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ORIGIN OF RED CLAYS AROUND MISKOLC (NORTH HUNGARY)

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ABSTRACT

During the geological mapping of the northern foreland of the Bükk Mountains red clay occurrences were identified by the Department of Mineralogy and Geology, University of Debrecen. Red clays are underlain by Triassic limestone and Miocene, mainly Sarmatian siliciclastic sediments containing tuffaceous bentonites. Based on the sedimentological (grain-size distribution), mineralogical (micromineralogy, X-ray diffraction, thermal analysis) and major element geochemical (ICP-OES) examinations, it can be established that the studied red clays belong to the less weathered, so-called „reddish clay" type. Montmorillonite prevails among clay minerals, while kaolinite is subordinate. Considering the strong sedimentological and mineralogical similarity with the underlying Miocene sediments, the red clays were probably originated by the weathering of the Miocene sediments under a moderately warm and dry climate. Taking lithostratigraphic features into account, the examined red clays can be classified as members of the Pliocene-Pleistocene Tengelic or Kerecsend Red Clay Formation.

Key words: red clay, Bükk Mountains, Pliocene, Pleistocene

INTRODUCTION

The Department of Mineralogy and Geology in the University of Debrecen has been carrying out the geological mapping of the hilly northern foreland of the Bükk Mountains (Northern Hungary) at a scale of 1:10000. In the course of the field surveys we have identified several patch-like red clay occurrences in the surrounding of Miskolc (Fig. 1). Their thickness and extension are significantly smaller than that of the Southern Transdanubian red clays therefore it has not been observed in detail yet.

The examined clays are brownish red and reddish brown (according to the Munsell Colour System: 2,5YR3/6, 5,0YR3/6, 5,0YR4/4), based on grain-size distribution they are clayey silts and silts with the mixing of various quantity of fine sand.

The red clays are underlain by karrened Triassic limestone (Bükkfennsík Limestone Formation) and Sarmatian siliciclastic sediments (Sajóvölgy Formation) the latter involving acidic tuffs and tuffaceous bentonite beds. They can be found at elevations between 190 and 350 m. Due to the mixing with colluvium and partly to their re-accumulation, it is difficult to explain their genesis. Possible source material might be the Jurassic iron rich pelagic sediments and the insoluble residue of Triassic limestones, however, also the reddening of Pannonian and Miocene sandy-clayey sediments, rhyolitic tuffs, tuffites and aeolian loesses seems to be probable.

The aims of our examinations concerning the less-known red clays are the following:

- sedimentological, mineralogical and geochemical characterization,
- determination of the degree of weathering,
- comparison the red clays of karstic limestone areas with red clays of their hilly Miocene foreland and clearing their relationship,
- comparison of the red clays with other Hungarian occurrences in order to evaluate their age,
- determination of the source material of red clays.

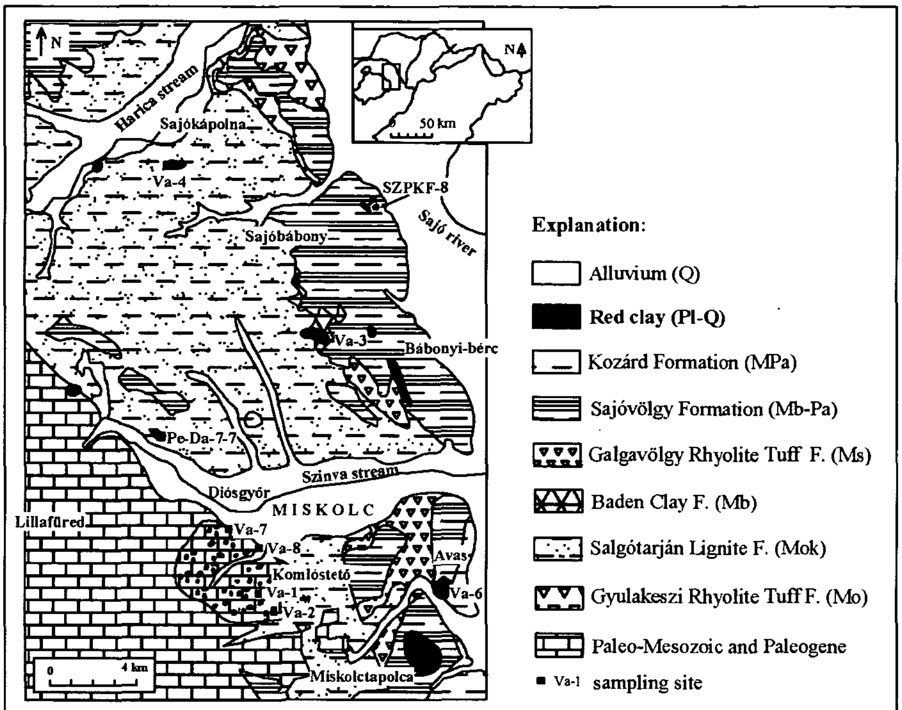
RESEARCH HISTORY

The red clay occurrences of Bükk Mts. characterised by some 10000 m^2 extension and by 0,5-10 m thickness had remained hidden before as geological investigations concentrated mostly on the Mesozoic basement and Miocene sediments neglecting the young formations. Moreover, publications mostly refer to the red clays found in the Central Bükk Mountains and not in its forelands.

Firstly Kerekes (1936) mentioned them supposing that the yellowish and reddish loam-like formations of the Bükk Mountains are originated from eolian material. Jámbor (1959) examining sink hole fillings near the Istállós-kő identified „yellow loess without lime" underlain by „red clayey loess without lime" then „red clay containing tuff quartz" deposited directly on the limestone. He supposed that the latter, containing idiomorphic dihexagonal quartz, ilmenite, zirkon, tourmaline and amphibole in the fraction coarser than 0,1 mm, derived from the rhyolite tuff, tuffite and sand occurring on the Great Plateau of the Bükk Mts. Comparing with cave fillings, he classified them as Rissian-Würmian. He stated that the red clays are not terra-rossa types.

The monograph of the Bükk Mountains edited by Balogh (1964), only shortly touch upon the post-Pannonian terrestrial formations of the Bükk Mountains like multiply transported and weathered rhyolite tuffs and yellow loess-like silts blown out from rhyolite tuff and accumulated in sink holes.

In the course of the 1:25000 scale geological mapping of the Bükk Mountains Pelikán (1992) described doline filling red clays in the surroundings of Répáshuta, Bükkszentkereszt and the Őr-kő. These contain dominantly kaolinite, subordinately illite and smectite as clay minerals. The author



Q: Quarter, Pl-Q: Pliocene-Quarter, MPa: Miocene Pannonian, Mb-Pa: Miocene Badenien-Pannonian, Ms: Miocene Sarmatian, Mb: Miocene Badenien, Mok: Miocene Ottnangian-Karpatian, Mo: Miocene Ottnangian

Fig. 1. Geological map of the hilly surrounding of Miskolc showing the red clay occurrences.

records similar sediments in limestone fissures. According to his verbal communication, the red clays are probably derived from the weathering of rhyolite tuffs which is proved by ilmenite and quartz crystals with sharp edges in the coarse fraction. On the basis of shallow drillings the most frequent covering sediment of the Bükk is a loess-like clayey-sandy silt, containing predominantly illite as clay mineral.

Red clay fillings including bone fossils have been described in several places in the caves of the Bükk Mountains (Hajnóczy cave, Köves-Várad, Horvát-lik, Diósgyőr-Tapolca cave etc). The oldest vertebrate fossils are 700 thousand years old (Kordos 2002), however they mark only the age of the accumulation, while the red clays can be significantly older. In the Hajnóczy cave red clays comprise illite as the most important clay mineral (29-34 %), montmorillonite (0-14 %) and kaolinite (0-16 %) are subordinate. In the Horváth-lik, near Uppony, where fossils from the Riss/Würm interglacial were identified (Füköh and Kordos 1977, 1978), two types of clays occur, the lower one contains kaolinite and illite in nearly equal amounts, while the upper one is characterised by smectite, illite/smectite and illite association. The mineral composition of the former one resembles the kaolinitic weathering crusts of Upper Eocene and Lower Miocene formations (Kozák et al. 1998), while the latter is probably derived from some of the Miocene tuff layers of the Borsod Basin (Viczián 2002).

The construction geological drillings of Miskolc explored usually 1-5, maximum 10 m thick red and reddish brown clay, frequently mixed with limestone debris, however, unfortunately, no information was published on their areal extension, genetic and stratigraphic relations (Juhász 1979).

Significant red clay occurrences, presumably derived by the weathering of volcanic rocks and aeolian material, can be found in the southern foothills of the Bükk and Mátra Mountains (Kerecsend Red Clay Formation) covering Pannonian lignite containing sequences (Császár 1997). In recent years the formation was investigated in detail including provenance. On the basis of mineralogical examinations the authors proposed that the smectite-rich red clays are derived from the older smectite vertisol blanket of the volcanic rocks of the Mátra Mountains (Berényi-Üveges et al. 2002, 2003).

The mineralogical composition, paleogeographical features and the geomorphological situation of red clays were examined by Schweitzer (1993) and Schweitzer and Szöőr (1997). Investigating several occurrences all around the country, they differentiated two groups on the basis of thermal analysis and geochemical aspects, the „typical red clays" of older Pliocene age containing disordered kaolinite and the smectite-rich „reddish clay" of younger Pliocene and Old to Middle Pleistocene age.

Owed to their extent, thickness and fossil content the most well-known red clays of Hungary can be found in southern Transdanubia (Tengelic Red Clay Formation). They have been recently investigated in detail by Koloszár and Marsi (1997).

In the last years Fekete and his colleagues have published several studies about Hungarian red clay localities (Fekete et al. 1997, Fekete and Stefanovits 2002, Fekete 2002) examining 61 soil profiles all over the country. They divided red clays into six groups based on detailed analysis (grain size distribution, X-ray diffraction, thermal analysis and main element concentrations), genesis and geographical situation

MATERIALS AND METHODS

During the geological mapping of the northern and eastern foreland of the Bükk Mountains we identified the setting and red clay occurrences of the area. Red clay samples were taken of the Mesozoic karstic and Miocene foothill areas. Special care was taken collecting autochtonous formations. The samples were collected from surface outcrops and from the upper 6,6 m interval of the SZPKF-8 bentonite exploration drilling (Fig. 1).

To compare clays covering Mesozoic and Miocene surfaces we pointed out two profiles: the artificial outcrop in Komlóstető and the SZPKF-8 bentonite exploration drilling (Kozák et al. 2003). To detect their provenance we collected samples from the underlying formations (Mesozoic limestone, Sarmatian silt, respectively), from the potential source rocks of the area (Sarmatian tuffaceous bentonite, typical loess), from the weathering crust of some Mesozoic sedimentary rocks containing high amount of iron, like the Csipkéstető Radiolarite Formation (Csipkés hill) and Berva Limestone Formation (Cserépfalu, Hór valley) containing 23,11 and 10,66 % iron.

All the samples were analysed by traditional sieving and Pappfalvi hydrometring method to characterise the grain-size distribution of the sediments. Statistical grain-size parameters were calculated by the methods presented by Gyuricza et al. (1999). The fraction above 0,08 mm was examined on 100 grains per samples by optical microscope determining the characteristic mineral components. The orientating quick analysis was only prepared for the light minerals in consideration of the high amount of igneous quartz to confirm and complement the information gained by XRD analysis.

The powdered material was analysed by XRD and thermal analysis methods in order to recognise the clay mineral content. X-ray diffraction analyses were carried out on a Philips PW 1730 diffractometer under the following conditions: Cu anti-cathode, 40 kV and 30 mA tube current, graphite monochromator, goniometer speed 2 °/minute. The more detailed research of the clay minerals was performed on the <5 μm clay mineral fraction separated by Atterberg cylinders. From the separated clay-mineral fraction oriented preparations were made and these were investigated by XRD in an untreated state, treated with ethylene-glycol (60 °C/9 hours), and heated (490 °C/4 hours). Semi-quantitative results were made by the methods of Hardy and Tucker (1988) and other authors, which are particularly useful for comparative purposes. In these methods the intensities of the individual clay components are measured on an oriented mount, after various treatments (air dried, glycol solvated and heated to 490°C) and the sum is normalized to 100%. Results were compared with quantitative data obtained by thermal analysis.

The thermal analyses were completed by Derivatograph PC, simultaneous TG, DTG, DTA recording, in a ceramic or corundum crucible, with a heating speed of 10 °C/minute up to 1000 °C and with Al_2O_3 as inert material. Both the XRD and the thermal analytical examinations were made in the laboratories of the Hungarian Geological Institute.

In order to determine the major elements the powdered samples were mixed with double quantity of lithium-metaborate. Then this treated sample was dissolved in 0,2 mol/dm^3 HCl. The element content was determined by ICP-OES. The determination of Si was made gravimetrically; the samples were heated with HF when Si evaporated as SiF_4. The total iron content was determined by spectrophotometry as well. Sample preparation, gravimetric and spectrophotometric measurements were carried out in the Department of Minrealogy and Geology while ICP-OES analysis were performed in the Regional Analytical Centre of the Agricultural Centre (Debrecen Universtiy).

For the better genetic interpretation of the results obtained, we used numerous works publishing the results of similar analysis on other Hungarian red clay localities (Fekete and Stefanovits 2002, Fekete 2002, Kovács 2004).

RESULTS AND DISCUSSION

Grain-size distribution

According to the Bárdossy's scale (1961) the examined materials are clayey silts, silts, sometimes sandy clayey silts. The percentage distribution of grain size fractions and the most important statistical parameters can be seen in Table 1.

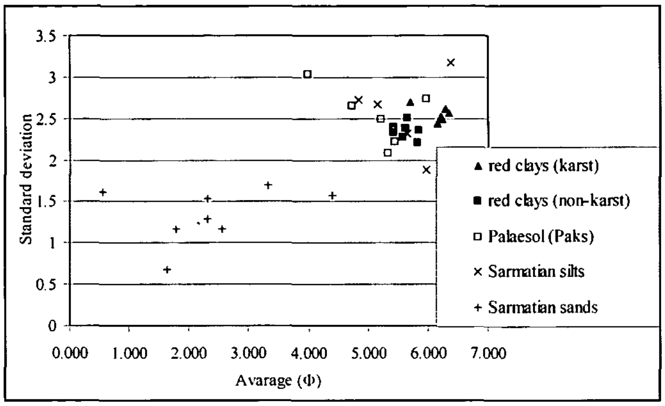


Fig. 2. Comparing red clays with the underlying Sarmatian sediments based on the avarage and the standard deviation of grain size distribution. Source of data: red clays, this study; palaesols, Pécsi et al. 2002; Sarmatian sediments: Kozák et al. 2003.

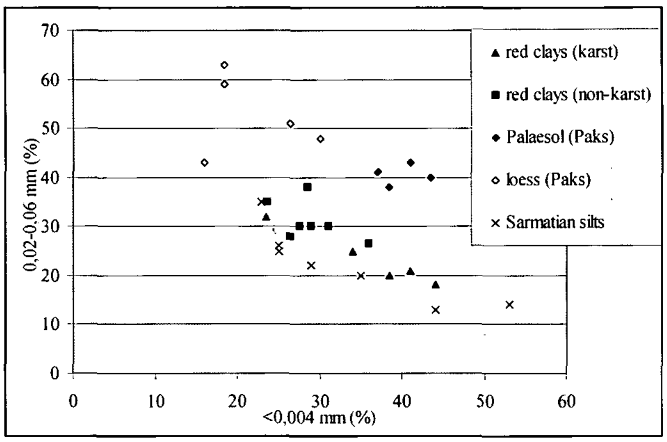


Fig. 3. Comparision of the fine fractions of red clays with loesses, paleosols (Paks) and Sarmatian silts based on the amounts of the <0.004 and 0,02-0,06 mm fractions. Source of data: red clays, this study; palaesols, Pécsi et al. 2002; Sarmatian sediments: Kozák et al. 2003.

The analysed red clay samples show characteristic bimodal distribution. The first mode is in the clay fraction (<0,002 mm: 13-28 %), the second is between 0,013 and 0,035 mm presenting some variability among the samples. The standard deviation is between 2,09 and 2,67 so they are weakly sorted, the skewness is slightly positive (0,21-0,39) and the kurtosis is approximately normal (0,80-1,14).

The grain size distribution of red clays of the karst and Miocene areas are basically similar, although the former ones contain 5-10 % more clay fraction and their second mode is finer (0,016 mm).

The standard deviation - average diagram (Fig. 2) presents that the grain size distribution of red clays is largely similar to that of the underlying Sarmatian shallow marine silts and the covering colluvium.

From the aspect of judgement of aeolian sediments as source material, the rate of the so-called loess fraction (0,02-0,06 mm) is very important. In the Paks loess profile the palaeosols derived from loess preserve their loess fraction (Pécsi et al. 2002), which is usually 30-50 % in the case of every loess derived paleosol. In Fig. 3 it can be seen that the

Table 1. Grain size distribution (v %) and the most important statistical parameters of red clays.

Sampling sites		<0.002 mm	0.002-0.004 mm	0.004-0.008 mm	0.008-0.016 mm	0.016-0.032 mm	0.032-0.063 mm	0.063-0.1 mm	0.1-0.25 mm	>0.25 mm	Average ()	Standard deviation	Skewness	Kurtosis
Drilling on Miocene surface														
Sajókeresztúr, Upper cellars SZPKF-8 bentonite drilling	0,8-1,0 m	19	6	12	17	17	18	8	3	0	5.608	2.385	0.278	0.977
	1,6-1,8 m	18	11	11	16	17	19	7	1	0	5.572	2.277	0.387	1.143
	2,4-2,6 m	21	7	9	15	18	24	5	1	0	5.42	2.335	0.308	0.908
	3,4-3,6 m	17	9	11	13	21	19	7	1	2	5.65	2.503	0.319	1.097
	4,4-4,6 m	16	8	10	14	18	23	8	2	1	5.43	2.225	0.304	0.94
	5,4-5,6 m	13	5	10	17	18	25	9	2	1	5.318	2.086	0.293	1.088
	6,6-6,8 m*										5.664	2,613	0,257	0,803
Outcrops, pits on Miocene surface														
Sajókápolna (southern border)	Va-4 1,0 m	15	7	7	12	15	16	18	9	1	4.533	2.668	0.304	0.902
Miskolc Bábonyi-bérc	Va-3 1,45-1,6 m	26	10	10	10	15	19	9	1	0	5.587	2.44	0.298	0.893
Miskolc Pereces cellars	Pe-Da-7-7 2,5 m	20	8	12	16	19	15	8	2	0	5.621	2.391	0.271	0.974
Diósgyőr (Mária street)	Va-7 1,0 m	13	5	6	12	13	13	12	10	16**	2.953	3.177	0.038	1.139
Diósgyőr cemetery(pit)	Va-8 2,0 m	18	8	18	14	17	17	5	2	1	5.815	2.208	0.286	1.055
Miskolc South Avas (pit)	Va-6 2,0 m	17	10	10	17	22	12	8	3	1	5.456	2.367	0.225	1.098
Karstified limestone surface														
Komlóstető (doline)	Va-1 1,8 m	9	3	9	18	14	13	7	4	23**	0.682	3.705	-0.311	1.286
Komlóstető karren surface	Va-2 0,15-0,2 m	27	9	14	13	21	13	3	0	0	6.144	2.441	0.268	0.83
	Va-2 0,5-0,6 m	28	10	15	19	13	10	5	0	0	6.232	2.498	0.209	0.844
	Va-2 0,9-1,0 m	28	10	13	20	14	11	4	0	0	6.211	2.523	0.271	0.825
	Va-2 1,3-1,4 m	12	6	9	6	9	10	11	11	26**	1.591	3.788	0.086	1.042

* underlying silt

**admixture of fragments of the underlying rock

Table 2. Mineral composition of red clays (bulk sample) in %, X-ray diffraction analysis.

Sampling sites		montmorillonite	illite/montm.	illite	muscovite	kaolinite	chlorite	quartz	K-feldspar	plagioclase	amorphous	goethite	hematite	calcite	dolomite	gypsum	Hb(001)mm	Hb(001)illite
Drilling on Miocene surface																		
Sajókeresztúr, Upper cellars **SZPKF-8** bentonite **drilling** (red clays)	0,0-1,0 m	18	5	9	0	3	0	55	5	8	4	0	2	0	0	0	0,7	0,26
	1,6-1,8 m	17	4	9	0	0	2	56	3	9	5	0	2	0	2	0	0,82	0,53
	1,8-2,0 m	17	5	8	0	2	0	57	4	10	4	2	0	0	1	0	0,43	0,3
	2,4-2,6 m	18	4	9	0	3	0	51	5	11	5	2	1	0	0	0	0,54	0,54
	3,4-3,6 m	19	5	9	0	0	2	58	4	7	5	0	2	0	0	0	0,66	0,51
	4,4-4,6 m	20	4	4	0	0	2	63	3	6	6	2	0	0	0	0	1,0	0,86
	5,4-5,6 m	19	6	9	0	0	2	57	3	7	5	0	0	0	0	0	1,37	0,47
SZPKF-8 drilling (underlying Sarmatian sediments)	6,6-6,8 m	20	6	10	0	0	4	51	4	9	6	2	0	0	0	0	1,11	0,71
	10,6-10,8 m	15	3	0	10	3	0	52	4	13	4	0	0	0	0	0	0,79	0,22
Outcrops, pits on Miocene surface																		
Sajókápolna (southern border)	Va-4 1,0 m	21	0	5	0	0	3	57	2	5	5	2	1	0	0	0	1,04	0,36
Miskolc Bábonyi-bérc	Va-3 1,45-1,6 m	26	0	5	0	0	3	51	2	5	0	1	1	0	0	6	0,95	0,58
Miskolc Pereces cellars	Pe-Da-7-7 2,5 m	23	0	4	0	3	0	54	1	6	6	2	0	0	0	0	1,17	0,56
Diósgyőr (Mária street)	Va-7 1,0 m*	13	0	5	0	2	0	70	1	3	3	2	1	0	0	0	1,04	0,46
Diósgyőr cemetery (pit)	Va-8 2,0 m	27	4	6	0	0	3	43	1	6	5	2	1	0	0	0	1,04	0,28
Miskolc South-Avas (pit)	Va-6 2,0 m	20	0	4	0	3	0	58	2	5	3	2	1	0	0	0	0,7	0,52
Karstified limestone surface																		
Komlóstető (doline)	Va-1 1,8 m*	12	3	7	0	0	4	46	2	3	4	2	1	16	0	0	0,9	0,3
Komlóstető (doline)	Va-1 Fe cemented concretion	0	4	0	0	0	0	66	2	4	7	4	10	3	0	0	-	-
Komlóstető karren surface	Va-2 0,15-0,2 m	16	6	10	0	3	0	48	1	4	6	4	0	1	0	0	0,41	0,65
	Va-2 0,5-0,6 m	20	7	9	0	0	3	46	2	5	5	3	0	0	0	0	0,78	0,49
	Va-2 0,9-1,0 m	25	0	6	0	6	0	46	2	5	6	3	1	0	0	0	0,68	0,63
	Va-2 1,3-1,4 m*	4	2	4	0	14	0	7	1	0	3	3	0	62	0	0	0,74	0,43

Made by Kovács-Pálffy, P., Geological Institute of Hungary, XRD laboratory
* admixture of rock fragments of the underlying rock

red clays and loesses are different from each other in a diagram illustrating 0,02-0,06 mm fraction and the fraction below 0,004 mm. The amount of the 0,02-0,06 mm fraction of red clay is significantly lower than that of the loesses and loess derived paleosols at Paks. As compared with the Sarmatian silts, the slightly higher amount of the 0,02-0,06 mm fraction in the red clays can be explained by the re-working of Karpatian and Sarmatian nearshore (firstly lower shoreface facies) sediments, the grain size distribution of which is very similar to that of the loess (Csiga-hill profile, Nagybarca, Dávid et al. 2005). Based on these considerations, the aeolian origin of red clays does not seem to be probable or it cannot be prevailing.

Micromineralogical analysis

The amounts of the grains above 0,08 mm are 2-5 % in the case of the samples which do not contain fragments of the underlying rocks. The most frequent mineral of the coarse fraction is igneous quartz (50-80 %), which is a variably rounded and weathered material, underwent mostly longer transportation. Rarely broken fragments of fresh, finely crystallized dihexagonal quartz can be found as well. They are more frequent, but still subordinate in the karstic localities. The latter observation proves the admixture of acidic tuffs. Among smaller grains some feldspar fragments were also found.

Rounded Paleo-Mesozoic quartzites, siliceous slates and quartz referring longer transportation are less frequent (0-15 %). Their varieties are missing from the red clays of karst areas, instead of them limestone and calcite fragments occur.

Limonite and hematite concretions are very frequent (5-50 %) in all of the samples.

It is remarkable that mainly tuff origin quartz was described on the Great Plateau which was not covered by Late Miocene sediments (Jámbor 1959 and Pelikán 1992). Consequently in the coarse fraction of red clays lying on the uplifted plateau of the Bükk Mountains (at elevation 800-900 m) tuff quartz is more frequent, while in the Miocene foreland (at elevation 190-350 m) it is very rare. Komlóstető (250 m), locating at the margin of the Bükk Mts., is about the transition containing more tuff quartz. It suggests that the source material of Bükk red clays is rhyolite tuff and tuffite, while on the Miocene surface the source materials are rather Miocene tuffaceous siliciclastic sediments.

X-ray diffraction

The results of the XRD analysis of 19 powdered bulk samples (Table 2) show that except of phyllosilicates the most frequent mineral of red clays is quartz (43-63 %) while plagioclase feldspar (3-11 %) and K-feldspar (1-5 %) are typical. The systematic presence of the amorphous material is significant (4-6 %). Among clay minerals montmorillonite prevails (16-27 %), illite (4-10 %) is subordinate, while kaolinite (0-6 %) and chlorite (0-3 %) is accessorial. The high proportion of quartz, montmorillonite, feldspars and amorphous material together with micromineralogical data reflect tuffaceous origin. High amount of quartz may be derived from Miocene shallow marine sands and silts.

The mineral composition of red clays of the Mesozoic karst and the Miocene area do not differ from each other

Table 3. Clay mineral composition of red clays (<5 μm) in %, X-ray diffraction analysis.

Sampling sites		**montmorillonite**	**chlorite/montm.**	**illite/chlorite**	**illite**	**muscovite**	**kaolinite**	**chlorite**	**Hb(001) mm untreated**	**Hb(001)mm eg****	**v/p mm eg****
Drilling on Miocene surface											
Sajókeresztúr, Upper cellars	0,8-1,0 m	38		5	28		16	13	1,52	1,32	0,43
SZPKF-8 bentonite **drilling**	2,4-2,6 m	46		4	29		12	9	1,36	1,24	0,55
(red clays)	4,4-4,6 m	66		3	14		10	7	1,48	1,48	0,65
SZPKF-8 drilling (underlying	6,6-6,8 m	41		4	27		15	13	1,84	1,4	0,51
Sarmatian sediments)	10,6-10,8 m	66			2	22	6	4	1,52	1,08	0,92
Sajókápolna (southern border)	Va-4 1,0 m	58		3	23		8	8	1,44	1,20	0,55
Miskolc Bábonyi-bérc	Va-3 1,45-1,6 m	58		3	17		12	10	1,64	1,52	0,61
Miskolc Pereces cellars	Pe-Da-7-7 2,5 m	59		3	13		15	10	1,56	1,64	0,60
Diósgyőr (Mária street)	Va-7 1,0 m*	18		6	61		8	7	0,60	1,08	0,49
Diósgyőr cemetery (pit)	Va-8 2,0 m	41		5	27		16	11	1,56	1,56	0,49
Miskolc South Avas (pit)	Va-6 2,0 m	43		5	26		14	12	1,52	1,52	0,47
Karstified limestone surfaces											
Komlóstető (doline)	Va-1 1,8 m*	20	12	5	32		16	15	1,16	1,36	0,00
Komlóstető karren surface	Va-2 0,15-0,2 m	11		5	40		24	20	0,61	0,71	0,00
	Va-2 0,5-0,6 m	7		6	46		22	19	1,1	0,31	0,00
	Va-2 0,9-1,0 m	21	9	4	14		33	19	1,40	1,08	0,18
	Va-2 1,3-1,4 m*	6			6		88		1,56	1,12	0,58
Weathering crusts of iron-rich	Hór valley	1		1	90		2	1			
Jurassic rocks	Csipkés hill	55		2	17		14	12	1,04	1,56	0,84

Made by Kovács-Pálffy, P.,Geological Institute of Hungary, XRD laboratory
* admixture of rock fragments of the underlying rock ** treated with ethyleneglycol

according to the examination of bulk samples taken. However, regarding the fine fraction (< 5 μm) there are significant differences between the red clays covering the karstified basement and its Miocene foreland (Table 3). In the samples taken of karst basement the montmorillonite content is lower while kaolinite, chlorite and illite contents are higher. The difference points to the mixing of older subtropical weathering crusts developed on the surface of the limestone. The higher proportion of kaolinite prove more advanced weathering during the development of the weathering crust.

On the basis of the high proportion of montmorillonite, feldspars and of amorphous material, the red and reddish brown clays in the surrounding of Miskolc are probably derived from the Sarmatian shallow marine siliciclastic sequence containing bentonites and volcanoclasts with acid and neutral composition. The width at half height of 001 basal reflection of montmorillonite, Hb (001) shows large degree of disorder. This points to the possibility of that montmorillonite is derived not only from the devitrification of tuffaceous material, but also from the further weathering during the formation of red clay. The clay mineral content of the weathering crust of Jurassic iron containing pelagic rocks is very different from that of the red clays, those comprise 90 % illite (Hór valley sample).

Studying the vertical changes of clay mineral composition and the width of half height (Hb) of montmorillonite of the samples collected from the SZPKF-8 bentonite drilling, it can be seen that the red clays and the underlying Sarmatian silts are considerably similar (Fig. 4 and Fig. 5A). The clay mineral content does not change at the bedding plane (6,6 m). Mineralogical change can be found at a depth of 9 m, below which muscovite occurs and Hb of montmorillonite and illite decreases. Higher values and also higher variability of Hb values point to the effect of weathering above the 9 m limit in the borehole. Based upon the facts mentioned, the close connection between the red clays and Sarmatian sediments can be established.

Studying the section of the depression filling red clay on karren limestone surface in Komlóstető it can be seen that more or less homogeneous smectite-rich red clay occurs from the surface down to 1,2 m and below that yellow clay lies including limestone debris. The latter contains a small amount of quartz, illlite and montmorillonite, but it is rich in kaolinite. The X-ray diffraction patterns of the two types can be seen in Fig. 5B. Between these beds there is a sharp bedding surface with no mineralogical transition. The high kaolinite content of the yellow clay refers to a warmer and more humid climate and to stronger weathering. The mineral content of it is remarkably similar to that of the red and yellow clays interbedding between coarse clastic sediments lying on Carboniferous limestone and slate near Lázbérc (Uppony Mountains), which is older than the overlying sequence, probably Eggenburgian or Ottnangian (Kozák et al. 1998). Consequently, at Komlóstető the yellow clay mixed with limestone debris is likely to be of Lower Miocene age. The overlying red clay according to the above mentioned similarities is presumably derived from the Miocene siliciclastic sequence containing acid tuffs, which formerly had covered certain parts of the Bükk Mountains. The red clays, investigated in the centre of the mountains, on higher level, contain less quartz and more illite and kaolinite (Pelikán 1992, Fekete 2002, Füköh and Kordos 1977, 1978). The difference can be explained by the lack of the underlying clastic sediments, by stronger weathering and by mixing with older weathered material.

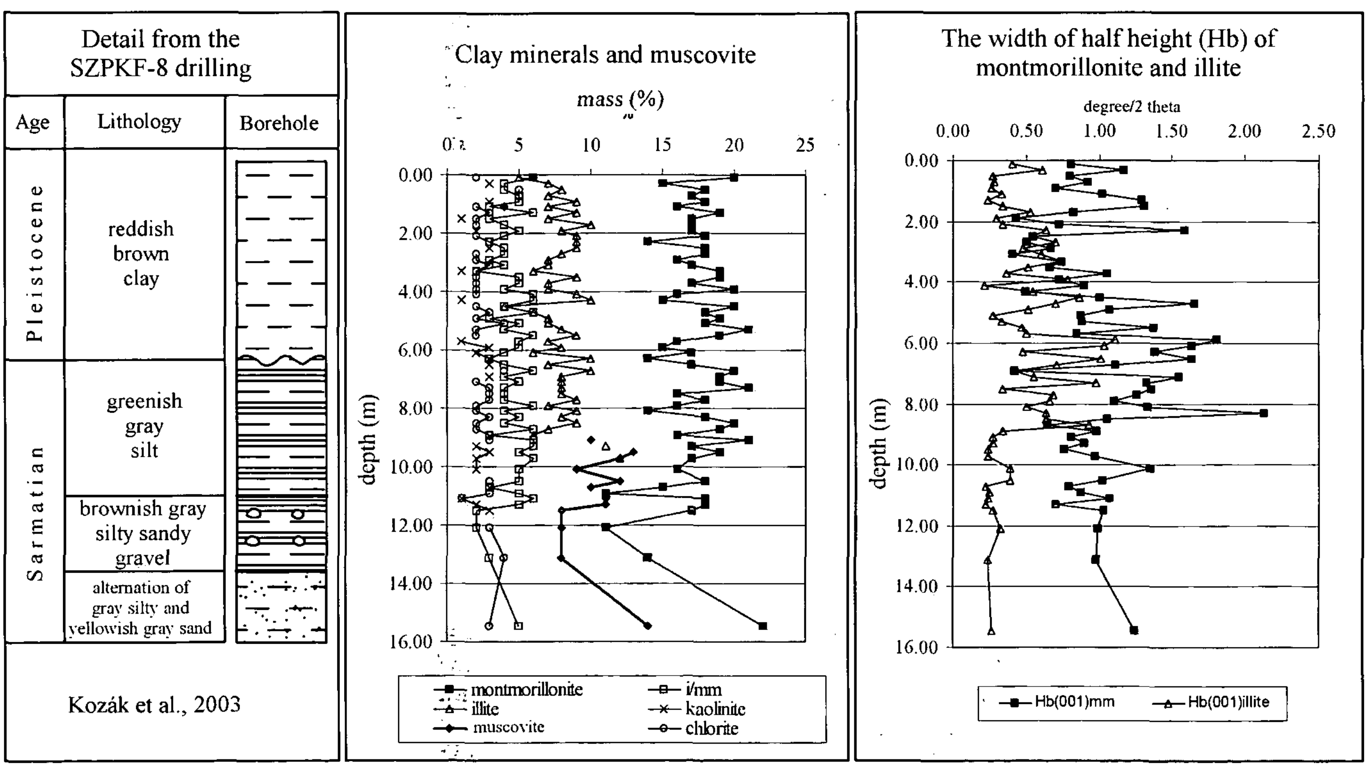


Fig. 4. Clay mineralogical features of the upper part of the SZPKF-8 bentonite drilling including the red clays.

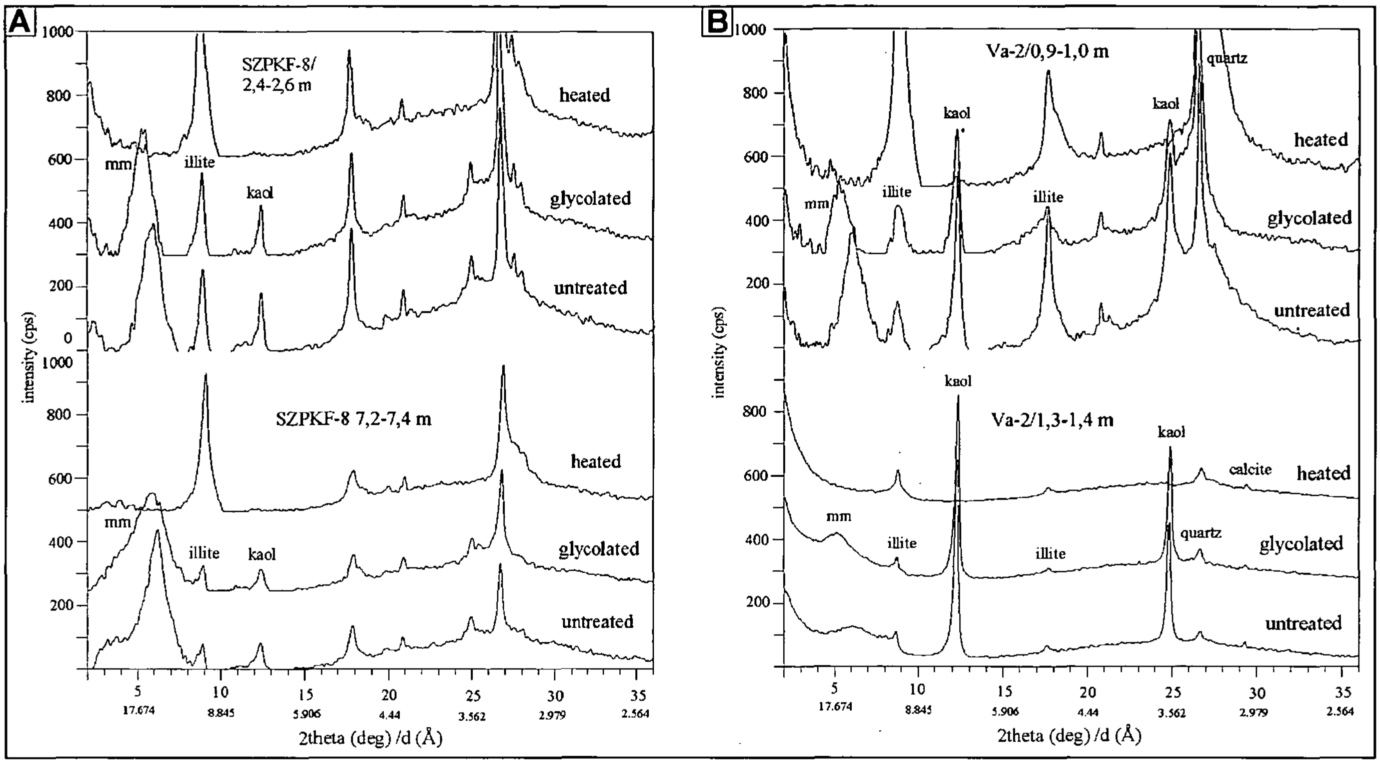


Fig. 5. X-ray diffraction patters of *(A)* the <5 µm fraction of red clay (SZPKF-8 2,4-2,6 m) and Sarmatian silt (SZPKF-8 7,2-7,4 m); *(B)* the <5 µm fraction of red clay (Va-2 0,9-1,0 m) and the underlying yellow clay (Va-2 1,3-1,4 m) at Komlóstető.

Comparing the clay mineralogy of red clays in the surrounding of Miskolc and in other Hungarian red clay localities (Fekete et al. 1997, Fekete and Stefanovits 2002, Fekete 2002, Kovács 2004) (Fig. 6) the examined formations can be classified as immature weathering crusts which can be separated well from the stronger altered red clays of the Transdanubian Mountain Range and the Northern Borsod Karst, comprising bauxite minerals and kaolinite. In the Transdanubian Hills (Tengelic Red Clay Formation) and in the pediments of the North Hungarian Mountain Range red clays with similar degree of weathering and with high smectite contents cover rhyolite tuff, Sarmatian and Pannonian siliciclastic formations often containing bentonite bearing beds. These are of late Pliocene, early and middle Pleistocene age (Viczián 2002).

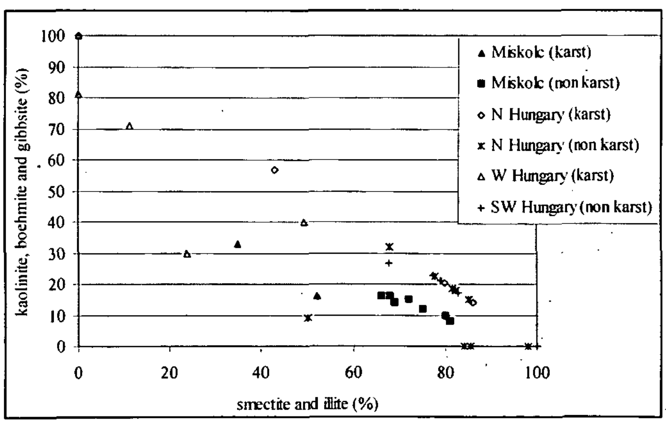


Fig. 6. The clay and bauxite mineral composition of the fine fraction of Hungarian red clays (excluding other components). Source of data: Miskolc, this study; others: Fekete and Stefanovits (2002) and Fekete (2002).

The mineral content of the red clays located in the study area is remarkably similar to SE Transdanubian Tengelic Red Clay Formation investigated in detail in the Udvari-2A and Diósberény-1 boreholes (Koloszár 1997, Földvári and Kovács-Pálffy 2002). The mineralogical criteria, including the high montmorillonite and typically low carbonate contents determined by the authors match with almost all of our samples which presumes very similar genesis and age. There are, however, lower kaolinite and goethite contents in the SE Transdanubian occurrences.

Thermal analysis

The control thermal analytical examinations carried out on some of the samples (SZPKF-8. drilling 0,8-1,0 m, 3,8-4,0 m, 7,2-7,4 m, 15,4-15,5 m, Va-2 0,9-1,0 m, 1,3-1,4 m, Va-6 2,0 m, see Fig. 7.) basically confirmed the results of X-ray diffraction: the high montmorillonite (mainly of Ca type) and lower illite contents. However, conspicuous difference appeared in the values of kaolinite contents. Thermal analysis detected 9-13 % of kaolinite, while X-ray diffraction showed only 0-6 %. The significant difference may be explained mainly by different methods of quantitative analysis, and also by the presence of disordered kaolinite. Disordered kaolinites called „kaolinite without basal reflection" or „degraded kaolinite" are mentioned in the literature (Bidló 1980), typically in the case of red clays deposited on Triassic and Jurassic carbonate formations.

Based on the analysis, the colour of the red clays is caused by the significant amount of goethite, the contents of which oscillates between 5 % and 12 %. Goethite contents

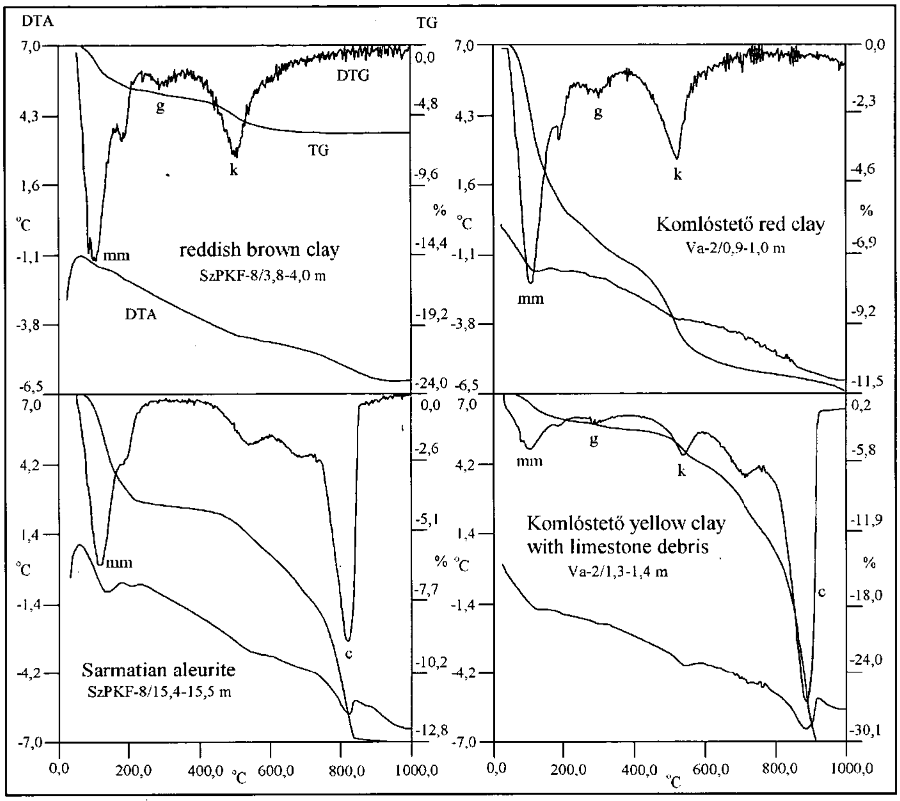


Fig. 7. DTA, TG and DTG curves of red clays and their potential source materials (mm: montmorillonite, k: kaolinite, g: goethite, c: calcite).

obtained by thermal analysis are substantially higher than those obtained by XRD (see Table 2) and slightly higher than it might be expected on the basis of Fe_2O_3 values (see Table 4). The difference partly can be explained by the assumption that a portion of goetheite is amorphous or poorly cystalline, which cannot be detected on the X-ray diagram. The high amounts of goethite formed by the weathering of various andesitic volcanic products (andesitic tuff, flow breccia, placer) occurring in the Sarmatian sequence.

Comparing the analytical results of the drilling SZPKF-8, it can be seen that the clay mineral contents of red clays are basically similar to the Sarmatian silts. The only differences are the lower kaolinite and goethite contents of the latter sample (15,4-15,5 m), marking reddening. The high montmorillonite content of red clays was probably inherited from the Sarmatian sediments. It is also confirmed by the similarity of the low temperature section of derivatograms – among others the dual dehydration of Ca-montmorillonite.

The Sarmatian sediments – examined in several drilling cores and outcrops – usually contain no or only a small amount of carbonate. Signs of dissolution of calcareous shells occurring in some of the layers indicate the dominance of carbonate poor facies. Nevertheless we could observe some well defined carbonate rich strata. The significant carbonate content of the sample from 15,4-15,5 m (SZPKF-8 drilling) can be explained by this local phenomenon. Comparing the samples from Sajóbábony (SZPKF-8 drilling), Komlóstető and the Avas hill, considerable similarity can be observed indicating the same origin. Although higher kaolinite and goethite contents of samples from the karst area can be explained by more intense weathering, probably in an earlier period.

In the central parts of the Bükk Mountains red clays with very low montmorillonite and high kaolinite content (e.g. Berva) and with high montmorillonite content can also be found (Hollóstető, Szarvaskő, Miskolctapolca). According to our opinion the former ones might be of Lower Miocene or older while the latter ones might be of Pliocene-Pleistocene aged.

According to the opinion of Földvári, M. (personal communication) the clay mineral content of red clays examined in this paper fit best for that of the red clays located in volcanic or tuffaceous environments (Salgótarján, Szurdokpüspöki, Gyöngyöstarján).

According to the nomenclature of Schweitzer and Szöőr (1997), introduced mainly on the basis of thermal analytical examinations the red clays studied by us around Miskolc can be classified as „reddish clays". Schweitzer (1993) mentioned typical red clays from Görömböly-Tapolca (near the karst surface of Miskolctapolca). On the other hand we found red clays rich in montmorillonite with a thickness of 3 m in the Avas hill which is probably derived from Miocene material. The sample from Görömböly-Tapolca shows that the older kaolinite rich red clays covering Triassic karst surfaces could have been transported to the Miocene foreland for short distances.

Major elements

The results of major element analysis can be seen in Table 4. For comparison some characteristic results of the Tengelic Red Clay Formation are presented at the bottom of the table (after Kovács 2004). Two indices representing the degree of weathering are displayed besides the element ratios and these refer to the paleoclimate as well.

The major element concentrations can be best interpreted when they are compared to the values of the widely used post Archean Australian Shale (PAAS, after Taylor and McLannon 1985) which reflect the average chemical composition of marine fine sediments. The PAAS-normalized spider diagrams for red clays and potential bedrock samples are shown in Fig. 8. Compared to the PAAS only SiO_2 contents are enriched due to the high amount of detrital components, however the concentrations of MgO and alkalies are significantly lower. It can be seen that the major element concentrations of red clays show moderate dispersion both in Miocene and karst areas. Only the MnO contents vary significantly the concentrations of which, however, are very low. Two samples (SZPKF-8/4.4-4.6 m, Va-7) differ a little from the others caused by the admixture of some detrial components. Red clays lying on karstified limestone and Sarmatian

Table 4. Major element composition and indices showing the grade of weathering of red clays (%).

Sampling sites		SiO_2	TiO_2	Al_2O_3	Fe_2O_3	MnO	MgO	CaO	Na_2O	K_2O	LOI	Sum.	CIA*	$SiO_2/(Al_2O_3+Fe_2O_3)$
Drilling on Miocene surface														
Sajókeresztúr, Upper cellars **SZPKF-8** bentonite **drilling**	0,8 - 1,0 m	67,5	0,85	13,1	5,09	0,08	0,97	0,76	0,72	1,46	10,3	100,80	82,6	3,71
	2,4-2,6 m	67,4	0,85	12,8	4,86	0,08	1,06	0,74	0,86	1,6	8,21	98,56	78,1	3,80
	4,4-4,6 m	75,1	0,59	8,56	3,63	0,07	0,6	0,51	0,29	0,99	8,44	98,79	82,0	6,15
	6,6-6,8 m	70,1	0,84	13,6	4,38	0,07	1,04	1,18	0,44	1,6	5,55	101,2	79,7	
	10,6-10,8 m	74,4	0,85	11,4	3,17	0,04	1,18	1,06	0,84	1,75	2,85	100,63	77,7	
Outcrops, pits on Miocene surface														
Sajókápolna (southern border)	Va-4 1,0 m	68,1	0,69	11,82	4,64	0,05	0,92	0,77	0,58	1,36	9,49	98,41	79,8	4,14
Miskolc Bábonyi-bérc	Va-3 1,45-1,6 m	60,2	0,88	14,2	5,49	0,1	1,05	0,92	0,69	1,37	11,8	96,47	81,3	3,05
Miskolc Pereces cellars	Pe-Da-7-7 2,5 m	69,5	0,84	12,4	4,74	0,02	0,79	0,60	0,53	1,18	8,26	98,84	84,2	4,05
Diósgyőr (Mária street)	Va-7 1,0 m**	76,0	0,61	9,28	3,70	0,07	0,61	0,54	0,39	0,90	7,07	99,13	84,5	5,86
Diósgyőr cemetery (pit)	Va-8 2,0 m	62,91	0,85	14,61	5,74	0,03	0,93	0,77	0,59	1,01	12,0	99,43	84,6	3,09
Miskolc South Avas (pit)	Va-6 2,0 m	67,8	0,90	12,7	4,72	0,13	0,85	0,83	0,65	1,45	10,1	100,05	82,8	3,90
Karstified limestone surface														
Komlóstető (doline)	Va-1 1,8 m**	59,8	0,94	12,3	4,96	0,09	0,87	6,23	0,52	1,37	11,3	98,37	83,6	3,46
	Fe cemented concretion	81,6	0,99	9,95	3,61	0,09	0,60	0,52	0,74	1,63	0,28	99,95		
Komlóstető karren surface	Va-2 0,15-0,2 m	68,6	0,97	13,2	4,92	0,1	0,90	1,31	0,35	1,56	8,65	103,06	77,6	3,79
	Va-2 0,5-0,6 m	70,3		13,3	4,82	0,09	0,87	0,94	0,30	1,5	6,27	101,54	81,6	3,87
	Va-2 0,9-1,0 m	55,9	0,87	16,6	6,73	0,04	0,92	0,86	0,40	1,35	13,4	96,96	87,5	2,40
	Va-2 1,3-1,4 m**	19,1	0,31	9,35	3,16	0,01	0,28	51,7	0,26	1,03	3,75	90,22		1,52
Comparative samples														
Felsőtárkány (SW Bükk)	own sample	52,7	0,37	13,3	6,42	0,15	0,62	2,24	0,12	1,85	15,1	92,86	84,4	2,68
Szekszárd (SW Transdanubia)	Kovács, J. (2004)	62,1	0,94	17,4	6,7	0,05	2,61	0,9	0,7	2,4	5,73	99,53		2,58
Szulimán (SW Transdanubia)	Kovács, J. (2004)	55,0	0,93	19,0	6,8	0,09	1,66	1,7	0,3	1,9	12,5	99,88		2,13
Beremend (Villány Mts.)	Kovács, J. (2004)	25,6	1,02	36,1	17,3	0,14	0,62	0,5	0,0	0,1	16,9	98,28		0,48

Made by Kovács, B.,University of Debrecen, Agricultural Centre, Regional Analytical Centre and Papp, I., University of Debrecen, Department of Mineralogy and Geology

* CIA (Chemical Index of Alteration) = $[Al_2O_3/(Al_2O_3+CaO'+Na_2O+K_2O)]\times 100$, where CaO' is the Ca in the silicate phase (Nesbitt and Young 1982, 1984, Varga, et.al. 2002)

** admixture ot rock fragments of the underlying rock sediments don not differ from each other. Only the yellow clay (Va-2/1.3-1.4 m), deposited on karstified limestone, differs significantly from the red clay samples as a consequence of its different origin.

The ratios of Al_2O_3 to TiO_2 and Fe_2O_3 are rather constant both in the case of red clays and in the underlying Sarmatian silts (Table 4) which indicate that red clays are derived from the Sarmatian formations.

One of the most accepted index of weathering degree is the CIA (Chemical Index of Alteration, after Nesbitt and Young 1982, 1984, Varga et al. 2002) which measures the degree of weathering of the examined materials relative to unaltered feldspars:

$$[Al_2O_3/(Al_2O_3+CaO^*+Na_2O+K_2O)]\times100$$

(in molar proportion), where CaO* represents the CaO content of the silicate fraction. CaO* values were calculated according to the recommendations of Fedo (1995). Except for the samples mixed with limestone debris (marked in Table 4), the red clays contain less than 1 % calcite (or 2 % dolomite, respectably).

The weathering tendencies and the connection between the major elements and the mineral fraction can be illustrated on A-CN-K triangular plot (see Fig 9. after Nesbitt and Young 1984, 1989, Varga 2005). It can be seen on the diagram that all red clay samples are dispersed in a relatively small field, the CIA indices (Table 4) vary between 77.6 and 87.5 which together indicate similar origin and strong weathering. Red clay samples are located between the smectite and illite zone on the diagram which is in agreement with the mineral composition presented before. The karst and near karst localities (Va-1, Va-2, Va-7) turn out to be weathered a little stronger than the Miocene ones which is in conformity with the mineralogical results obtained by the XRD method. The red clays of the SZPKF-8 drilling are practically identical with the Sarmatian near shore silts (marked by grey signs on the triangular plot). Their similarity refers to a narrow genetic connection and to the fact that the red clays arose by the light alteration of silts partly originated from tuffs during previous marine alteration. Compared to the Sarmatian bentonitic rhyolite tuffs the plots of Sarmatian silts and red clay samples are shifted approximately parallel with the A-CN axis according to the ideal weathering trends. The little shift toward the illite zone is due to the fact that detrital sediments of different origin also took part in their formation.

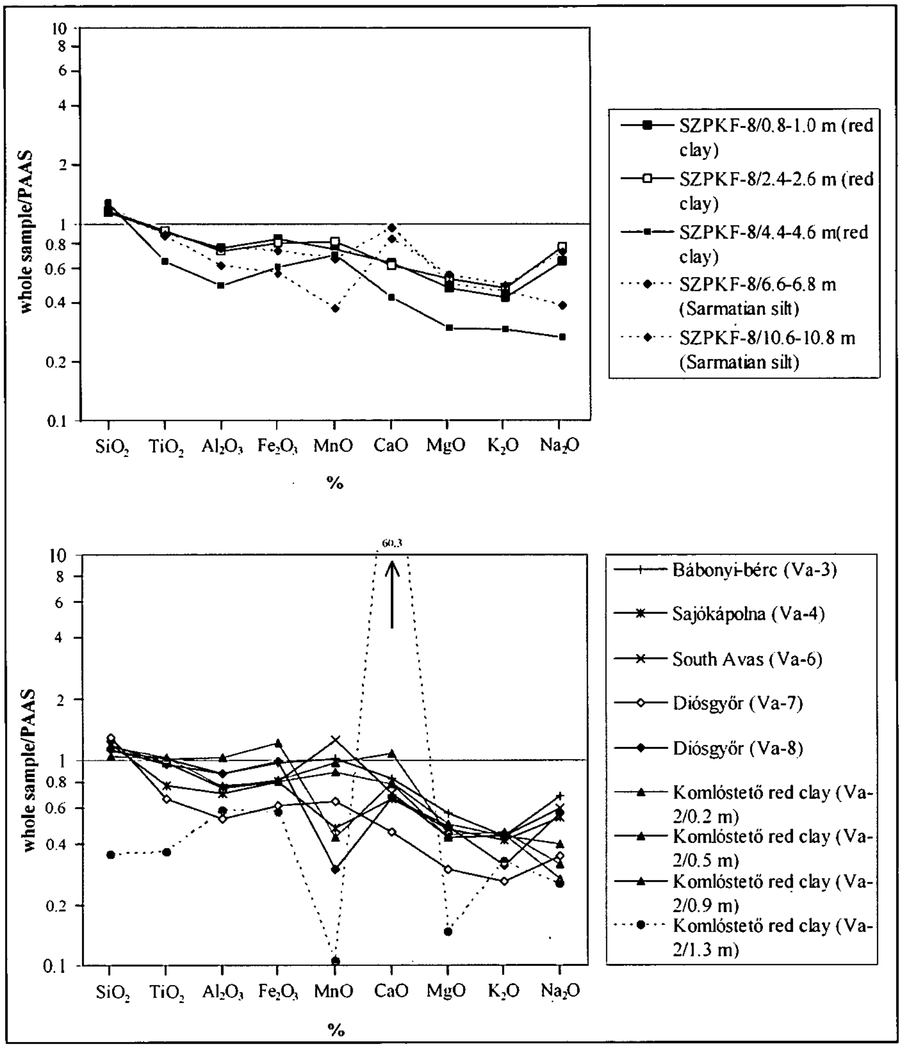


Fig. 8. PAAS-normalized major element spiderdiagrams of red clays (PAAS after Taylor and McLannon 1985).

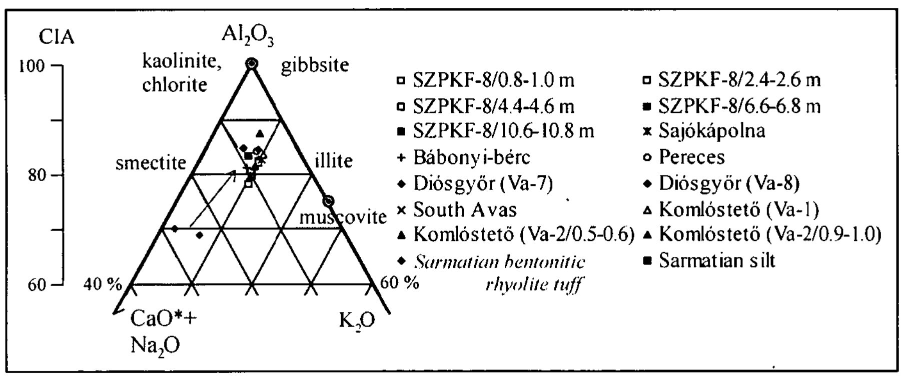


Fig. 9. A-CN-K triangular plot of red clays and source materials (Nesbitt and Young 1982, 1984). Arrow indicates weathering trend.

According to Schlesinger (1991) the $SiO_2/(Al_2O_3+Fe_2O_3)$ quotient approximately shows the climatic conditions of weathering. The limiting values are the following: <1.61: tropical climate, 1.61-3.15: subtropical climate, 3.15-3.77: warm moderate and >3.77: moderate climate. Most of the materials indicate warm moderate climate, two samples from the Bükk Mts. (Va-2/0.9-1.0 m and Felsőtárkány) indicate subtropical while the yellow clay indicates tropical climate. The significant variability of the quotient can be explained partly by the fact that SiO_2 content is highly affected by the variable quartz contents derived from the (later) mixing with detrital components.

Schweitzer (1993) and Schweitzer and Szöőr (1997) differentiated two types of clays, the „typical red clay" and the „reddish clay" (usually brownish red, reddish brown coloured), based on their appearance, mineral and geochemical composition. According to their opinion the age of the former is lower Pliocene, and of the latter late Pliocene to middle Pleistocene. Examining some of the major elements they have found that it is possible to separate the red clay types on the basis of their Fe_2O_3 - Na_2O and Fe_2O_3 - K_2O relations, respectively. Most of our samples displayed on the Fe_2O_3 - Na_2O diagram (Fig.10) are classified as „reddish clay", however, they are closer to the border line on the Fe_2O_3 - K_2O diagram showing an unclear result. The difference can be explained by the different behaviour of Na and K during weathering. While Na is being leached, K may accumulate in clay minerals. Consequently our samples rather belong to the „reddish clays", or are transitional between the two types. The results show, however, the restricted value of this method for the discrimination.

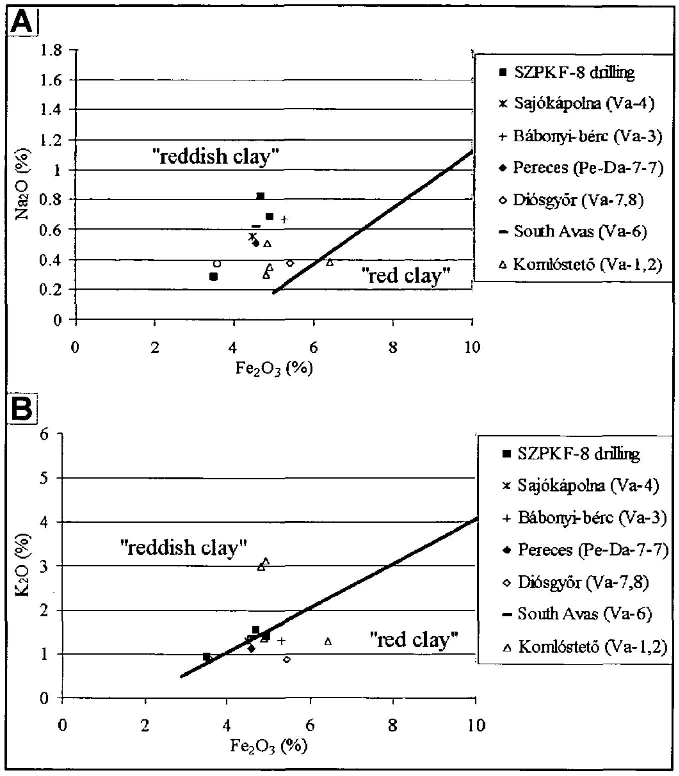


Fig. 10. Distinguishing "red clays" and "reddish clays" based on alkali oxides as a function of Fe-oxides, according to the discrimination diagrams of Szöőr (1993).

CONCLUSIONS

The comparative characterisation of a less well-known red clay formation by sedimentological, mineralogical and geochemical examinations is given in the paper. Based on the results obtained the following conclusions are drawn:

- The examined red and reddish brown clays collected from 10 localities in the surroundings of Miskolc, are basically similar and they are paraautochtonous formations. The difference between red clays underlain by karstic Mesozoic limestone and Sarmatian sediments is moderate, although the former one is slightly finer and contains more kaolinite, especially in the fine fraction. It marks more advanced weathering.
- Sedimentological, mineralogical and geochemical similarities between the underlying Miocene sediments and the red clays indicate that the source material of the examined materials is probably the Miocene shallow marine, bentonite containing sequence. The mixing with older kaolinite rich weathering crusts occurs only on the karstified limestone surface of the Bükk Mountains. Based on the XRD mineralogical results the possibility of the derivation of red clays from the weathering crust of Jurassic iron containing rocks and their terra rossa origin are excluded. Aeolian origin is not excluded but it can be subordinate considering the grain size distribution and mineral contents of the red clays.
- The examined red clays belong to the slightly weathered type ascompared to other Hungarian localities. The considerable similarity with underlying sediments refer to moderate alteration during warm, semi-arid periods in the course of which alkali and alkali-earth metals were leached, iron was oxidized, less stable silicates weathered, montmorillonite and illite became more disordered, but the basic clay mineral composition hardly changed. This is the process called by Kubiena rubefication while the more intense tropical weathering would be called lateritisation (cited by Fekete 2002).
- Based on lithostratigraphical analogies the age of the clays may be late Pliocene, early and middle Pleistocene, but mixing with older red clay formations is not excluded. According to the subdivision used by Schweitzer and Szöőr (1997) the majority of the examined sediments are „reddish clays".
- Taking lithostratigraphic features into account, the examined red clays can be classified as members of the Pliocene-Pleistocene Tengelic Red Clay Formation or of the Pliocene Kerecsend Red Clay Formation, however, because of the similarities between them it is difficult to make a final decision even in the case of these mapped and mineralogically, geochemically well defined red clays.

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