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The origin of the raw material of basalt polished stone tools in Hungary

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Abstract: Basalt is a very common rock type among the Hungarian archaeological polished stone tools. The popularity of this raw material is due to, on the one hand, the good mechanical qualities and, on the other hand, the common occurrence of the raw material in the Carpathian Basin. The appearance, size and finish of these stone tools can be very different. The main aim of our study was to determine and characterise the different types of polished stone tools and localise the source region of the different types of basalt in Hungary. Macroscopical, petrographic microscopical and geochemical studies were made on archaeological as well as geological samples. Geochemical studies were made by Prompt Gamma Activation Analysis (PGAA) which is a relatively new, sensitive non-destructive analytical method; therefore it may be useful on archaeological finds. Apart from possibly determining the source of basalt raw material, the application of this relatively new technique on basalt was one of the main objectives of the current studies.
Basalt samples from the outcrops and polished stone tools macroscopically identified as basalt were compared. We could separate three groups according to the PGAA and the macroscopical and microscopical investigation. First and second(?) type is similar to the basalt from Mecsek; the third type is similar to the late Miocene to Quaternary alkaline basalt type.

Key words: polished stone tools; basalt; PGAA method; raw materials

Introduction

Basalt is a very popular raw material of polished stone tools in the Neolithic-Late Bronze age period in Hungary. It is a massive, hard and resistant rock type occurring fairly common in the Carpathian Basin. In the framework of the current study, 14 basalt polished stone tools and basalt rock types from different geological sources from Hungary were investigated. The archaeological age of the finds is ranging from the Neolithic to the Early Iron Age. 13 samples from basaltic occurrences were collected for study and we compared the material of the archaeological implements with the samples from the outcrops. Our aim was to identify the source of the raw materials with traditional petrographic methods and a relatively new, non-destructive method geochemical, prompt gamma activation analysis (PGAA).

Basalt polished stone tools from the Carpathian-Pannonian area

On the NW-side of Hungary, the Little Hungarian Plain and the Balaton Highlands, there are important source areas of basalt raw materials. Therefore in this part of Hungary in the archaeological collections it is very often used as raw material for polished stone tools (Nikl 1998, Schléder, Biró 1999, Szakmány et al. 2001). Schléder and Biró studied polished stone tools from Baranya County, from 18 finds had 5 basaltic raw material, which were grouped to 3 part. One of the all groups was measured by neutron activation analysis (NAA). According to NAA he supposed the source of the basalt is the Hungarian Little Plain, but didn't preclude the possibility of the origin from Balaton Highlands, Nógrád-Gömör Unit or Bánát region. Nikl (1998), in her thesis studied 7 basalt stone tools from Tolna County and she found similar origin than Schléder, only 1 is from the Mecsek Mts. The Mihaldy collection, collected basically from Northern Transdanubia in the 19th century, comprised 378 polished stone tools; from these, more than 30 % have basaltic raw material. The exact origin of the individual pieces in this collection is unknown, but we know, that the bulk of the pieces were collected in the Bakony Mountains and its environs (Horváth, T. 2001). Most of the material was studied macroscopically but some thin section were prepared already for our former studies (Szakmány et al. 2001). According to the big size of the tools, the large quantity of pieces, the texture and mineral composition, these are possibly from local raw material sources (Balaton Highlands and/or Little Hungarian Plain). In the basalt polished stone tools from Bicske-Galagonyás, clinopyroxene with aegirine-augite core and Ti-augite rim was observed (Szakmány 1996), which is characteristic for the basalt of Nógrád-Gömör unit (Dobosi 1989, Dobosi, Fodor 1992).

On the East Side of the country, Biró studied a Late Neolithic axe workshop from Aszód, Papi földek (Biró 1992) and Judik et al. (2001) defined some rock types from this site exactly. In this collection the most frequent raw material type is basalt and dark glassy andesite. Basalt samples containing phlogopite could be related in respect of origin to Medves-Karancs (Nógrád-Gömör

Unit) by the microscopical observation. At the Szarvas-Endrőd archaeological site, only one basaltic stone tool was identified, and the source of the raw material is unknown yet (Starnini et al. 1998).

In Slovakia, basaltic raw material is relatively rare among the polished stone tools, probably due to frequency of competing greenschist. It is rather connected to some cultures, 1 or 2 pieces in an assemblage, for instance in Bajč (Méres et al. 2001). The basalt stone tools belong mostly to the group of alkali basalt, which were supposed to originate from Central Slovakian young volcanic area (Illášová 2001).

The origin of the studied polished stone tools

In this study, basalt stone tools from several Hungarian archaeological collections were selected from the different parts of the country. Some of them were treated in other studies as well using other methods, mainly traditional petrographic investigation. One part of the examined samples originated from the Mihäldy collection of the Laczkó Dezső Museum (Veszprém). We examined 2 pieces from Wosinszky Museum (Tolna County) and 1 piece from private collection from Mórágy (Tolna County), which were treated by other analytical methods by Nikl in her MSc thesis (Nikl 1998). We got some samples for non-destructive measurements from the Hungarian National Museum. 2 polished stone tools originated from the site Tápé-Lebő, which were studied by Hargita Oravecz and Sándor Józsa and 2 samples from Late Neolithic settlements around Szentgál (Bakony Mts.) (Fig. 1., Table 1.).

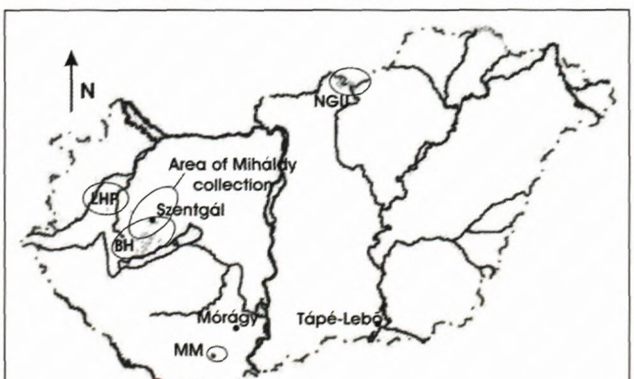

Fig. 1. The localities of studied samples and main basalt occurrences in Hungary: Balaton Highlands (BH), Little Hungarian Plain (LHP) Nógrád-Gömör Unit (NGU), Mecsek Mts (MM).

Basalt occurrences in Hungary and in the Carpathian-Pannonian Region

In Hungary there are 4 main areas, where significant basalt occurrences suitable for tool-making can be found: notably, in the Mecsek Mts (MM)., the Balaton Highlands (BH), the Little Hungarian Plain (LHP) and in the Nógrád-Gömör Unit (NGU) (see Fig. 1.).

The most typical basalt formation period comprises Late Miocene to Quaternary alkaline volcanism in Hungary apparent in several isolated volcanic fields in the Carpathian-Pannonian Region. The outcrops of these volcanic fields can be found extending from Burgenland in Austria, trough the Little Hungarian Plain, the Balaton Highlands and the Nógrád-Gömör Unit to the Persany Mts. This volcanic activity produced almost exclusively basaltic rocks. The alkaline mafic volcanism began at the western (Burgenland) and central Pannonian Basin in the Late Miocene (10-12 Ma), culminating at 3-5 Ma at each volcanic field. The last eruptions occurred only a few hundreds of thousands years ago at Brehy (Sk) and in the Persany Mts. (Balogh et al.1986, Harangi 2001). The general mineral composition of these basalt types is olivine, clinopyroxene as phenocryst, in the groundmass with plagioclase, clinopyroxene, olivine, ilmenite, Ti-magnetite and apatite as accessories. 60 eruption centres are known in the Balaton Highlands. The area of the basalt outcrops in Nógrád-Gömör Unit is smaller, but the volume is similar to the former. The differences in the mineral composition are the occurrence of phlogopite, nepheline and the green-core in the clinopyroxene, which is caused by high pressure (Embey-Isztin et al. 1998).

In the Mecsek Mountains, another type of basalt is occurring in Hungary, originating from the Early Cretaceous period. The Cretaceous volcanic activity produced extensional-related magmatism along the southern margin of the European continental plate. K/Ar radiometric age data indicate that the paroxysm of the volcanic activity occurred between 135 and 100 Ma. Two volcanic series have been recognised in the Mecsek Mts.: the more important series for our studies is the ankaramite-alkali basalt-trachybasalt effusive series, the other one is a Na-basanite-phonotephrite-tephriphonolite-phonolite subvolcanic suite (Harangi et al. 1993). The magma was flown partly below the seawater, it is proved by pillow lava structure and lava breccias, other part of the lava became solid than subvolcanic body or dyke (Viczián 1966, Harangi et al. 1993). The large thickness of the basaltic lava flows (somewhere over 1000 m) reflects the intensity of the volcanic activity (Bilik 1974). The load structure basalt connects to the Na-rich series. The phenocrystals of the alkali basalt are about 5-15-volume %. The quantity of basic plagioclase is more than that of clinopyroxene. The quantity of olivine is few or it is missing. The groundmass is composed of neutral plagioclase, clinopyroxene and Fe-Ti-oxides, mostly ilmenite (Harangi et al. 1993). We can also find these volcanic rocks in the Upper Cretaceous conglomerate in the Mecsek Mts. The gravel size is up to 10-15 cm, which could be good for stone implement preparation.

Analytical methods

Petrographical investigation

We made groups on base of the macroscopical features and we selected some of them for further analyses.

Microscopical thin sections were made of each group and thin sections from previous studies were also studied. All of the samples, which were studied microscopically were measured by PGAA.

Prompt gamma activation analysis

Chemical composition was investigated by prompt gamma activation analysis (PGAA), carried out at the Budapest Research Reactor, Hungary. This measurement technique is based on the detection of prompt gamma rays originating from neutron radiative capture or (n, ?) reaction. The output of the reactor is 10 MW. The emitted gamma rays (prompt gamma rays) are recorded simultaneously in a spectrum. The detection of the gamma rays is made by HPGe-BGO detect system. The spectrums are evaluated by Hypermet-PC program. Elements are detected according to their energies and intensities (Molnár et al. 1997). The intensity is proportional to the concentration of the element and the energies are characteristic to the elements. The method is suitable for determination of all the elements, however with different sensitivities. PGAA measurements give reliable data for main components of basalt (SiO_2, TiO_2, Al_2O_3, Fe_2O_3, MnO, MgO, CaO, Na_2O, and K_2O) and some trace elements (B, Sc, V, Cr, Sm, Eu, Gd and Dy) moreover we can detect Cl too. Since the neutron beam come through on the material of samples, we can get average chemical composition from the full thickness, about from 1×1 cm or 2×2 cm area. The time of measurements depends on the size of the samples, the long of our measurements are about 1000 sec-24000 sec. The advantages of method are absolutely non-destructive and the measurement not required preparations, therefore same identical sample can be used for other chemical analysis later. This method was applied on greenschist and blueschist polished stone tools, and gave good results on the different type of greenschist, which are from several places in the Carpathian-Pannonian region (Szakmány-Kasztovszky 2001).

Results of petrographical analysis

Macroscopically, the archaeological basalt samples have grey or dark-grey, black colour, the cut surface is usually darker, almost black. They are massive fine-grained, homogenous rocks. The surface is sometimes heavily altered; therefore it has a lot of tiny holes because of the dissolution of olivines and pyroxenes. We can see small black (pyroxene) and dark green (olivine) phenocrystals in the grey matrix. In general, the smaller finds have more elaborate surface than the larger pieces.

Mineral composition of alkali basalt is characterised by plagioclases and clinopyroxenes, olivine, amphibole, and ore minerals are also present. We can distinguish three groups of basalt on the basis of microscopical features. The first group has fluidal texture (Fig. 2.). The phenocrysts are clinopyroxenes and sometimes a few olivines, too (Fig. 3). The second group is similar to the first group, but it has smaller pyroxene phenocrystals. The third group includes a lot of olivine phenocrysts, but their size and quantity are smaller than clinopyroxenes (Fig 4.). In the groundmass, the plagioclase forms the skeleton in all three cases, and there are small-size opaque minerals, clinopyroxenes, apatites, chlorites and variable quantity of glass among the plagioclase laths.

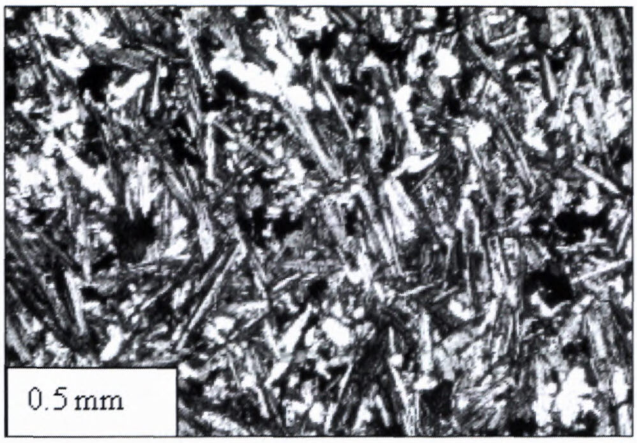

Fig 2. Fluidal texture of the basalt polished stone tools. Characteristic to the first and second group of basaltic polished stone tools.

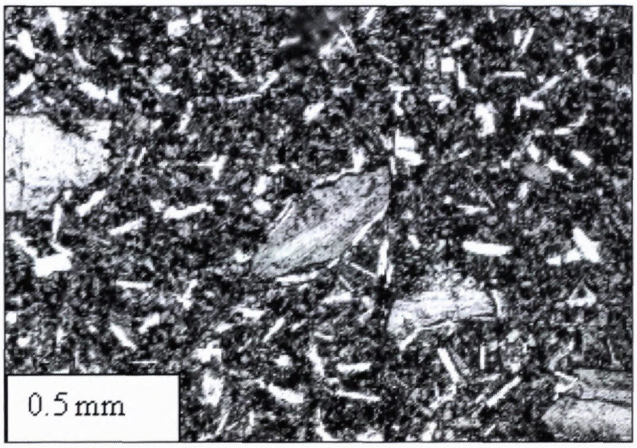

Fig. 3. Clinopyroxene phenocrysts in fine grained matrix of first group of basaltic polished stone tools.

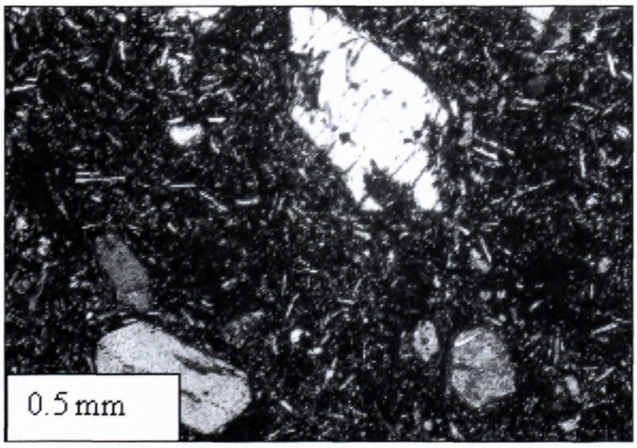

Fig. 4. Large olivine and small sized clinopyroxene phenocrysts in fine grained and glassy matrix of the third group of basaltic polished stone tools.

We studied some samples from geological outcrops from Balaton Highlands, Little Hungarian Plain, Nógrád-Gömör Unit and Mecsek Mts. The average mineral composition is plagioclases, clinopyroxenes, olivine, amphibole, ore minerals, apatite and glass. The basalt from Nógrád-Gömör Unit differs from the other young basalts in the green-core of the clinopyroxene and it has nepheline and phlogopite. The samples from Mecsek have fluidal texture and the quantity of olivine is low or it is missing.

Result of chemical analysis

The chemical composition of polished stone tools and the samples from outcrops were measured by PGAA, altogether on 27 samples (Table 2.). We made different diagrams from the results of PGAA. As a result of the measurements we got two groups of data by some types of diagrams. The two groups are separated by the TiO_2-Fe_2O_3, Sm-Gd, TiO_2/Al_2O_3-Na_2O/K_2O, TiO_2/Al_2O_3-Sm diagrams (fig. 5, 6, 7, 8). From the 14 archaeological samples 2 belong to different group by trace elements (fig. 6., Sm-Gd) and by major elements (fig. 5, 7).

Discussion

We could formulate three groups among the measured rock samples on bases of the different investigations.

Macroscopical investigation did not show a significant difference between the samples. Moreover, the archaeological implements are typically heavily altered on the surface, because of being buried for a long time. The differences in texture and minaral composition observed in the microscopical studies did not show any connection with the previous macroscopical grouping. Seemingly meaningful groups could be made on the basis of microscopical observation and PGAA.

Comparing the microscopical observation with PGAA results, we can define three groups among the basalt polished stone tools and samples from geological occurrences. Two groups were quite clear and they were observed both in the geological and archaeological sample set. Group 1 correspond to Mecsek, group 3 correspond to young volcanites from the Carpathian-Pannonian unit. Group 2 was only present in the archaeological set and from a site in SE Hungary: its unique features can be explained several ways

1. they belong to a so far not investigated source (Transylvania(?), Southern territories(?))
2. they belong to a variety of the groups established, i.e. Mecsek, the textural features of which are very similar to this group.

The first and second group have similar microscopical features, but in the second group there are no phenocrysts. Maybe this is the reason of the difference observed in the rare earth elements results of PGAA. The second group has two pieces from the archaeological finds, which are similar to the samples from Mecsek in major elements, but different in rare earth elements on bases of PGAA. The first group is comparable to samples from Mecsek in chemical composition and in microscopical features, too, therefore we think this group is from the Mecsek. The third group is similar to the young basalt types in results of PGAA, but we could not make further dissection within group. The mineralogical distintive features of Nógrád-Gömör basalts were not observable in our archaeological sample set. Further chemical differentiation within the young volcanites may need further studies, for example the mineral chemistry of the rock-forming minerals by electron-microprobe analysis. Though chemical analysis did not differentiate regions within the young volcanic range, we suppose that the "third group" is from the Balaton Highlands or Little Hungarian Plain, because the archaeological sites investigated are nearest to former raw material sources.

Among the basalt tools, the young basalt is typical on the North part of Transdanubia and North-Eastern Hungary. Lower Cretaceous basalt is typically occurring in

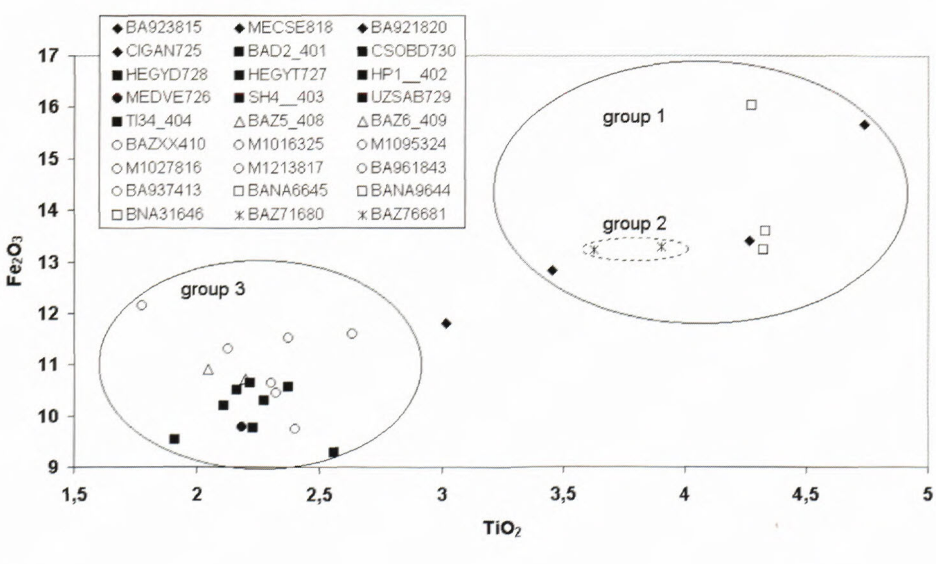

Fig. 5.
Fe_2O_3 versus TiO_2 diagram of analysed basaltic polished stone tools (open and X signs) and samples from outcrops (fill signs). The origin and legend of the individual sample signs see the table 1.

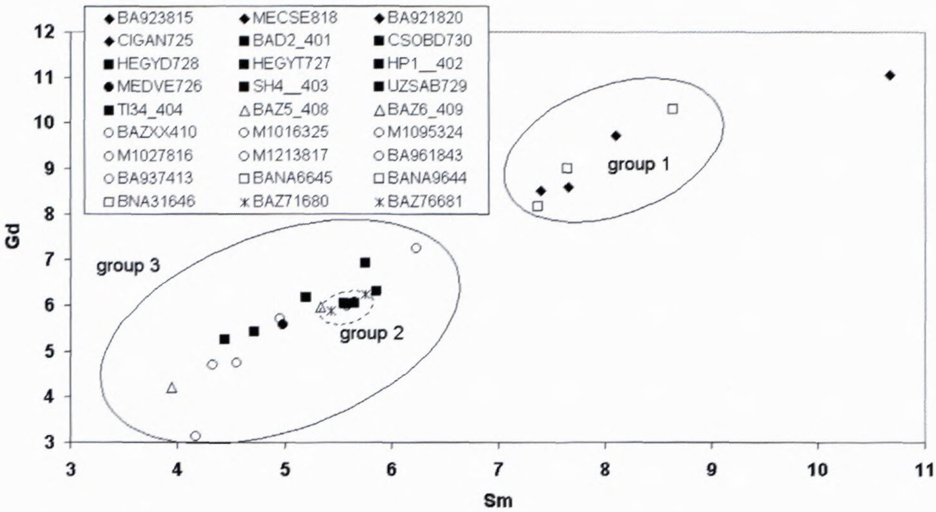

Fig. 6.
Gd versus Sm diagram of analysed basaltic polished stone tools (open and X signs) and samples from outcrops (fill signs). The origin and legend of the individual sample signs see the table 1.

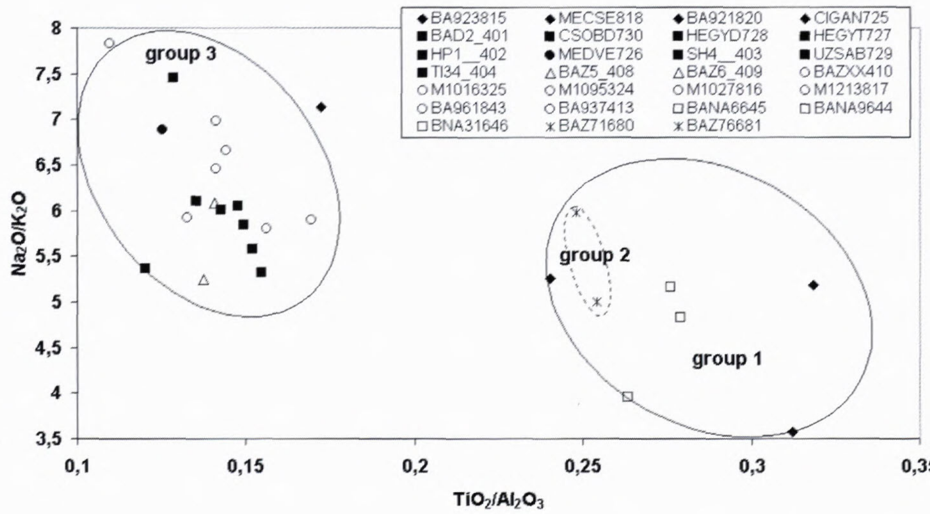

Fig. 7.
Na_2O/K_2O versus TiO_2/Al_2O_3 diagram of analysed basaltic polished stone tools (open and X signs) and samples from outcrops (fill signs). The origin and legend of the individual sample signs see the table 1.

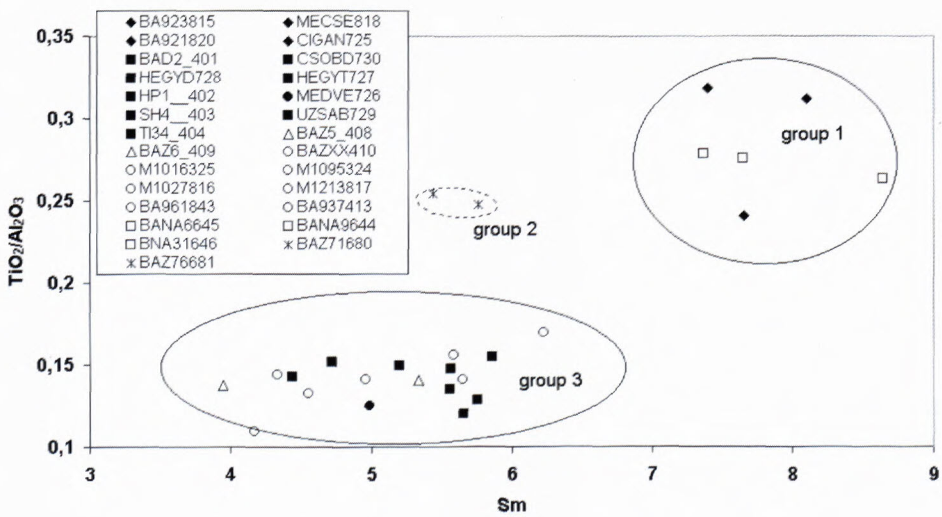

Fig. 8.
TiO_2/Al_2O_3 versus Sm diagram of analysed basaltic polished stone tools (open and X signs) and samples from outcrops (fill signs). The origin and legend of the individual sample signs see the table 1.

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the Southern part of Transdanubia. Here we can find two basalt raw material types (groups 1 and 2, respectively).

Conclusion

1. 27 samples were investigated (14 from polished stone tools, 13 from outcrops). The samples are from different geological/archaeological age and locality.

2. We get three groups by geochemistry and petrography. Two of them are clearly distinct and correspond to, on one hand, to Mecsek Cretaceous basalt (Group 1), on the other hand, to young alkaline basalt (Group 3). Group 2 shares more features of the former, its interpretation, however, needs further studies.

3. The distance of transport between the archaeological sites and the raw material sources was relatively short, except in the Great Hungarian Plain, where raw material from all basalt raw materials sources could be observed.

4. Within the groups established, no further (regional) differences could be observed by the methodology applied. This can be due to the homogeneity of the specific quality of basalt used for tool making or the homogeneity of the chemical composition of the Late Cenozoic volcanic event as well.

Acknowledgements

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Table 1. The signs, localities and origin of the studied samples.

Sign of samples	Origin	Locality
BA921820	outcrop	Mecsek
BA923815	outcrop	Mecsek
BA937413	55.937	Mihåldy-collection
BA961843	55.961	Mihåldy-collection
BAD2_401	outcrop	Badacsony
BANA6645	Nikl A 6.s.	Tolna County
BANA9644	Nikl A 9.s.	Tolna County
BAZ5_408	Szentgál 2/2	Hungarian National Museum (Szentgál)
BAZ6_409	Szentgál 2/1	Hungarian National Museum (Szentgál)
BAZ71680	7.1951.363	Hungarian National Museum (Tápé-Lebő)
BAZ76681	7.1951.276	Hungarian National Museum (Tápé-Lebő)
BAZXX410	55.1xx1	Mihåldy-collection
BNA31646	Nikl A. 31.s.	Tolna County
CIGAN725	outcrop	Cigányhegy (Mecsek)
CSOBD730	outcrop	Csobánc
HEGYD728	outcrop	Hegyesd
HEGYT727	outcrop	Hegyestű
HP1__402	outcrop	Haláp
M1016325	55.1016	Mihåldy-collection
M1027816	55.1027	Mihåldy-collection
M1095324	55.1095	Mihåldy-collection
M1213817	55.1213	Mihåldy-collection
MECSE818	outcrop	Mecsek
MEDVE726	outcrop	Medves (Nógrád)
SH4__403	outcrop	Ság-hegy
Ti34_404	outcrop	Tihany
UZSAB729	outcrop	Uzsabánya

wt %	BA921820	BA923815	BA937413	BA961843	BAD2_401	BANA6645	BANA9644	BAZ5_408	BAZ6_409	BAZ71680	BAZ76681	BAZXX410	BNA31646	CIGAN725
SiO_2	42,20	44,75	45,39	44,74	45,08	46,65	45,31	47,60	43,43	46,08	47,37	44,57	41,96	43,54
TiO_2	4,74	4,27	2,13	2,63	2,22	4,33	4,32	2,05	2,20	3,63	3,90	2,32	4,27	3,45
Al_2O_3	15,20	13,40	16,04	15,54	17,24	15,54	15,67	14,90	15,63	14,63	15,37	16,46	16,24	14,37
Fe_2O_3	15,67	13,39	11,30	11,60	10,64	13,58	13,23	10,92	10,73	13,22	13,27	10,45	16,02	12,82
MnO	0,25	0,41	0,14	0,22	0,20	0,20	0,23	0,18	0,18	0,19	0,13	0,19	0,15	0,19
MgO	4,26	6,34	8,43	7,85	7,33	4,45	4,81	9,15	10,64	7,11	5,04	8,45	4,69	6,45
CaO	9,84	9,15	9,56	10,45	8,75	7,87	8,63	8,86	10,01	8,14	7,93	10,00	9,86	10,79
Na_2O	2,99	3,15	3,04	3,46	4,79	3,26	3,39	3,44	3,40	3,62	3,14	3,77	2,29	3,32
K_2O	0,58	2,04	2,88	2,43	2,66	1,57	1,77	1,80	2,68	2,35	1,86	2,68	1,67	1,94
H_2O	4,13	3,02	0,90	0,84	0,81	2,40	2,57	0,87	0,96	0,87	1,85	0,88	2,77	2,90
ppm														
B	11,3	7,6	5,7	10,8	6,1	4,5	7,1	4,7	4,3	11,1	6,1	6,1	3,7	6,9
Cl	70	35	651	960	1042	88	49	553	581	178	94	749	101	707
Sc	29,2	25,0	1,0	34,4	30,8	24,6	22,1	39,2	25,3	39,5	1,0	36,5	26,9	24,0
V	<100	<100	<100	<100	234	316	330	340	<100	419	355	332	362	374
Cr	<200	<200	656	<200	561	524	<200	715	562	424	381	672	<200	620
Sm	8,1	7,4	4,5	6,2	5,8	7,4	7,6	3,9	5,3	5,8	5,4	5,6	8,6	7,7
Eu	3,0	2,3	3,6	5,2	3,3	0,1	4,5	3,4	0,1	0,5	0,7	3,2	1,5	4,1
Gd	9,7	8,5	4,7	7,2	6,9	8,2	9,0	4,2	6,0	6,2	5,9	6,1	10,3	8,6
Dy	<1	<1	<1	<1	<1	<1	<1	12,0	<1	<1	<1	<1	<1	<1

wt %	CSOBD730	HEGYD728	HEGYT727	HP1__402	M1016325	M1027816	M1095324	M1113326	M1213817	MECSE818	MEDVE726	SH4__403	TI34_404	UZSAB729
SiO_2	46,65	46,41	44,15	48,83	48,02	44,64	43,61	50,55	46,88	44,69	44,80	48,85	41,19	45,94
TiO_2	2,11	2,56	2,37	2,17	1,78	2,40	2,37	0,94	2,30	3,02	2,19	2,23	2,27	1,91
Al_2O_3	15,61	16,55	16,07	15,18	16,19	15,36	16,46	19,27	16,31	17,49	17,45	14,66	15,23	15,87
Fe_2O_3	10,20	9,28	10,57	10,52	12,14	9,75	11,52	8,11	10,64	11,80	9,79	9,76	10,30	9,53
MnO	0,17	0,15	0,20	0,19	0,24	0,19	0,20	0,11	0,19	0,20	0,20	0,18	0,24	0,19
MgO	8,46	6,47	8,59	7,11	5,84	11,08	8,37	6,90	6,68	3,92	7,06	9,11	5,54	9,61
CaO	8,16	9,32	10,20	7,94	7,33	9,76	9,72	7,80	8,63	6,42	9,16	8,63	17,30	7,77
Na_2O	3,69	2,88	3,42	3,89	5,05	3,42	3,94	2,85	4,62	5,50	4,62	3,61	3,50	3,49
K_2O	2,41	2,43	2,64	2,12	2,78	2,38	2,71	1,06	2,36	1,63	2,26	1,97	2,34	1,87
H_2O	2,32	3,72	1,59	1,87	0,51	0,88	0,89	2,27	1,12	5,27	2,23	0,76	1,85	3,67
ppm														
B	5,0	2,6	5,1	3,5	1,1	6,2	2,7	8,1	5,5	7,2	2,9	10,0	3,0	5,5
Cl	778	822	378	89	1130	659	742	325	954	123	856	482	792	153
Sc	26,6	32,5	25,4	27,1	0,0	28,4	21,0	30,9	20,5	11,3	27,6	35,5	31,1	26,1
V	216	294	350	303	<100	<100	260	<100	<100	<100	283	291	372	222
Cr	601	498	655	762	<200	<200	569	602	<200	<200	657	882	632	643
Sm	5,6	5,9	5,6	4,4	4,2	5,6	4,3	1,5	5,0	10,7	5,0	4,7	5,2	5,7
Eu	1,6	2,2	1,6	9,2	0,0	1,3	0,0	2,9	1,5	4,5	2,4	0,1	6,5	7,4
Gd	6,0	6,3	6,0	5,2	3,1	6,0	4,7	1,8	5,7	11,1	5,6	5,4	6,2	6,0
Dy	<1	<1	<1	<1	<1	<1	<1	<1	<1	<1	9,2	<1	<1	<1

Table 2. Major and trace elements results of studied samples.

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