IVAN KRAUS — TIBOR ĎURKOVIČ*

# POSSIBLE DETERMINATION OF SOURCE AREAS ON THE GROUNDS OF TRACE ELEMENTS IN CLAY SEDIMENTS

*(Figs. 1—8)*

Abstract. Examined are clay sediments of the East Slovakian basin for the frequency of B, Ga, Pb, Zr, V, Ni, Cr, Ba, Sr. Most attention is paid to differentiation of two areas most significant for the East-Slovakian basin — the nonvolcanogenic (particularly the Flysch Belt) and the volcanogenic provenances. Vanadium, nickel and chromium are the most important geochemical indicators.

Резюме: В предлагаемой статье авторы рассматривают содержание B, Ca, Pb, Zr, V, Ni, Cr, Ba, Sr в глинистых седиментах восточнословацкого бассейна. Главное внимание авторы обратили на решение проблемы влияния двух главнейших областей сноса, которые находятся в восточнословацком бассейне — неовулканического и вулканогенного происхождения. По их предложению особенно важными геохимическими индикаторами тут являются V, Ni и Cr.

## *Introduction*

The authors deal with the possibilities of determining source areas in sedimentary basins of the West Carpathians on the grounds of the results of mineralogical-geochemical examination of clays. Most attention is paid to the clays of the East-Slovakian basin (Fig. 1), regarded as the western part of the Transcarpathian Inner Depression by R. Rudinec — J. Slávik (1970). The basin formed during the Eggerburgian — Pliocene. As for lithology, it has volcano-sedimentary character. The regional distribution of products of the volcanic activities is traceable in the Badenian, Sarmatian, and Pliocene. Among clastic sediments, clays of various petrographic types are in freshwater, brackish and hypersaline facies mostly in the central parts of the basin. Sands and gravels concentrate mainly in marginal facies. Coal sedimentation proceeded in the Upper Sarmatian and in the Pliocene. It was most extensive in the Podvihorlatská kotlina depression.

Two methods were applied for studying the influence of source areas upon the genesis of clay sediments. The first consists in detail mineralogical examination. It was applied by I. Kraus — E. Šamajová (1973) for the Neogene basins in the West Carpathians. The authors based their investigations upon the publications dealing with the saturation of montmorillonite by $K^{1+}$ facilitating differentiation of volcanogenic and nonvolcanogenic source areas (E. C. Weawer 1958, G. I. Teodorovič et al. 1965, G. V. Karpova — E. P. Sebjakova 1968). In clays redeposited from nonvolcanogenic source areas is the so-called "degraded" montmorillonite formed by alteration of micas, mainly muscovite. After saturation with KOH its basal reflex 13,0—15,0 Å is reduced to 10 Å. The change is stable, the basal reflex will not increase to 17,5 Å neither after repeated saturation with glycerine. In clays redeposited from volcanogenic source areas montmorillonite formed mostly by alteration of volcanic

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glass. After saturation with KOH its basal reflex remains practically unchanged. In contrast to the former, glycerine causes increase to 17,5 Å. Mineralogical examination of clays with unambiguously identifiable source area according to the texture of parent rocks preserved, the existence of montmorillonite of volcanogenic and nonvolcanogenic origin was testified in more samples from the Neogene basins of the West Carpathians.

The other method is based upon the study of distribution of trace elements in clays. controlled by the composition of source areas. Compared was the distribution of some trace elements in shales of a probable source area (the Flysch Belt in East Slovakia) and their distribution in the Neogene clays redeposited into the East-Slovakian basin from the Flysch Belt (Fig. 1).

On the grounds of mineralogical data we may regard parent rocks from two source areas as most significant for the East-Slovakian basin. The first source area is represented by the rocks of volcanogenic origin. intensively damaged by hypergenic processes. Such clays are mostly composed of minerals from the group of montmorillonite or kaolinite. Characteristic is the presence of cristobalite, while quartz is either subsidiary or missing in the fraction 0.2 micrometers. The second source area comprises rocks of nonvolcanogenic origin with clastic sediments predominant. Redeposited clays of this provenance are polymineral in compositon, containing illite, kaolinite, eventually admixture of montmorillonite, chlorite and minerals of mixed I-M structures. They are characterized by increased content of quartz.

A comparison with the clays of the Pozdišovce formation (with a decisive provenance influence of the Flysch Belt), and mineralogical analysis of shales of the Magura and Dukla units in East Slovakia, resulted in a supposition that for the substantial portion of clay sediments of the East-Slovakian basin, among nonvolcanic source areas most important would be the Flysch Belt. Carbonate rocks, particularly of the Humenné Mesozoic are to be considered as well, only so far there are no reliable criteria for differentiating the influence of the Mesozoic source areas mostly with carbonates, and that of the Paleogene source areas of the Flysch Belt and the Paleogene of Central Carpathians, in the clay sediments. The extend and influence of the source areas with prevalence of metamorphites and granitoid intrusives, either from the west (the Čierna Hora, Branisko, Spiškogemerské rudohorie mountains, the Zemplín horst) or from the east (the Užhorod horst) are still insufficiently investigated and the problems should be paid more attention in the future.

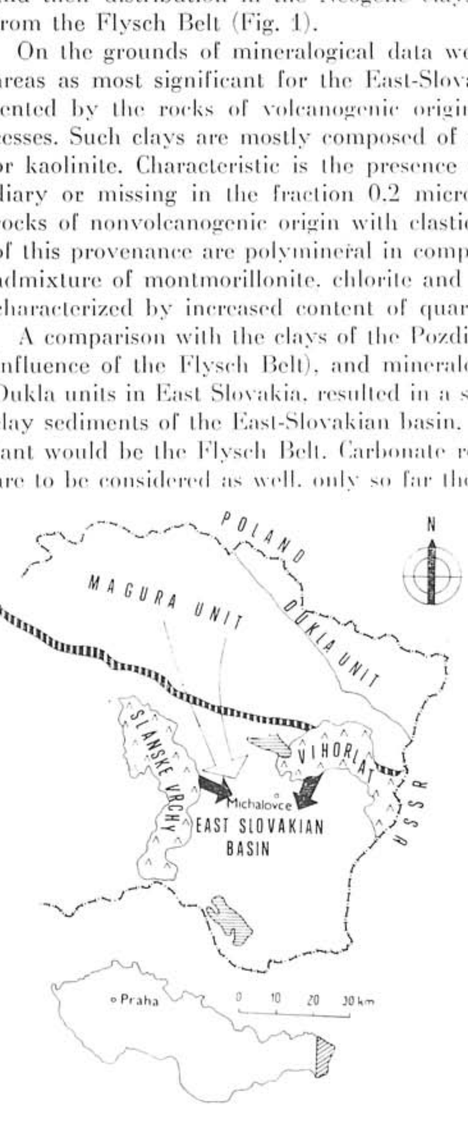


Fig. 1. Scheme of geologic structure of the area studied. 1. Klippen Belt, 2. Pre-Tertiary formations, 3. Neovolcanic rocks, 4. Generalized direction of supply from neovolcanic areas, 5. Generalized direction of supply from nonvolcanic areas.

Confronted are data on the frequency of trace elements in the shales of the Dukla and Magura units of the East-Slovakian Flysch Belt (T. Ďurkovič 1969, 1973) and information on the distribution of trace elements redeposited into the East-Slovakian basin from the nonvolcanogenic and volcanogenic source areas (I. Kraus 1973).

### *Boron*

The distribution of boron in clays is controlled by facies, by mineralogic composition, by geotectonic history of the respective area, by diagenesis and petrographic character of sediments. Some authors (C. T. Walker 1963, I. A. Alexina 1972) regard this trace element as a sensitive indicator of even small alterations in salinity of water in marine basins. Others emphasize the role of the rocks in the source areas in controlling the content of boron in sediments (D. A. Spears 1965, R. D. Cody 1971). Distinctly negative is the standpoint of G. D. Nicholls — D. H. Loring (1962). In H. Harder's (1970) opinion, boron in clay sediments cannot positively indicate slight alterations in the salinity of water, caused by a sudden transgression or by brackish environment. Concentrations of boron in clays may only illustrate the mean value of salinity during the sedimentation process.

The content of boron in brackish and marine clays of the East-Slovakian basin was not markedly affected by its distribution in the rocks of source areas. It is testified by a good accordance between the mean content of boron in the shales of the Magura and Dukla units, and in clays redeposited from this source area into the East-Slovakian basin. The mean content of boron in shales of the Magura and Dukla units is 123 ppm, in the clays redeposited into the East-Slovakian basin from nonvolcanic source areas 122 ppm (Tab. 1). In the East-Slovakian basin compared may be the contents of boron in clays redeposited from the Flysch Belt, only of a different facies. The results give evidence of a marked differentiation. The mean

Tabelle 1. Statistical data for trace elements of studied rocks

| | Dukla Unit shales | | Magura unit shales | | Clays in East-Slovakian Basin redeposited from nonvolcanic source areas | | Clays in East-Slovakian Basin redeposited from nonvolcanic source areas | |
|---|---|---|---|---|---|---|---|---|
| | N | X | N | X | N | X | N | X |
| B | 79 | 123 | 28 | 123 | 46 | 122 | 44 | 46 |
| Ga | 104 | 13 | 34 | 11 | 46 | 12 | 44 | 19 |
| Pb | 28 | 51 | 34 | 29 | 46 | 13 | 44 | 17 |
| Zr | 104 | 239 | 34 | 283 | 46 | 138 | 44 | 164 |
| V | 104 | 119 | 34 | 101 | 46 | 195 | 44 | 93 |
| Ni | 104 | 61 | 34 | 53 | 46 | 54 | 44 | 29 |
| Cr | 104 | 137 | 34 | 133 | 46 | 128 | 44 | 61 |
| Ba | 104 | 272 | 34 | 213 | 46 | 273 | 44 | 246 |
| Sr | 104 | 133 | 34 | 121 | 46 | 190 | 44 | 113 |

N — number of specimens, X — arithmetic mean content of boron in freshwater clays redeposited from nonvolcanic source areas into the East-Slovakian basin, is 57 ppm. In contrast to that, the mean content of brackish-marine clays redeposited from the Flysch Belt into the East-Slovakian basin, is 149 ppm. This is a very important evidence of the fact that the distribution of boron is controlled by alterations of facies and not by the rocks in the source areas.

An approximately linear correlation has been found between the contents of boron and gallium in the shales of the Magura and Dukla units, i. e. in the shales of the Flysch Belt source area (Fig. 2). But the clays of the East-Slovakian basin derived from a nonvolcanogenic source area, do not show any linear trend in the diagram. So far it is not possible to explain satisfactorily the post-resedimentation change of the linear correlation B/Ga.

### *Gallium*

Data on the distribution of gallium in the clays of the East-Slovakian basin are an important contribution to information on their facies history and on the role of the source areas, in spite of some authors regarding the relation between gallium and some other specific clay mineral as unevidenced. Frequent are opinions about gallium not recording the change in facies evolution of sediments. R. J. W. Mc Laughlin (1959) presents a very low isomorphic substitution of $Ga^{3+}$ for $Al^{3+}$ in the structure of kaolinite. G. D. Nicholls — D. H. Loring (1962) stated preferred concentra-

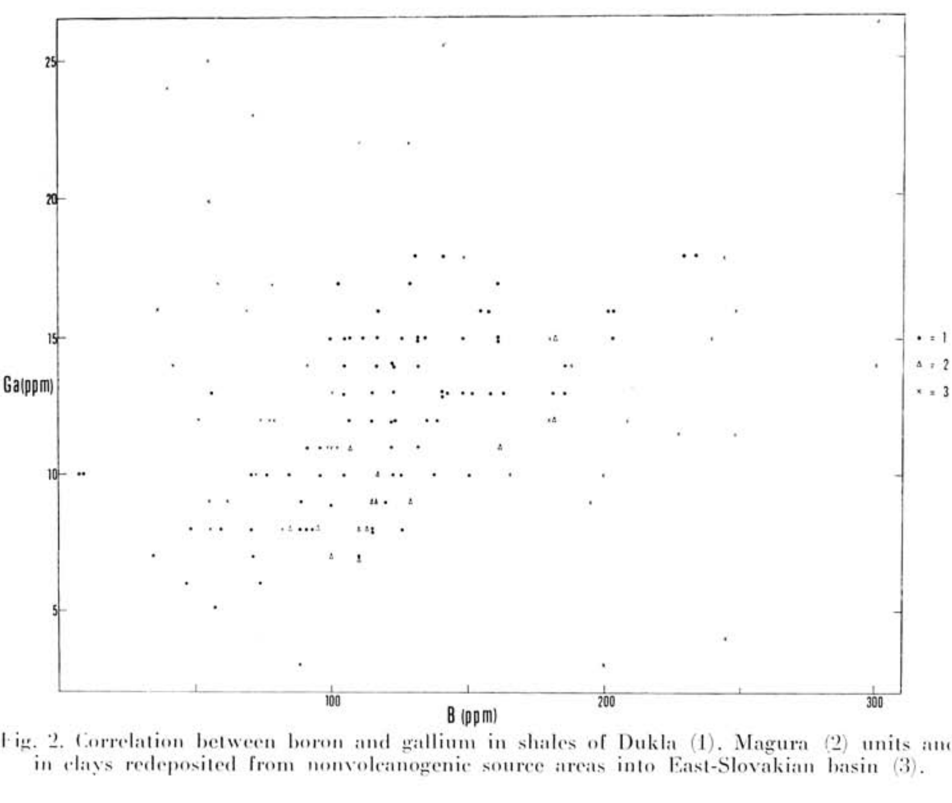


Fig. 2. Correlation between boron and gallium in shales of Dukla (1), Magura (2) units and in clays redeposited from nonvolcanogenic source areas into East-Slovakian basin (3).

tions of gallium in illite in the Carboniferous sediments of Great Britain. D. M. Hirst (1962) presumes that gallium participates in the structure of the three most frequent clay minerals-illite, kaolinite, montmorillonite. He regards the concentrations of gallium in kaolinite as most probable. Interestingly, the authors determining the gallium content in non-separated clays, most frequently did not prove its differentiation in dependence upon facies.

In the Neogene basins of the West Carpathians the highest gallium content is restricted to clays of the kaolinitic type (29—33 ppm), and them medium values are concerned with montmorillonite (18 ppm). This is why we were interested in the gallium content in illitic clays. It may be comprised into the discussion about the

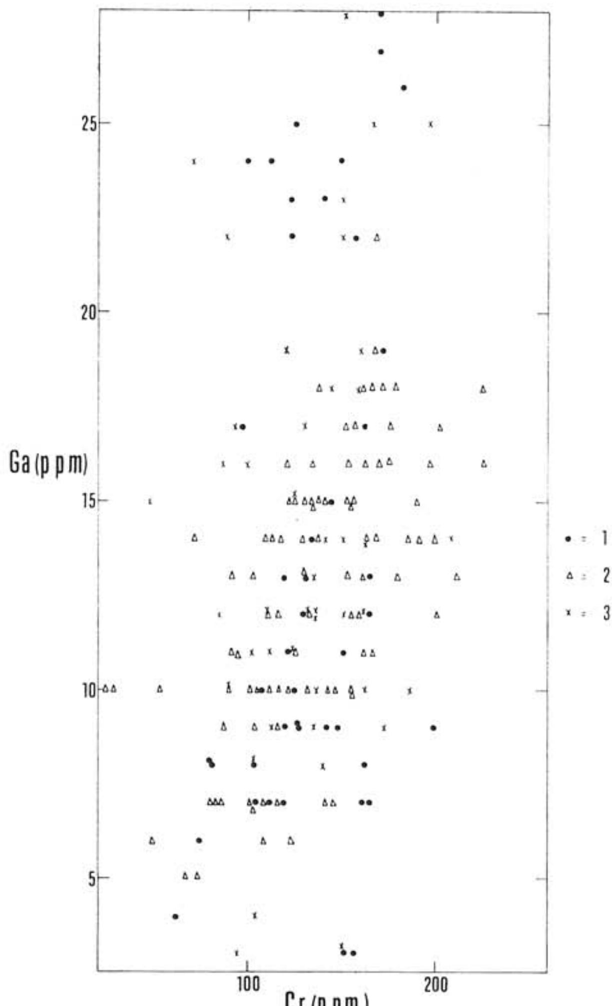


Fig. 3. Correlation between gallium and chromium in shales of Dukla (1), Magura (2) units and in clays redeposited from nonvolcanogenic source areas into East-Slovakian basin (3).

structure of the clay mineral preferred by gallium. The results of I. Kraus (1973) are in accordance with the opinion about isomorphic substitution of gallium for $Al^{3+}$ of the source area as well as in the clays of nonvolcanic origin from the East-Slovakian gallium. It is illustrated by the illitic clays of the East-Slovakian basin redeposited from the Flysch Belt, and by the shales with illite predominant in the sediments of the Flysch Belt (Tab. 1). Consequently, in the East-Slovakian basin, the differentiation of gallium contents in clays must be controlled by the respective source areas. The differentiation is a function — first of all — of mineralogical composition. The clays redeposited from nonvolcanic source areas and the shales of the Flysch Belt are composed mostly of illite, with low average gallium content. The clays redeposited from volcanogenic source areas consist of kaolinite or montmorillonite and have a markedly higher average gallium content. Information based upon mineralogic composition of clay sediments facilitate distinguishing clays as for their origin from different source areas.

An inconspicuous linear correlation is between gallium and chromium in the shales of the source area as well as in the clays of nonvolcanic origin from the East-Slovakian basin (Fig. 3).

## *Lead*

The position of lead in the hypergenic stage is known just partially, and few data are available about its distribution in sedimentary rocks. V. T. Pogrebnoj et al. (1971), M. V. Kobelev — S. V. Nečaev (1971) found an increased lead content in the weathering crust on granitic rocks and a decreased content on the ultramafic rocks. The mean lead contents in the shales of the Magura and Dukla unit are markedly variable. Considerably different are lead contents in the clays of the East-Slovakian basin, redeposited from nonvolcanic source areas, and particularly from the Flysch Belt. G. D. Nicholls (1967) refers lead to trace elements concentrating preferably in a deep-sea environment. I. Kraus (1973) means that extremely high concentrations of lead in clays appear in connection with kaolinite, owing to its high concentrations in acid intrusive rocks.

## *Zirconium*

It is equally distributed in the shales of the Flysch Belt and in the clays of the East-Slovakian basin redeposited from nonvolcanic source areas (Fig. 4). Most likely it is due to zirconium transported in clay minerals in a form of detritus. It is also evidenced by the highest zirconium concentrations in sands, among sediments. The distribution of zirconium is controlled by the nature of the parent rocks in the source areas more than that of other trace elements. The mean zirconium content in clays — 150 ppm (A. P. Vinogradov 1957) — is identic with its content in intrusive rocks. In the clays of the East-Slovakian basin the zirconium content varies around this mean value. In shales of the Magura and Dukla unit the content is much higher (Tab. 1).

## *Vanadium*

At the weathering of intrusive and metamorphosed rocks, most vanadium concentrates in clay minerals, Fe hydroxids, and in organic matter. D. M. Hirst (1962) stated preferred concentration of vanadium in the structure of montmorillonite. G. D. Nicholls — D. M. Loring (1962) found vanadium in the structure of kaolinite and illite. K. B. Krauskopf (1956) basing upon his experiments, regards adsorption of vanaidum by means of clay minerals as little effective. In contrast to that, H. A. Tourtelot (1964) regards the sorption of vanadium as the most important form of its bond. In Z. A. Janočkina's (1966) opinion, for vanadium most important is migration in the form of finely dispersed suspension.

The distribution of vanadium in the clays of the Neogene basins with respect to different source areas is important for more reasons. First of all there is an extremely differentiated content of vanadium in the clays of the East-Slovakian basin. In the future it might serve a criterium in distinguishing sediments redeposited from the Flysch Belt and from the East-Slovakian neovolcanites. I. Kraus (l. c.) determined the mean content of vanadium 195 ppm in the clays of the East-Slovakian basin redeposited from nonvolcanogenic source areas, while in the clays redeposited from the East-Slovakian neovolcanites — only 93 ppm. Different values of the mean content of vanadium are also reflected in character of frequency of this trace element (Fig. 5). In histograms for vanadium of clays redeposited from the East-Slovakian neovolcanites, are two maxima: one around 60 ppm and the second around 140—200 ppm. In histogram for vanadium of clays redeposited from nonvolcanogenic source areas, is only one maximum around 180 ppm.

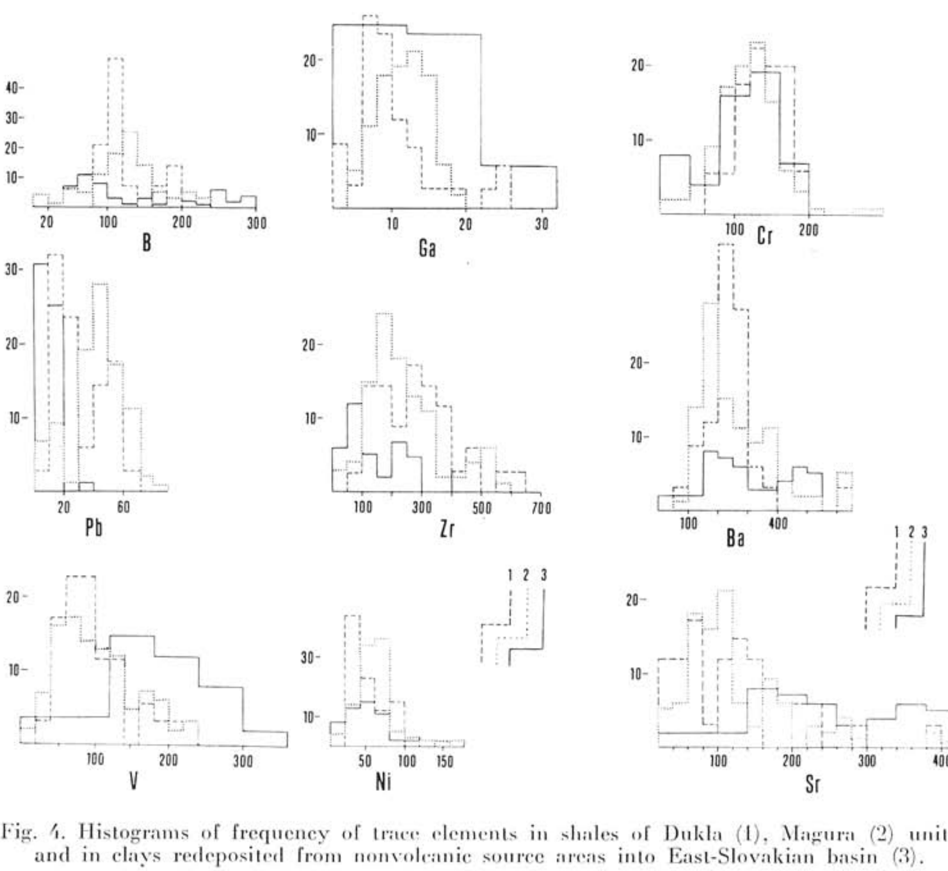


Fig. 4. Histograms of frequency of trace elements in shales of Dukla (1), Magura (2) units and in clays redeposited from nonvolcanic source areas into East-Slovakian basin (3).

Interestingly, the mean content of vanadium is higher in the clays of the East-Slovakian basin, redeposited from nonvolcanic source areas than in shales of the Magura and Dukla units (Tab. 1). The cause of the phenomenon cannot be explained so far. It is known that besides other factors, the concentration of vanadium is also affected by the content of C org. (H. A. Tourtelot (1964).

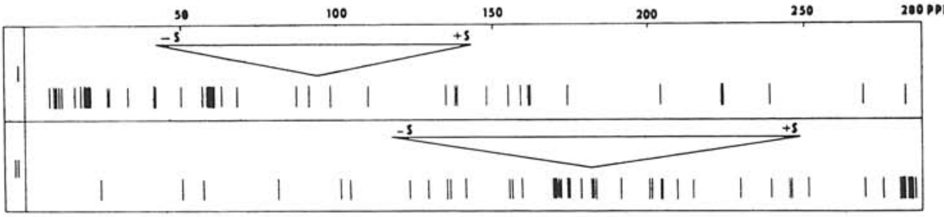


Fig. 5. Distribution of vanadium in clays redeposited from different source areas. 1. Clays redeposited from neovolcanites into East-Slovakian basin. 2. Clays redeposited from nonvolcanogenic source areas into East-Slovakian basin. S-standard deviation.

## *Nickel*

Among sedimentary rocks clays show highest concentrations of nickel. Its mean content — 25 ppm (A. P. Vinogradov 1957) is somewhat lower than in clay sediments of the Neogene basins in the West Carpathians — 32 ppm. In the shales of the Magura and Dukla units in East Slovakia, the mean content of nickel is considerably higher (Tab. 1).

Alterations in the distribution of nickel in the clays of the Neogene basins in the West Carpathians depend upon the respective source areas. The mean content of nickel in the clay of nonvolcanogenic origin is markedly higher than in the clays of volcanogenic provenance. In the East-Slovakian basin nickel is an indicator facilitating distinguishing the clays redeposited from the East-Slovakian neovolcanites (29 ppm) and from nonvolcanic source areas (54 ppm). From this standpoint the distribution of nickel in the clays of the Inner Depressions (Fig. 6) shows highest concentrations with vanadium and chromium in the periods of the weakest volcanic activity — in the Eggenburgian, Karpatian and Pliocene. The concentrations were lowest during the culminating volcanic activity — in the Badenian, Sarmatian, Lower Pannonian. This is an evidence of a generally decreased content of V, Ni, Cr in the clays formed during the Neogene by decomposition of the products of susequent volcanism.

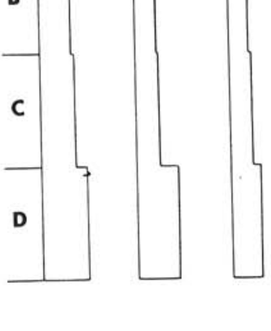


Fig. 6. Mean contents of vanadium, nickel and chromium in clays of Inner Depressions in Central Slovakia in separate stratigraphic stages. A-Pliocene, B-Upper Sarmatian — Lower Pannonian, C-Upper Badenian — Lower Sarmatian, D-Eggenburgian — Karpatian.

*Chromium*

There is a good accordance among the mean contents of chromium in the clays originating from source areas composed of rocks of the same type. In fact, there is only a slight difference in the contents of chromium in clays redeposited into the Neogene basins of the West Carpathians from the Central-Slovakian (57 ppm) and East-Slovakian (61 ppm) neovolcanites. Analogous situation is in the Neogene clays redeposited from the nonvolcanogenic source area into the East-Slovakian basin (128 ppm) —if compared with the shales of the source area proper, i. e. of the Magura and Dukla units (137 and 133 ppm). The data testify to the fact that the distribution of chromium in the clays of the Neogene basins in the West Carpathians is controlled first of all by the nature of source rocks and by the course of the weathering process in original source areas.

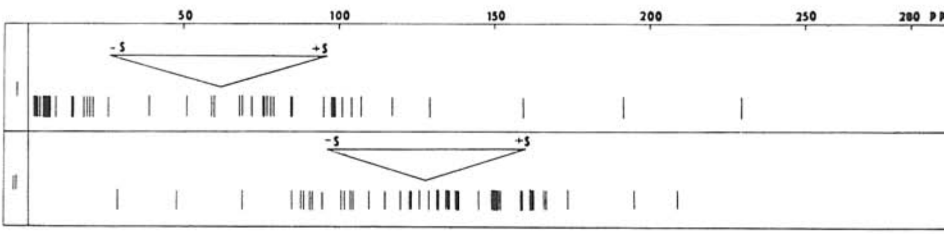


Fig. 7. Distribution of chromium in clays redeposited from different source areas. 1. Clays redeposited form neovolcanites into East-Slovakian basin. 2. Clays redeposited from nonvolcanogenic source areas into East-Slovakian basin. S — standard deviation.

Character of the distribution of chromium (Fig. 7) shows equal regularity in clays redeposited from the East-Slovakian and Central-Slovakian neovolcanites, and a marked difference in the frequency in clays redeposited from nonvolcanogenic source area. Most samples of clays redeposited from volcanogenic source areas show values below 50 ppm in contrast to most samples of clays redeposited from nonvolcanogenic source areas showing the values within 90—165 ppm. This is an immediate cause — as it was stated in vanadium and nickel — of considerably higher contents of the trace elements and of a different nature of distribution in the clays of nonvolcanogenic provenance than in the clays of volcanogenic origin. Different nature of distribution indicates different forms of the bond of chromium in parent rocks from the above source areas.

A. P. Vinogradov (1962) quotes the mean content of chromium in granites 25 ppm, in clays 100 ppm, in basalts and gabbro 200 ppm. In the Neogene basins of the West Carpathians, I. Kraus (l. c.) proved chromium as a sensitive indicator of the general nature of parent rocks participating in forming clay minerals in the original weathering crusts. Consequently, the rocks of probably intermediary and basic types were most significant among source rocks, decomposition of which gave rise to flysch shales. This opinion is rather meant as an illustration of the extent and possibilities of geochemical research in sedimentary rocks. It is, however, a fact that by means of the association of V — Ni — Cr the influences of volcanogenic and nonvolcanogenic source areas may be distinguished in the clay sediments of the East-Slovakian basin.

An approximately linear correlation is among V/Ni + Cr in the shales of the Flysch Belt in East-Slovakia (Fig. 8) and in the clays of the East-Slovakian basin, redeposited from volcanogenic and nonvolcanogenic source areas (Fig. 9).

### *Barium*

For the study of the rules of geochemistry of barium in hypergenic stage very important are its isomorphism with K, high concentration in acid intrusive rocks, and its ability of entering colloid systems of negative charge. G. D. Nicholls — D. H. Loring (1962) emphasize that the bond of barium in clay sediments is facilitated by sorption on the surface of the individual particles of clay minerals. Owing to close ionic diameters between Ba and K, more frequent is association of barium with illite.

The mean contents of barium in clays from individual source areas show no distinct connection the distribution of barium in clay sediments of the Neogene basins in the West Carpathians and in the Flysch Belt of East Slovakia is even and has extremely broad dispersion (Fig. 4). Most trace elements in clays of volcanogenic origin concentrate in a short interval characteristic particularly of B, Ga, Pb, Ni, Cr. In barium is a reversed tendency. Frequency of samples in the interval of 40—340 ppm is very regular, without any pronounced maximum.

### *Strontium*

In sediments of different genesis and lithologic composition most attention is paid to mutual relations between Ca/Sr. More authors (S. K. El Wakeel — J. P. Riley 1961, K. K. Turekian — J. L. Kulp 1956) regard carbonates as the main bearer of strontium in recent marine sediments. On the other hand, D. M. Hirst (1962), G. D. Nicholls — D. H. Loring (1962), basing upon the ratio Sr/Ca, suppose that strontium may be restricted only to clay minerals in some cases.

In the area studied, the contents of strontium and barium are broadly variable. The mean contents of strontium in shales of the Magura and Dukla units are very close to

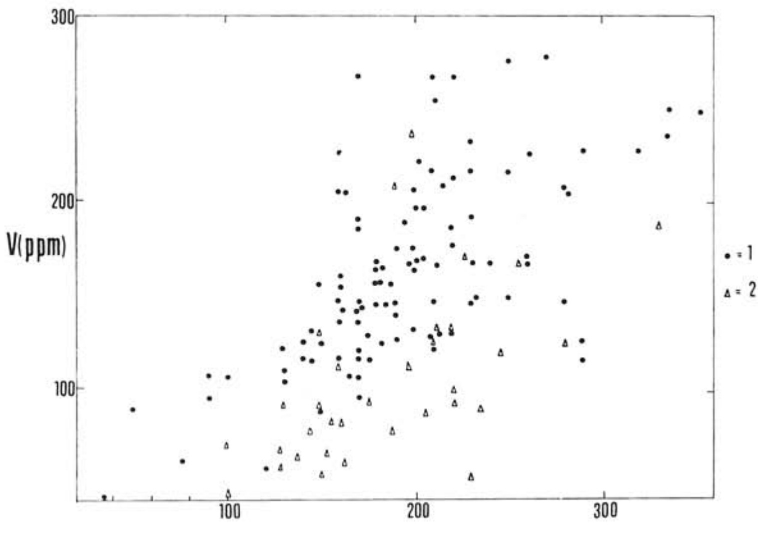


Fig. 8. Correlation between V/Ni+Cr in shales of Dukla (1) and Magura (2) units.

each other (Tab. 1). A good accordance is between the mean content and the character of distribution of strontium in clays redeposited from the Central-Slovakian and East-Slovakian neovolcanites. Most likely the increased content of strontium in clays redeposited into the East-Slovakian basin from nonvolcanic source areas will depend upon the increased $CaCO_3$ content in the rocks of the source area, besides other factors. T. Ďurkovič (1973) got evidence on the dependance correlating Sr/CaO contents particularly in the Mafeov beds of the Magura unit.

## *Conclusions*

The purpose of this article is to inform about the frequency of B, Ga, Pb, Zr, V, Ni, Cr, Ba, Sr, examined by spectrochemical quantitative analysis in clay sediments of the East-Slovakian basin and and in shales of the Flysch Belt in East Slovakia. The authors concentrated to studying the distribution of the trace elements with respect to the influence of the source areas, to finding trace elements able of indicating the

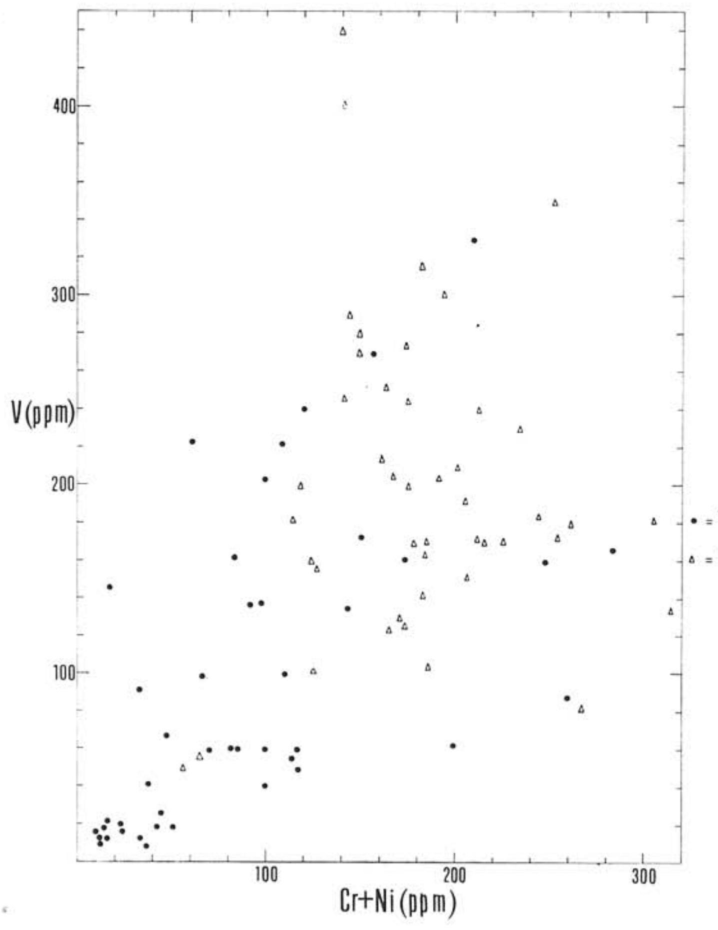


Fig. 9. Correlation between V/Ni+Cr in clays redeposited from neovolcanites (1) and from nonvolcanogenic source area (2) into East-Slovakian basin.

nature of original parent rocks in the sedimentation basin. From this view, the elements studied may be divided into two principal groups: 1. Trace elements indicating first of all changes in the nature of the environment of deposition and thus also the facies evolution of the sedimentation basins. The distribution of these trace elements is not decisively affected by the nature of rocks in the original source areas. Among the elements studied such are boron, gallium, and partially strontium. 2. Trace elements whose distribution in clay sediments is affected first of all by the nature of parent rocks in the original source areas, and the role of facies is unimportant for their distribution. Such are Pb, Zr, V, Ni and Cr among the elements studied.

Most attention was paid to differentiating the influence of two most important source areas in the East-Slovakian basin — the nonvolcanogenic and volcanogenic provenances. In this respect most significant geochemical indicators are V, Ni, Cr. The values of their mean contents are very close to each other in clays redeposited from related source areas like the Central-Slovakian and East-Slovakian neovolcanites. However, if their frequency in clays from source areas of a different composition is compared, then the mean content of these trace elements is at least twice higher and the character of distribution different in the clays of the nonvolcanogenic provenance from clays of the volcanogenic provenance. V, Ni and Cr are comparatively most favourable for distinguishing the influence of the volcanogenic and nonvolcanogenic source areas in clay sediments of the East-Slovakian basin.

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