

RELATIONS BETWEEN THE CLAY MINERAL AND ORGANIC MATTER CONTENTS IN THE SEDIMENTS OF THE SOUTH GREAT PLAIN, HUNGARY

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INTRODUCTION

Our investigations aimed to find relations between the clay mineral contents and the soluble and insoluble organic matter content of rocks since these are of great importance from the points of view of formation and migration of oil.

For this purpose the core samples of the boreholes drilled in the Hódmezővásárhely—Makó Trench, *i. e.* sandstones, aleurites and clay-marls were used. The strata sequence of the boreholes Makó-1, Makó-2 and Hód-I discovered the deep-lying internal sedimentary sequences of the Neogene basin in the South Great Plain. The samples derive partly from the Upper, partly from the Lower Pannonian, as well as partly from the Miocene.

DETERMINATION AND DISCUSSION OF THE CHANGE OF MINERAL COMPOSITION

The quantitative determination of clay minerals was carried out from the fractions of less than ten microns after removing the carbonates and organic matter. The procedure was carried out by means of chemical methods. The quantities of montmorillonite, micas, chlorite, kaolinite, amorphous material, quartz, K- and Na-feldspars were determined.

The method of determination was summarized by ALEXIADES and JACKSON [1966]. The method and calculations were modified according to the problems arisen. The determination of the mica-content is based on the measurement of K-content. The quantities of quartz and feldspars were calculated on the basis of the K- and Na-content of the rest remaining after the fusion by pyrosulphate. The amorphous material was determined on the basis of the SiO_2 and Al_2O_3 content remained after the solution in 0.5 N NaOH. The determination of kaolinite is based on the difference of SiO_2 and Al_2O_3 contents occurring between the solution in 0.5 N NaOH after heating treatment at 110° resp. 585°C. The chlorite content was computed from the water content decrease following 300 and 950°C taking into consideration also the water content of the other clay minerals, as well. The determination of vermiculite and montmorillonite is based on the measurement of ion exchange capacity.

The method does not throw light upon the fact that montmorillonite is a discrete phase or it is present in form of interstratified illite-montmorillonite. Based on the X-ray diffractometric investigations carried out by J. MEZŐSI, however, in these samples montmorillonite occurs only in interstratified structure.

In the boreholes the mica and chlorite contents predominate though in some samples considerable amount of montmorillonite could also be detected. The quantity

of kaolinite is small, it does not exceed 5 per cent. The quartz content varies around 20 per cent. The quantity of Na-feldspar is greater than that of the potash feldspar but remains below 10 per cent. Only the coarse detrital Miocene sediments represent exceptions in which the quantity of Na-feldspar amounts to more than 10 per cent.

The average quantities of each components were computed to the Upper, and Lower Pannonian as well as to the Miocene samples.

The Miocene samples were divided into two groups, *i. e.* the pelitic sediments between 4176 and 4565 m and the coarse-grained sediments between 4565 and 4803 m, respectively. Concerning the two types of Miocene samples, the average quantities of minerals were also computed.

*Average of percentual quantities of mineral components
in the sediments of different ages*

TABLE 1

Sample	Na-feldspar	K-feldspar	Quartz	Amorph mat.	Kaolinite	Montmorillonite	Chlorite	Mica
Upper Pannonian	3.2	1.1	17.4	5.6	3.0	15.9	19.9	26.0
Lower Pannonian	5.1	1.4	18.4	2.7	1.4	10.4	30.2	28.0
Miocene	8.3	0.4	20.2	2.2	1.6	10.1	30.4	30.7
Miocene pelites	4.1	0.6	19.9	2.2	2.0	8.5	39.8	29.5
Miocene coarse sediments	11.4	0.3	20.4	2.1	1.2	11.2	23.5	31.8

It is worthy to note that the quantity of the minerals reflecting most sensitively the diagenetic changes, *i. e.* that of kaolinite, montmorillonite, chlorite and amorphous material, except micas, shows good agreement rather between the Lower Pannonian and Miocene, than between the Upper and Lower Pannonian samples.

Based on the differences of the average quantities of mineral components showing between the Upper and Lower Pannonian samples it would be a false conclusion that the diagenetic changes took place under relatively small depth of burial and during the subsequent burial only insignificant transformations followed since on the basis of other investigations at the boundary of the Upper and Lower Pannonian the change of the sedimentation environment and that of the source area, respectively, should be taken into account. Thus, in addition to the average values the investigation of the quantities of individual minerals as a function of depth prove to be also important.

It is to be noted here that when investigating the quantitative average values of minerals as a function of the depth it seems so that the great differences in the average values can be attributed to the change of the source area while the trends of changes can be related to the diagenetic effects in the Lower and Upper Pannonian.

The decrease of amorphous material parallel with the increase of crystallinity may also be in connection with the diagenetic transformation.

Regarding the average values of the pelites of Miocene age the diagenetic transformations are continued. If, however, we consider the dependence of depth

only the continuous increase of the mica content can be proved. The quantities of other minerals and that of the amorphous material either remains unchanged or show trends opposite to those observed so far.

Since as a function of depth mica, chlorite, kaolinite and montmorillonite show the most characteristic changes, further their genetic relationships can also be assumed the mica + chlorite/kaolinit + montmorillonite quotient was also calculated.

Parallel with the increasing mica + chlorite content the quantities of kaolinite and montmorillonite decrease. Greater deviations are found only in the coarse-clastic Miocene samples.

It has been assumed that the mica + chlorite/kaolinite + montmorillonite quotient marked by *K* in the following, proves to be characteristic of the diagenetic transformations. In case of the *K*-quotient the average values as well as the depth-dependence were studied both in the Upper and Lower Pannonian and in the Miocene samples.

Similarly to the clay minerals there is a definite difference in the average values between the Upper and Lower Pannonian samples, while the average value of the Lower Pannonian and Miocene samples are in accordance. Investigating the Miocene

TABLE 2
*Values of mica + chlorite/kaolinite + montmorillonite quotient (K);
the regression coefficients and the value of K extrapolated
to "O" m in the sediments of different ages*

Sample	K average	Regression coefficient	<i>a</i>
Upper Pannonian	2.30	$1.77 \cdot 10^{-3}$	0.91
Lower Pannonian	4.96	$2.61 \cdot 10^{-3}$	-5.08
Miocene	4.91	$-8.09 \cdot 10^{-3}$	41.24
Miocene pelites	6.41	$-8.06 \cdot 10^{-3}$	41.24
Miocene coarse sediments	3.65	$7.34 \cdot 10^{-3}$	-30.34

pelites separatedly, the average value increases in the following order: Upper Pannonian, Lower Pannonian, Miocene pelites. To draw any kind of conclusions concerning the diagenesis itself the dependence on depth should also be investigated. In the Upper and Lower Pannonian the value of *K* increases parallel with the depth, but the measure of increase is different and this is fairly reflected also by the regression coefficients.

Consequently, when taking into consideration the dependence on depth it can be concluded that diagenetic changes really take place in the Upper and Lower Pannonian. It is obvious, however, that the measure of diagenesis is different in the two sub-stages. Regarding all the Miocene samples and the Miocene pelites the *K*-value decreases parallel with increasing depth, *i. e.* in this case the diagenesis characteristic of the Upper and Lower Pannonian, cannot be taken into account. In the coarse sedimentary Miocene samples the *K*-value increases parallel with the

depth, but the correlation coefficient is in this case only 0.3, thus the dependence of K as a function of depth cannot be evaluated. Based on the values of K as a function of depth the value extrapolated to "0" m can be computed and this has been marked by a . These values do not really reflect the composition of clay minerals transported originally into the sedimentary basin. On the basis of the a -values it can be assumed that though the diagenetic changes should be taken into account, these changes cannot be regarded to be continuous, *i. e.* these take place in certain depth intervals but in other depth intervals these do not occur or take place only in restricted manner.

CHARACTERIZATION OF THE ORGANIC MATERIAL OF THE SEDIMENTARY ROCKS INVESTIGATED

The organic matter syngenetic to the mineral components of the sediments being either of plant or animal origin will be transformed during sedimentation and during the subsequent diagenesis. This transformation is essentially coalification and bituminization process which take place together in the nature. The fact that out of the processes mentioned above the coalification or hydrocarbon formation are characteristic of the given area, depends on the conditions predominating within the sedimentary basin, on the facies of sediments and last but not least on the composition of the starting organic matter.

Concerning the processes proceeded during transformation the investigation of the organic matter insoluble in organic solvents, *i. e.* of the kerogen may provide useful information.

It is known that three types of kerogen were distinguished by FORSMAN and HUNT [1958]:

1. *coaly type* — it resembles to the matters of peats, lignites and other coals thus it can be derived from lignine-bearing plant materials;
2. *non-coaly oil-shale type* — rich in lipids and proteins, it may be derived from algae, bacteria, etc;
3. *coaly oil shale type* — this is transition between the two previous types regarding both its origin and features.

Hydrocarbons derive from kerogens of non-coaly oil shale type. The genesis of methane, however, may be different.

To decide whether the kerogen of the core samples deriving from the deep basin parts of the South Great Plain played any role or not in the formation of the hydrocarbons discovered in the neighbouring areas, first of all the type of kerogen has to be known.

Since the composition, moreover the structure of the kerogens of different types considerably differ from each other it can be assumed that by means of determination of some characteristic functional group the possibility to classify the kerogens may be provided.

We started from the assumption that in case of kerogen of coaly type (which may derive from lignine-bearing plant material) the quantity of methoxy groups is relatively higher even if during the coalification process ever more of the about 17 per cent methoxy groups were cut during geological times. Further, knowing the geological conditions in can be assumed that the decomposition products of the plant material which got the sediments did not reach the anthracite state.

On the contrary, in case of kerogen of non-coaly oil shale type very low methoxy group content can be expected on the basis of the assumed starting organic matter.

The validity of this supposition is probalized by the investigations of SEMENOV *et al.* [1955]. Among others, they determined the methoxy group content of the kukersite kerogen of Estonia and this proved to be about 0.2 per cent.

It is to be noted, however, that in addition to the percentual determination of the methoxy group in possession of the C- and H-contents of the kerogen and of their quotient further evaluations of different points of view are provided.

Our investigations were carried out on the organic matter extractable by organic solvents, *i. e.* on bitumen as well as on kerogens remaining after the removal of mineral components by means of chemical methods. The determination of the methoxy groups was carried out by the method suggested by ZEISEL and modified by us.

The quantity of the methoxy groups varies between 2.48 and 1.60 per cent in the Lower Pannonian sediments of the investigated section of the Hód-I borehole (2885-5812 m). Its value is 1.14 per cent in the lowermost sample which may be assigned to the Lower Pannonian while in the Miocene sediments values between 0.78 and 0.32 per cent were found.

In the core samples of the borehole Makó-2 considerably smaller deviations were found in the methoxy group contents of the sediments of different ages. It varies between 1.65 and 1.18 per cent in the Lower Pannonian and between 0.64 and 0.27 per cent in the Miocene. The values of the two samples lying at the boundary of the Miocene and Lower Pannonian proved to be 1.28 resp. 1.57 per cent.

Without trying to draw definite limits among the types of kerogen on the basis of the percentual quantity of the methoxy groups or assign the kerogen of the investigated sediments into groups only on the basis of these data, the following is to be noted. In case of both boreholes the organic matter of the younger sediments is rather of plant origin resp. the coalification processes had been of predominating role in its transformation. In the older sediments, however, the original organic matter had been of other composition and the bituminization processes predominated in its transformation.

Further, on the basis of the quantity of methoxy content the insoluble organic matter of the core samples can be assigned to the group of kerogens of coaly oil shale type. It is to be noted, however, that in this case the kerogen of the core samples of Miocene age ought to be assigned to the group of kerogens of non-coaly oil shale type, this is to be verified, however, by the investigations to be done.

In the course of transformation of kerogen soluble organic matter also develops, consequently the soluble organic matter content (in the following bitumen) of the rock samples was also analysed.

The bitumen content was extracted in Soxhlet apparatus. Subsequent extraction was applied: first the ground rock sample was extracted by chloroform in order to get the Bit-A; then having treated the sample with hydrochloric acid the benzene-ethanol extraction was carried out. The extract obtained in this way is the Bit-C.

The extracts Bit-A and Bit-C were divided into three fraction by means of column chromatography.

In the sedimentary rocks of the "Hódmezővásárhely—Makó" Trench the Bit-A content varies between 0.020 and 0.249 per cent. Its value shows increasing trend as a function of increasing depth. This is in harmony with the statement of EREMENKO, *i. e.* the quantity of the neutral bitumen increases with the increasing age. The value of Bit-C changes nearly in the same interval with the only difference that the trend of change proves to be decreasing as a function of depth. This can be related probably to the gradual transformation of the organic matter.

Investigating separately the quantities of Bit-A for the Upper and Lower Pannonian it can be stated that its value hardly changes in the Upper Pannonian, while it shows increasing values as a function of depth in the Lower Pannonian.

The changes observed in the Miocene rock samples of the boreholes Hód-I and Makó-2 relate to two different petrological formations. In the borehole Hód-I the pelitic sequences predominate, while in the borehole Makó-2 coarse clastic formations were found. The quantity of Bit-A increases in the Hód-I samples and decreases in the Makó-2 samples when regarding only the Miocene sequence.

The increase following in the Lower Pannonian may be in connection with the more progressed measure of the continuous transformation of the organic matter and with the strengthening of the secondary character of Bit-A.

When grouping the changes of the Bit-C values also according to geological ages the following statements can be made: in the Upper Pannonian its quantity increases, in the Lower Pannonian it shows decreasing tendency, in the Miocene, however, changes of different character can be observed, similarly to those found in case of Bit-A. During the Miocene the quantity of Bit-C increases in the Hód-I samples and decreases in those of Makó-2. This change can be probably explained by the different measure of the Bit-C → Bit-A transformation as well as by the different petrological formations of the two sequences.

According to the investigations of TISSOT [1971] the quantity of the hetero-compounds of great molecular weight decreases as compared to that of the compounds poor in hetero-atoms or containing no heteroatoms. He assumed that the following transformation takes place: kerogen → compounds containing O, N and S hetero-atoms (acid bitumen) → hydrocarbons, resins, asphalts (neutral bitumen).

The experiments carried out by LOUIS and TISSOT [1967] on the samples of the Paris Basin relate also to such transformation. In the course of experiments the samples were treated under such pressure-temperature conditions which prevail in the deepest part of the basin. They observed the increase of the quantity of neutral bitumen and assumed that this proceeds through the formation of the acid bitumen.

In our investigations the Bit-C corresponds to the acid, the Bit-A to the neutral bitumen, thus according to TISSOT the kerogen → Bit-A → Bit-C transformation was assumed.

On the basis of the core sample investigations, however, this transformation cannot be so unambiguously followed. The quantity of the Bit-A and Bit-C of each samples is affected by the environmental factors, by the quantity of the organic matter got the sediments, by its quality and by the possible migration. Taking into account all these factors it can be stated that the change of the Bit-C value may in connection with consecutive character of the transformation itself. In the Upper Pannonian the kerogen → Bit-C, while in the Lower Pannonian the Bit-C → Bit-A transformations might prevail. The change followed in the Miocene samples of the borehole Hód-I may relate to the acceleration of the Bit-A-formation process.

The average quantities of bitumens (regarding both Bit-A and Bit-C) are considerably higher in the core samples of the borehole Hód-I than in those of Makó. This difference in quantities can be deduced from the positions of the boreholes in the sedimentary basin. On the basis of the borehole Hód-I. in the Hódmezővásárhely-Makó trench a hydrocarbon-bearing pelagic facies of negative redox potential prevailed with a water depth of several hundred metres.

The boreholes of Makó show littoral characteristics while the borehole Hód-I shows farther and deeper features on the basis of bitumen content, too.

The formation of the quantities of Bit-A and Bit-C was explained by the thermal

degradation of the organic matter. To explain the changes, however, it is insufficient to take into consideration only the chemical transformations since other factors may also play important role in the qualitative formation of the organic matter, as has been mentioned earlier, as well. One of the most important factors is the migration, thus in the course of our investigations we tried to throw light upon the primary or secondary character of the organic matter.

As to our assumption, to decide the primary or secondary character of the organic matter the quotient Bit-C/Bit-A together with the Bit-A and Bit-C values can be used. Since Bit-A consists of much more mobile components, the Bit-A values have to show maxima in the sites where the organic matter can be considered to be secondary (Fig. 1.).

Due to its more polarized character the Bit-C is able to adsorption, *i. e.* it is bound rather to the place of formation, consequently it can be assumed that its greater quantities relate to the primary character of the organic matter. If this assumption can be accepted within certain limits, the value of the Bit-C/Bit-A quotient will be low in case of secondary organic matter and will high in case of primary organic matter. This can be in connection with the mobility of Bit-A and immobility of Bit-C, respectively. The quotient itself alone is, however, insufficient to decide this problem. In case when the Bit-A value is high, but that of Bit-C is also high, *i. e.* the value of the quotient does not differ from the average it is less probable that in the samples there is an organic matter of secondary character (though the value of the quotient relates to this fact). Thus, taking into account the values of Bit-A and Bit-C it can be stated that high Bit-C/Bit-A quotient relates to the primary, the low one to the secondary character of the organic matter. Organic matter of ideally pure primary and pure secondary character was found in no core samples, consequently the denomination of primary and secondary denote a predominantly primary and predominantly secondary character. Between these two extreme states an intermediary group can be distinguished which, on the basis of the relative values of Bit-A and Bit-C can be assigned to the groups of "rather primary" resp. "rather secondary".

Taking all these into consideration it can be stated in general that the organic matter is rather of primary character in the Upper Pannonian, and rather of secondary character in the Lower Pannonian.

In case of the Miocene samples, however, the differences observed so far, also occur. In the Miocene core samples of the borehole Hód-I predominantly primary organic matter was found while in those of the borehole Makó-2 the organic matter of primary character becomes predominant only in a depth below 4560 m. Between 4560 and 4160 m, *i. e.* to the Lower Pannonian — Miocene boundary, the organic matter is mostly of secondary character. It is worthy of note, however, that in the borehole Hód-I the organic matter of primary character becomes predominant nearly in the same depth interval (4540—4880 m).

As it has been mentioned earlier, according to the transformation kerogen → Bit-C → Bit-A, a genetic relation between Bit-A and Bit-C can be assumed. Due to other factors influencing the quantity of the organic matter, this relation cannot be unambiguously followed.

On the basis of the factors used to distinguish the primary and secondary character of the organic matter and taking into account only the samples of primary organic matter content, a relationship was found between Bit-A and Bit-C.

The points lie on a straight line when plotted Bit-A against Bit-C (Fig. 2.).

In case of the borehole Hód-I the samples containing primary organic matter lie along two straight lines. Taking into account the distribution according to ages it can

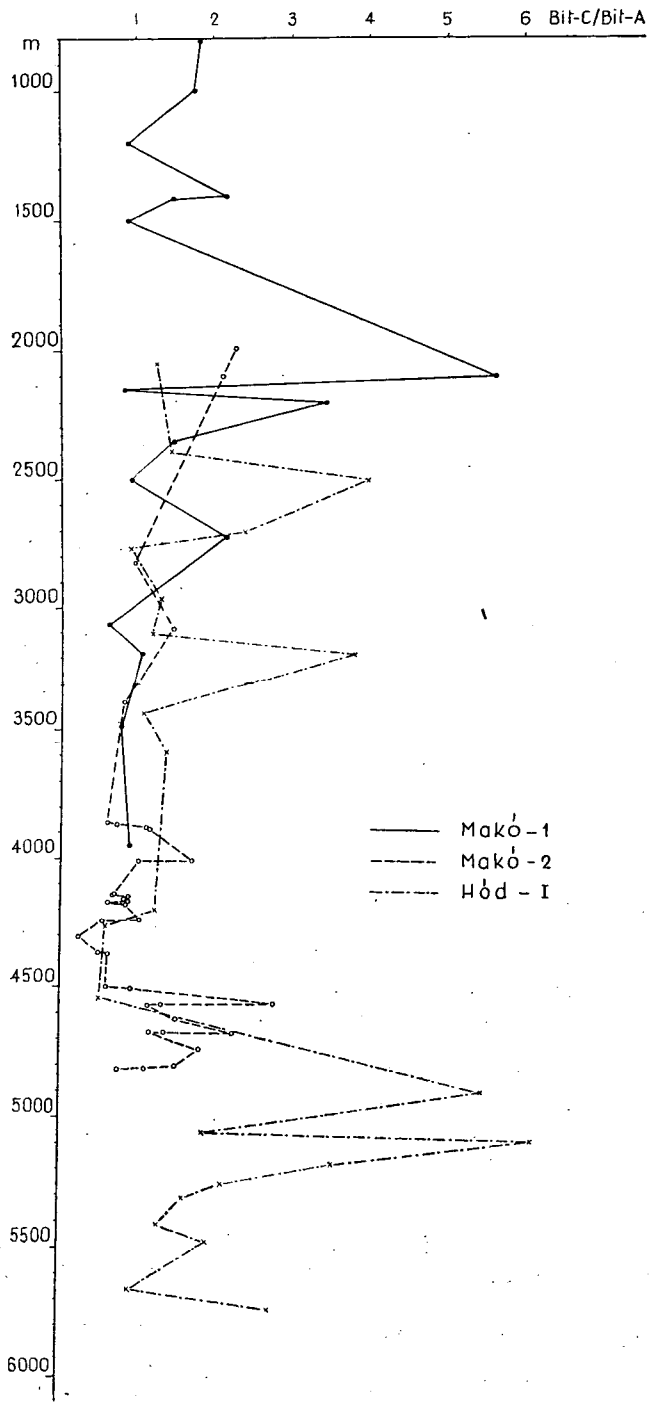


Fig.1. Dependence on depth of the Bit—C/Bit—A

be stated that the Miocene samples lie along the line of steeper rise, the samples deriving from the Lower Pannonian, however, lie along the line of less steeper rise.

In the borehole Makó-I the samples of Upper and Lower Pannonian ages were investigated. In case of the samples being assigned to the Upper and Lower Pannonian the rise of the straight lines is nearly the same and this may relate to a hydrocarbon genesis of similar character.

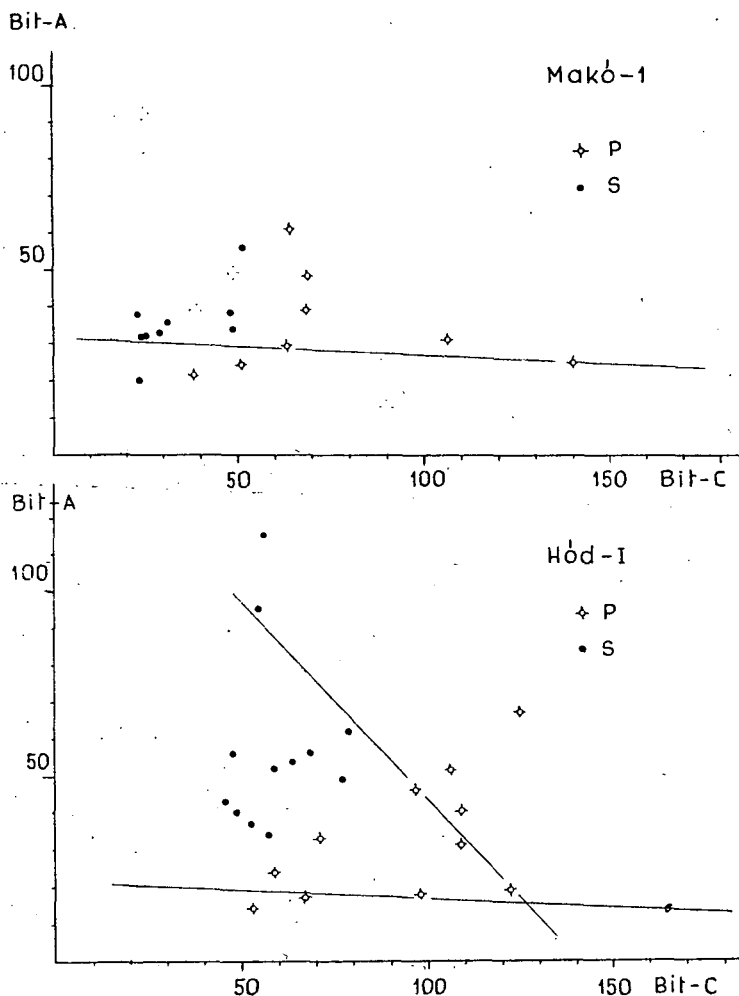


Fig.2. Change of Bit—A against Bit—C

On the basis of the two lines obtained for the core samples of the borehole Hód-I it can be assumed that hydrocarbon genesis of different character proceeded in the Miocene and in the Lower Pannonian.

Due to the migration of the organic matter not only the value of the Bit-C/Bit-A quotient will change but a relative difference in the bitumen composition also occurs.

This can be explained by the differences in mobility of the three fraction obtained in the course of column chromatographic separation of bitumen (in the first fraction the hydrocarbons, in the second the neutral resins, in the third the acid resins are found). It is to be noted here, that the separation by means of column chromatography was carried out in the samples where this proved to be possible from the quantity of the organic matter.

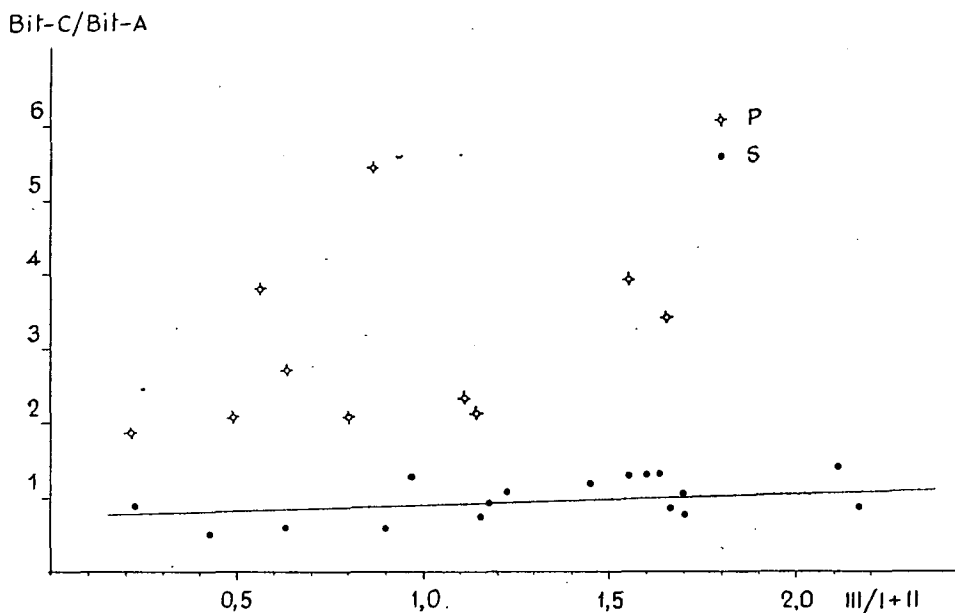


Fig.3. Relations of Bit—C/Bit—A and of the quotients of fractions obtained by column chromatography

According to our investigation results to separate the primary and secondary character of bitumen the column chromatographic fractions can also be used in addition to the quantity of the chloroform soluble bitumen. On the basis of these factors and of theoretical considerations the Bit-C/Bit-A quotient should be in relation with the quotient obtained from the fractions III/I+II.

When plotting the Bit-C/Bit-A quotient against the fractions III/I+II the core samples containing the organic matter of secondary character lie along one line in case of both boreholes and this may verify the relation between the two quotients in the changes during migration. (Fig. 3.)

As to our assumption the organic matter content of the core samples is in connection with the clay mineral content.

Since the adsorption capacity of the clay minerals is determined by their cation exchange capacity (in the following CEC), the change of the Bit-A and Bit-C was investigated as a function of CEC. The relation between the Bit-A and CEC verifies our assumption on the separation of primary and secondary bitumens. Relation between the two parameters may exist only in the samples which contain primary bitumen. In case of the samples, however, the Bit-A might emigrate, thus between Bit-A and Bit-C there may be a relationship that all the primary Bit-A lie on a line

of certain rise and on the are a under this line respectively, when plotted Bit-A against CEC (Fig. 4.).

As it was expected, between the CEC and Bit-C containing the polar groups in greatest quantity there is an unambiguous relationship: the quantity of Bit-C increases parallel with the CEC. Some of the samples show irregular behaviour, *i. e.* the

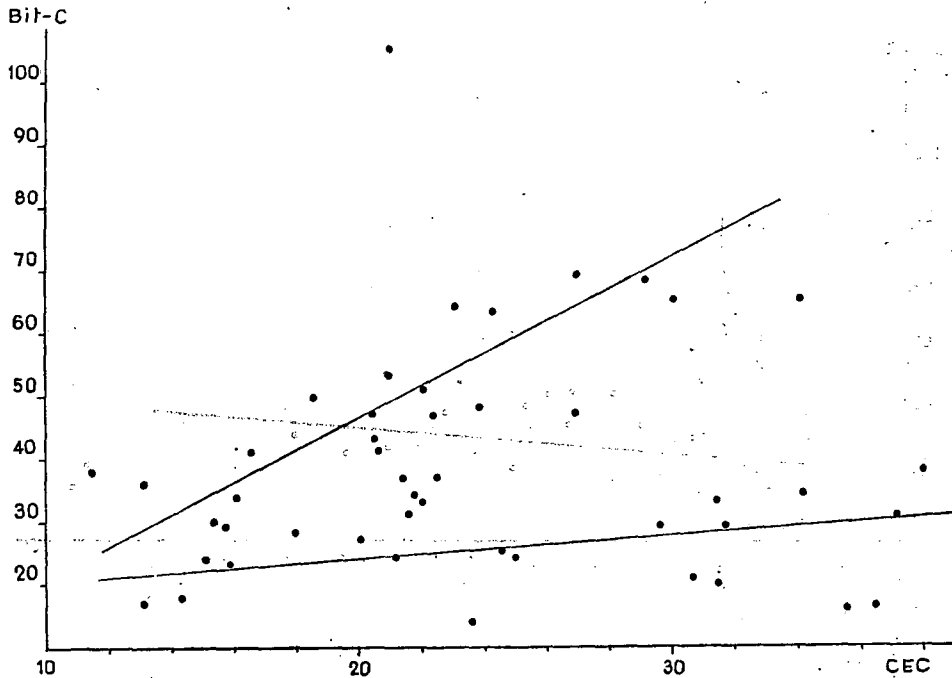


Fig. 4. Relation between Bit—C and CEC (cation exchange capacity)

quantity of Bit-C increases parallel with the CEC but this increase is of smaller measure than that observed in major part of the samples. This double distribution can be explained neither by the difference in the rock types, nor by the geological ages.

Further, the change of the quantity of the soluble organic matter was investigated as a function of the *K*-quotient characteristic of the diagenesis of clay minerals (Fig. 5.).

Part of the samples shows exponential increase with increasing *K*-value, the other part shows linear increase in the Bit-C quantity. The Upper Pannonian samples lie on the exponential, the Lower Pannonian and Miocene lie on the linear line.

The sharp difference between the Lower Pannonian and Miocene and Upper Pannonian samples, respectively, is reflected also by the geological conditions. On the basis of the macroscopic description the Upper Pannonian samples contain brown coal strips, humic substance and plant remnants. It can be assumed that in the Upper Pannonian a coal-forming while in the Lower Pannonian a sapropelic facies can be expected.

The exponential and linear character of the changes observed in the Upper

Pannonian, Miocene and Lower Pannonian samples respectively, can be explained by the difference of microbiological activity between the coal-forming and sapropelic facies.

The exponential change observed in the Upper Pannonian can be probably explained by the fact that in case of coal formation organic matter develops which restrain the diagenetic transformation of the clay minerals.

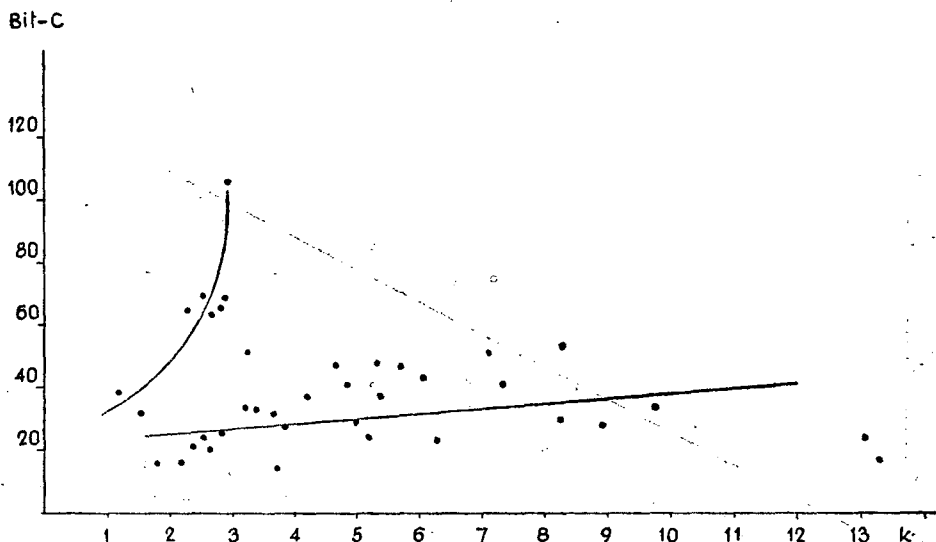


Fig.5. Relation between Bit—C and the quotient mica + chlorite/kaolinite + montmorillonite (K)

CONCLUSIONS

When evaluating the results of investigations, the following conclusions can be drawn:

1. On the basis of the dependence on depth of the quantities of each clay minerals, as well of their mutual relations diagenetic transformations can be assumed in the area in question. In the course diagenesis the mica and chlorite contents increase while the kaolinite and montmorillonite contents decrease, thus the K -quotient can be used to characterize the grade of diagenesis.

2. The quantity of the methoxy groups, together with the C- and H-contents and with their quotients allow to draw conclusions to the degree and character of the transformation.

3. A genetic relationship can be assumed between Bit-A and Bit-C, according to the transformation kerogen \rightarrow Bit-C \rightarrow Bit-A. As to our investigations the transformation cannot be unambiguously followed since in certain samples the quantities of Bit-A and Bit-C are highly affected by environmental factors, by the quality of the organic matter of the sediments, and by the possible migration.

4. It is assumed that to determine the character of the organic matter the Bit-C/Bit-A quotient resp. the Bit-A and Bit-C values can be used.

5. There is a relationship between the bitumen content and the cation exchange capacity of clay minerals influencing their adsorption capacity. This relationship verifies that the organic matter is bound to the fine-grained clayey components.

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