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Physical and chemical properties of hydrocarbon-bearing sediments (Outer Western Carpathians, Czech Republic)

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Abstract: The present study deals with the impact of hydrocarbons on changes in the physical properties of the sediments of the Ždánice-Hustopeče strata in the flysch sedimentary cover in the south-eastern part of the Czech Republic. The discussed changes are mainly concerned with contents of natural radioactive elements Th, U and K. The subjects of the study are rocks from deep boreholes collected down to the depth of 3.3 km. The boreholes are situated in an area of local hydrocarbon occurrences and are divided into two groups — with and without hydrocarbon indications. Physical properties of sandstones and claystones from the two groups are compared. It has been concluded that the distribution of U and K in the Ždánice-Hustopeče Formation indicates shifts in contents of these elements which are undoubtedly due to the presence of hydrocarbons.

Key words: petrophysical data, flysch sedimentary cover, statistic evaluation, oil deposit, deep boreholes, prospective indicators.

Introduction

In East Moravia between the villages of Nevojice and Ždánice a hydrocarbon deposit can be found at a depth of nearly 1 km. The reservoir is located in a fractured and weathered surface part of Upper Proterozoic granitoids and in Lower Miocene sandstones and conglomerates overlying the crystalline basement. The seal was formed during the Late Miocene by the tectonically emplaced Ždánice thrust sheet, which attains the thickness of 750 to 870 m over the hydrocarbon field (Krejčí 1993). To study prospective indicators of a buried hydrocarbon deposit in near-surface portions of overlying flysch sediments, several shallow boreholes were drilled above the deposit and in its vicinity.

The objective of investigations was to find out whether there is a significant difference in the studied parameters between the boreholes above the deposit and the boreholes situated at a distance of 2–3 km. Research conducted in other countries revealed various changes in sediments over hydrocarbon deposits, including radioactivity, for example, Shideler & Hinze (1971) or Saunders et al. (1993), which prompted the assumption that similar petrophysical changes in overlying beds might be present in the Ždánice deposit, though the deposit proper occurs at a depth of approximately 1 km under the surface.

Our research results (Matolín et al. 2007) confirm changes of the physical and chemical parameters in near-surface flysch sediments due to hydrocarbon contamination over the Ždánice oil deposit. In this case, cores from ‘shallow’ boreholes were used as a comparatory material. Interpretation of results yielded information on changes to the physical and chemical properties of rocks in the vicinity of the deposit delimited in this way. It can be summed up that the contents of potassium and uranium are generally lower in the overlying area. Despite a considerable distance from the deposit, the existence of phenomena causing changes in contents of radioactive elements was confirmed. Contents of sulphur in claystones and sandstones as well as magnetic susceptibility and mineralogical density can be regarded as reliable indicators of an oil deposit. The deposit body can be identified on the basis of physical-chemical parameters measured on specific rock samples from surface or shallow boreholes; however, in most evaluated parameters this is not reliable.

Following this experiment it was decided to evaluate the available petrophysical data from deep boreholes in analogical geological areas using the methodology verified in investigations of so-called shallow boreholes. Selected measurements of physical properties on cores from deep boreholes drilled in the sediments of the Carpathian Foredeep and Outer Group of Nappes of the West Carpathian Flych Belt were used for this purpose. The objective was to find links between the presence of hydrocarbons and changes in the physical-chemical parameters of rocks. The study material was obtained within the framework of the geophysical database Grant of the Czech Geological Survey — Geofond (Dědáček et al. 2003). Over the past decades the geological formations of the Carpathian Neogene Foredeep and Carpathian Flysch Belt were subject to intensive reconnaissance and exploratory drilling with extensive state investment. That is why a relatively large set of measurements was obtained and archived, and is now being transferred into an electronic database administered by the Czech Geological Survey — Geofond Praha. On the territory of the Czech Republic there are approximately 30,000 samples from drill cores including coded petrographic data.

Petrophysical measurement methods

Archived petrophysical measurements were evaluated. Petrophysical measurements are laboratory measurements on specimens of rocks for mineralogical and bulk density, porosity, specific magnetic susceptibility and contents of Th, U and K.

Petrophysical measurements archived in the database of the Czech Geological Survey — Geofond have been conducted since the mid-sixties on cores of most core-drilled deep boreholes in the territory of the former Czechoslovakia. The methodology is governed by analogical principles, though updated in the course of time. Measurements conducted at different times are therefore comparable.

Density measurements — the parameters measured are bulk density (Do), mineralogical density, that is the density of the mineral component of a rock without pores (Dm), and open porosity. In tables it is designated as (Por). It is a proportional volume of communicating pores — a scalar quantity. Measurements were carried out by the triple weighing method using kerosene as the saturating medium. It should be noted that porosities of rocks dried at 100 °C are considered.

The standard deviation of one measurement is $\pm 0.001\ \mathrm{g \cdot cm^{-3}}$ for Dm, $\pm 0.003\ \mathrm{g \cdot cm^{-3}}$ for Do, and ±0.02 % for porosity.

Magnetic susceptibility was measured on the KLY kappabridges with the sensitivity of 10^{-8}. It is a dimensionless material parameter (tensor), its mean value (SUSC), however, is used for observations of material changes.

Th, U and K contents and the total gamma activity were determined using the scintillation spectrometry. U was determined through its daughter nuclide ^{226}Ra. The mean measurement errors were: Th ± 0.8 ppm, Ura ± 0.4 ppm and K ± 0.2 %.

Investigated deep boreholes and drilled tectonic-stratigraphic units

Data for our investigation were taken from numerous measurements of physical properties on cores from deep boreholes, drilled in the sediments of the Carpathian Foredeep and Carpathian Flysch Belt and reaching the Ždánice-Hustopeče Formation whose sediments are evaluated. The relatively extensive area is shown in Fig. 1. Apart from the hydrocarbon occurrences known as the Ždánice deposit there were numerous indications in other localities. Among the most significant ones is the Lubná area where an oil field occurs in Precambrian (Cadomian) Brunovistulicum granitoids under the ovethrust sediments of the Carpathian flysch nappes. Moreover, drilling reached Paleozoic rocks of the Bohemian Massif, Cambrian and Devonian (old-red) clastic rocks (?), Devonian carbonates, lower Carboniferous (Culm facies) and sporadically coal-bearing lowermost upper Carboniferous (Namurian). From the Mesozoic and Tertiary rocks making up the autochthonous cover of the Bohemian Massif, the presence of Jurassic carbonate and pelitic-carbonate sediments, Paleogene limestones and Miocene sediments of the Carpathian Foredeep was proved by drilling.

The Carpathian Flysch Belt investigated by deep drilling in south-east Moravia consists by Pouzdřany, Ždánice and Zdounky Units belonging to the Outer (Menilite-Krosno) Group of Nappes and by the Magura Group of Nappes.

The Ždánice Unit in the studied area is composed of Paleogene to Lower Miocene sediments (Ždánice-Hustopeče, Menilite and Němčice Formations). Older sediments (Upper Jurassic to Upper Cretaceous) are known from the surrounding area of this unit (Chlupáč et al. 2002). Some of these boreholes revealed manifestations of hydrocarbons, some did not. A few boreholes offered only weak indications.

Boreholes indicating the presence of hydrocarbons

Bařice 1 — without hydrocarbon presence, Karlín 1 — productive, gas, Krumvíř 1 — no hydrocarbons, Letošov 1 — weak indications, Lubná 1 — productive, Lubná 2 — weak indications, Lubná 5 — productive, Lubná 7 — weak indications, Mouchnice 1, 2 — indications, N. Mlýny 1 — no indications, Němčičky 1 — contentious indications, Uhřice 1 — hydrocarbon occurrences, Vranovice 1 — contentious indications, Snovídky 2 — contentious indications, Ždánice 2 — strong indications, Ždánice 4 — no indications, Křepice 5 — no indications, Bučovice 1 — no indications, Bulhary 1 — no indications, Kobeřice 4 — no indications, Kobylí 1 — productive, Nikolčice 2a — productive, Sedlec 1 — weak indications, Uhřice 6 — productive, Žarošice 1 — weak indications, Žarošice 2 — productive.

The boreholes have been processed in two groups:

- Productive boreholes, with hydrocarbon indications including those classified as contentious or bearing only weak traces (in short ‘productive’).
- Boreholes without hydrocarbon indications (‘non-productive’).

Comparison of the physical parameters in psammites and pelites from the two processed groups indicated potential changes of physical parameters due to the presence of hydrocarbons. Hydrocarbon indications are related to the entire borehole, that is the Ždánice-Hustopeče strata reached by the borehole which produced the evaluated samples.

Tectonic position and stratigraphic classification of evaluated samples

Each borehole reached the Ždánice-Hustopeče strata as had been planned. The cores from these boreholes provided a sufficient number of samples (mainly of psammites), which does not apply to the Miocene of the Carpathian Foredeep or the underlying Menilite and Němčice Forma-

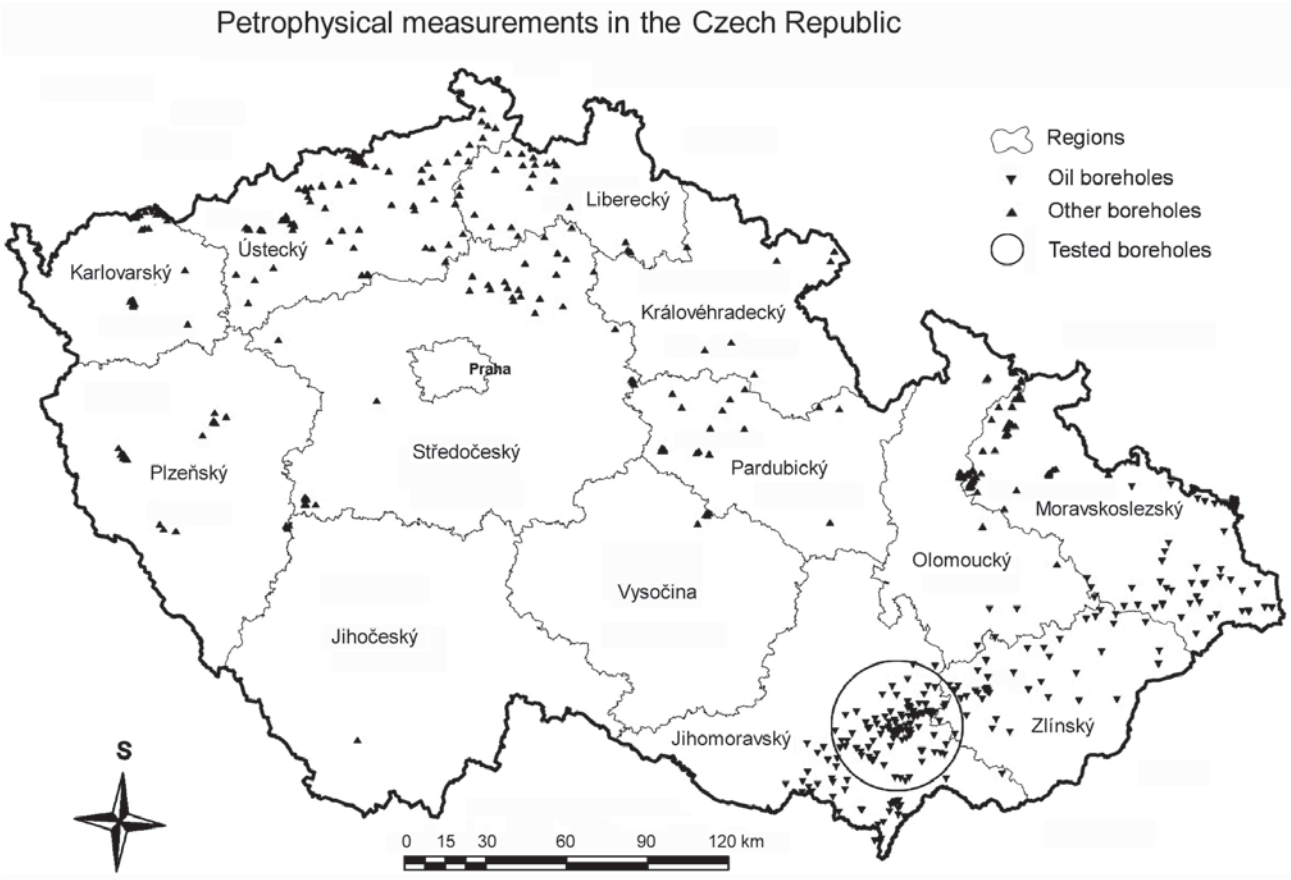

Fig. 1. The study area with deep boreholes.

tions. We assume that if there are hydrocarbon occurrences or indications in some layers, the other reached stratigraphic levels are likely to be at least partially geochemically 'contaminated', including the investigated Ždánice-Hustopeče Formation. Our considerations are therefore based on the assumption that hydrocarbon 'contamination' is more obvious in borehole group 1 as compared with group 2 although some weak contamination effects cannot be excluded in group 2. Indications of epigenetic metasomatism of clastics ('contamination') due to hydrocarbons can be statistically evaluated by comparing two sets (pairs of sandstone sets and pairs of claystone and siltstone sets).

The Ždánice-Hustopeče Formation is assigned to the Upper Oligocene (Chattian) to Lower Miocene (Aquitanian). It is an identical lithostratigraphic unit investigated in 'shallow' boreholes between the Ždánice and Nevojice villages. The thicknesses are considerably higher in deep boreholes, with a high probability of a close, direct contact with hydrocarbons. In deep boreholes the strata was drilled at depths between 5 and 3300 m under the recent surface. The greatest depth — over 3 km was reached in boreholes Sedlec 1 (the stratum is represented by samples from the interval 1000–3300 m). The real thickness of the Ždánice-Hustopeče strata is approximately 1200–1300 m. In individual boreholes the strata is often tectonically scaled and thus it is encountered in the borehole profile in more intervals (e.g. borehole Kobylí 1), or double thicknesses (Sedlec 1).

The Ždánice-Hustopeče Formation is developed in psammitic (Ždánice Sandstone), pelitic (Hustopeče Marl) and psammitic-pelitic rocks (alternating sandstones and claystones).

Methodology of processing

Routine Excel programs were used for processing, enabling selection from database and calculations of statistical parameters and creation of histograms. Parameters Kd, Ud and Drad were used for evaluation of hydrocarbon indications as defined by Saunders et al. (1993). The authors published a theoretical explanation of the differences in radioactive element contents observed in sediments over the hydrocarbon deposit and outside it. They assume that by action of organic acids and CO_2 clayey and other minerals are destructed in the vicinity of the deposit, and in consequence U and K are released and drifted away. The contents of radioactive elements in affected sediments are therefore lower than in sediments not affected by this process. The authors also assume that Th remains immobile in these processes. In their opinion the following parameters can be regarded as indicators:

$$Kd = [Ka-(Kmo/Thmo)\times Tha]/Ka$$
$$Ud = [Ua-(Umo/Thmo)\times Tha]/Ua$$
$$Drad = Ud-Kd$$

Ka, Ua, Tha are contents of radioactive elements in samples of the investigated sedimentary unit containing hydrocarbons or altered by their products, Kmo, Umo a Thmo are contents of these elements within the same unit outside the deposit accompanying the alteration. According to the authors negative Kd and Ud values indicate the presence of hydrocarbons. However, their findings related to arid climate, and therefore the theoretical model need not be of a general value and the distribution of radioactive elements may not be so simple (see Gnojek 1976; Borovec 1985; Fiala 1989).

Processing of archived and derived data is based on statistical comparison of two corresponding sets (pairs of sandstone sets and pairs of claystone and siltstone sets) from the so-called productive and non-productive area, as explained above. In correlation matrices for individual parameters we give the correlation coefficient R and the critical value for 95 % probability for N number of samples (for the minimal value N at different numbers of measurements of individual parameters).

The crucial problem in evaluating the changes of physical parameters due to hydrocarbon 'contamination' is the assessment of the significance of differences between averages of productive and non-productive boreholes. Statistical distribution of parameters is often asymmetric, very different from the Gaussian distribution, as the distribution of values from 'shallow' boreholes was analysed by tests. Using standard significance tests of differences in arithmetic means is not without problems. It appears that to solve this problem it is best to evaluate the character of the distribution of individual parameter values by means of histograms, as was found out in processing of 'shallow' boreholes.

Presentation of results

The results of the evaluation are presented in tables and histograms appended to this paper. The parameters from 'productive' (code A) and 'non-productive' (code O) boreholes are compared.

Tables 1 and 2 contain statistical data on the physical parameters including dimensionless parameters Kd, Ud and Drad in the sense of Saunders et al. (1993). Each table contains data from productive and non-productive boreholes. In the matrix for dimensionless parameters average values characteristic for the investigated lithotypes must be used. An average of all available samples of a particular lithotype is taken.

Table 1: Physical parameters of sandstones.

	Productive boreholes — Code A										Non productive boreholes — Code O									
	Do	Dm	Por	SUSC	Th	Ura	K	Ud	Kd	Drad	Do	Dm	Por	SUSC	Th	Ura	K	Ud	Kd	Drad
	(g.cm^{-3})	(g.cm^{-3})	(%)	(10^{-6})	(ppm)	(ppm)	(%)				(g.cm^{-3})	(g.cm^{-3})	(%)	(10^{-6})	(ppm)	(ppm)	(%)			
AM	2.486	2.716	8.4	110.5	6.1	2.3	1.5	–0.47	–0.30	–0.17	2.422	2.713	10.6	119.1	6.2	3.1	1.7	0.07	0.07	0.00
SD	0.140	0.021	4.9	42.2	2.9	1.2	0.6	1.30	1.05	1.51	0.182	0.052	6.5	40.7	2.8	1.2	0.4	0.46	0.46	0.42
ME	2.530	2.718	6.6	99.0	5.3	2.0	1.5	–0.19	0.05	–0.16	2.489	2.721	8.3	110.0	5.9	3.0	1.7	0.12	0.13	0.01
Q3	2.601	2.727	12.4	132.0	7.8	2.6	1.9	0.15	0.28	0.23	2.547	2.731	15.4	150.8	7.2	3.7	2.0	0.42	0.31	0.18
Q1	2.374	2.705	4.5	80.0	4.2	1.6	1.1	–0.63	–0.40	–0.58	2.301	2.704	5.8	85.7	4.5	2.3	1.4	–0.18	–0.01	–0.22
No	108	108	108	97	87	87	87	87	87	87	70	70	70	62	55	55	55	55	55	55

AM — Arithmetic mean, SD — Standard deviation, ME — Median, Q3, Q1 — Quartiles, No — Number of observations.

Table 2: Physical parameters of calcareous claystones.

	Productive boreholes — Code A										Non productive boreholes — Code O									
	Do	Dm	Por	SUSC	Th	Ura	K	Ud	Kd	Drad	Do	Dm	Por	SUSC	Th	Ura	K	Ud	Kd	Drad
	(g.cm^{-3})	(g.cm^{-3})	(%)	(10^{-6})	(ppm)	(ppm)	(%)				(g.cm^{-3})	(g.cm^{-3})	(%)	(10^{-6})	(ppm)	(ppm)	(%)			
AM	2.527	2.720	7.1	177.4	9.6	3.3	1.9	–0.45	–0.04	–0.42	2.397	2.703	11.2	170.7	9.9	5.0	1.7	–0.20	–27.46	27.25
SD	0.087	0.045	3.5	41.3	2.8	2.0	0.6	0.86	0.59	0.91	0.188	0.105	6.7	43.3	3.0	4.3	0.8	0.86	87.08	86.39
ME	2.536	2.729	6.2	183.0	9.9	2.9	2.0	–0.36	0.08	–0.45	2.430	2.730	9.7	171.5	9.2	3.8	2.0	0.11	0.18	0.06
Q3	2.600	2.750	9.7	200.0	10.9	3.5	2.3	–0.09	0.20	–0.14	2.558	2.755	15.9	195.3	10.8	5.4	2.3	0.31	0.31	0.42
Q1	2.475	2.704	4.0	165.3	8.0	2.4	1.5	–0.72	–0.08	–0.72	2.280	2.703	6.1	152.8	8.4	2.8	1.6	–0.32	–0.33	–0.24
No	84	84	84	82	65	65	65	65	65	65	69	69	69	64	53	53	53	53	53	53

AM — Arithmetic mean, SD — Standard deviation, ME — Median, Q3, Q1 — Quartiles, No — Number of observations.

Table 3: Correlation of the physical parameters of sandstones.

	Productive boreholes — Code A						**Non productive boreholes — Code O**					
	Do	**Dm**	**Por**	**SUSC**	**Th**	**Ura**	**Do**	**Dm**	**Por**	**SUSC**	**Th**	**Ura**
Dm	0.380						0.240					
Por	–0.991	–0.251					–0.962	0.024				
SUSC	0.431	0.507	–0.376				0.340	0.046	–0.330			
Th	0.234	0.329	–0.198	0.579			0.314	–0.007	–0.308	0.755		
Ura	0.080	0.010	–0.082	0.151	0.211		0.157	0.105	–0.126	0.191	0.206	
K	–0.090	0.131	0.110	0.484	0.404	0.128	0.009	–0.017	–0.003	0.333	0.360	0.273
	Critical value for α = 0.05 is 0.208						Critical value for α = 0.05 is 0.300					

Table 4: Correlation of the physical parameters of calcareous claystones.

	Productive boreholes — Code A						**Non productive boreholes — Code O**					
	Do	**Dm**	**Por**	**SUSC**	**Th**	**Ura**	**Do**	**Dm**	**Por**	**SUSC**	**Th**	**Ura**
Dm	0.017						0.354					
Por	–0.898	0.424					–0.862	0.165				
SUSC	0.240	0.455	–0.021				0.422	0.227	–0.323			
Th	–0.051	0.297	0.186	0.400			–0.144	0.218	0.293	0.150		
Ura	–0.130	–0.476	–0.111	–0.354	–0.094		–0.471	–0.438	0.260	–0.220	–0.236	
K	0.094	0.312	0.059	0.396	0.354	–0.115	0.233	–0.195	–0.366	0.181	–0.589	0.333
	Critical value for α = 0.05 is 0.260						Critical value for α = 0.05 is 0.273					

Tables 3 and 4 contain correlation matrices for individual parameters with the correlation coefficient R and critical value for 95 % probability given for N samples (always for the minimal value N at different numbers of measurements of individual parameters). The distribution of parameters which exhibited differences between productive and non-productive boreholes in preliminary evaluation are documented by histograms — Figs. 2, 3, 4, 5 and 6.

Discussion and conclusions

Parameters dependent on sediment texture and structure

Bulk density and porosity undergo compaction and lithification in dependence on the depth. The differences between productive and non-productive boreholes (in both sandstones and claystones) (Tables 1 and 2) are due to different depths of occurrences of processed samples under the recent surface. Compaction and lithification, as shown earlier, are mainly dependent on the depth of 'burial' in the sedimentary basin (Ondra & Hanák 1988). It took place predominantly in the period before tectonic deformation. It is closely related to the basin (collector) properties of sediments. The discussed impact of hydrocarbons on these parameters cannot be evaluated here, though it cannot be excluded.

Parameters independent of texture and structure — contents of radioactive elements

a) The differences in the distribution of Ura in sandstones are manifested by a shift to lower contents in pro-

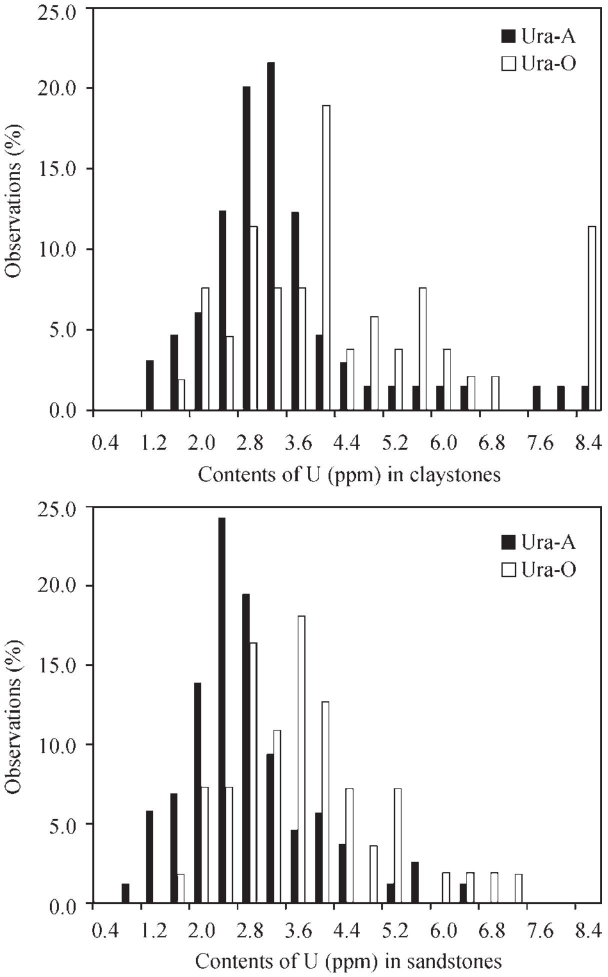

Fig. 2. Uranium contents — deep boreholes.

ductive boreholes as opposed to non-productive boreholes (Fig. 2).

b) The same trend can be observed in claystones and siltstones. It must be noted, however, that particularly in pelites Ura can be related to the primary organic element, which need not coincide with migrating oil and gas. It is confirmed by the highly significant negative correlation (Table 4) between Dm and Ura in claystones from productive and non-productive boreholes. The organic element in claystones binding Ura reduces the Dm value. In sandstones, however, this phenomenon is not of importance, or is not present at all, as the correlation relationships Dm and Ura (Table 3) are contrasting. The contents of the organic element binding Ura in sandstones are substantially lower. In claystones with siltstones there are extreme contents of Ura attaining 25 ppm, most probably due to the content of the organic element. The claystone-siltstone set exhibits extreme Kd and Drad values observed in average values (Table 2) due to samples from the borehole Vranovice with very low contents of potassium and extreme contents of thorium.

c) Positive Drad values — Saunders et al. (1993) are indicative of hydrocarbons. Fig. 3 shows that 12.6 % of sandstone samples from productive boreholes exhibit Drad values higher than 1, in non-productive boreholes only 1.8 %. This difference in the distribution of Drad is considered to be due to the presence of hydrocarbons in 'productive' boreholes. These manifestations are indicated by random choice of samples more numerous in productive boreholes.

d) The histogram for the distribution of K in sandstones (Fig. 4) shows that 23 % of samples from productive boreholes exhibit contents of K lower than 1 %. In the second group (from non-productive boreholes) there are no samples with contents of K lower than 1 %.

e) On the other hand, it is difficult to interpret the differences between the two groups in the histogram for Th contents in sandstones (Fig. 5).

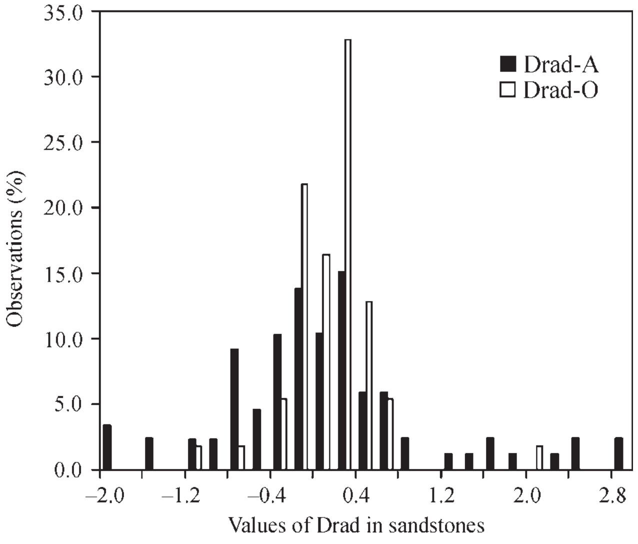

Fig. 3. Bar chart for Drad values — deep boreholes.

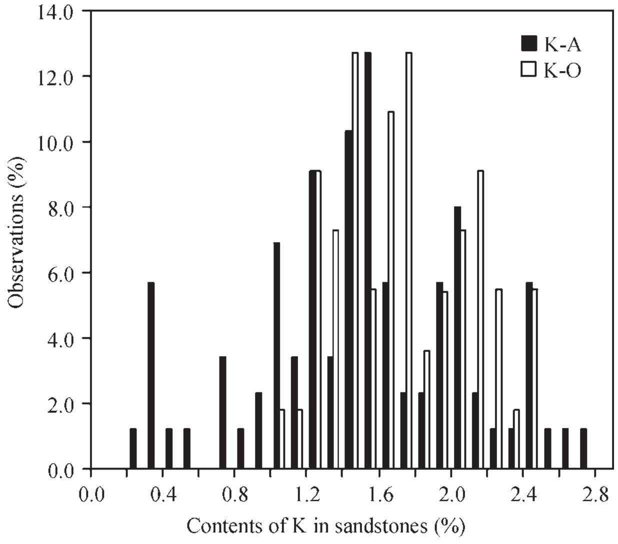

Fig. 4. Bar chart for potassium — deep boreholes.

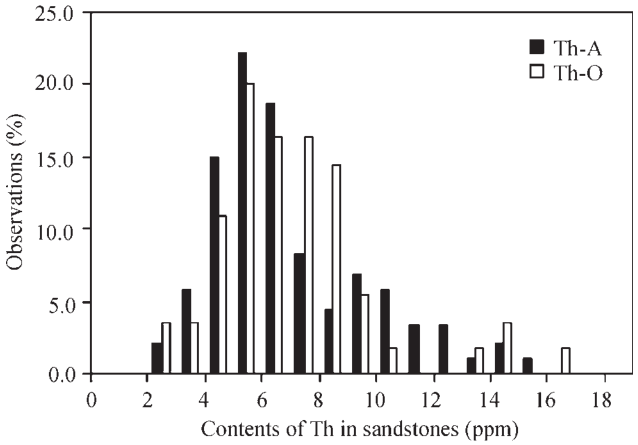

Fig. 5. Bar chart for thorium — deep boreholes.

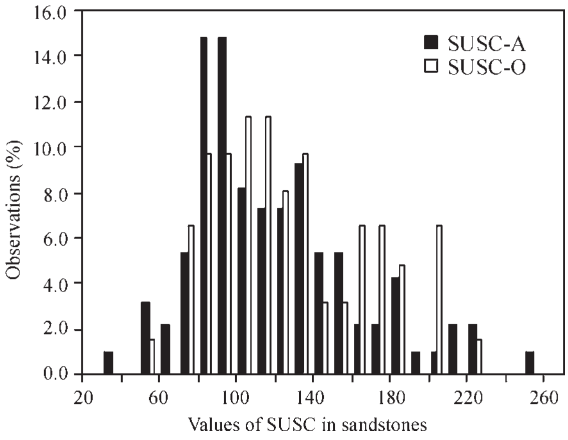

Fig. 6. Bar chart for magnetic susceptibility — deep boreholes.

In conclusion it can be summed up that the analysis of the distribution of Ura and K in the Ždánice-Hustopeče Formation reached by deep boreholes indicates changes in contents of these elements due to the presence of hydrocarbons. Both observed elements are in the process of epigenesis forced out due to the presence of hydrocarbons. It is obvious in psammites, and in accordance with the assumptions of Saunders et al. (1993). Indications of this phenomenon are much stronger in deep boreholes as compared with near-surface portions as deep boreholes reach the contaminated portions nearer the source of hydrocarbons, or even the samples come directly from deposit accumulations. It is obviously necessary for recording the geochemical contamination by means of the used methodology. In other studied parameters the effect of the contact with hydrocarbons has not been observed in the studied set of samples.

On the basis of measurements of the physical-chemical parameters of rock samples from shallow and deep boreholes in the Ždánice Unit a methodology can be recommended for employment in oil prospecting:

- Make use of 'shallow' boreholes in seismic shooting for collection of rock samples for determination of radioactive elements and contents of sulphur.
- Adapt the gammaspectrometric probe for shallow logging in these boreholes.
- Specific petrophysical measurements in selected horizons and lithotypes.

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