with ore occurrences of Mo, Ta-Nb, rare-earth elements (Nechaev et al., 2019). According to the geochemical zoning, the deposit is located within the Dnipro weakly differentiated chalcophile-siderophile geochemical region, which is characterized by numerous local concentrations of Cu, Ni, Co, Mo, Mn, Sc, V (Galetskiy, 1984). According to the landscape-geochemical zoning of the territory of the USh (Mitskevych, 1971), the deposit is located within the Steppe region (Ingul-Ingulets steppe subregion), where in the humus horizons of geochemical landscapes directly connected with the



**Fig. 1.** Dnipro brown coal basin (Radzivil et al., 1987; Kyryliuk, Shevchenko, 2023): 1 – coal-bearing deposits of the Buchak suite; 2 – boundaries of the Dniprobass; 3 – deep faults; 4 – contours of the Verkhniodniprovske deposit

crystalline rock basement, the elements Mn, Ni, Ti, V, Cr, Zr, Cu, Pb, Zn, Y, Sr form the most stable secondary dispersion halos, while Co, Be, Sn, Ga, La, Ba form less stable ones.

A similar spectrum of elements was found in the coal of the Verkhniidniprovske deposit: Mn, Ni, Ti, V, Cr, Mo, Zr, Cu, Pb, Ga, Ge, Be, Sc, Y, Yb, La.

The degree of concentration of the elements, their qualitative composition and distribution features were determined both by the crystal-chemical properties of the atoms of the elements and by the physico-chemical and facies conditions of peat bog formation.

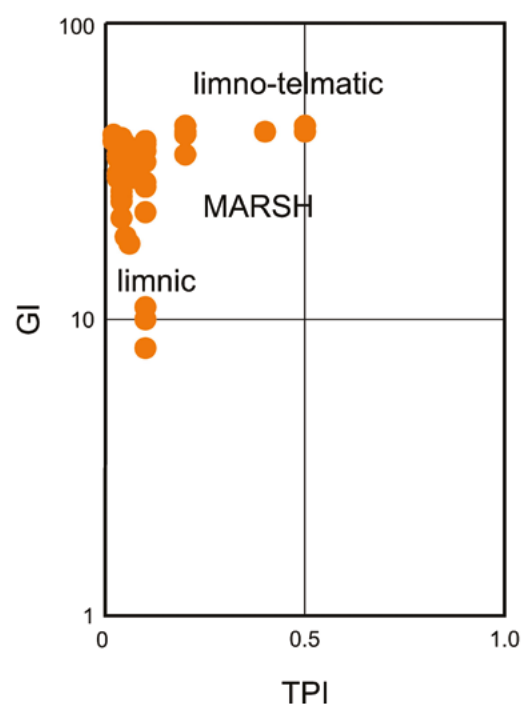
## Coal petrographic studies

The classification developed by M.O. Ignatchenko (Ignatchenko, Zaitseva, 1978) for brown coal was used to establish the features of the relationship between rare elements and microcomponents of brown coal during microscopic examination. An important feature of the classification is the ability to clearly subdivide organic microcomponents depending on the source material, morphology, structure, and degree of transformation of plant residues. The studied coal was divided into lignite, huminite-gelinite, fusinite, and liptinite groups. The primary role belongs to the components of the huminite-helinite and lignite groups. Within the groups, microcomponents are divided depending on the raw plant material, the anatomical and morphological features of the plant residue and the degree of preservation of the botanical structure.

The lignite group is represented by yellow and yellowish-orange tissues that originated from the remains of coniferous plants. The huminite-helinite group, which plays a major role in the composition of the coal, includes various red, brownish-red tissues of angiosperms (xylinite, parenchyme, felinite). Structural fragments are observed quite rarely, atrite is the most common. Sometimes dopplerinite is found, filling the cavities of cracks. The components of the fusinite group are observed sporadically and are represented by single fragments of semifusinite. The prevalence of ortho- and nigrosclerotinite is characteristic for the brown coal of the studied deposit. The composition of the liptinite group includes various components: suberinite of different states of preservation, suberinite-atrite, resinite, cutinite bordering leaf blades, and, rarely – microexinite. It is necessary to note the significant distribution of the structureless microcomponent – bituminite-desmite.

Mineral components are represented by quartz, clayey material, often of kaolinite composition, muscovite, biotite, glauconite, pyrite. Minerals of feldspar, amphiboles and pyroxene groups are less common. Accessory minerals are quite common: tourmaline, zircon, rutile, garnet, magnetite, ilmenite, staurolite, etc.

The assessment of the paleoenvironment of peat formation based on the microcomponent composition of coal by the method of S.F.K. Diessel (Diessel, 1992) indicates that peat accumulation occurred in the zone of the lacustrine-marsh coastal lowland under conditions of high flooding and erosion-tectonic paleovalleys with a noticeable influence of the marine environment (limno-telmatic conditions, marsh), less often in overgrown lakes (limnic conditions) (Ivanova et al., 2021) (Fig. 2).

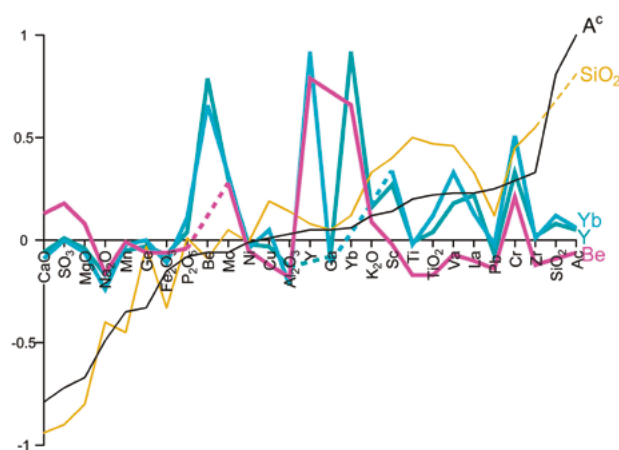


**Fig. 2.** Diagram of the reconstruction of the conditions of peat accumulation in the Verkhniidniprovske deposit of the Dniprobas

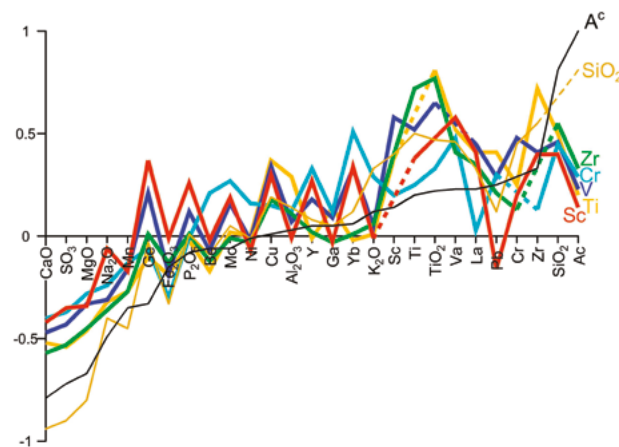
The chemical influence of the seawater on the peat bed is indicated by the ratio  $\text{Ca}^{++}/\text{Mg}^{++}$  (Werner's coefficient (Werner, 1954)), which averages 5.6, and the Leifman-Vassoevich coefficient (Vassoevich, Leifman, 1979), which averages 0.54. These indicators suggest the formation of peat beds from plants of terrestrial origin under the influence of marine conditions.

## Results and their discussion

On the correlation matrix of the data of ash content ( $A^c$ ), chemical composition of ash and spectral analysis, geochemical associations (Yudovich, 1978) of rare and trace elements Yb-Y-Be (at the level of correlation coefficients  $r = 0.79$  and higher) (Fig. 3) and Ti-Zr-Sc-V-Cr (at the level of  $r = 0.48$  and higher) (Fig. 4) were identified. The remaining elements do not form stable associations.



**Fig. 3.** Correlation profiles of the elements of the Yb-Y-Be association



**Fig. 4.** Correlation profiles of the elements of the Ti-Zr-Sc-V-Cr association

Elements of the geochemical association Yb-Y-Be have almost zero correlations with ash content and silica, insignificant relationships or their absence with other ash-forming components. At the same time, positive correlations are observed with humic acids ( $r$  up to 0.23), parenchyme ( $r$  up to 0.37), parenchyme-atrite ( $r$  up to 0.34), felinite ( $r$  up to 0.15), felinite-atrite ( $r$  up to 0.26) and microcomponents of the liptinite group ( $r$  up to 0.26). This means that these elements have significant coalphilic

properties. Migrating in solutions in the form of elemental cations, complex carbonates and fluorine anions (Kraynov, Shvets, 1980; Yudovich, Ketris, 2002), they are sorbed by the organic matter (OM) of coal or form organometallic compounds with it (Manskaya, Drozdova, 1964; Mitskevych et al., 1983; Yudovich, Ketris, 2002). In addition, they can be impurity elements in minerals of terrigenous origin: rock-forming – amphiboles (hornblende), feldspars, biotite, accessory – garnet, zircon (Lazarenko, Vinar, 1975; Shcherban, 1986). These elements came to the peat bed with groundwater and surface water in the form of suspensions, the solid phase of which was formed as a result of erosion of crystalline rocks and weathering crust of acidic composition. Their presence within the same association can be explained by the common source of their input and the common mechanism of their transfer from the source to the peat bed.

Elements included in the geochemical association Ti-Zr-Sc-V-Cr have high correlations with silica, quite high relationships with ash content, high inverse relationships with calcium, magnesium and sulfur oxides. These elements are associated with terrigenous material of basic (Ti, V, Cr) and acidic (Zr, Sc) composition accumulated during the peat stage. The presence in the coal of rock-forming and accessory minerals-carriers of elements of this association (pyroxenes, rutile, ilmenite, zircon) and minerals in which they are found as impurities (amphiboles, feldspars, biotite, garnet, magnetite, tourmaline) (Lazarenko, Vinar, 1975; Shcherban, 1986) confirms this conclusion.

Positive correlations of these elements with bitumen (Ti, V,  $r = 0.27$ ; Zr,  $r = 0.18$ ; Sc,  $r = 0.44$ ; Cr,  $r = 0.15$ ), dopplerinite (Zr,  $r = 0.17$ ; Ti,  $r = 0.11$ ), parenchyme (Zr,  $r = 0.31$ ; Ti, Sc,  $r = 0.11$ ), semifusinite (Ti,  $r = 0.21$ ) and lignitite (Sc,  $r = 0.52$ ), indicating the possibility of their presence in the coal in a non-mineral sorbed form or in complexes with OM (Manskaya, Drozdova, 1964; Yudovich, Ketris, 2002). They entered the peat bed in the form of elemental anions, oxyanions, carbonate and hydroxocomplex anions (Kraynov, Shvets, 1980; Yudovich, Ketris, 2002). These elements are probably related by the synchronicity of their accumulation.

The elements Pb, Ga, Cu, which form an unstable association (Fig. 5), have positive correlations with ash content, silicon and aluminum oxides, negative correlations with calcium and magnesium oxides, sulfuric anhydride and sulfur. They could have come as impurity elements with ter-

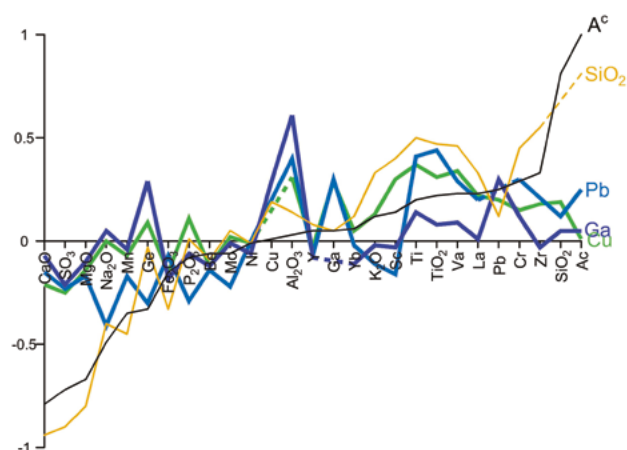


Fig. 5. Correlation profiles of Pb, Ga, Cu

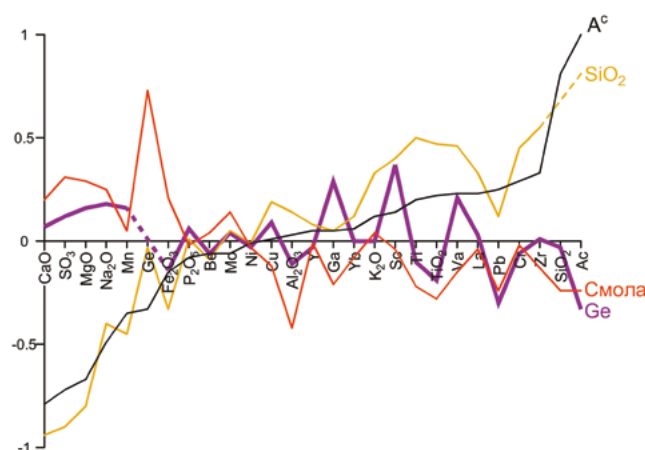


Fig. 6. Correlation profile of Ge

igenous rocks from acidic to basic composition. These elements can be associated with aluminosilicates (kaolinite, muscovite, biotite) and oxides (magnetite). The positive correlation of these elements with Ti ( $r$  up to 0.41) suggests the presence of their impurities in accessory minerals of titanium (ilmenite). In solution, these elements could have entered the peat bed mainly in the form of elemental cations and hydroxocomplex anions (Kraynov, Shvets, 1980; Yudovich, Ketris, 2002). Their positive correlation with felinite (Pb, Ga,  $r$  up to 0.18), sclerotinite (Pb,  $r = 0.26$ ) and bituminite-desmite (Cu,  $r = 0.20$ ) indicates the possibility of their presence in the coal OM in the sorbed form and in the form of organomineral complexes, especially complex compounds of the chelate type (Manskaya, Drozdova, 1964; Yudovich, Ketris, 2002).

Ge has a significant negative dependence on ash content and small and negative correlations with ash-forming components. At the same time, there is a high positive correlation ( $r = 0.73$ ) between Ge and the resin content of the benzene extract (Fig. 6) and a significant positive dependence on the content of lignitite ( $r = 0.19$ ), xylinitite ( $r = 0.24$ ), felinite-atrite ( $r = 0.34$ ) and sclerotinite ( $r = 0.33$ ). This certainly confirms the predominant association of Ge with the OM of brown coal, where it can be found in a non-mineral sorbed form, in the form of simple and complex humates of the chelate type, as well as Ge-organic compounds (Yudovich, Ketris, 2004). These elements could be delivered to the peat bed with the solution mainly in the form of oxyanions (Kraynov, Shvets, 1980; Yudovich, Ketris, 2002;). The high correlation of

Ge with the resin content of the benzene extract allows to obtain germanium in the technological process of separating bitumen from coal.

Mo is characterized by the absence of dependence on ash content and ash-forming components and a positive relationship with potassium oxide (Fig. 7), which indicates the possibility of its occurrence as an impurity in potassium feldspars, muscovite, biotite. Mo could enter the peat bed in the form of elemental anions and oxyanions (Kraynov, Shvets, 1980; Yudovich, Ketris, 2002). In coal, where a positive relationship of molybdenum with lignitite, lignitite-atrite and parenchymite ( $r$  up to 0.20) is recorded, it can be found in a physically sorbed non-mineral form and in the form of organic complexes containing oxycompounds in the ortho position (Manskaya, Drozdova, 1964; Yudovich, Ketris, 2002).

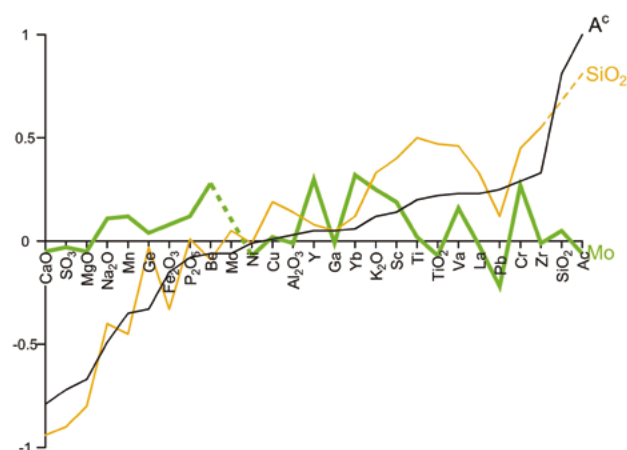


Fig. 7. Correlation profiles of Mo



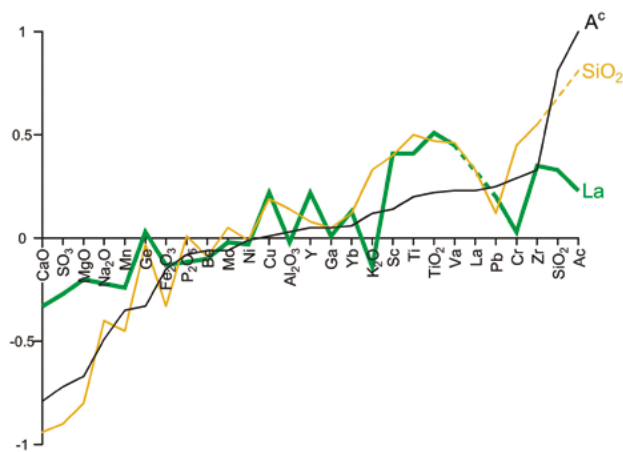


Fig. 8. Correlation profiles of La

La has a positive relationship with ash content and silica, and a negative relationship with other ash-forming components (Fig. 8). This indicates its input with terrigenous material of acidic composition. It can be present as an impurity in silicates (albite, muscovite) (Mitskevich, 1976; Shcherban, 1986). The high positive relationship of La with Ti ( $r = 0.41$ ) suggests the presence of an admixture of La in accessory titanium minerals (ilmenite, rutile). Lanthanum could enter the peat bed with the solution in the form of simple cations, complex carbonate and fluorine anions (Kraynov, Shvets, 1980; Yudovich, Ketris, 2002). The positive correlation with bitumen ( $r = 0.29$ ) and such microcomponents as xylinite and parenchymite ( $r$  up to 0.15) indicates the possibility of lanthanum occurrence in the OM of coal, both in the sorbed form and in the form of organic complexes.

Mn has negative correlations with silicates and ash content and positive correlations with calcium, magnesium and sulfur oxides (Fig. 9).

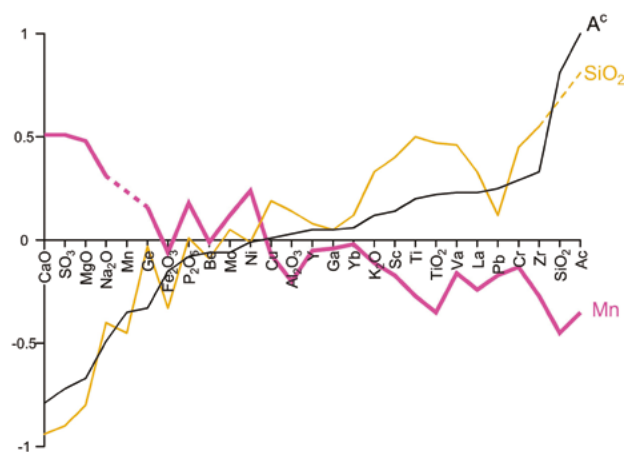


Fig. 9. Correlation profiles of Mn

This suggests its presence as an impurity in carbonates and iron sulfides. The Mn admixture is inherent to micas (biotite, muscovite), hydromicas (glauconite) and some accessory minerals (garnet, staurolite, ilmenite, magnetite) (Lazarenko, Vinar, 1975). Migration of Mn, which is released during weathering of crystalline rocks, is possible not only in mineral form, but also in solutions in the form of elemental cations and hydroxocomplex anions (Kraynov, Shvets, 1980; Yudovich, Ketris, 2002). Mn is slightly positively correlated with humic acids ( $r = 0.15$ ), phyllinite and sclerotinite ( $r = 0.12$ – $0.13$ ). According to (Manskaya, Drozdova, 1964), manganese may be present in coal in the form of organometallic complexes.

Ni does not correlate with ash content and ash-forming components (Fig. 10), except for  $K_2O$  oxide. It can be found in a non-mineral sorbed form in clay minerals, micas, and also as an impurity in some accessory minerals (staurolite, magnetite) (Lazarenko, Vinar, 1975). Nickel enters the peat bed in the form of elemental cations and hydroxocomplex anions (Kraynov, Shvets, 1980; Yudovich, Ketris, 2002). It is positively correlated with bituminite-desmite ( $r = 0.20$ ). It can probably be found in coal in sorbed form and in the form of humates (Yudovich, Ketris, 2002).

According to the authors, the main amount of elements in coal is accumulated at the stage of sedimentogenesis. It is during peat formation that OM is characterized by high reactivity, with the highest sorption and ion exchange properties. At this stage, optimal conditions for contact and interaction with chemical elements introduced into the peat bed were created.

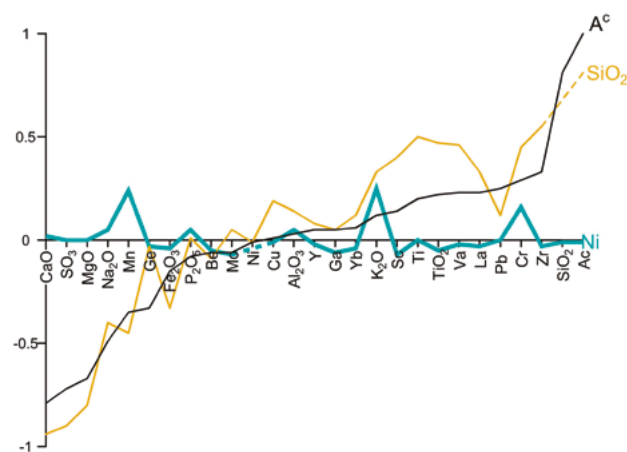


Fig. 10. Correlation profiles of Ni

As mentioned above, all the elements considered migrated in solutions in the form of simple ions, oxyanions and complex compounds whose ligands were hydroxide, carbonate or fluoride ions.

However, it should be noted that natural waters contain a large number of natural organic compounds (humic and fulvic acids). Due to their chemical structure and the presence of functional groups, they can form complexes with migrating metals, which vary in strength and solubility and are capable of moving over long distances (Manskaya, Drozdova, 1964; Kravynov, Shvets, 1980). They, like the above-mentioned forms of migratory elements, can be introduced into the peat bed and absorbed by the peat OM.

## Conclusions

It was found that the source of rare and trace elements in brown coal is the crystalline rocks of the USh and their weathering crust. They entered the peat bed with groundwater and surface water in the form of suspensions, the solid phase of which was formed as a result of erosion of crystalline rocks and weathering crust. The considered elements also migrated in solution in the form of simple ions, oxyanions and complex compounds whose ligands were hydroxide, carbonate or fluorine ions. Their migration is also possible in the form of soluble organic complexes.

The considered elements are found in coal as impurities or main components in minerals dispersed in the coal substance, in the form of finely dispersed inclusions sorbed on the coal surface, and in the form of organometallic compounds.

For the first time, an analysis of the correlations of the considered elements with the microcomponents of the coal OM of the lignite, huminite-helite, fusinite and liptinite groups is presented.

The most coalphilic elements are Be, Yb, Y, Ge, Mo, Sc. Elements such as Ti, Zr, V, Cr, Pb, Ga, Cu, La, Mn, Ni are more often associated with terrigenous material, which does not exclude their presence in coal in sorption and organically bound form.

The high correlation of Ge with the resin content of the benzene extract allows to obtain Ge in the technological process of separation of bitumen from coal.

*The work was carried out at the Institute of Geological Sciences of the National Academy of Sciences of Ukraine within the framework of the research on the topic "Coal-Bearing and Shale-Bearing Formations of Ukraine and Related Mineral Resources of Strategic Importance (code: III-3-24)" for 2024-2027 (code for program classification of expenditures and lending to local budgets 6541030).*

Вивчено геохімічні особливості вугілля Верхньодніпровського родовища Дніпровського буровугільного басейну, що знаходиться в межах Придніпровського мегаблоку Українського щита. Визначено положення родовища в межах щита за металогенічним, геохімічним та ландшафтно-геохімічним районуванням. Показано, що джерелом надходження у вугілля рідкісних та розсіяних елементів є кристалічні породи Українського щита. З допомогою кореляційного аналізу проведено оцінку асоційованості розглянутих елементів. Виділено дві стійкі асоціації: Yb-Y-Be та Ti-Zr-Sc-V-Cr. Виявлено шляхи міграції елементів у торфовище та можливі форми фіксації елементів в органічній речовині вугілля. Вперше представлено аналіз кореляційних зв'язків розглянутих елементів з мікрокомпонентами органічної речовини вугілля груп лігнітиту, гумініто-геліту, фюзиніту та ліптиніту. Найбільш вуглефільними є елементи Be, Yb, Y, Ge, Mo, Sc. Такі елементи, як Ti, Zr, V, Cr, Pb, Ga, Cu, La, Mn, Ni, частіше пов'язані з теригенним матеріалом, що не виключає їх присутності у вугіллі в сорбційній та органічно пов'язаній формах. Виявлено високу кореляційну залежність Ge від вмісту смоли бензольного екстракту, що дозволяє отримати германій у технологічному процесі виділення з вугілля бітуму.

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