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MIXED-LAYER ILLITE/SMECTITES AND CLAY SEDIMENTATION IN THE NEOGENE OF THE PANNONIAN BASIN, HUNGARY

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Abstract: Shales and clay rocks of Neogene age were studied from 38 boreholes in the Pannonian Basin. The effect of geological factors on the transformation of smectite to illite was studied. These factors include the origin of the sedimentary material, facies and postdepositional tectonic evolution. Partial basins and relative structural heights within the Pannonian Basin were considered separately.
Terrigeneous clastic rocks deposited in marine or lacustrine environments are the most suitable rock types for the determination of the degree of diagenesis. Clays of *in situ* or redeposited kaolinitic weathering crusts found at the bottom and in particular horizons within the Neogene sequence usually contain low smectite proportions. Illite/smectites in weathered alluvial and continental clays of the uppermost stratigraphic horizons are of variable composition. On the other hand, smectite proportions in volcanogenic sediments are higher than those in terrigeneous ones in the shallow and transitional zones of diagenesis.
In continuously subsiding deep depressions the depth of the transitional zone of diagenesis is in the interval of about 2.0 to 3.5 km. In relatively uplifted areas the transitional zone may start at a depth of several hundred meters and its lower boundary may be found at about 1.5 to 2.0 km subsurface depth. This is due to the erosion of several hundred meters from the upper part of the sedimentary column and to differences in the thermal history of the sequences.

Key words: Neogene, lithofacies, Pannonian Basin, illite/smectite, XRD.

Introduction

In this paper the results of studies on diagenetic transformation of smectite to illite are presented. These studies were part of the hydrocarbon prospection and exploration carried out by the Hungarian Geological Survey in the Neogene fine-grained clastic rocks of the Pannonian Basin. The degree of the diagenesis was systematically determined by vitrinite reflectance and clay mineral analysis. The results of the clay mineralogical investigations were published so far only for a limited number of boreholes from the Hungarian part of the basin (Viczián 1982, 1985, 1992; Hámor-Vidó & Viczián 1994). Similar results from the Transcarpathian Basin (East Slovakia) were published by Franců et al. (1990a), Šucha et al. (1993) and from the Danube Basin (South Slovakia) by Šucha et al. (1994). The Slovak and Hungarian parts of the Pannonian Basin were compared with the Vienna Basin by Franců et al. (1990b).

Using the classical terminology of Müller (1967), the main phases of the smectite to illite transformation belong to the shallow and deep burial phases of diagenesis. These zones may be identified with the zones of litho- and katagenesis according to the classification of Balogh & Bérczi (1991, Figs. 7, 12). Only few samples from the lowermost part of the deepest boreholes (e.g. Hód-I, Doboz-I) can be classified as metagenetic according to this subdivision.

The variation of the smectite proportion (S %) with subsurface depth generally follows the classical model of Burst (1969; see also Révész et al. 1991), i.e. S % decreases simultaneously with the increase of depth forming three zones of transformation: zones of high, transitional and low smectite contents. The essential driving forces of diagenesis are increasing temperature and variations in the ionic composition of the pore waters (see the review of Viczián 1994).

Thermal aspects of the illite/smectite diagenesis were discussed in the Pannonian Basin by Viczián (1985) and in the Transcarpathian and Vienna Basins by Franců et al. (1990a). The results relating the three basins were later summarized by Franců et al. (1993) and Viczián (1994). All these investigations have shown that differences in the course of transformation do not completely disappear when plotting S % data vs. temperature or R_o % instead of subsurface depth. This means that there are other factors which may be responsible for the variation of S % with subsurface depth.

Among these other factors the effect of the pore water composition is not yet sufficiently studied in the Pannonian Basin. The importance of the initial mineralogical composition of the sediments was implicitely included in the remarks of Kübler (1984, Ch. III.3). He pointed to the possibility that S % may also rapidly change as a result of rapid changes in the bulk mineralogical composition at stratigraphic boundaries. The question of the initial composition was briefly discussed by Viczián (1984). Srodon (1979) and Šucha et al. (1993) demonstrated the difference between the behavior of shales and bentonites during diagenesis which is clearly a consequence of the difference in the initial composition of these rocks.

The aim of this paper is twofold: (1) to present as far as possible a complete data set of S % values obtained so far from the Hungarian part of the Pannonian Basin, and (2) to study the influence of geological factors of their variation during diagenesis, like the initial composition of the sedimentary material and later tectonic evolution of the basin.

Methods and materials

X-ray diffraction analyses of the smectite proportion (S %) in mixed-layer illite/smectites have been carried out on the < 2 μm grain size fraction. All measurements were carried out in the Hungarian Geological Survey. The applied X-ray diffraction method is based on determinative diagrams published by Srodon (1980). Description of the method and typical X-ray diagrams were presented in a previous publication (Viczián 1992). In many cases the diffraction patterns can be interpreted assuming that a single sample contains individual grains of variable smectite proportion (e.g. 40 to 60 %). In such case the range of S % values is shown in the diagrams of Figs. 2 to 15 by horizontal lines instead of point-like symbols.

The samples were collected from 38 boreholes (their location is shown in Fig. 1). 17 of the 38 boreholes were hydrocarbon producing wells, from which only a few core samples were available. The further 21 research boreholes were drilled by the Survey, and except for two boreholes (Mikepércs-1 and Nagyberény-1) continuous core sampling was performed. The field description of the cores of hydrocarbon exploratory boreholes, their age determination and stratigraphic classification were done by experts of the petroleum industry and the Hungarian Geological Survey. The geological relations of the Neogene fill of the Pannonian Basin were summarized by Dank & Jámbor (1987), Jámbor (1989, including lithostratigraphic nomenclature) and by Hetényi (1992, stressing the aspects of petroleum geology).

The samples were classified into four genetic types. Three types were already distinguished by Viczián (1984). By considering textural and paleontological features even more detailed genetic classification can be made (see e.g. Jámbor 1980) which is, however, not necessarily reflected in the bulk mineralogical composition (Viczián 1970). The simple subdivision into four genetic types proposed here focuses on the starting material of a possible further diagenetic transformation. It can be made with rather high reliability considering the characteristic bulk mineral composition of the samples:

1 – Terrigeneous clastic sediments. This is the most widespread sediment type among the marine and lacustrine basin sediments. The components originate from the redeposition of older sediments and from the not too intense weathering in the source area. Typical minerals are muscovite (illite) 2M, chlorite, quartz and generally two feldspar types (low-plagioclase and K-feldspar). In the < 2 μm grain size fraction the dominant clay mineral is mixed-layer illite/smectite. The typical iron mineral is pyrite.

2 – Kaolinitic weathering crust. It is the product of weathering during the continental period preceeding the Neogene transgression, found at the bottom of the Neogene sequences, sometimes *in situ*, but more frequently in a redeposited form. The formation is characterized by the kaolinite + illite (2M and 1Md) clay mineral composition, quartz and by the lack of swelling clay minerals and chlorites. Feldspar is also frequently missing or is present in very low amounts.

3 – Volcanogenic sediments. This is the material of tuffitic horizons produced by the intensive Neogene volcanism and of sediments dominated by products of weathering of the volcanic terraines. Na-smectite is the dominant clay mineral, other clay minerals such as

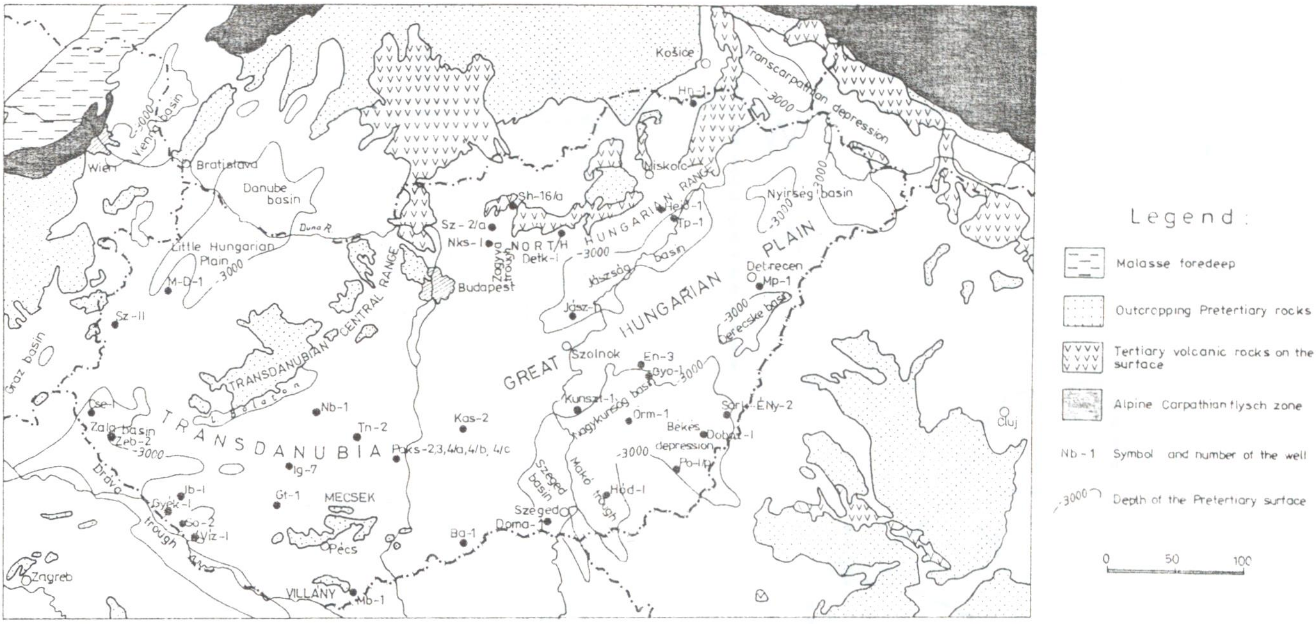

Fig. 1. Outline map of the studied wells. The geological map was compiled by Royden and Sandulescu (in Royden & Horváth 1985).

illite, kaolinite and chlorite are less important. Volcanic glass, opal-CT, zeolites, especially clinoptilolite and mordenite are frequent. The plagioclase is a high temperature modification. Sometimes a little biotite and amphibole can also be found. The quartz contents in the redeposited volcanogenic sandstones can be extremely low (e.g. borehole Tiszapalkonya-I, Upper Pannonian lignite-bearing formation).

4 – Sediments affected by weathering. All the three former sediment types may be modified by the impact of weathering in the course of sedimentation in lacustrine, floodplain or semi-arid to arid continental environments. Examples are the Upper Pannonian silty clay, marl, lignite-bearing and variegated clay complexes as well as the Lower Pleistocene red clay formations. These formations are partly oxidized (with goethite contents). Sometimes chlorites contain vermiculitic interlayers but usually no detectable modification of the mineralogy can be observed except oxidation. This group was distinguished because it was suspected that weathering and soil-forming processes may have influenced the initial S % values of the sediments.

Results: variation of the smectite proportion (S %) in different parts of the Pannonian Basin

Within the territory of the Pannonian Basin deep partial depressions and relatively emerged areas can be distinguished (see depth contour lines in Fig. 1). These areas differ in their history of subsidence. Deep partial depressions can be considered as more or less continuously subsiding basins filled by several km thick sediments columns. In relatively emerged areas the thickness of the sediments may be substantially lower (several hundreds of meter) due to low sedimentation rates or erosion and lack of sedimentation. In this chapter the variation of S % in the boreholes deepened in different regions of the basin will be examined.

From some of the boreholes we have well sampled complete sections while from others only sporadic data are available. Boreholes located close to each other and also those having similar stratigraphic relations were studied together. The aim was to determine the characteristic trend diagrams of the diagenetic transformation on the basis of reliable data for different parts of the basin. Sections are discussed starting from the W and going towards the E. For regional geological relations first of all the map enclosures of the monograph edited by Royden & Horváth (1985), the Pannonian map series compiled by Jámbor (Csiky et al. 1986-1987) and the worms-eye map of the Cenozoic formations edited by Tanács & Rálisch (1990, 1991) were consulted.

Basin of the Little Hungarian Plain (Fig. 2)

From the Little Hungarian Plain only one continuously sampled section, the Szombathely-II (Szh-II)

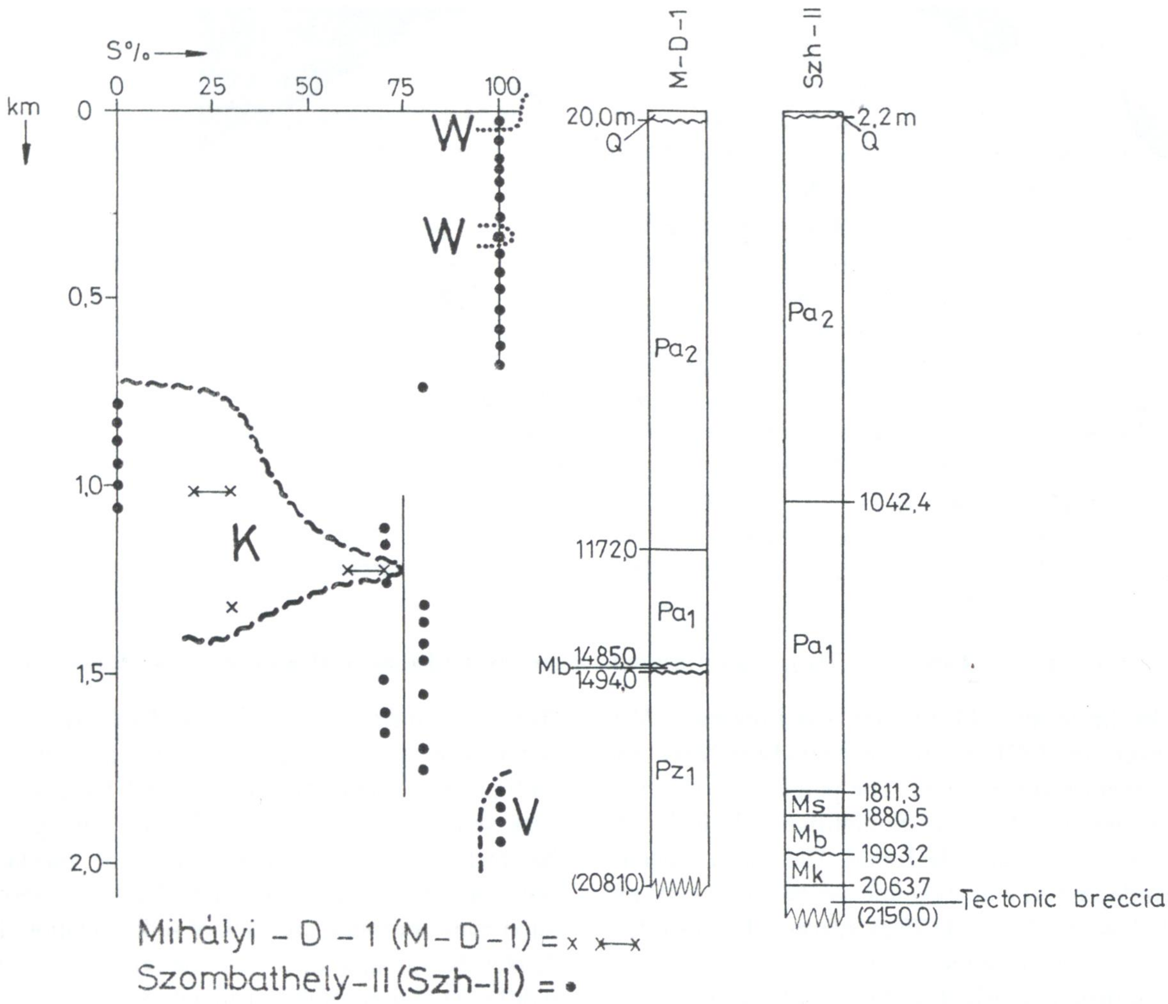

Fig. 2. Basin of the Little Hungarian Plain (Kisalföld).

Legend to the Figs. 2–16

[crystalline symbol]	Crystalline rock	Pa2	Upper Pannonian	E3	Upper Eocene	T	Triassic generally
		Pa1	Lower Pannonian	K	Cretaceous, generally	T3	Upper Triassic
[volcanic symbol]	Volcanic rock	Ms	Miocene, Sarmatian	K3	Upper Cretaceous	T2	Middle Triassic
		Mb	Miocene, Badenian	K2	Middle Cretaceous	T1	Lower Triassic
~~	Unconformity	Mk	Miocene, Karpatian	K1	Lower Cretaceous	Pe	Permian, generally
K ⌒⌒	Kaolinitic weathering crust	Me	Miocene, Eggenburgian	J	Jurassic, generally	Pz1	Lower Paleozoic
W	Weathered	Mo	Miocene, Ottnangian	J3	Upper Jurassic	Pk	Precambrian
V - • -	Volcanogenic	Ol	Oligocene, generally	J2	Middle Jurassic		
Q	Quaternary formations	Ol2	Upper Oligocene	J1	Lower Jurassic		

borehole was studied. Some sporadic samples from the Mihályi-Dél-1 (M-D-1) borehole are also available. Both wells explored a Neogene sediment sequence of medium thickness (1.5–2 km).

Badenian, Sarmatian and Lower Pannonian at the depth interval of 1.8–2.0 km are near-shore, shallow marine, low carbonate conglomerates and sandstones containing redeposited, volcanogenic clastic material. This may cause the maximal S values (100 %) while in the overlying terrigeneous sediments only S = 70–80 % can be found. The samples in the depth interval 0.8–1.1 km of Szh-II are kaolinitic, those of the M-D-1 borehole are transitional between terrigeneous and kaolinitic types (nearly equal amounts of kaolinite and chlorite in the clay fraction).

The trend diagram of the transformation was drawn disregarding the volcanogenic and kaolinitic samples. Here the transformation is rather slow, presumably

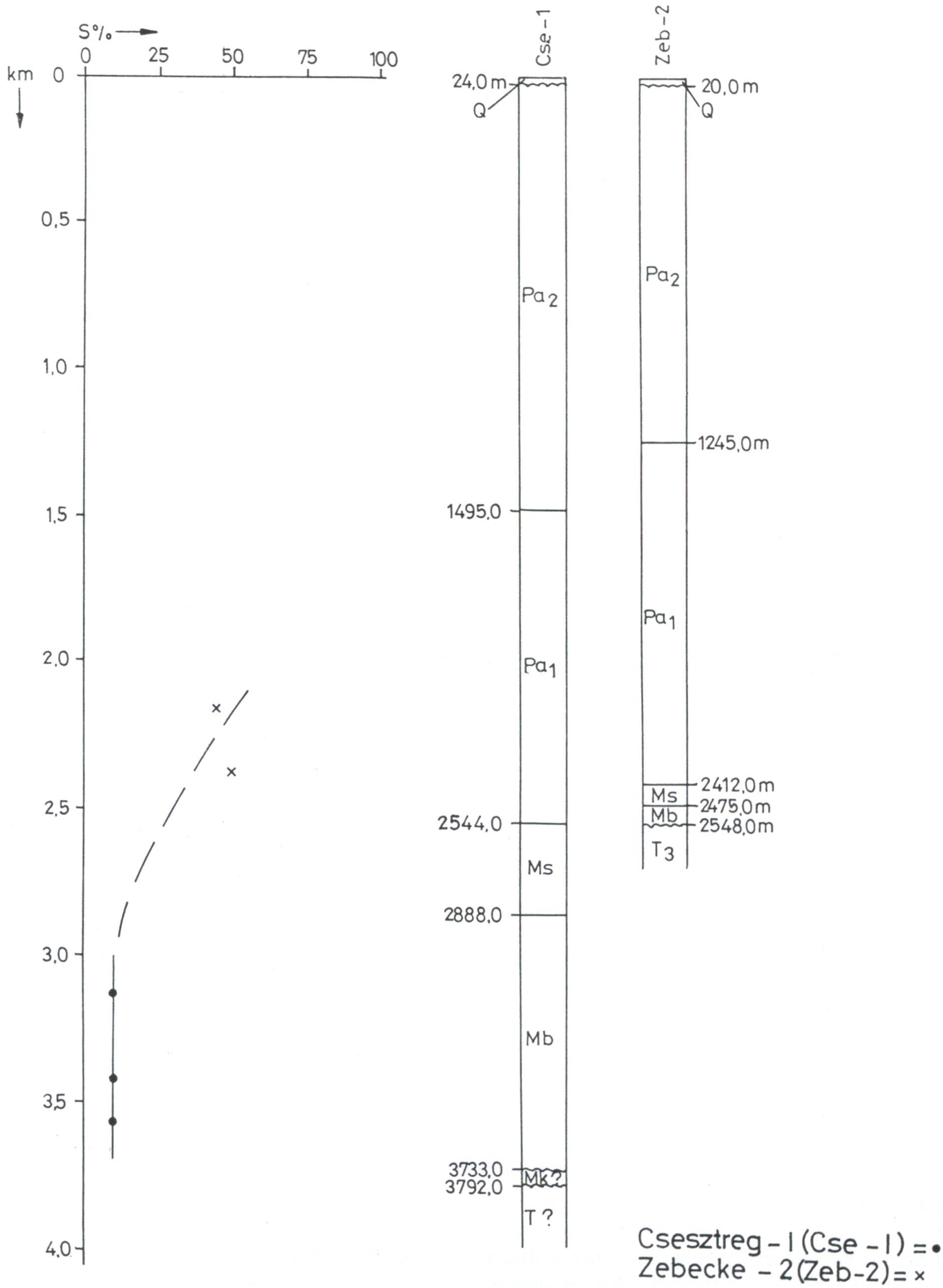

Fig. 3. Zala Basin.

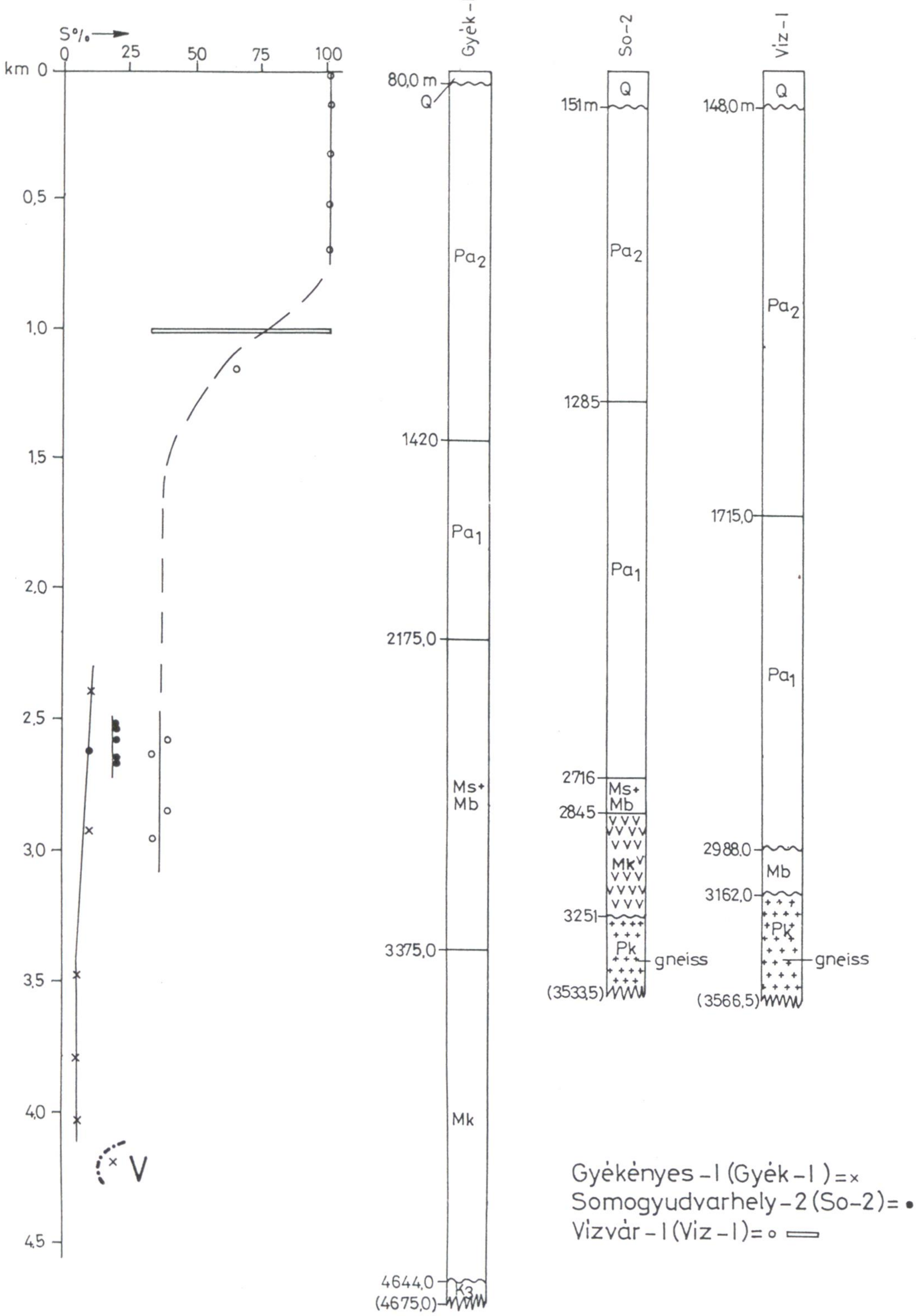

Fig. 4. Dráva Basin.

because it is the "coldest" part of the Pannonian Basin. In the Szh-II borehole the transformation of smectite starts under a depth of 0.7 km but only its upper zone is recovered by the Neogene sequence of the borehole.

Zala Basin (Fig. 3)

From the Zala Basin area only a few data are available, obtained from two boreholes, Csesztreg-I (Cse-I)

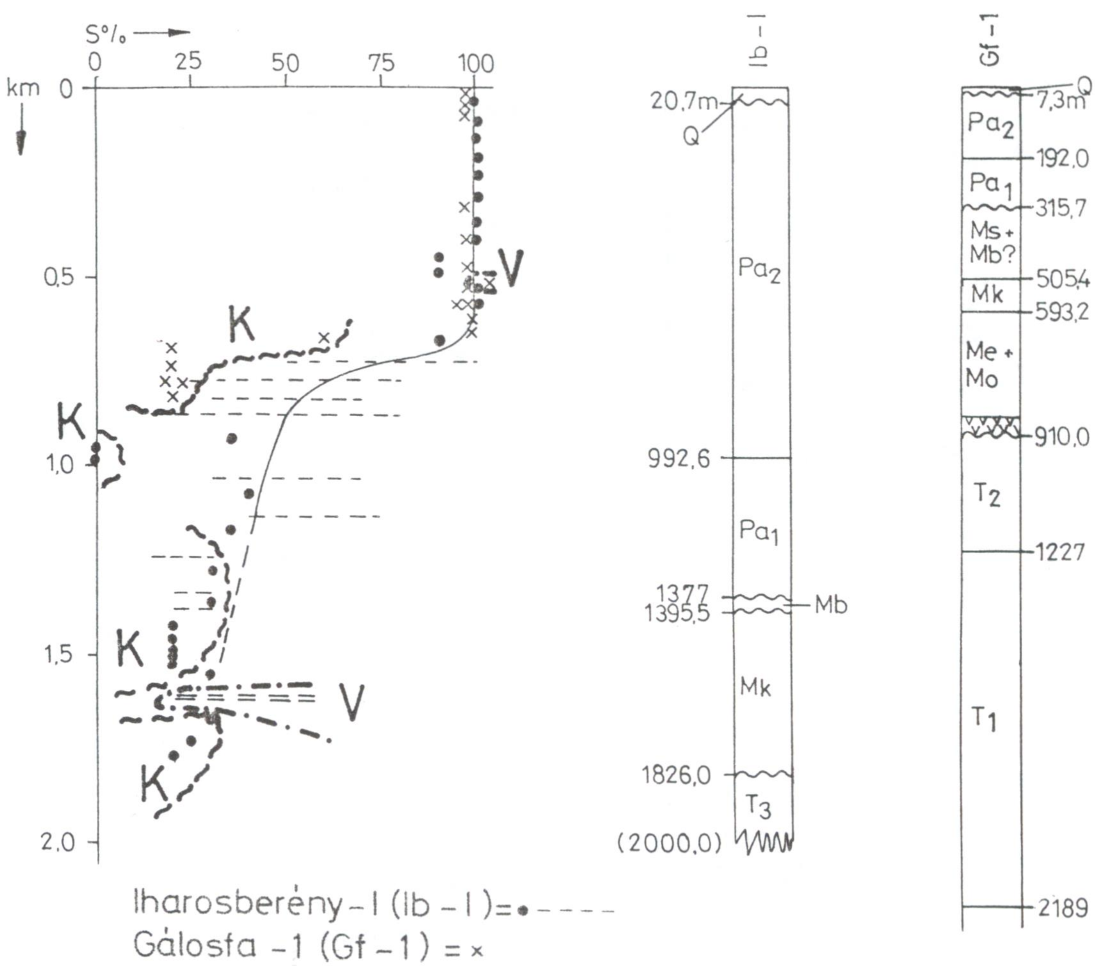

Fig. 5. NE marginal region of the Dráva Basin.

and Zebecke-2 (Zeb-2). In both boreholes the Pannonian sequences are of about the same thickness (ca. 2.5 km), while the total thickness of the Neogene is significantly different. In the borehole Zeb-2 both Sarmatian and Badenian are made up of thin, shallow marine, near-shore formations. In the Badenian clayey marl with gravel intercalations, marl, calcareous marl, limestone breccia and lithothamnic limestone were formed upon which, with continuous sedimentation Sarmatian marl, siltstone and sandstone layers were deposited. On the other hand, the Cse-I borehole was bored in the central area of the Őrség deep basin where thick Badenian and Sarmatian sequences were formed.

In spite of the above differences the data of the two boreholes, with more or less uncertainties, may be connected with a common trend line, because the thickness of the Pannonian is very similar in the two sections. In the Cse-I borehole the strong diagenetic transformation and low S % values (S = 10 %) can be accounted for by the comparatively old, Badenian age and the strong tectonic stress (several sliding planes, steep layer dippings: Kőrössy 1988).

Dráva Basin (Fig. 4)

From the central deep zone of the Dráva Basin the samples of three boreholes were studied. The deepest part was explored by the Gyékényes-I (Gyék-I) borehole that cut acoss a more than 4.6 km thick Neogene layer sequence. The older, Karpatian-Badenian samples are unusually strongly transformed (S = 5-10 %). The lowermost Karpatian core, because of its tuffaceous material, displays somewhat higher value (S = 20 %).

The two other boreholes, Somogyudvarhely-2 (So-2) and Vízvár-I (Víz-I) cut across Neogene sequences of about the same thickness, i.e. somewhat more than 3 km. In the Víz-I borehole the diagenetic transfomation starts at a depth between 0.7-1.0 km but its progress, due to the lack of samples, could not be followed. In the Víz-I borehole at a 2.5-3.0 km depth interval we have still S = 35-40 % while in the So-2,

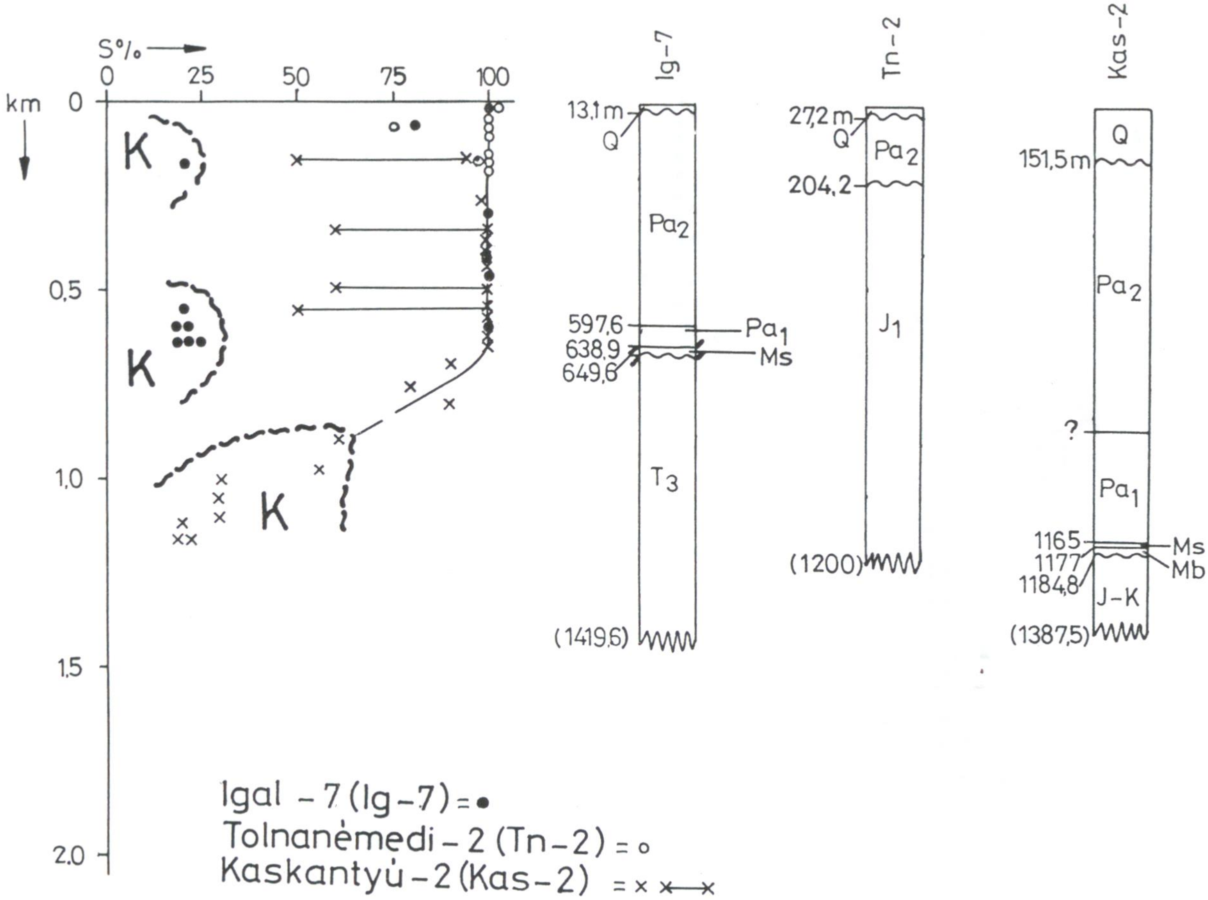

Fig. 6. Shallow basin parts in Central Transdanubia and in the Danube-Tisza Interfluve.

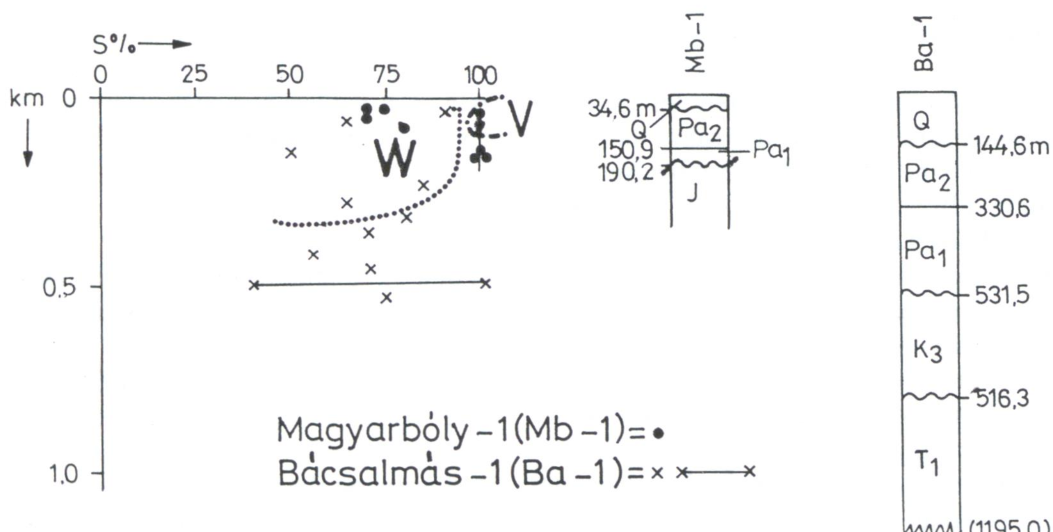

Fig. 7. S foreland of the Mecsek Mts., S part of the Danube-Tisza Interfluve.

at 2.5-2.7 km it is only S = 20 %. Since both of the sample series are of nearly the same age, Lower Pannonian, the difference could not be interpreted so far.

NE marginal region of the Dráva Basin (Fig. 5)

The Iharosberény-I (Ib-I) borehole can be still classed to the Dráva Basin differing, however, from the previous ones with its much thinner Neogene layer sequence (< 2 km). The borehole was deepened at the W part of the elevated Inke structure. This "high block range towards E ... passes over to the elevated foreland of the N Mecsek Mts." (Kőrössy 1989, p. 6). For this reason the Ib-I borehole will be discussed together with the Central Transdanubian Gálosfa-1 (Gf-1)

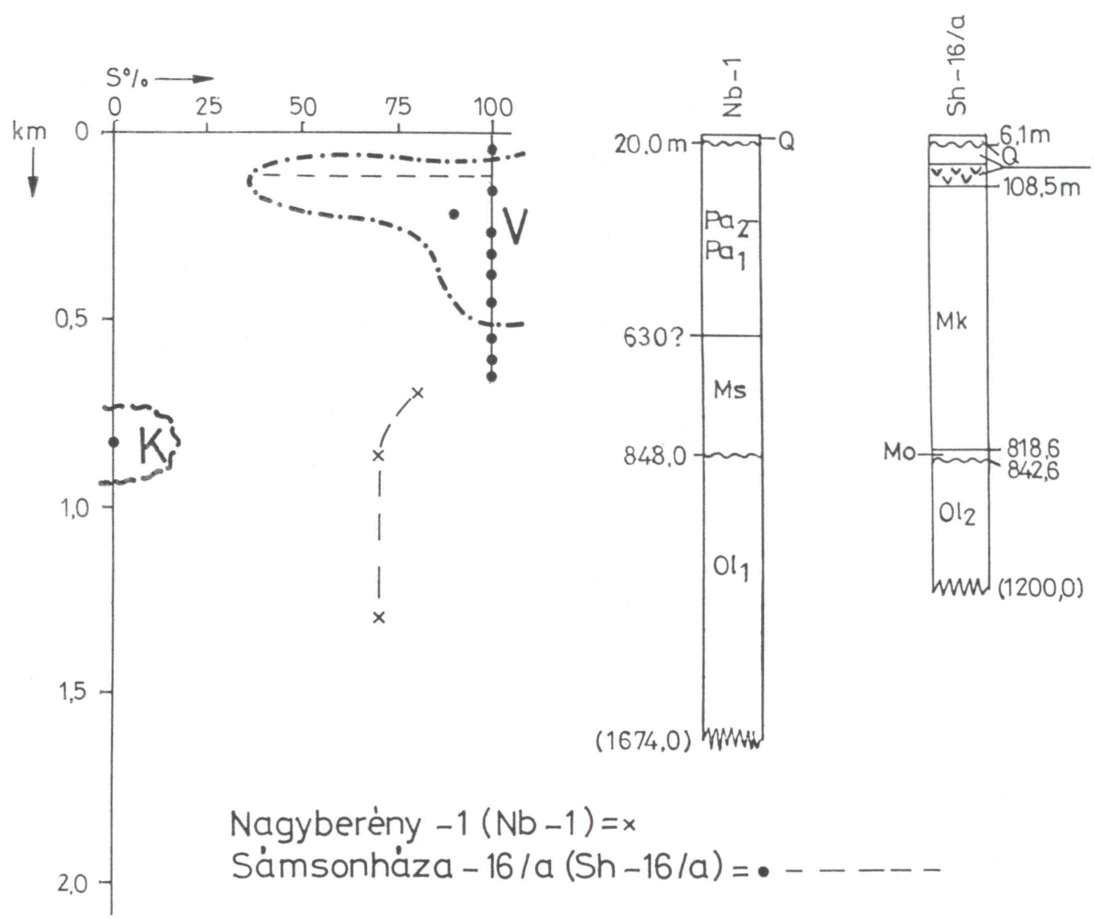

Fig. 8. Paleogene based Neogene basins with thin Miocene volcanites.

borehole. The Neogene sequences of this region generally start with kaolinite-rich formations which, however, may be of different ages.

The sequences of the Gf-1 borehole is similar to that of the Ib-I because the lower part of it includes kaolinite-rich layers of low S % (clayey, gravelly sand, gray, green, red variegated clay). These in the Ib-I are of Karpatian while in the Gf-1 they are of Eggenburgian and Ottnangian ages, respectively. The S % values in these kaolinite-rich layers are below 30 %, in a single sample of transitional kaolinitic-terrigeneous composition in the borehole Gf-1, S = 60 %. Prolongation of the diagenetic trend line into the depth interval of these kaolinitic layers would be misleading, though the difference between the terrigeneous and kaolinitic layers in the Ib-I borehole is less pronounced than in Gf-1. Two samples in the Ib-I borehole represent the kaolinite-rich formation at the Lower to Upper Pannonian boundary (see also borehole Szh-II, Fig. 2). In the Ib-I borehole two samples of volcanogenic material have higher S % values (up to 60 %) than the overlying sample (S = 30 %) which is mostly terrigeneous (at 1.6 km depth). Here the transformation begins at a depth of about 0.7 km. From the Gf-1 borehole it can only be stated the transformation had not yet started at the depth of 0.65 km.

Shallow basin parts in Central Transdanubia and in the Danube-Tisza Interfluve (Fig. 6)

In the Kaskantyú-2 (Kas-2) borehole very thin Badenian and Sarmatian and thick Pannonian were deposited on the surface of the Mesozoic. These formations are kaolinite-rich below about 1.0 km, the Upper Pannonian is terrigeneous, the Lower Pannonian between 9.0 and 9.7 km depths is of transitional composition. Kaolinite-rich samples have low S % values (< 30 %), those of transitional kaolinitic-terrigeneous composition contain mixed-layers with S = 55–60 %. It is difficult to decide in this case, whether the decrease of S % is due to the increasing kaolinite content or to the progress of diagenesis. In the Igal-7 (Ig-7) borehole the contact between the Mesozoic basement and overlying Sarmatian and that between the Sarmatian and Lower Pannonian is of tectonic type with traces of hydrothermal activity. Accordingly, in this latter borehole the hydrothermal formation of kaolinite is possible. In the Upper Pannonian, at about

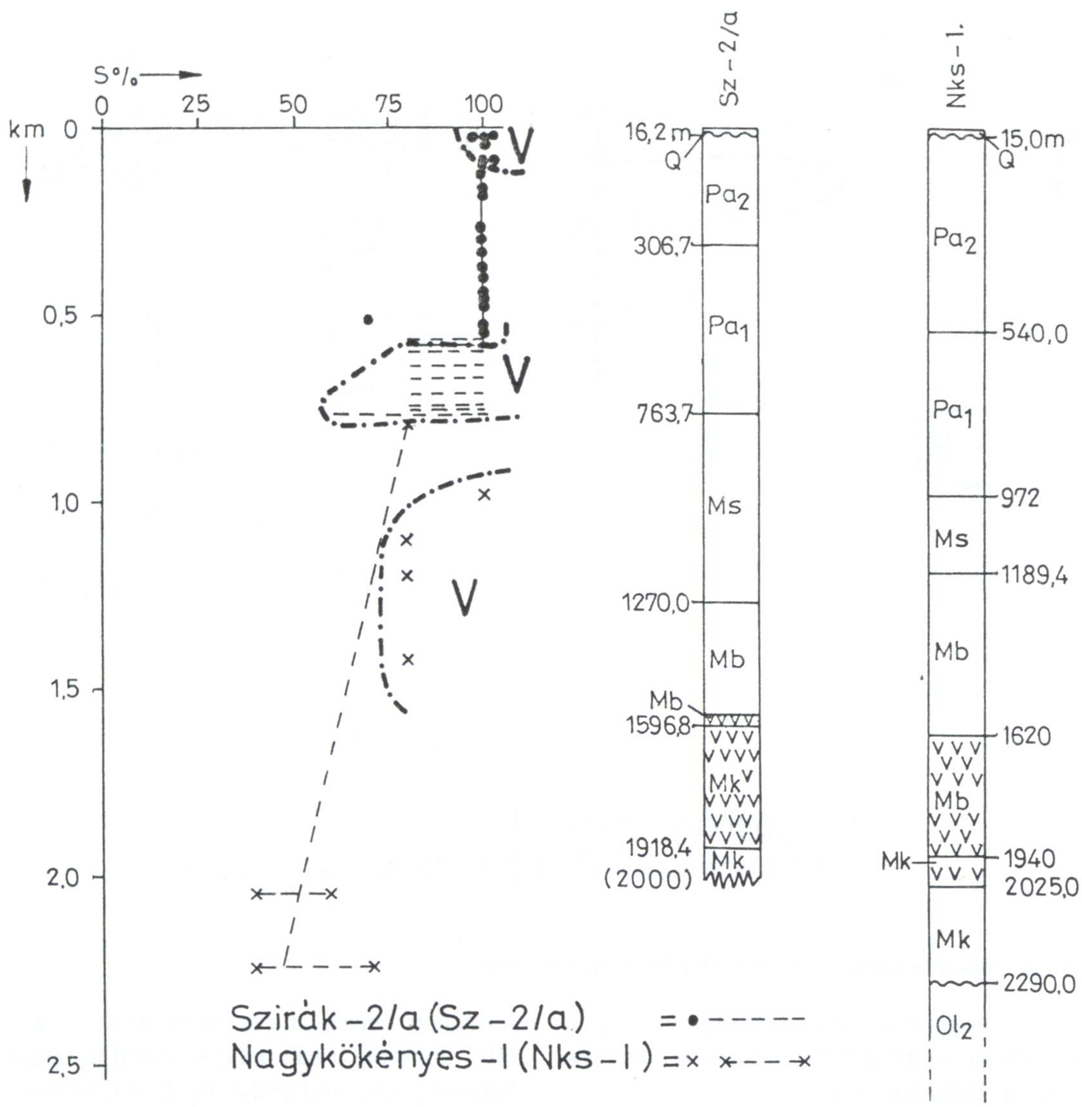

Fig. 9. Paleogene based Neogene basins with thick Miocene volcanic formations.

170 m depth a single kaolinite-rich sample with S = 20 % was found. This is a special clayey ankeritic dolomite-rock of presumably shallow lacustrine origin.

In these boreholes only the Upper Pannonian-Quaternary sequence can be interpreted from the point of view of diagenesis. Transformation in the Kas-2 borehole starts at 0.7 km. Concerning the Ig-7 borehole we can only conclude that till 0.5 km no diagenetic transformation could be observed.

The very thin (ca. 200 m thick) Upper Pannonian of the sequence of the Tolnanémedi-2 (Tn-2) borehole was deposited directly on an elevated horst of the Mesozoic basement. Diagenetic transformation cannot be observed and no redeposited kaolinitic weathering crust can be found, S is almost exclusively 100 %.

S foreland of the Mecsek Mts., S part of the Danube-Tisza Interfluve (Fig. 7)

Two borehole profiles were studied from this area, Magyarbóly-1 (Mb-1) and Bácsalmás-1 (Ba-1, see Fig. 7). Here comparatively thin Pannonian and Quaternary sequences were deposited upon the surface of the Mesozoic basement. Practically at both sites, only the Lower Pannonian can be diagenetically interpreted. In the Upper Pannonian and Quaternary siltstones, silty clays and variegated clays terrigeneous sediments were altered by the contemporary surficial weathering, which generally resulted in decrease of S %. Thin layers of volcanogenic sediments with S = 100 % are present in the Upper Pannonian of the Mb-1 borehole.

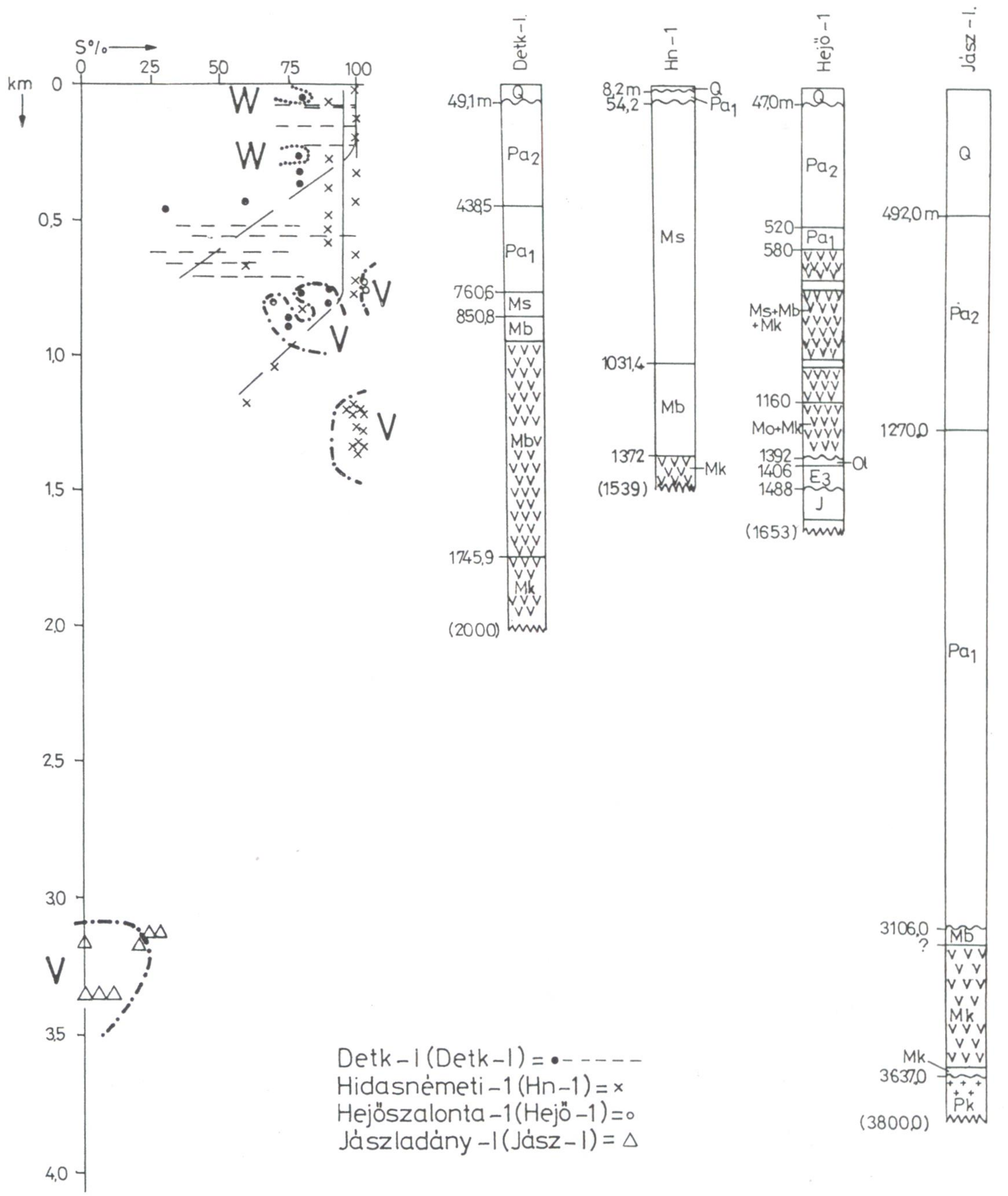

Fig. 10. Neogene basins with thick Miocene volcanic formations (N Hungary).

Paleogene based Neogene basins with thin Miocene volcanites (Fig. 8)

Two, comparatively remote boreholes were studied where the not too thick (1-1.5 km), rather old Neogene is underlain by Oligocene formations. At both sites, however, there is unconformity at the Paleogene-Neogene boundary. The Nagyberény-1 (Nb-1) borehole is located S of the Lake Balaton, in a deep basin on the S side of the Balaton structural line. According to Kőrössy (1990) here the Paleogene clastic sediments "are unconformably covered by Karpatian and younger basin sediments".

The other borehole, Sámsonháza-16/a (Sh-16/a) was deepened in the N end of the Zagyva Graben (Hámor 1985). Here, in spite of the relatively old Miocene age, the diagenetic transformation does not start till about 0.7 km. In Hámor's opinion (1985) in the Schlier Formation, in general, redeposited volcanogenic material may be present. Between 108 and 452 m the samples contain much opal-CT and smectite that may indicate volcanogenic origin. One sample

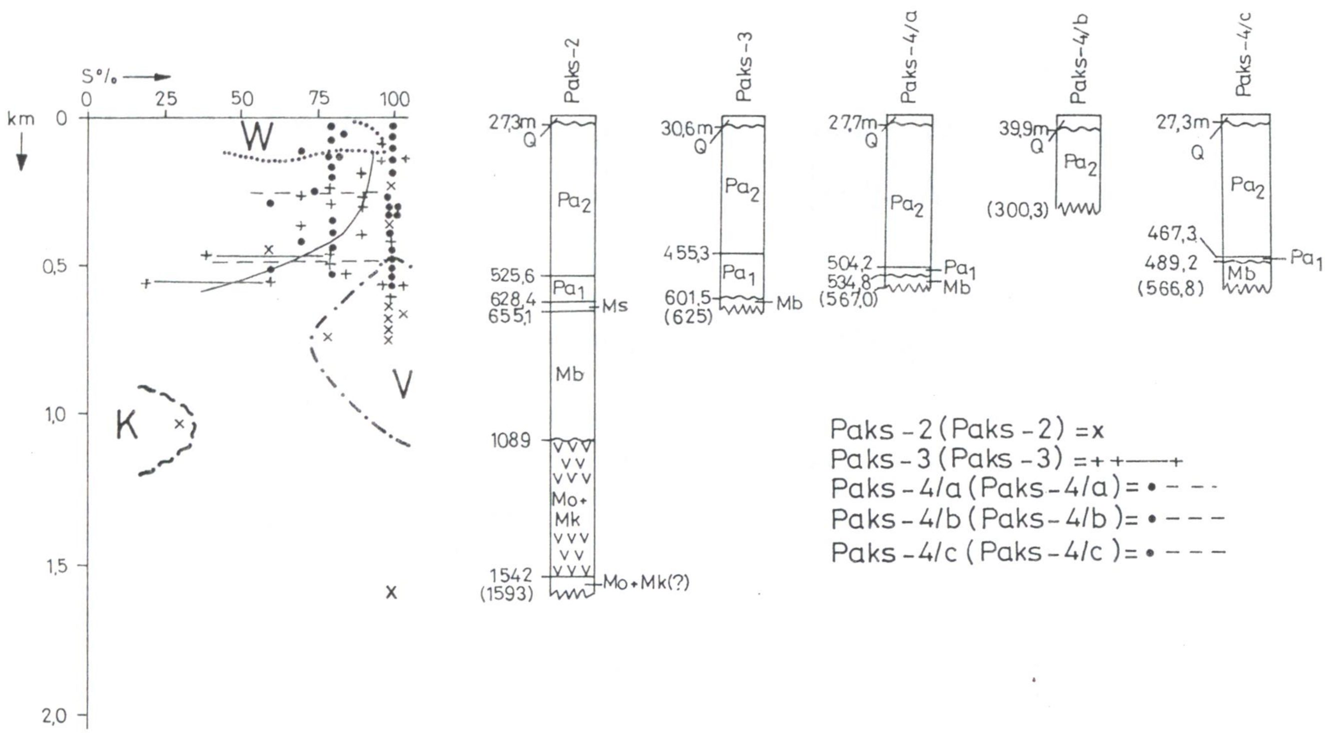

Fig. 11. Neogene basins with thick Miocene volcanic formations (Paks, Central Hungary).

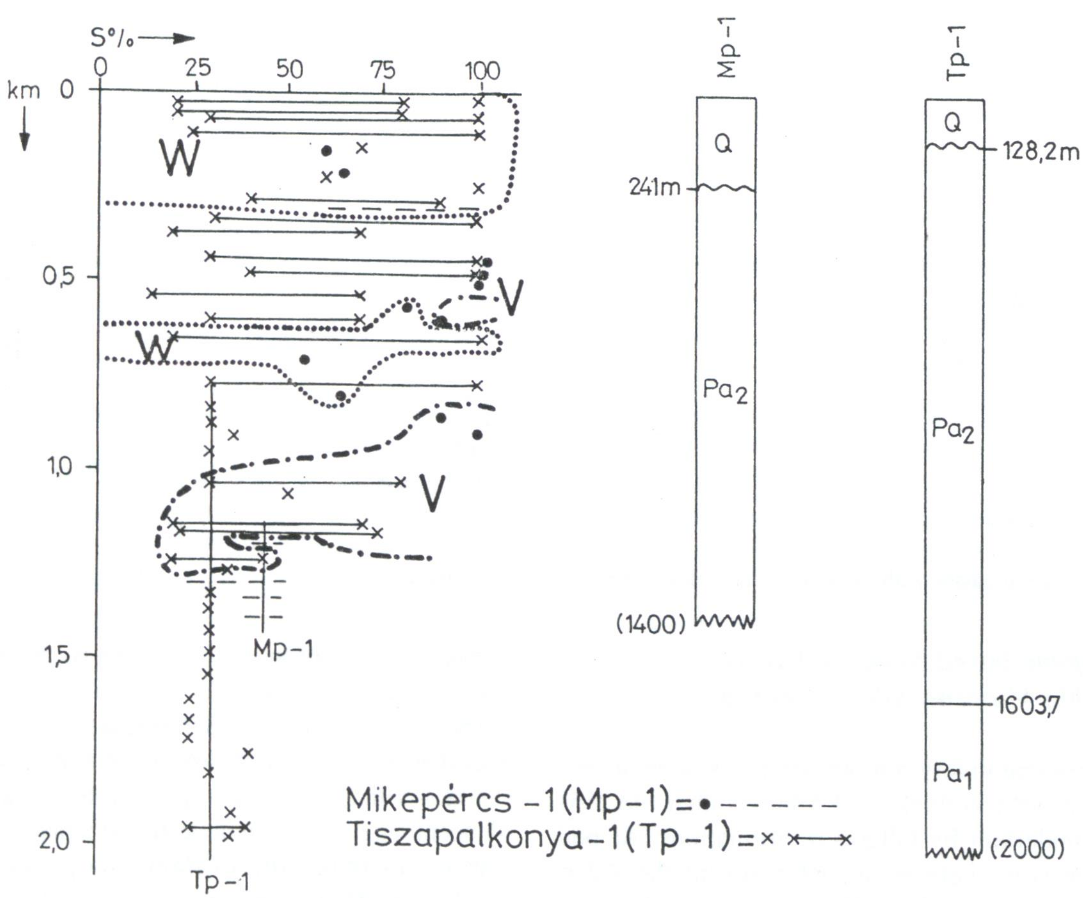

Fig. 12. NE Tisza Basin.

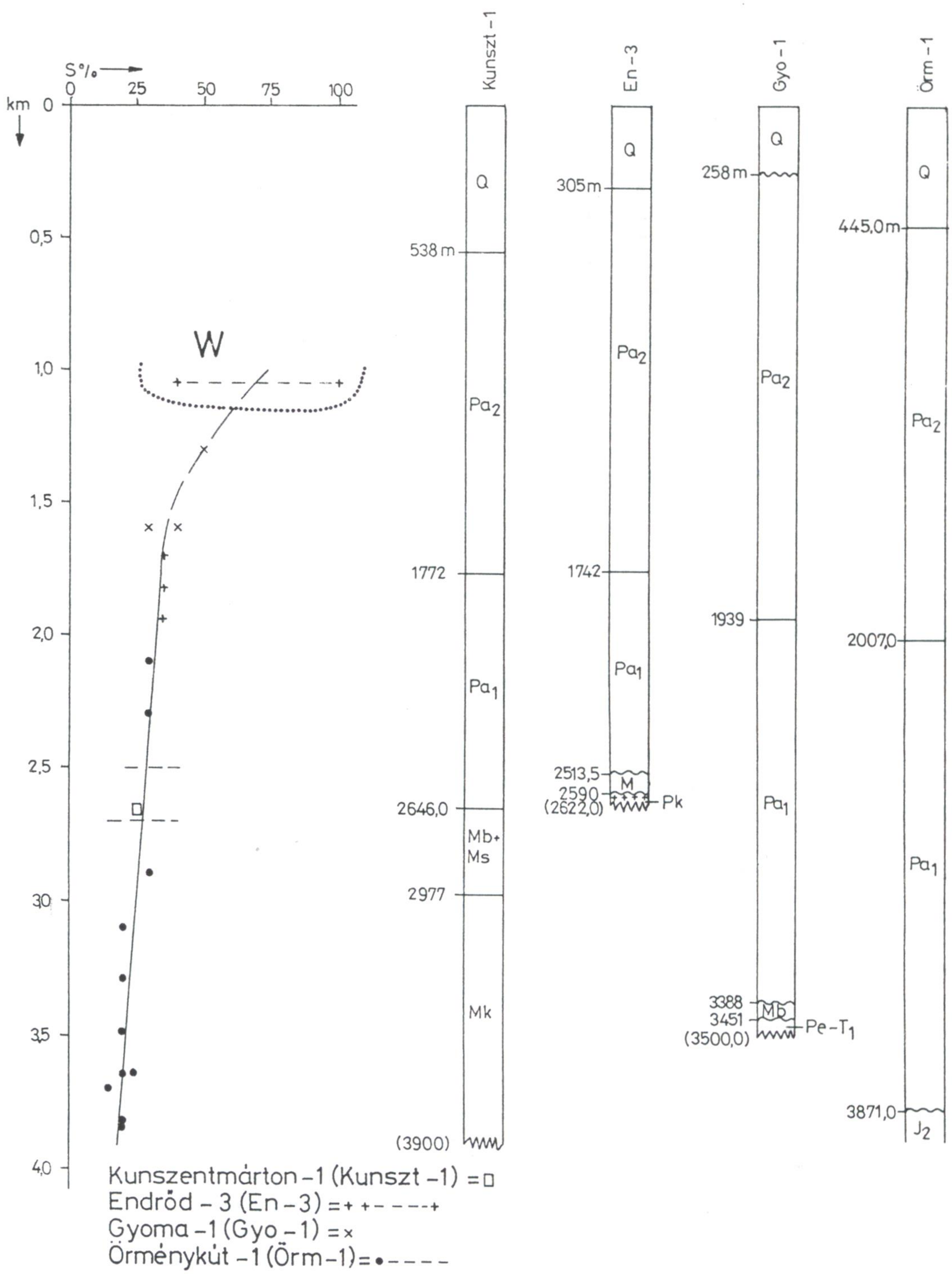

Fig. 13. Nagykunság Basin.

belonging to the Ottnanginian coal formation represents the illite + kaolinite association (S = 0 %).

The two Neogene samples studied from the Nb-1 borehole, found at greater subsurface depth, fall into the zone of the starting diagenetic transformation in the depth interval 0.7–0.85 km, and this fits well into the lower continuation of S values of the Sh-16/a borehole.

Paleogene based Neogene basins with thick Miocene volcanic formations (Figs. 9, 10)

Here layer sequences will be discussed where the comparatively thin Badenian-Pannonian series overlies a rather thick Miocene volcanic and sedimentary sequence as well as Oligocene sedimentary rocks.

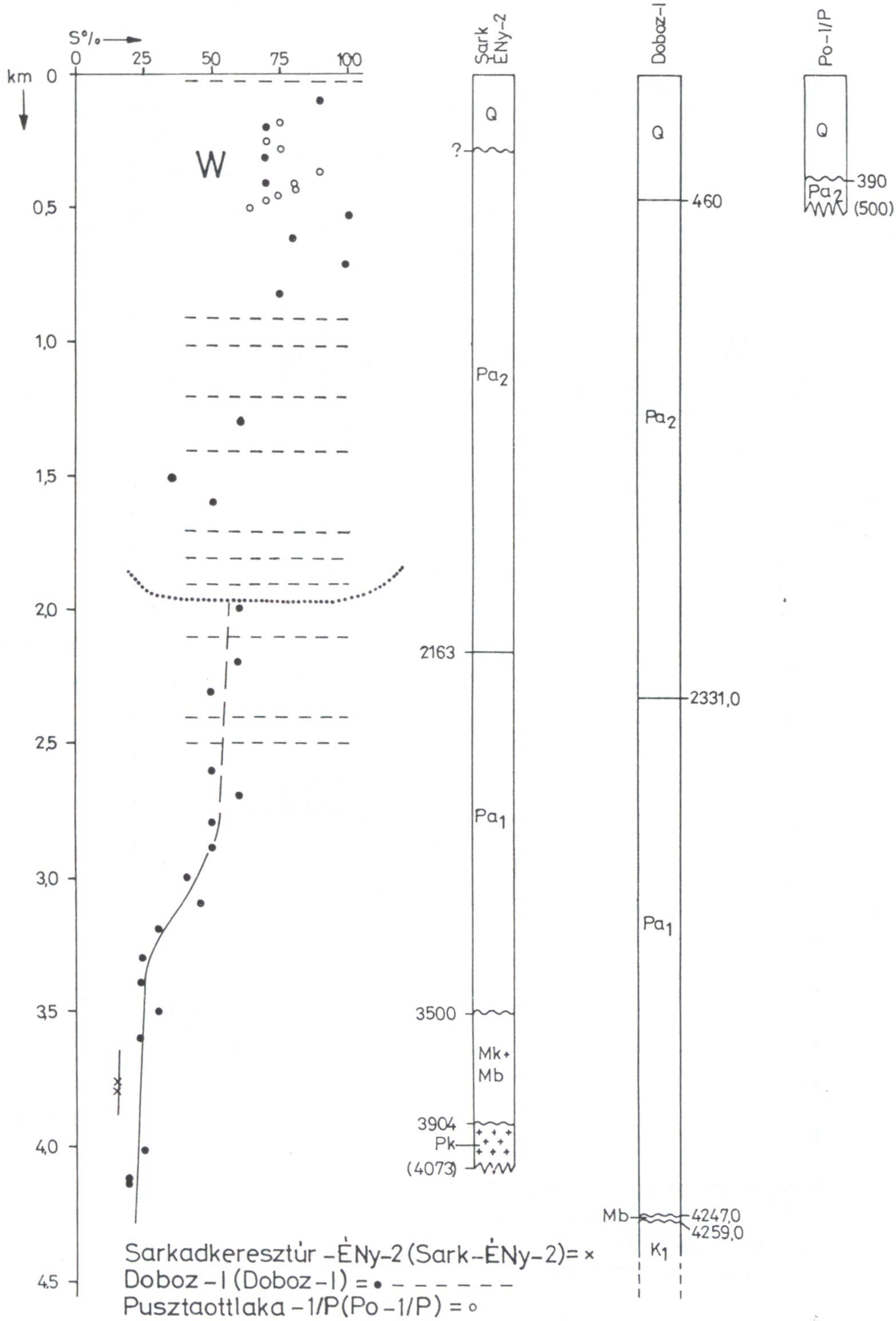

Fig. 14. Békés Basin.

Two boreholes of this type, Szirák-2/a (Sz-2/a) and Nagykökényes-I (Nks-I) were drilled in the Zagyva Graben close to each other, so their data are almost each other's continuation (Fig. 9). In these layer sequences there are sedimentary rocks containing redeposited volcanogenic materials in various proportions. In the zone of shallow diagenesis, however, S % values are nearly the same (in Sz-2/a) or probably somewhat higher (in Nks-I) than those in the terrigeneous clastic rocks (80–100 %).

Considering only the clearly terrigeneous clastic layers, it can be stated that no diagenetic transformation took place in the Sz-2/a borehole, until 0.6 km. In the Nks-I borehole there are only three evaluable data, falling into the depth interval 0.8–2.3 km, and it seems that they are in the zone of transformation (S = 40–80 %).

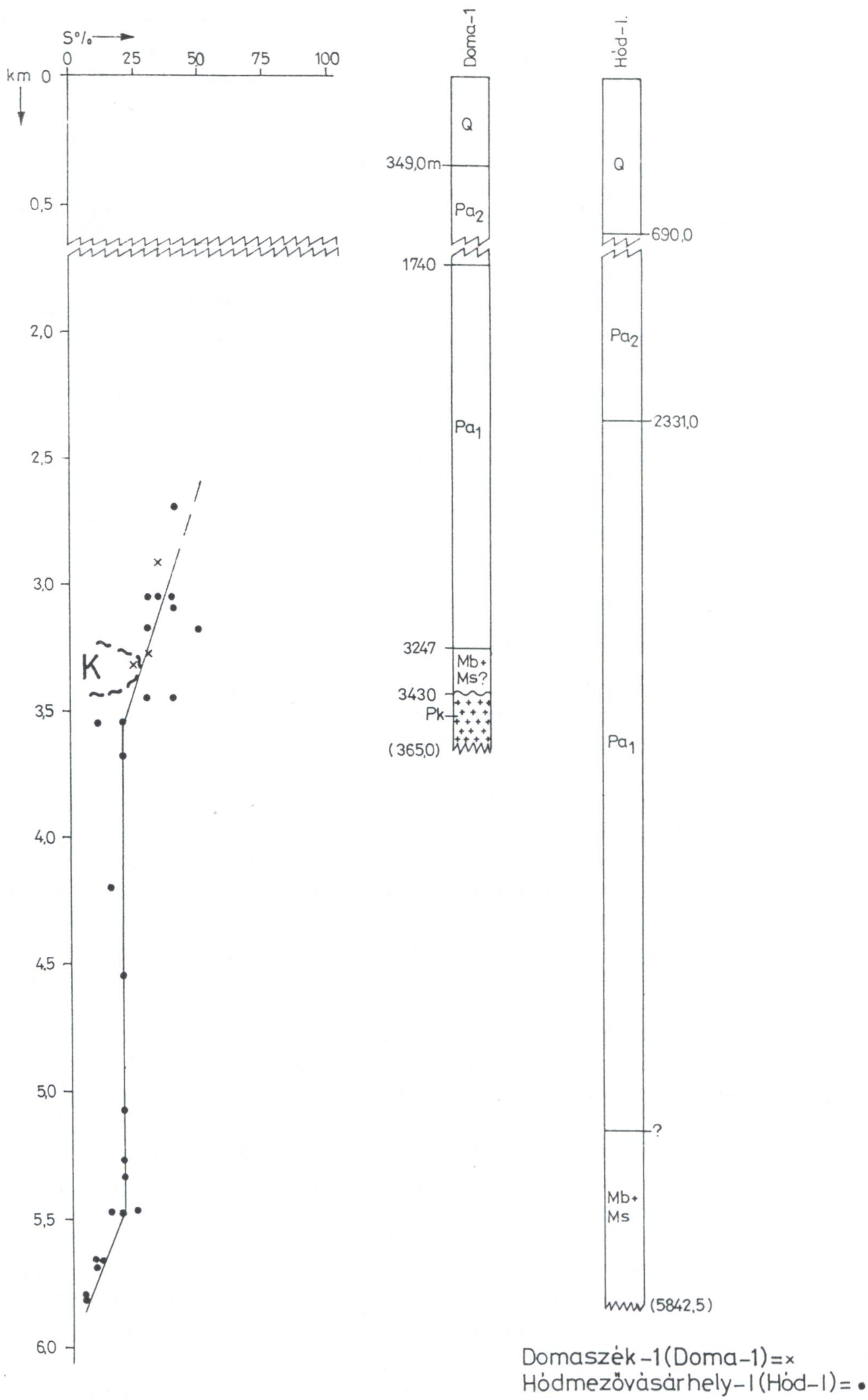

Fig. 15. Makó Trough, Szeged Basin.

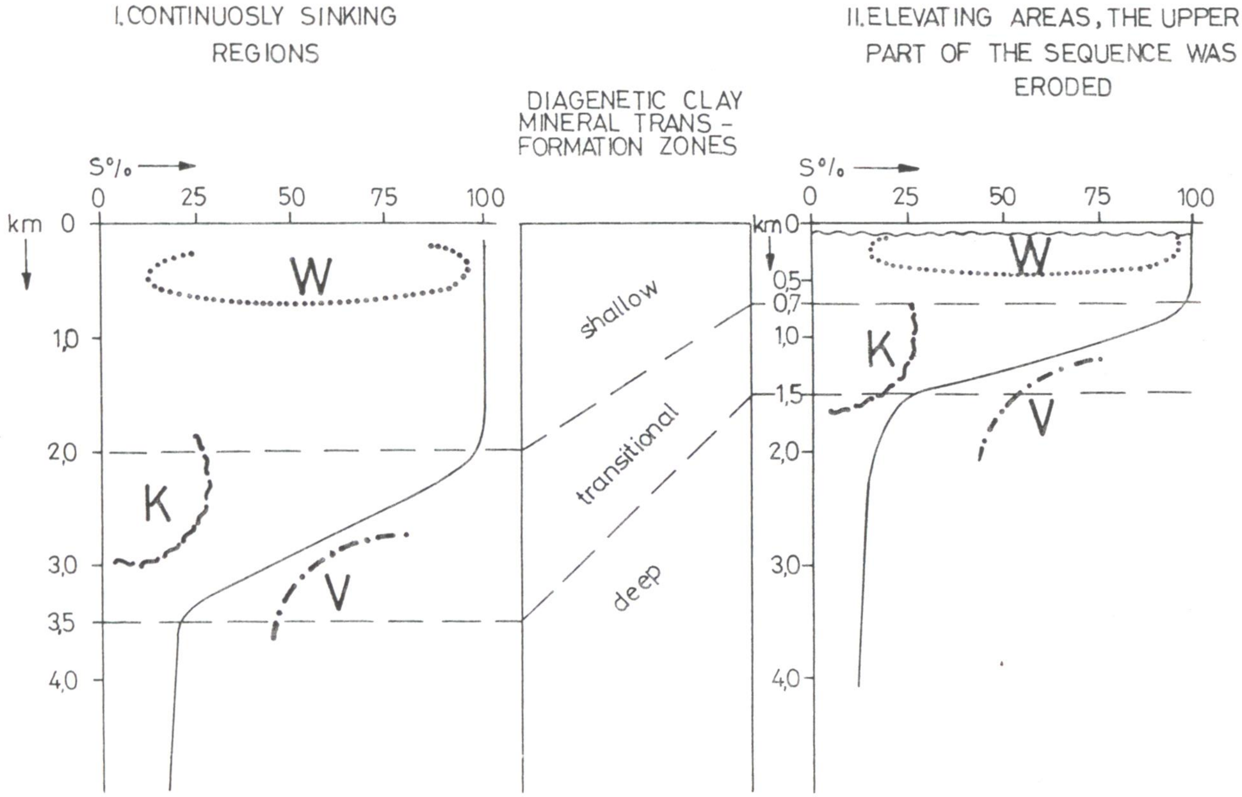

Fig. 16. Summary of S % versus depth diagrams: Variation of the smectite proportion (S %) in function of depth, genetic type of clay and basin evolution in Neogene formations of the Pannonian Basin.

The thick (more than 800 m) Miocene sequence of the Hejőszalonta-1 (Hejő-1) borehole is characterized by the alteration of acidic volcanic tuff and clayey marl (Fig. 10). Here, from a depth of ca. 0.7 km redeposited andesitic sand samples were studied in which S = 100 %.

Neogene basins with thick Miocene volcanic formations (Figs. 10, 11)

In the Hidasnémeti-1 (Hn-1) borehole (Hernád Valley, Fig. 10), the Lower and Middle Badenian sediments above the Karpatian dacite tuff contain redeposited volcanogenic sediments (S = 100 %). The degree of diagenesis can be determined in the Upper Badenian and Sarmatian terrigeneous clastic formations above ca. 1.2 km depth. The transformation starts at a depth of ca. 0.7 km.

The Detk-I borehole drilled in the S foreland of the Mátra Mts. cut across a very thick Karpatian and Badenian volcanic sequence (Fig. 10). The overlying Badenian and Sarmatian sedimentary sequences also contain large amounts of redeposited volcanic material in which, as compared with the overlying terrigeneous layers, S is relatively high, 70-90 %. Only the terrigeneous Pannonian samples above 0.7 km can be evaluated from the point of view of diagenesis (except for two samples that may have been affected by weathering). Here it seems that the transitional zone starts very soon, at a depth of about 0.3 km. From the top of the Pannonian, however, a sequence of several hundred meters thickness was eroded. According to Jámbor (1989; Tab. 1), the erosion of the Upper Pannonian is characteristic of a large part of the basin except for certain, continuously subsiding areas. It seems that here, in the S foreland of the Mátra Mts. the extent of the uplift was especially strong.

The Jászladány-I (Jász-I) borehole, in the central zone of the Jászság Depression, explored a more than 400 m thick pyroclastic sequence belonging to the Karpatian dacite tuff formation that was subsided to great depth (Fig. 10). Directly upon the pyroclastic sequence thin Badenian shales of terrigeneous origin contain illite/smectites with S = 25 %. This value is characteristic of the deep burial zone of diagenesis. In the pyroclastics immediately beneath this (lower part of core No. 2) S = 0–20 % while ca. 180 m deeper (core No. 4) S is only 0–10 %. These volcanogenic illite/smectites are typical neoformational minerals. The character of the mixed-layer minerals is similar to those of the meta-bentonites.

The region around Paks (central Hungary, see Fig. 11) is another Miocene volcanic centre. In the Paks region the pre-Tertiary basement is significantly deeper than in the surroundings (deeper than 1.5 km). These boreholes are situated close to each other and their layer sequences are practically identical.

The Paks-2 borehole has the longest and thus the most completely sampled sequence starting with Ottnangian (?)- Karpatian (?) sediments, and above this, between 1089 and 1542 m, a stratovolcanic sequence of the same age. The Badenian and the thin Sarmatian stages are represented by typical volcano-sedimentary rocks and accordingly, by S = 100 % values. The data point at 1.03 km depth and S = 30 % represent a kaolinite-rich clay marl which was deposited in an alluvial environment. Though being within the volcano-sedimentary complex, this rock presumably belongs to the kaolinite-rich type.

In the Paks-4/a and -4/c boreholes Badenian and in the Paks-3 borehole the lower part of the Lower Pannonian are volcanogenic. Above this the Pannonian sequence of the Paks-3 borehole as well as that of the Paks-4/a, -4/b and -4/c boreholes consists of terrigeneous clastic rocks. In its upper part the influence of the Upper Pannonian weathering may be supposed to be shown by the oxidized character of these sediments. Here the S % values are, however, only slightly lower than those in the non-oxidized rocks of the same depth interval.

No exact degree of diagenesis can be determined even from the terrigeneous clastic rocks because of the high scattering of the S % values. An approximate trend line can be drawn, which shows that at Paks the diagenetic transformation of illite/smectites starts at about 0.5 km depth. The shallow position of the transformation may be explained by the erosion of the upper part of the Pannonian sequence.

NE Tisza Basin (Fig. 12)

Two boreholes were studied, Tiszapalkonya-1 (Tp-1) drilled in the E part of the Jászság Basin, and Mikepércs-1 (Mp-1) drilled at the N margin of the Derecske Depression. Since these boreholes were not very deep (2.0 and 1.4 km, respectively), they reached only the upper part of the Neogene sequence, mainly the Upper Pannonian lignite formation and the discordantly overlying variegated clays. In both boreholes the Quaternary is of significant thickness. There is another unconformity between Upper Pannonian and Quaternary. The Quaternary formations and the upper part of the lignite-bearing series may have been influenced by terrestrial weathering as it can be supposed on the basis of facies, yellow colours and goethite contents. Fields of oxidized samples are shown in Fig. 12 (marked with "W" in the depth intervals 0–0.3 km and 0.55–0.8 km). The samples have very variable composition in respect of S % but the same wide range of variability can also be observed in the non-oxidized samples of the same depth interval.

An interesting feature of these sequences is that there are stages in the lignite formation that contain much redeposited volcanic material. In these layers illite/smectites have higher S % values than those in normal terrigeneous sediments.

In the Mp-1 borehole the transformation begins ca. below 0.5 km but it cannot be determined exactly because of the discintinuity of the data. In the Tp-1 borehole the transitional zone starts already below 0.3 km. The transition ends at about 0.8 km and beneath this the S ≈ 30 % value remains constant till the bottom of the borehole (2.0 km). The shallow depth of the diagenetic transformation in the Tp-1 and Mp-1 boreholes can be due partly to the erosion of the upper part of the Upper Pannonian variegated clay formation. Considering the depth of the transitional zone Tp-1 is more uplifted than Mp-1 by several hundreds meters.

Nagykunság Basin (Fig. 13)

In the Nagykunság Basin we have a relatively continuous Lower Pannonian sample series from the Örménykút-1 (Örm-1) borehole (2.1-3.9 km). This series is well complemented by some other data from the Kunszentmárton-1 (Kunszt-1), Endrőd-3 (En-3) and Gyoma-1 (Gyo-1) boreholes. In the En-3 borehole, the uppermost studied sample at 1050 m was probably influenced by terrestrial weathering. In other boreholes of the Nagykunság Basin the base of the Upper Pannonian variegated clay formation also reaches greater depths (e.g. Örm-1: 1052 m, Kunszt-1: 800 m). This formation may have been influenced by subaerial weathering.

In the Nagykunság Basin the transitional zone ends at a depth of 1.6 km and there is only a small change in the 1.6 to 3.1 km interval where S decreases from 35 % to 20 %.

Békés Basin (Fig. 14)

In the Békés Basin a continuous sample series was examined from the Doboz-I borehole which cut across a more than 4 km thick Neogene + Quaternary sequence (Viczián 1992). In the section of the borehole above ca. 1.9 km subaerial weathering can be supposed on the basis of the presence of Quaternary sediments and of the Upper Pannonian variegated clay formation. Some samples even in the underlying Upper Pannonian marly siltstones contain goethite and are of yellow to brown colour. In this interval many samples show considerable variability in the S % values and the average of S % is clearly lower than 100 %.

Below the depth of ca. 1.9 km until 3.3 km the transitional zone of the diagenesis of illite/smectite can be found. It can be divided into two parts:

- 1.9–2.8 km: S is relatively constant, the high variation within the samples decreases downwards;
- 2.8–3.3 km: S decreases to 25 %, there is more uniform composition within the samples.

From the depth of 3.3 km downwards the composition is stabilized at about S = 25 % (deep burial zone).

Two samples fall into this deep burial zone from the Sarkadkeresztúr-ÉNY-2 (Sark-ÉNY-2) borehole in which S = 15 %, that is by ca. 10 % less than in the same depth of the Doboz-I borehole. The lower S value is probably due to the much older, Early (?) or Middle Miocene age, while in the Doboz-I borehole, at the same depth the age is only Early Pannonian.

The clayey rocks of the Pusztaottlaka-1/P (Po-1/P) borehole belong to the Upper Pannonian variegated clay formation and Quaternary (Viczián 1982). Values of S = 60–90 % agree well with the data obtained from the Doboz-I borehole from the same formations. These S % values were probably influenced by terrestrial weathering.

Makó Trough and Szeged Basin (Fig. 15)

The so-far deepest borehole in Hungary, Hódmezővásárhely-I (Hód-I) was drilled in the axial line of the Makó Trough. We have measurements from its 2.7–5.8 km (Lower Pannonian-Badenian) section. The S values are rather scattered till 3.5 km but it is visible that we are near the lower end of the transitional zone of diagenesis. Below this depth, till about 5.5 km the composition is stabilized at about S = 20 %. In the lowermost part of the borehole, between 5.5 and 5.8 km the value of S decreases to 5–10 %.

The Domaszék-1 (Do-1) borehole was drilled W of the relatively elevated Algyő structure. Data from this borehole fit well into the trend of the lower part of the transitional zone observed at Hód-I (S = 25–35 %). In this borehole a single Badenian kaolinitic sample displays somewhat lower S value than two other samples.

Discussion of the results

Kaolinite-rich sediments

In fine-grained kaolinite-rich sediments either there is no swelling clay mineral at all, or S % is low (usually < 30 %). In a few samples of transitional composition between terrigeneous and kaolinitic types S = 50–60 % can be found (e.g. Figs. 2, 5 and 6). Usually S % values in kaolinite-rich layers are lower than those indicated by the trend line of the terrigeneous clastic formations (e.g. Figs. 2, 5, 6 and 8). Since these formations are generally at the bottom of the layer sequences, one should be careful attributing the sudden decrease of S % to diagenesis. In case of gradual decrease of S % with depth, however, both factors may be responsible for the low S % values.

Kaolinite in these sediments may have various geneses. Kaolinitic basal formations are widespread in Transdanubia and in the Danube-Tisza Interfluve in different stratigraphic levels (Ottnangian to Lower Pannonian), depending on the starting time of the Neogene transgression. Exceptionally, kaolinite may also be formed by hydrothermal activity (Ig-7, Fig. 6). Kaolinite-rich horizons presumably formed by rapid redeposition of kaolinitic material can also be found within the sedimentary sequences (e.g. at the boundary of Lower and Upper Pannonian, see Figs. 2 and 5). Rarely, kaolinite may be enriched in Upper Pannonian lacustrine carbonate rocks (Ig-7, Fig. 6). In all these genetic types kaolinite is well or medium-well crystallized (width of the 001 reflection at half height: 0.27–0.50° 2Θ).

Volcanogenic sediments

The volcanogenic sediments originally contained smectite with S = 100 %. In the shallow burial zone of diagenesis both terrigeneous and volcanic sedimentary rocks have S % values practically equal to 100 % (e.g. Gf-1 in Fig. 5, Mb-1 in Fig. 7 and Sh-16/a in Fig. 8). There is a difference in the crystallographic character of the smectite minerals which is, however, not expressed in the numerical value S = 100 %. (Perhaps in the case of terrigeneous sediments S = 80–100 % would be more appropriate.)

In the transitional zone of diagenesis S % values of volcanogenic sediments are consistently higher than those of terrigeneous sediments in the same depth. This was observed in many boreholes in various parts of the Pannonian Basin, e.g. in Sz-II (Fig. 2), Gyék-I (Fig. 4), Ib-I (Fig. 5), Hn-1 and Detk-I (Fig. 10), Paks (Fig. 11), Mp-1 and Tp-1 (Fig. 12). In some cases the data points of volcanogenic and terrigeneous sediments seem to fit into the same trend line or only slight differences may be observed. This was the case in the boreholes of the Zagyva Graben (Sz-2/a and Nks-I in Fig. 9). This may be due to relatively low proportions of volcanogenic material in the sediments.

In the deep zone of burial the differences between the S % values of the individual rock types disappear, or illite/smectites in volcanogenic formations may be even less expanded than in terrigeneous clastic ones (e.g. Jász-I, Fig. 10). All these observations show that smectites formed from volcanic rocks are more stable during diagenesis and they change in the shallow and transitional zones slower than mixed-layer structures found in terrigeneous clastic sediments.

The redeposited volcanogenic sedimentary material is especially characteristic of the Karpatian, Badenian and Sarmatian sediments covering the volcanic sequences or alternating with them, at certain places even a part of the Lower Pannonian is of the same type (e.g. Sz-2/a, Paks-3). In the N part of the Great Hungarian Plain, even the Upper Pannonian lignite-bearing series may contain much redeposited volcanogenic material (e.g. Tp-1, Mp-1, Sz-2/a).

Effects of Upper Pannonian-Quaternary weathering

The smectite ratio (S %) in the upper part of the Upper Pannonian and in the Pleistocene varies within a wide range (20 to 100 %). As these formations are alluvial and continental deposits, it can be supposed that subaerial weathering and soil forming processes played an important role in the determination of the composition of illite/smectites. Some sort of illitization process may have taken place. The exact nature of the process, however, is not yet clear. One has to consider that this period of time is characterized by extreme climatic variations. From the present study it seems to be sure that there is no significant difference between S % values found in goethite-bearing, oxidized and non-oxidized sediments, the variability of S % can be equally high in both cases.

Effects of uplift and erosion

After the Pannonian (Rhodanian and Romanian phases) the large part of Transdanubia (except for the Little Hungarian Plain), the central and W part of the Danube-Tisza Interfluve, and areas adjacent to the North-Hungarian mountain range were significantly uplifted and a several hundred meters thick sediment column was eroded from the upper part of the Upper Pannonian (Rónai 1974; Jámbor 1989). As a result of the uplift and erosion the already formed depth zones of clay mineral diagenesis were also "uplifted". The upper boundary of the transitional zone can be found in these areas at depths of several hundred meters, and the lower one at a depth of 1.5 to 2.0 km. Examples are Kas-2 (Fig. 6), Hn-1 and Detk-I (Fig. 10), Paks (Fig. 11), Tp-1 and Mp-1 (Fig. 12).

Conclusions

The conclusions are summarized in Fig. 16 in which the possible fields of the various genetic groups in the S % vs. depth diagram and the effect of two ideal cases of basin evolution are shown. Continuous curves represent the course of the diagenetic transformation in rocks of terrigeneous clastic origin. Depth values of diagenetic zones are only tentatively shown as they are also highly dependent on the geothermal conditions.

References

Balogh K. & Bérczi I., 1991: Diagenesis. In: Balogh K. (Ed.): *Sedimentology I. Akadémiai Kiadó*, Budapest, 373–400 (in Hungarian).

Burst J.F., 1969: Diagenesis of Gulf Coast clayey sediments and its possible relation to petroleum migration. *AAPG Bull.*, 53, 1, 73–93

Csíky G., Erdélyi Á., Kárpáti-Radó D. & Kőrössy L., 1987: Pannonian s.l. formations of Hungary. 4 sheets, 1:500,000. *Hung. Geol. Inst.*, Budapest (in Hungarian).

Dank V. & Jámbor Á., 1987: Allgemeine geologische Merkmale der Ablagerungen des Pannonien s. str. (Kunság-Stufe) in Ungarn. *Annals of Hung. Geol. Inst.*, 69, 9–25 (in Hungarian).

Franců J., Müller P., Šucha V. & Zatkalíková V., 1990a: Organic matter and clay minerals as indicators of thermal history in the Transcarpathian depression (East Slovakian Neogene Basin) and the Vienna Basin. *Geol. Carpathica*, 41, 5, 535–546.

Franců J., Šucha V., Viczián I. & Johns W.D., 1993: Expandability as related to diagenesis and geothermal conditions in the West Carpathian-Pannonian region. *Geol. Soc. Amer., North-Central Section, 27th Annual Meeting, Rolla, Missuori. Abstracts with Programs* 25, 3, 17.

Franců J., Viczián I. & Šucha V., 1990b: Illite-smectite expandability as diagenetic temperature indicator in the Neogene basins of the Carpathian and Pannonian region. *Abstracts, 11th Conf. Clay Min. Petr., České Budějovice.*

Hámor G., 1985: Geology of the Nógrád-Cserhát area. *Geologica Hung., Ser. Geol.*, 22, 1–307 (in Hungarian).

Hámor-Vidó M. & Viczián I., 1993: Vitrinite reflectance and smectite content of mixed-layer illite/smectites in Neogene sequences of the Pannonian Basin, Hungary. *Acta Geol. Hung., Ser. Geol.*, 36, 2, 197–209.

Hetényi M., 1992: Organic geochemistry and hydrocarbon potential of Neogene sedimentary rocks in Hungary. *J. Petroleum Geol.*, 15, 87–96.

Jámbor Á., 1980: Palaeogeographically significant Pannonian facies units in and around the inselbergs of the Hungarian part of the Carpathian Basin. *Földt. Közl.*, 110, 3–4, 498–511.

Jámbor Á., 1989: Review of the geology of the s. l. Pannonian formations of Hungary. *Acta Geol. Hung.*, 32, 3–4, 269–324.

Kőrössy L., 1988: Hydrocarbon geology of Zala Basin, Hungary. *Ált. Földt. Szemle*, 23, 3–162 (in Hungarian).

Kőrössy L., 1989: Hydrocarbon geology of Dráva Basin, Hungary. *Ált. Földt. Szemle*, 24, 3–122 (in Hungarian).

Kőrössy L., 1990: Hydrocarbon geology of SE Transdanubia, Hungary. *Ált. Földt. Szemle*, 25, 3–53 (in Hungarian).

Kübler B., 1984: Les indicateurs des transformations physiques et chimiques dans la diagenese. Température et calorimétrie. In: Lagache M. (Ed.): *Thermométrie et barométrie géologiques. Soc. Franc. Min. Crist.*, vol. 2, Ch. 14.

Müller G., 1967: Diagenesis in argillaceous sediments. In: Larsen G. & Chilingar G.V. (Eds.): *Diagenesis in sediments. Elsevier*, Amsterdam, 127–177.

Révész I., Szabóné Drubina M., Tóth K., Viczián I. & Balogh K., 1991: Pelites deposited in water. In: Balogh K. (Ed.): *Sedimentology II. Akadémiai Kiadó*, Budapest, 181-227 (in Hungarian).

Rónai A., 1974: Size of Quaternary movements in Hungary's area. *Acta Geol. Hung.*, 18, 1-2, 39-44.

Royden L.H. & Horváth F. (Eds.), 1985: The Pannonian Basin - a study in basin evolution. *AAPG Memoir*, 45, 1-394.

Srodon J., 1979: Correlation between coal and clay diagenesis in the Carboniferous of the Upper Silesian Coal Basin. *Int. Clay Conf. Oxford, 1978*, 251-260.

Srodon J., 1980: Precise identification of illite/smectite interstratifications by X-ray powder diffraction. *Clays and Clay Miner.*, 28, 401-411.

Šucha V., Elsass F. & Vass D., 1994: Diagenetic illitization of smectite from the shales of the Danube Basin. *Acta Geol. Hung*, 37, 1-2.

Šucha V., Kraus I., Gerthofferová H., Peteš J. & Sereková M., 1993: Smectite to illite conversion in bentonites and shales of the East Slovak Basin. *Clay Miner.*, 28, 243-253.

Tanács J. & Rálisch E., 1990: Worms-eye map of Cenozoic formations of Hungary, 1:500,000. *Hung. Geol. Inst.*, Budapest.

Tanács J. & Rálisch E., 1991: Explanations to the worms-eye map of Cenozoic formations of Hungary (1:500,000). *Hung. Geol. Inst.*, Budapest (in Hungarian).

Viczián I., 1970: Faziesprobleme im ungarischen Pannon auf Grund des Tonmineralgehalts. *Ber. Dtsch. Gesell. Geol. Wiss., B. Mineral. Lagerstättenforsch.*, 15, 1, 75-82.

Viczián I., 1982: An expanding mixed-layer clay mineral in Upper Pannonian to Pleistocene fine-grained clastic rocks of the borehole Pusztaottlaka I/P (SE Hungary). *Annual Rept. of Hung. Geol. Inst. 1980,* Budapest, 449-456 (in Hungarian).

Viczián I., 1984: Zur Ausgangsphase der diagenetischen Mineralreaktion Illit/Smektit - Illit. *Z. Geol. Wiss.*, 12, 469-475.

Viczián I., 1985: Diagenetic transformation of mixed-layer illite/smectites in deep zones of the Pannonian Basin (Hungary). *5th Meeting of the European Clay Groups, 1983, Prague*, 135-140.

Viczián I., 1992: Clay minerals of a thick sedimentary sequence in SE part of the Pannonian Basin (Hungary). *Geol. Carpathica, Ser. Clays*, 1, 27-30.

Viczián I., 1994: Smectite-illite geothermometry. *Földt. Közl.*, 124, 3, 367-379 (in Hungarian).