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GEOLOGICAL CONDITIONS OF THE UPPER PANNONIAN OIL-SHALE DEPOSIT RECOVERED IN THE BALATON HIGHLAND AND AT KEMENESHÁT

Á. JÁMBOR and G. SOLTI

ABSTRACT

In the course of systematic geological investigations carried out in the Transdanubian Central Mountains, in the shallow borehole west from Pula (Balaton Highland area), immediately under the surface, an about 20 m thick, organic-rich laminated formation occurred which, according to examinations, proved to be a characteristic oil-shale deposit. During geological studies we came to the conclusion that this oil-shale was deposited in the basalt crater lakes developed in Pliocene time, when special conditions enabled the formation of this characteristic sedimentary facies. After having disclosed these circumstances of development, in the environs of the villages Kemeneshát and Gérce, in the basalt area of the Little Hungarian Plain's central part, in an earlier basalt crater, about 70 m thick oil-shale deposit was recovered.

May be that, a systematic examination of Hungary's sediment series will result the recovering of further oil-shale deposits.

1. PRELIMINARIES

In Hungary, oil-shale prospecting has no traditions. It was I. VITÁLIS [1946] who mentioned for the first time a 50-cm-thick "bituminous brown coal" intercalation in Miocene volcanites in the southwestern part of the Mátra Mountains and published the respective results of chemical analyses:

Moisture	2.74%	Moisture	3.79%
Ash content	38.16%	Semi-coke	56.70%
Heat of combustion	4816 kg/cal	Tar	26.7 %
Calorific power	4551 kg/cal	Water as a decomposition product	2.11%
Sulphur content	7.51%	Gas and losses	11.4 %
Hydrogen content	4.6%		
Carbon content	34.71%		
O+N	12.24%		

Regarding the precise composition of the rock characterized by I. VITÁLIS, only the work of E. SZÁDECZKY-KARDOSS and P. TAKÁCS can be taken for a basis, who came to the conclusion that this was no brown coal but a "boghead rock", including alginite, eualginite, bituminite, fusite and their minerals. The fossil algae were mainly of *Botryococcus* origin.

To the end of 1973, in the frame of the geological reambulation of the Transdanubian Central Mountains, a survey-borehole was sunk to reveal the existing stratigraphic conditions on the southern margin of Mt. Kab, in the northern foreland of the Upper Triassic dolomite fault block, northwest of Pula. In borehole Put-1, overlying the Pliocene basalt, a 38.0 m thick, when dry greenish grey, very fine-grained, organic-rich and rather tuffitic formation, loose, light laminated and in wet state dark green, was traversed. Derivatographic analyses (M. FÖLDVÁRI) gave a striking result: the rock was burnt, smelling disgustingly of stearine and leaving behind very little ash. On our request, DR. F. GÓCZÁN, analyzing the rock, found that the residual material obtained after palynological treatment, consisted mostly of *Botryococcus braunii* KÜTZ., planktonic brown algae of microscopic size.

On the basis of these informations, the Research Department of the Central Geological Office of Hungary ensured significant funds for further investigations of the above formation. After carrying out this activity, we turned to the Mining Research Institute. We feel greatly indebted to DR. P. TAKÁCS, Head of Department at the Mining Research Institute, for the informations he gave us, which has had an incentive influence on further research. He called our attention to the fact, that these oil-shale deposits, if available in substantial amount, would enable the development of a significant oil industry, *i. e.* organic chemistry.

In their expert opinion, P. TAKÁCS, MRS. J. ARATÓ and MRS. L. BELLA maintained firmly that, from technological view-point, the oil-shale found in the vicinity of Pula is a characteristic one and as such, it can be used as starting material on distillation for oil.

Beside technological analyses, geological mapping results of the Pula region were estimated, too, and after their comparison with the simultaneously obtained laboratory data, the following geological, paleogeographical and geohistorical and genetical conclusions could be deduced.

2. GEOLOGICAL CONDITIONS OF THE OIL-SHALE DEPOSITS AT PULA

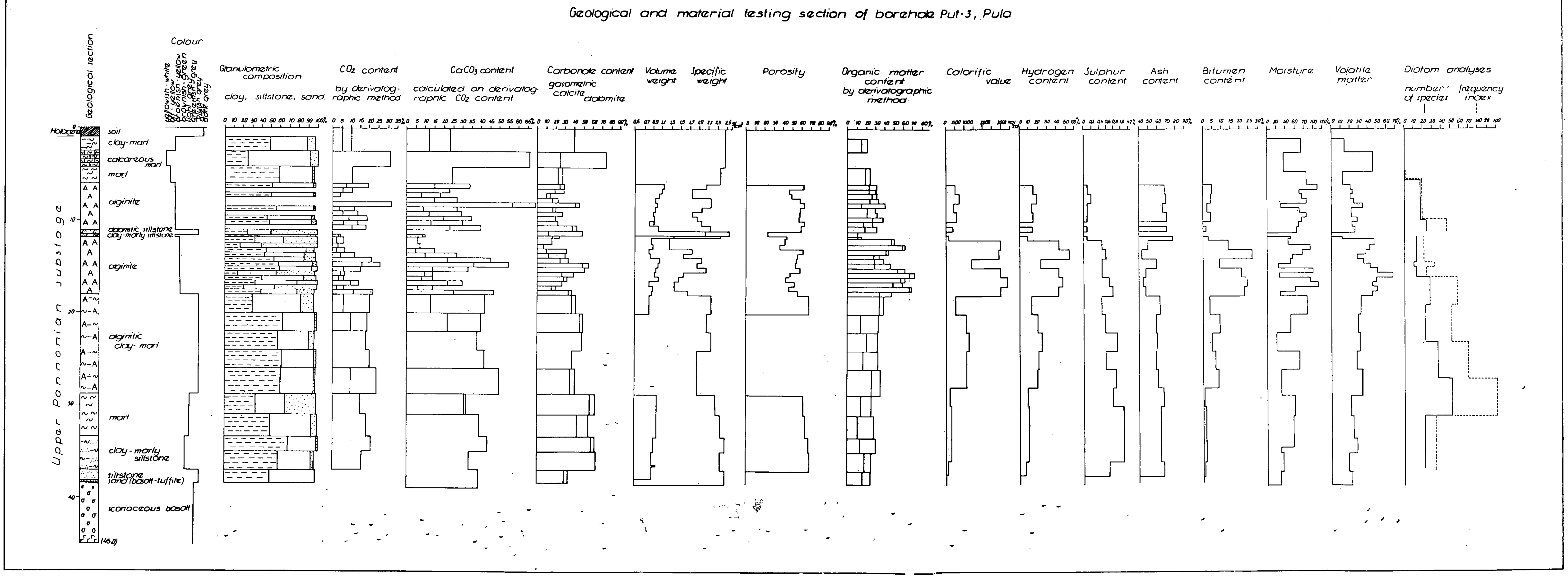
About 500 m west of Pula, the oil-shale sequence covers the second basalt lava flow with local unconformity *(Fig. 1)*. Drilling in 1973 proved the presence of sedimentary rocks (sand, siltstone, calcareous silt) between the second and third basaltic beds.

As a consequence of gas emanations, the texture of the uppermost 8 metres of the basalt, underlying the oil-shale, became scoriaceous. This member is followed by a 34.2-m-thick (4.2 m to 38.4 m) Upper Pannonian formation, called "oil-shale member of Pula" which can be subdivided into four parts. It should be remarked that in Hungary this was the first case when above the basalt no Pleistocene sequence occurred.

The lowermost group of strata starting with the 5 cm of basaltic sands, contains siltstones, clay-marls with siltstone and one marl bed. The second group consists of alginitic clay marls, the third one of clay marls with alginite, and with dolomitic intercalations, while the fourth, *i. e.* youngest one, is made up of marly, and clay-marl strata. The whole member is characterized by greyish-green, green-grey,that is, by reductive colours, and a more or less thinly laminated structure.

In borehole Put-3 *(Fig. 1)* directly above the basalt, 5.0 cm of arched and cross-laminated sand was deposited, containing light-grey basalt grains of 0.1—2.0 mm diameter, from which the oldest pelitic rock of the "oil-shale member of Pula" has developed with alternating changes in lithology, forming a 1.3-m-thick layer which on the whole may be denoted as siltstone (Photo 1, Plate I). In the lowermost 10 cm of the siltstone 5 arched cross-laminae of basaltic sands of 30° inclination and each 1 cm thick, are intercalated. The siltstone layer consists of differently colour-

Fig. 1.



Geological and material testing section of borehole Put-3, Pula
Geological section
Colour
Granulometric composition
clay, siltstone, sand
CO2 content
by derivatographic method
CaCO3 content
calculated on derivatographic CO2 content
Carbonate content
gasometric
calcite
dolomite
Volume weight
Specific weight
Porosity
Organic matter content by derivatographic method
Calorific value
Hydrogen content
Sulphur content
Ash content
Bitumen content
Moisture
Volatile matter
Diatom analyses
number of species
frequency index
Holocene
soil
clay-marl
calcareous marl
marl
alginite
dolomitic siltstone
clay-marly siltstone
alginite
alginitic clay-marl
marl
clay-marly siltstone
siltstone
sand (basalt-tuffite)
scoriaceous basalt
Upper Pannonian substage
(45.0)

red (grey, light-grey, off-white, greenish grey) and distinctly horizontal and parallel laminae with an average thickness of 1 to 30 mm (mostly 5 mm and maximum 300 mm and made up of various materials. The material of each lamina was not separately analyzed, nevertheless even after examination in the field it was evident that the lighter parts were richer in unconsolidated calcareous silt.

Above the siltstone (between 28.8 m and 37.0 m) follows a light greenish-grey, laminated siltstone member with clay marl which we divided into two parts at 33.4 m, the upper part being richer in lime. This rock consists of 1 to 150 mm, more frequently 2 mm thick laminae (Photos 2 and 3 Plate I). Lithologically, three different kinds of laminae can be distinguished. The siltstone laminae are 5 to 30 mm thick, consisting of readily identifiable, very fine grains of basalt tuff. The yellowish laminae are calcareous, while the greenish ones contain much clay but both are thinner than the siltstone layers. In the more calcareous laminae of parallel stratification, there are more or less compressed, hollow fragments of *Chara* stem sometimes in rock-forming quantity. These fossil debris are only of some tenth of millimetres size, their length attains 5—10 mm, and they are unoriented along the bedding plane. Limpid mono- and bivalves of Ostracoda situated parallel to stratification are not rare either. The inner part of bivalves is empty. At 36.1 m some carbonized leaves occur.

The layers, except for the 32.1 to 32.4 m interval, are completely horizontal. The afore mentioned interval still seems to have been produced by subsiding mud still in plastic state, which may be proved not only by the seemingly chaotic plication but also by its intraformationally brecciated texture (Photo 4, Plate II) as well.

This layer is followed by a properly alginitic subdivision (from 6.0 to 28.8 metres), quite different due to its slate green colour, differently laminated jointing and striking lightness, and in the fresh state (that is, 3—10 months after being exposed) to its characteristic smell reminding mainly of bensene; and, normally, they are conspicuous for being able to be ignited and for the flexibility of their laminae.

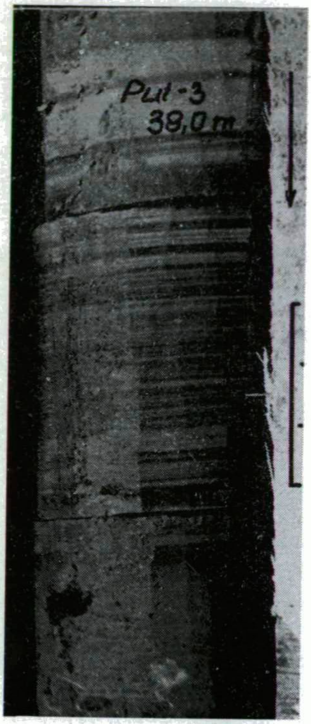
The sediments between 6.0 and 28.8 m are similarly laminated but differ from the underlying rocks, including only two predominant types of laminae. The laminae are easily separable and thinner, with a thickness only of 0.1 to 4.0 mm (Photo 6 Plate II). One type is roughly green in colour and, viewed under hand magnifying glass, it seems to be made up mainly of green and pale yellow, isolated or grouped globules. The other one with its pale green or greyish yellow colour is more calcareous. The laminae between 18.0 and 28.8 m include a significant amount of pelitic component.

In the alginitic sediments the siltstone with clayey marl (11.6 to 11.8 m) is overlain by siltstone (11.1 to 11.6 m) the cement of which is light grey dolomite. This latter is made up locally by fine sand, the grains of which predominantly result from erosion of basaltic tuff, however, some muscovite laminae of metamorphic origin can be observed, too.

The layers between 18.0 and 28.8 m within the alginitic subdivision, are horizontal, whereas from 6.0 to 18.0 m they show an inclination of 5—8°, perhaps being the consequence of a rearrangement after mud-subsidence. Between 15.5 and 15.6 m plication due to mud subsidence and/or intraformational brecciation could be observed (Photo 5, Plate II).

Between 6.0 and 11.1 m, the laminae after drying out — in contrary to those of the other parts in the sequence — are curved up as a burnt paper. At the same time, following deposition, along some bedding planes several arched desiccation

PLATE I



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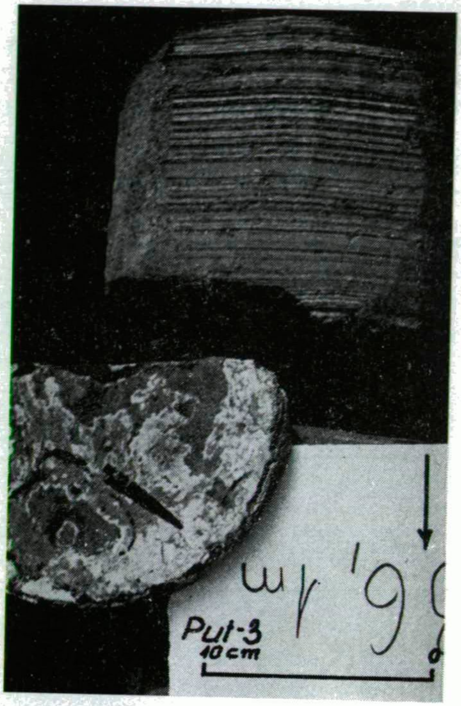

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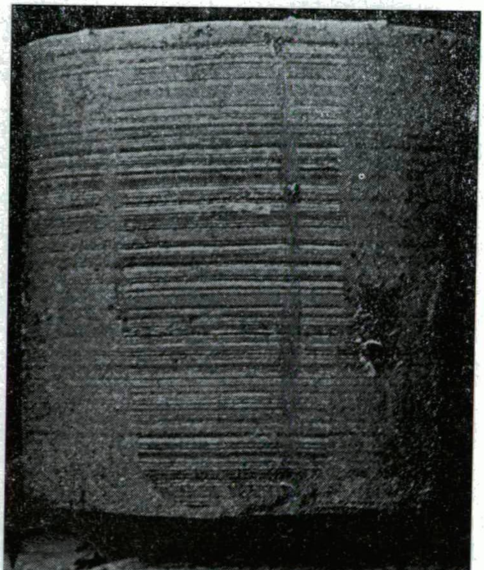

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cracks were formed. From 7.4 to 7.8 metres, again intraformational breccia can be observed, at 10.5 m with a somewhat compressed *Diptera* (?) fragment.

In the hanging wall of the alginitic layers (1.0 to 4.2 m) the lamination of the laminated marl, calcareous marl and clayey marl is not so distinct as in the alginitic part, since the quantity of the loose pelitic calcareous mud within the laminae — though significantly fluctuating — roughly surpasses that of clayey components. Thus we may find white, pale yellow, yellowish grey and grey laminae, respectively. In the lower part of the hanging wall (about 2 m in extension) many carbonized small plant-stems and herbaceous-like leave debris can be observed on the surface of some laminae. In the upper part of these sediments the slope is 1—2°, while in the lower part 5—8°. At last it is covered by a dark grey, 1 m thick, humus-soil.

Geological data have made it obvious that the alginitic shales, i. e. oil-shales, developed under rather special paleogeographical conditions, at the same time when the upper part of the *Congeria balatonica* sediments were deposited. Their present-day local stratigraphic situation is shown in *Fig. 4* As an evidence of borehole Put-1 the basalt of the foot-wall may be regarded as the filling of a crater channel. Taking into consideration the sequence found in borehole Kpt-1 (at Kapolcs) this basalt may be identified with the second basalt layer of the above sequence, while the immediately underlying basaltic sand may be regarded as a deposited layer of the 2nd basalt-tuff. The sediments accumulated simultaneously with alginitic rocks are known from boreholes Put-2 and Vgt-1 (Vigántpetend) in the region of the Tálod forest. Here the lower layers of freshwater limestone at Nagyvázsony can be correlated with the rocks overlying the oil-shale (1.0—4.2 m) in borehole Put-3. At the basis of limestone at Nagyvázsony, several dm thick bentonitized sediments (corresponding to the 2 nd basalt-tuff) are followed by a bluish-grey clayey marl, underlain in borehole Put-2 by alternating sequence of clayey marl, clayey siltstone and carbonaceous clay including brackish and freshwater fauna correlating well with the Upper Pannonian *Congeria balatonica* beds. These in turn, developed by continuous sedimentation from the *Congeria ungulacaprae* beds. Although the lower strata of freshwater limestones at Nagyvázsony are of the same age as the alginitic series of Pula, their development is different, because the latter was sedimented under special circumstances in a basalt crater lake.

In the region of the Tálod-forest also the hanging wall of the freshwater limestone of Vázsony is known. It is overlain by bentonitized sediments of the 3rd basalt-tuff, which, in turn, is covered by 1—8 m thick 3rd basalt. Then it is followed by Würmian loess.

Explanation of Plates

Sedimentologic phenomena of the oil-shale deposits in borehole Put-3 at Pula

Diameter of the drill cores is always 10 cm. The arrows show the direction of the foot-wall.

Plate I

Photo 1 Laminated siltstone at the beginning of the sequence
Photo 2 2nd layer: clay marly siltstone including plant debris
Photo 3 2nd layer: lamination of the clay marly siltstone (36.0 m)

Plate II

Photo 4 Mud subsidence in laminated marl
Photo 5 Mud subsidence in laminated alginite
Photo 6 Laminated alginite (9.2 m)

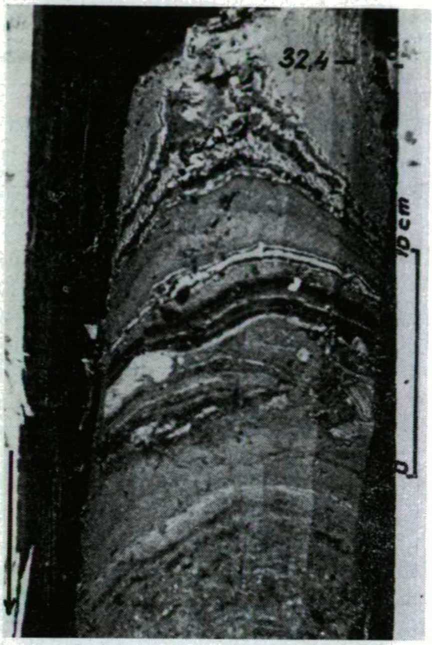

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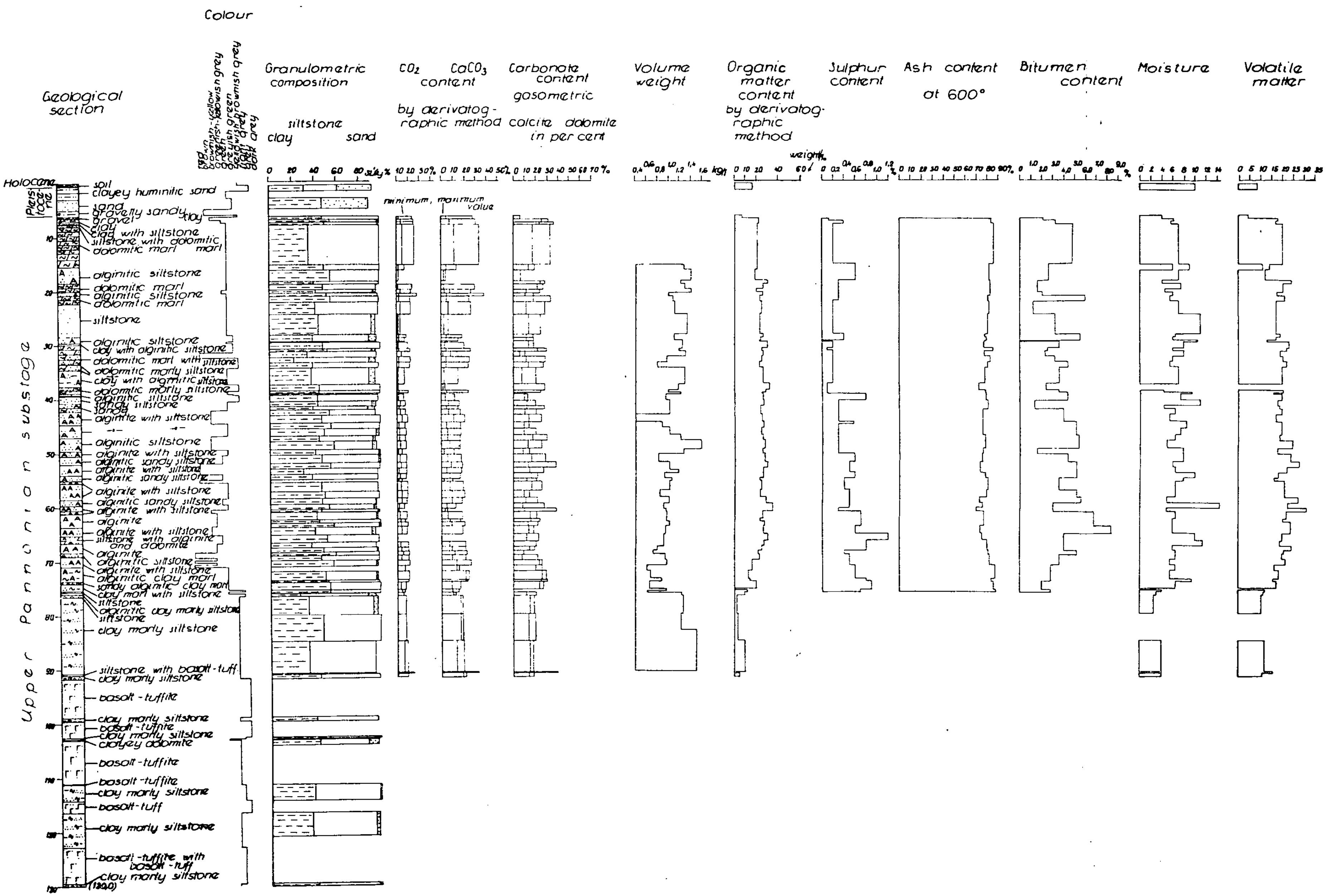
Fig. 2.

Geological and material testing section of borehole Gét-1, Gérce



The precise size of the lagoon surrounded by the ring-shaped tuff wall of the volcanic crater and impeding sediment transportation from the near-by brackish-water lake was examined by the Transdanubian Ore Prospecting Department of the Eötvös Geophysical Institute (Cs. TÓTH) with the aid of surfacial resistivity measurements. According to these investigations, the ring is elongated southwest of the Eger river's valley including also the sinter mound on the small hilltop soaring in the north above the valley bend and possibly having been active simultaneously with the deposition of the upper part of freshwater limestone sequence of Nagyvázsony.

The investigations of the special sedimentation conditions of the lagoon enclosed within the tuff-ring, enabled to determine the area of further studies.

3. GEOLOGICAL CONDITIONS OF THE GÉRCE OIL-SHALE

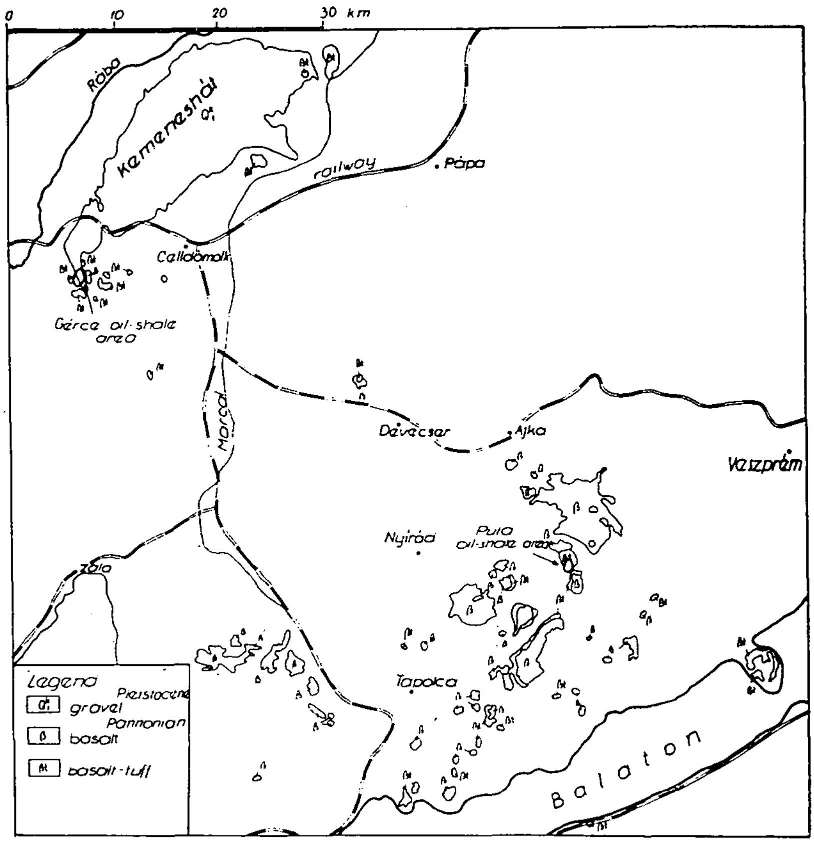
Thus, it was evident that, for the recovering of further oil-shale deposits it would be necessary to seek for similar tuff-rings as that of Pula and to drill their inner part. Earlier geological investigations had made easier this work. Thus, from the literature [L. LÓCZY, 1913, L. SZEBÉNYI, 1953, K. VARRÓK, 1953], the tuff rings of Sitke and Kemenesmagos are well-known *(Fig. 3)*.

The tuff-ring northwest of Gérce (reaching a thickness of 90 m) is filled up with sediments connected with the oil-shale member of Pula. Similarly to the sediments filling up the area at Pula, the series occurring within the tuff-ring at Gérce, can be divided in three parts, however, it may be remarked, that at this last place the basalt foot-wall is unknown.

In the environs of Gérce, in the deeper basement of the oil-shale recovered by borehole Kemt-1 at Kemenesmagos, 32 m below the oldest basalt tuff (52.7 to 55.7 m) a brackish water Mollusca fauna bearing clayey marly siltstone was found indicating the middle-top part of the Upper Pannonian and determined by M. M. KORPÁS-HÓDI *(Congeria neumayri* ANDR., *Unio sp.?*, *Pisidium sp.*, *Valvata obtusaeformis* LŐR., *Valvata sp.)*. Thus, the basalt tuff of Gérce is the younger being substantially higher. The diatoms and macroflora-bearing oil-shale of Gérce, as well as the palynological character of the basalt tuff, i. e. of the ring formed by it and filled up, proves its belonging to the Upper Pannonian. According to I. PÁLFALVY, the macroflora includes the following: *Cercidiphyllum crenatum* (Ung.) BROWN, *Equisetum sp.*, fragment of *Glyptostrobus, Gramineae, Liquidambar sp., Phyllostochys sp., Quercus sp., Q. cerris* type, carbonized fruits. The oldest subdivision of the infilling was traversed by the drilling between 90.8 and 130.0 m *(Fig. 2)*. If we consider the inclination of 50—70°, it would mean a thickness of about 20 m. This part is made up of alternating basalt tuff, clayey marly siltstone, dolomite marlstone and basalt tuff. The basalt tuffites consist of grain-fragments of different size and are, in terms of the nomenclature of sedimentary rocks, sands or siltstones with sand and small gravels. Their material is basalt, rounded or completely unrounded, scoriaceous, vesicular, aphaneritic or vesicular porphyritic. Naturally, only grains above 0.3—0.5 mm could be determined by the naked eye. Sometimes the arched cross-lamination of sands could be well observed. Here, the grains are well to fairly graded, in contrary to sediments originating from dispersion or tuffs fallen into water, which show a distinctly poor grading. In basalt tuffite layers (=washed-in basalt tuff fragments) also intercalations of basalt tuffs fallen into water were observed. There occur fragments reaching even 15 cm and every grain seems to be unrounded, with sharp contours, while the character of stratification within a layer could not be observed. Besides the basalt tuff fallen into water (114.2—116.4 m) also some other marks of a simultaneous volcanic activity could be recorded. In some water-fallen tuffs of silt size, disproportionately large, isolated and unrounded basalt fragments occurred, which had fallen as lapillies into an unconsolidated mud. Similar fragments occur in some pelitic sedimentary rocks of non-volcanic origin.

Tig. 3.

Transdanubian oil-shale deposits related to Pannonian basalts



Basalt tuffite layers — except for a single basalt tuff and a single dolomitic marlstone intercalation — alternate with clayey marly siltstones. On the basis of their lithology and texture they can be regarded as common Pannonian sediments. They show a conchoidal-laminated jointing and, fine muscovitic laminae frequently occur parallel to the joints. With unaided eyes no fossils can be observed.

In connection with dolomite marlstones of borehole Tihany T-62 [Á. JÁMBOR, 1973] mention should be made of the two dolomite layers interbedded with the water-fallen basalt tuff (102.8 to 103.0 m). Even these intercalations are of pale greenish grey colour, conchoidal-splintery, cryptocrystalline, hard, dense, unbedded rocks. The presence of dolomites may be connected with the excess of magnesium originating from the weathered material of basalt tuff glass.

This is conformably overlain by a "stratovolcanic basalt member", which may be correlated with the "oil-shale member of Pula". The boundary is only theoretical, because it was drawn there, where basalt tuff fragments upward in the sequence were last observed even with an unaided eye.

The oil-shale member starts with a 14 m thick clayey marly siltstone (76.5 to 90.8 m) consisting of 1—20 mm thick, parallel lamination and slightly different average grain size. Among the fine grains both basalt tuff grains and grains originating from metamorphic sources can be recognized. This layer is situated practically horizontal. The oil-shale sequence in the strict sense (76.5—7.0 m) may be divided into three parts, though emphasizing the fact that on account of their organic content being far more abundant than the average and their predominantly laminated texture and the low volume weight, they represent the same unit with close genetic relations. These series are similarly united by the frequency of fossils belonging to the sedge family, *Herbaceae* and, subdued, leaves of deciduous trees. The leaves of trees — except for some limonitized ones from the uppermost part (15.4—40.6 m) — are carbonized. Their substance is of black, lustrous, and flexible coal. Another common character of these sediments is the strong, chemical smell, different from that of lignites, which is well perceivable even half a year or a year after the core material was brought to the surface.

The first subdivision (76.5—68.3 m) is — in contrary to the younger part of the sequence, — characterized by frequent occurrence of pale yellowish-white, loose calcipelite laminae. The subdivision is made up of greenish-grey alginitic siltstones, but between 69.0 and 71.0 m the colour of every third lamina is pale red. The other layers consist of 0.1—1.0 mm thick, slightly undulating laminae, of greyish-green colour. These are represented by three types. The pale leaf-green and possibly the above-mentioned pale red ones are highly alginitic, while the grey sediments include plenty of clay and other mineral fragments, and the pale yellow ones are made up of calcipelite. Between 73.6 and 74.0 m, besides the small-grained sand laminae occur maximum 5 mm thick fine-grained sand laminae including basalt tuff and quartz material, while the earlier burrows of mud-eaters filled with sands (of 3—7 mm diametre) irregularly change their direction. Although the layers of this subdivision are practically horizontal, in their middle part of about 80 cm (69.0 to 71.0 m) the laminae show an inclination of 15—20°. This may be the consequence of slumping.

In the two lowermost layers of this subdivision (76.0—76.5 and 75.4—75.6, respectively) parallel to stratification, several, rather large, *Ostracoda* mono-valves were found.

The second subdivision (68.3—42.2 m) is the most alginitic part of the oil-shale with alginite-rich laminae. Thus it is also alginitic siltstone, alginite with siltstone and alginite with sand and siltstone, consisting of fine (0.1—0.5 mm thick and locally even 15 mm thick) laminae. Unlike the lowermost part, here calcipelite laminae are not present, however, the siltstone and fine-sandy laminae include more or less dolomitic marl components. As mentioned above, the alginite-rich laminae are of different green and pale brick-red colour, while the siltstone and sand laminae are grey. Brick-red laminae occur only in 4 layers (60.5—61.1, 55.0—55.5, 52.0—54.0, and 42.2—44.6 m). In the lower part of this subdivision (60.5—60.6 m) again a yellowish grey calcareous layer was found.

The laminae are generally horizontal, only in the uppermost layer (42.2—44.6 m) plication due to slumping can be seen. Between 61.1 and 63.8 m, small "unconformities" due to scour by subaquatic current may be observed.

The grains of the sandy laminae, made up of basalt and quartz, are similar to those of the lower parts. In contrary, the siltstone laminae including some tenth of millimetre thick muscovite laminae parallel to stratification are characteristic.

The uppermost part of the alginitic (=oil-shale) sequence considered in the strict sense, and traversed by drilling Gét-1 (42.2 to 6.4 m) contained dolomitic marly siltstone, siltstone with sand and dolomitic marl, and alginitic siltstone with more or less dolomitic marl.

The ratio of dolomitic marl increases upwards while the amount of alginitic rocks is diminishing. The uppermost (0.6 m thick) layer of the subdivision is mottled clay, which seems to be of bentonitic origin. It may be assumed, that it originated from a different transformation of basalt-tuff ejected from an eruption centre other than Gérce.

The dolomitic marl and dolomite-marly siltstone intercalations are not stratified, their colour is pale yellowish or pale brownish-grey. The texture of siltstone is, however, laminated. Alginite-rich laminae are green, greyish-green or pale brownish-red, while those of siltstone are grey of different shades. The laminae are horizontal or, as a result of slumping, chaotically plicated (34.5—37.5, 31.5—32.5, 20.0—20.7 m). It should be remarked, that though two layers in the 7.0—8.0 m interval are plicated, judging by their features, these seem to be rather the result of crioturbation movements.

In three dolomitic siltstone layers (37.5 to 39.2 and 32.5 to 3.4) dark green rounded bentonitic nodules (with a diameter of 1.0 to 5.0 mm) were observed, while in the siltstone bearing dolomitic marl (29.8 to 31.5 m) pieces of basalt tuff (with a diameter of 1.0 mm) occurred. Regarding its texture, the dolomitic siltstone (38.7 to 39.2 m) is an intraformational breccia but chaotically plicated. The sandy components are of two types: basalt tuff and quartz. In the siltstone layers regular, some tenths-of-mm-large muscovite laminae parallel to stratification occur frequently.

In the lowermost layer (at 39.4 m) the total surface of the drill core was covered by a yellowish dark-grey pyrit segregation. (This visual observation was confirmed by X-ray diffraction measurements by I. VICZIÁN.)

The alginitic (=oil-shale) sequence is unconformably overlain by an Early Pleistocene gravel-terrace of the Rába river covering almost uniformly the whole Kemeneshát area. It includes gravels, gravelly sandy clay, and sandy clay. The sand grains manifest rather well the aeolian influence. The 4.7-m-thick Pleistocene terrace is overlain by 1.1 m of black clayey sand (swamp facies) which in turn, is covered by pale grey sandy soil (0.6 m). These deposits have been assigned to the Holocene, as in the vicinity of the borehole, in the area enclosed by the tuff-ring minor pools becoming periodically swampy still occur even today.

If we compare the features of the Pula and Gérce deposits studied by field methods, the following results will be reached.

TABLE 1

Comparison of geological features of the alginitic basalt member hanging wall

Features	Pula	Gérce
Conformable laying stratigraphic cover	known theoretically	unknown
Mineralogical features of the surrounding layers	calcitic	dolomitic
The features of organic material	green, coarse alginite, the lower part rich in diatoms	green, and pale red fine-grained alginite, the lower part rich in diatoms
Plant debris	carbonized leaves, Chara stems	carbonized leaves
Laminae	green, grey, off-white	green, red, grey, off-yellow
Green alginite-rich laminae	occur	occur
Red alginite-rich laminae	are not present	occur
Laminae with much clay	characteristic	not characteristic
Laminae with much siltstone	not characteristic	characteristic
Low volume weight	characteristic	characteristic
Laminae with basalt-sand	are not present	characteristic
Calcipelite laminae	characteristic for the lower part	characteristic for the lower part
Average thickness of laminae	higher (0.1—50 mm)	lower (0.1—5 mm)
Plication due to subsidence	characteristic	characteristic
Intraformational breccia	occur	occur
Presence of muscovite	unobservable by naked eye	characteristic of the siltstone
Bentonite nodules (1—5 mm)	rare	in several layers frequent
Freshwater gastropodal fauna	rare	is not present
Ostracoda bearing siltstones	below the alginitic part	below the alginitic part
Siltstone layers	are present	are present
Initial siltstone layers	cross-stratified basalt-sand	cross-statified basalt-sand
Basement	basalt	unknow
Thickness of the basalt-sand layer	8 cm	some layers are of several metres thickness
Foot-wall of the basalt-sand	basalt	basalt-tuff
Thickness of the whole complex	40 m	90 m
Structure of the holder	basalt crater	basalt crater

4. RESULTS GAINED BY WELL-LOGGING MEASUREMENTS

According to prescriptions by the Central Geological Office, complex well-logging (including natural potential, resistivity, natural gamma, gamma-gamma and yield measurements) was performed in boreholes Put-3 and Gét-1, by the geophysical group of the Hungarian Geological Exploration and Drilling Company, Várpalota. Their interpretation was carried out by geophysicist SZ. URAY.

The resistivity logging of the basalt in borehole Put-3 could be carried out from 17 m down to the bottom of the hole, since the upper part of the hole was empty because of water swallowing by the basalt. In the measured intervals the three resistivity values were about 20 ohm/m, and varied slightly. It decreased only in the lowermost part, that is, in the sand layer overlying the basalt, to the value of 3—12 ohm/m.

On the basis of natural gamma measurements, the radioactive substances in the oil-shale formation of Pula do not reach the well-known low radioactivity of basalts (35 microroentgen/hour). It was, however, observed that, in the 38.4 and 16.0 m interval the value of natural gamma radiations decreased from 35 microroentgen/hour with slight fluctuations to 13 microentgen/hour. This decrease is parallel with the sulphur content, ash content and change of specific weight of the sediments, thus it may be connected with the pyrite and potassium content of the sediments. It is a well-known fact, that pyrite is inclined to fix uranium.

It is similarly difficult to explain the higher micro-roentgen/hour values (22—31) of the 16.0 to 4.0 m interval. Most valuable data were reached by gamma-gamma section. The 30,000—125,000 pulses/min value of the 4.0 to 28.0 m interval may be regarded as anomalous. A significant anomaly was observed between 6.0 and 15.0 m, where the oil-shale content also showed anomalous values (90,000—125,000 pulses/min).

The studied sections of the oil-shale sequence of Gérce, were similar. The gamma-gamma values between 8—23 m, however, were only 21,000, while from 23 to 76 m 60—65,000 pulses/min. The lower boundary, at 76 m, coincides with the setting in of anomalous organic content, but at the upper limit a deviation of 5 m was recorded. Anomalous values of organic matter begin to occur at 18 m already. At present, the cause of this deviation cannot be interpreted.

5. RESULTS OF TECHNOLOGICAL AND CHEMICAL ANALYSES

The analyses of alginitic layers are being run now according to well-developed and tested procedure. In the field, after sampling layer by layer or at an even closer spacing with longitudinal halving of the core samples, the low amount necessary to examining thin sections, volume weight, specific weight, diatom content, palynological and granulometric determinations, was separated and the rest ground to homogenize the samples for derivatographic and spectral analyses, Fischer's analysis method and other carbon-chemical tests, further on, for ash-content, carbonate and Ca—Mg content determinations. In view of its usefulness, Fischer's analysis, the carbon-chemical tests and volume weight determinations are of crucial importance. Without these data the quality and quantity of the raw material are indeterminable. Some other tests are necessary to enable further research concerning the oil-shale.

a) Analyses by derivatograph (M. Földvári) serve to demonstrate the presence of raw material difficult to observe by the naked eye. Thus, with its aid the whole organic matter, burning at 380—600 °C if organically bound, could be determined with relative precision. In the case of borehole Put-3, this is conform to the expectance, a quantity far over the sedimantary average. Taking into consideration 36 analyses, the value of each case surpassed 5 weight per cent, only in 3 cases was the value between 5 and 15 per cent. The results were as follows: in the subdivision from 38.4 to 20.0 m 22—25%, from 20.0 to 12.0 m 32—72%, and from 12.0 to 1.0 m 5—22%.

Raw material content determined by derivatograph may roughly correlate with the combustion value of oil-shale, its bitumen, volatile and hydrogen content, suggesting thus their close relationship. It is, however, striking, that the above-mentioned correlation is only approximate, in many cases being unambiguous. Thus on the basis of organic matter content the included volatile and bitumen can be determined only with a considerable error.

A significant difference presents itself in the organic matter to bitumen ratio between the Pula and Gérce sections. At Pula in 30% organic matter 4—8% is bitumen, and in 35—70% organic matter, the share of bitumen is 13—25%; while at Gérce the 25% organic matter contains only 1.2—5.5% bitumen. In both sections the organic matter content of the upper and lower part showed an identical character while in the middle part the organic content was less. The section of Gérce was even in this regard more homogeneous. These variations at Pula attain 12 to 70%, and at Gérce 3 to 35%. The differences of organic matter to bitumen ratio in oil-shale could be explained in both cases, at Pula and at Gérce, by the lack of accuracy in bitumen determinations or by the character of the organic substance. In any case, further studies are needed.

b) Under the term *qualification analyses* (P. TAKÁCS, MRS. ARATÓ, MRS. BELLA, MRS. KOVATSITS) the results of determinations of bitumen content, calorimeter tests, volatile content, humidity, sulphur and hydrogen content, and volume weight have been summarized.

The bitumen content of the above rocks was determined by the experts of the Mining Research Institute, taking as a basis the Hungarian standard obtained by Fischer's method. According to their data gained of samples taken layer by layer and then longitudinally halved, crushed and averaged, the bitumen content in borehole Put-3 (from 6.0 to 37.5 m) was 9.9—27.2 weight %, while in the section of Gérce (from 7.0 to 76.0 m), the weight per cent fluctuated between 1.2—8.4. The organic matter content of the upper and lower part in both sections contains significantly lesser bitumen as the middle part. The frequency of green laminae in the section of Pula, seen even by naked eye, is in close connection with the higher amount of bitumen. At Gérce both the green and red-brown laminae contain bitumen, and even in the dolomite marls of the upper part a considerable amount of bitumen is present. It may be remarked that, nearly half of the organic matter can be dissolved in a bensene-alcoholic substance. Thus it can be processed even in this way, provided that a greater part of the economically useful organic material disappears.

The volatile content shows a rough correlation with the quantity of bitumen: its amount in the section of Pula approximates 30%, while in the section of Gérce this figure is about 20%.

A similar trend was shown in both sections by the ash content of the rocks. In the case of Pula, when burning out occurs about 1000 °C, this value attains 65%; however, in the middle part of the section with higher bitumen content, it is only 40—50%. At Gérce, when the ignition is carried out at about 600 °C, this value attains 80%, and never decreases below 70%.

Moisture in both sections is about 7% and while at Gérce it scarcely changes, at Pula it shows significant variations.

Calorimetric value was determined only for samples of Pula. It presented along the section a very similar curve as that of the bitumen content with a value variation from 120 to 3350 kg/cal.

Very close connection with the calorimetric value, even a function of it, manifests the extremely high hydrogen content of the rock, changing along the section from 0.75 to 5.4%. A critical point for every fuel material is the sulphur content. The quality of oil-shales of Pula and Gérce is also in this regard very favourable with the low sulphur content, the amount of which decreases step by step: from 1.1 to 0.1 at Pula and from 0.9 to 0.1 at Gérce, respectively. It may be remarked, however, that the sulphur content shows no correlation with the ash or the volatile content. At the same time it changes parallel to the frequency of diatoms and the quantity of carbonate. The 0.1—1.1% sulphur content for oil-shales is strikingly low. According to H. FÜCHTBAUER and G. MÜLLER (1970), bituminous shales contain 3 to 20% pyrite (in average 8.2%), which would mean roughly 1.6—11.0% (that is 4.2%) sulphur content.

The distillation refinery gas yield of Pula was measured, too, from samples taken werhe the oil-shale was most abundant (6.0 to 18.0 m). The sample yielded 54.0 m^3 gas/ton, from which 13 volume weight per cent is CO_2+H_2S, 4 volume weight per cent is CO, 8 volume weight percent O_2 and the rest (75%) other materials (hydrocarbons). At Gérce such studies were not carried out.

Volume weight determinations are indispensable for the estimation of rescurces. The analyses were processed by mercurial method (G. SOLTI). In the section of Pula (6 to 20 m), apparently of highest practical value, the volume weight, according to expectations, is very low (0.80—1.14 kg/litre). The lowest values occurred in alginite and in the layers of higher organic content. Except for the 50 cm thick dolomitic silstone, the average value attained 0.95 kg/litre (thus at calculations, practically, it may be reckoned with 1 volume weight). The specific weight of rocks from Pula varies between 2.45 and 1.15 g/cm^3. There is a close connection between organic matter content and specific weight, which is evident if we take into consideration the specific weight conditions of silicate and organogenic minerals.

At Gérce, because of the higher ash content, even the volume weight data were higher: varying from 0.61 to 1.42 kg/litre in the 15.4 and 76.0 m interval and with an average value of 1.1 kg/litre, which is remarkably low when compared to the average of the Pannonian clay marl (1.8—2.2) or lignite (1.20 kg/litre). Obviously, the majority of the minute *Botryococcus* colonies is not filled with mineral pelites, hence the marked looseness of the rock.

Summing up the results of the above testing of materials, it may be concluded that, thebasalts or basalt tuffs in the sections of Pula and Gérce, are overlain by a formation (40—38.4 m that is, 7.0—76.0 m respectively) practically representing a kind of oil-shale. The oil-shale of Gérce is of lower quality while that of Pula is somewhat better, being of higher quality in the 6.0 to 18.0 m interval and reaching a "very high quality" in the 15.0 to 18.0 m one.

c) It should be mentioned here, that because of the presence of a particular facies, the two sections were subject to *spectral analyses*. According to P. ZENTAI, there were no striking values. Table 2 presents the extreme and average values.

d) For the purpose of *petrographic* characterization granulometric composition and calcite-dolomite ratio determinations were carried out, layer by layer, of the oil-shale sequences of Pula and Gérce. According to these, significant differences between the two series were revealed.

Taking into account the granulometric composition it may be stated that the clay content of borehole Put-3 varies from 14 to 60%. Below 18 m depth the share of the clay fraction shows an increasing trend, then, at 18 m, a sudden decline is observed to be followed further up, again, by an increase with significant fluctuations. In the 6 to 18 m interval more abundant in organic matter, the sand fraction was also considerably represented, though the sand grains themselves could not be observed in the rock. This may be due to coagulation of the organic matter. In fact, the rocks are made up mostly of siltstone fraction, which can be observed even by unaided eye.

The section of Gérce is characterized by higher ratio of the siltstone fraction. In the 20 to 60% interval the clay fraction increases up to 42 m (on the average it is about 40%). At this height it significantly declines but increases again up to the top.

TABLE 2

Trace element content in ppm, of the ash recovered from 19 samples of the 4.0—38.4 m interval, borehole Put-3, Pula, after ignition at 600 °C

Values	Elements B	Mn	Cu	Pb	Ga	Mo	V	Ti	Zn	Ni	Co	Sr	Cr	Ba	Li
Highest	160	4000	250	40	16	100	400	10 000	160	160	60	1000	160	1600	250
Average	85	2294	128	17	9	33	159	3 484	112	80	29	831	78	753	131
Lowest	40	1000	60	6	2.5	<6	40	400	40	40	10	600	40	250	60

Trace element content in ppm, of the ash recovered from 49 samples of the 6.4—90.6 m interval of borehole Gát-1, Gérce, after ignition at 600 °C

Values	Elements B	Mn	Cu	Pb	Ga	Mo	V	Ti	Zn	Ni	Co	Sr	Cr	Ba	Li
Highest	400	2500	150	60	60	100	250	10 000	250	250	60	1600	400	2500	6
Average	194	1622	67	22	30	12	105	7 367	70	104	24	813	146	922	6
Lowest	60	600	25	4	10	10	40	1 000	<160	16	10	60	40	400	6

The rest of the sediments is made up of siltstone; sand fraction in turn occurs only in some layers reaching a maximum of 5%.

The calcite content of the sediments at Pula show upward significant variations in the profile, decreasing from 63% to 23% and showing then a sudden increase in the final bed. At the same time, the dolomite content is about 5% up to the end, except for an increase to 8—21% in the organically enriched part of the profile where it becomes occasionally even predominant. In the final part, i. e. the calcareous marl sequence, the quantity of dolomite is significantly higher than that of calcite. Thus, the share of dolomite shows a gradual increase upward.

In the section of Gérce, the calcite content of the sediments varies between 15 and 26%, however, unlike at Pula, it shows a slightly increasing trend. The dolomite content varies between 3 and 18%, its ratio increases and from 54 m on, it gets in equilibrium with calcite. Thus the two sections show similar development. The most logical interpretation accounting for the difference in calcite content between the sequence of Pula and that of Gérce is to suppose that Pula must have had a calcium carbonate environment.

6. PALEONTOLOGICAL INVESTIGATION

Paleontological research had two aims. First, the age of sediments overlying the basalt had to be determined. This was all the more important, as no sediment was found in the immediate hanging wall of the Transdanubian basalt or in their closest vicinity and, therefore, the basalts were declared, on and on, to be partly or totally of Pleistocene age.

The other task was to determine the facies. M. HAJÓS identified 1—52 diatom species in various samples of the sequence. The number of species and also their frequency show an upward decrease. (This has been expressed with so-called frequency show an upward decrease. (This has been expressed with so-called frequency indices: "sparse" = 1, "fair" = 2, "frequent" = 3, "abundant" = 4.) As was mentioned above, pyrite, sulphur and carbonate presents a decrease with the reduction of the amount of diatoms, while dolomite increases with decreasing diatom content. The abundance of the diatomaceous microflora is an evidence of favourable life-conditions, the sufficiency of oxygen, insolation and mineral nutrients. Since the combustible organic matter content increases rather inversely to the content of diatoms, it is very possible that the conditions for the life of planktonic diatoms became, with the passing of time, more and more unfavourable, while *Botryococcus* were to find gradually more favourable conditions of living.

The diatomaceous flora proves impressively the presence of the Upper Pannonian. This statement, however, relies on paleoclimatic and water quality considerations rather than on biostratigraphic results. On the basis of the ranges of the species identified, the rocks may be both older or even younger, that is, of Pleistocene age. The microflora testifies to a rather warm February water temperature: 11 °C, slightly brackish water. The two phenomena combined could hardly occur in Pleistocene time.

According to E. NAGY, the palynological aspect of the samples from boreholes Put-1, Put-3 and Gét-1 shows a typical Pannonian floral assemblage. One of the most characteristic forms of this assemblage is the planktonic alga *Botryococcus braunii* KÜTZ., one of the main constituent of bituminite-rich solid rock. Besides the pelagic Botryococcus, the most significant role was played by the representatives of *Conifera* family (*Abies, Tsuga, Cedrus* etc.). Their presence suggests the one-time occurrence of Mediterranean-type coniferous forests in the closest or wider neighbourhood. At altitudes below those of the coniferous vegetation, there was a habitat composed of deciduous trees again of Mediterranean type (*Celtis, Carpinus, Zelkova, Ulmus, Juglans, Betula, Fagus, Quercus, Acer, Ilex, Liquidambar)*. The presence of swamp forests on the shore of the Pannonian lake is evidenced by the genera *Nyssa, Taxodium, Alnus, Carya, Pterocarya.* Accordingly, the vegetation of the neighbourhood testifies to a humid climate warmer than present-day's, i. e. to one of Pannonian rather than Pleistocene type.

According to I. PÁLFALVY, the microflora is mostly represented by sedges, though Upper Pannonian deciduous trees *(Liquidambar, Quercus)* also occur.

A rather great number of well-preserved remnants of *Ostracoda* have been recovered from the lower beds of both sections. On the basis of a sample from borehole Put-3, M. SZÉLES determined a typical Upper Pannonian, slightly brackish water, fauna.

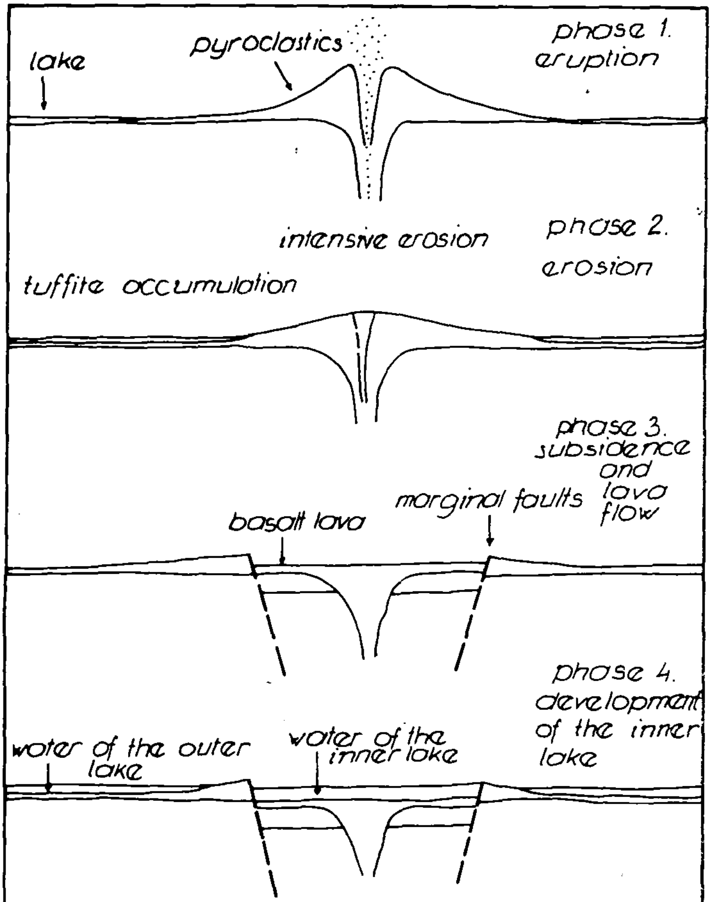
Again from borehole Put-3 (32.4 and 35.4 m) mollusc fragments have been recovered. These were determined by E. KROLOPP as *Radix aff. peregra* (MÜLL.), living in a freshwater environment, but occurring in both the Pannonian and Pleistocene rocks. *Pisidium sp.* from 32.4 m lived in fresh or slightly brackish water.

Summarizing the above, it may be concluded that the paleontological character of the alginite sequence overlying the basalt is closely connected with that of Upper Pannonian formation, though it does not contain any fossil enabling a firm chronostratigraphic assignment either to the Pliocene or the Pleistocene.

7. PALEOGEOGRAPHY AND GEOHISTORY

At the end of the middle Upper Pannonian, the area — taken *in sensu lato* — was covered by a system basalt tuffites a few metre to tens of metres thick. It is only around the eruption centres overlying the cross-laminated beds that true continental (that is, air-borne) basalt tuffs and agglomerate layer can be found and the first basalt lava bed is even more delimited *(Fig. 5)*. In most cases the first basalt tuff layer is present now only there, where during the Pannonian and the Pleistocene it was protected from weathering by the hard bed of first basalt lava flow. The first eruption cycle was soon followed by a second one, sometimes with quite new eruption centres as that of Pula and, as shown by analogy, that of Gérce as well. This eruption cycle consisted of a single intensive ejection, during which at the eruptive centre first a low volcanic cone was formed, then in the second phase, after the appearance of the basalt lava, the middle part of the original volcanic cone subsided along volcanotectonic fractures and its base was filled up with basalt lava. In the third phase of development, as a consequence of the continuous subsidence during the development of Upper Pannonian formation, lacustrine water intruded through the lithoclases of the tuff ring or through some channels above the more shallow parts, into the cooled volcanic body. Here, despite of shallow lakes of varying size becoming repeatedly swampy and extending over the whole Tapolca—Nagyvázsony basin. The quiet and low sedimentation was disturbed by basalt eruptions along deep fracture lines presently unknown. Although it is very unusual but it can be taken as a fact, that sedimentation was influenced by these eruptions only over a very limited area. The pyroclastic material of basalt reaching to the surface with the beginning of the first eruption was absorbed by the Upper Pannonian lake system. While over an area of several km^2 inwashed water-fallen tuffs few occurred, the positive volcanic forms — if any — were limited only to a few thousand m^2, i. e. very small areas. The thin tuff layers of inwashed or water-fallen origin were argillized

Fig. 5.



Stages of the Transdanubian basalt eruptions which developed the structure of the oil-shale holder under the water or partly above it; in fact, minor waves could sweep tuffs away to great distances within the lake system letting them completely disappear.

In the second part of the eruption, a significant volume of pyroclastics reached to the surface, and built up a volcanic cone arising above the lake. Simultaneously with eruptions torrents swept slurry flows from the mountains to the surface of the Pannonian lacustrine sediments, forming in the territory of the Balaton Highland arched cross-laminated the isolation, the rather significantly deep (5—10 m) water and average Upper Pannonian conditions, the intensive near-by weathering of basalt tuff glass resulted in abundance of nutrients, forming special sedimentation conditions. The low hydrodynamics of the Upper Pannonian lake was not able, or was so only in a very limited form, to transport the fine pelitic material and into the inner lake, thus the pelite mainly originated from the eroded inner side of the basalt ring (at Gérce locally reaching even a grain size of sand) of which the filling material is made up. Besides a significant quantity of calcipelite (precipitated as a result of the general bacterial activities), subdued dolopelite, further on perished planktonic algae — in the beginning with diatoms, later mainly Botryococcus forms — were added. The ratio of the three main components changed — on the basis of the size of lamination — according to seasons being a function of the insolation, distribution of precipitation and quantity of available nutrients. At present it is not known, which season is represented by the 0.1—5.0—mm-thick laminae of different composition. Obviously those rich in lime manifest the warm summer time. Considering the average thickness of laminae being 0.5 mm, the sequence at Pula may be of 50,000 years and at Gérce of 140,000 years.

Taking into consideration the increasing amount of dolomite material and desiccation structures within the section of Pula, the crater lakes showed a trend of drying out. In the rather deep water of the lake, protected from wind, the original lamination of the sediments was preserved under the water table covered by algal cultures (*Botryococcus* colonies). Under such conditions, the rolling waves could not develop their stirring and resedimenting influence. The effect of the rather steep immediate neighbourhood of great relative relief is not manifested well enough in the sediments. This may be due to the filter effect of the dense vegetation of sedge and reed (*Taxodium* etc.) in the shallow water along the shore of the lake, though it is abvious that several years after cooling out of the crater, its inner side was covered by a dense vegetation which impeded in a similar way the getting in of coarse material. In spite of this, the life of crater lakes was not completely undisturbed. Slumping activities took place periodically, mainly as a consequence of earthquakes related to volcanism.

The macrofauna is lacking in both series studied at Pula and Gérce, moreover those debris found in the section of Pula are of freshwater origin, which is not in agreement with the determinations concerning the "slightly brackish environment" of the diatom flora. The lack of brackish water molluscan fauna has to be interpreted by a special anaerobic bottom mud, while some freshwater debris could be introduced by nektoplanktonic transportation afloat.

Attention has to be called to the fact that the alginitic rocks cannot be fitted in the swampy zone system of coal-formation, which means that a deep origin can be proved only for some types. Theoretically, a mud with abundant algae, combustible in dry state, bearing much bituminite and with few other sediments may be deposited in a closed water where there is no sediment transportation and the accumulated material was affected by diagenetic processes under anaerobic circumstances and thus its organic content is not subjected to oxidation. The extension of bituminite-rich oil-shale formations as well, as several examples of other countries manifest this unanimously in Hungary. (BRADLEY, W. H., 1964, 1970; CANE, R. F., 1969; DUNCAN, D. C. and SWANSON, V. E., 1965; MÜLLER, G. and BLASCHKE, R., 1969).

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DR. ÁRON JÁMBOR
GÁBOR SOLTI
Hungarian Geological Survey
Népstadion, u. 14.
H-1442 Budapest, Pf. 106, Hungary